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¹ Colin, M., and Herold, A., *CR Acad. Sci. Paris, Serie C*, **269**, 1302 (1969).² Murray, J. J., and Ubbelohde, A. R., *Proc. Roy. Soc., A*, **312**, 371 (1969).³ Hayward, D. O., and Trapnell, B. M. W., *Chemisorption*, p. 3 (Butterworths, London, 1964).

Dependence of Ice Melting Electrification on Earlier Freezing Rate

MANY explanations have been offered for the electrification of clouds and precipitation involving effects such as the change of state of water between phases, selective ion capture, a thermo-electric effect in ice, the rupture of water films, induction charging when polarized particles collide, concentration cells and ion diffusion. These processes are probably not all independent, but often only one may have a dominant role¹. Dinger and Gunn² have reported that melting ice containing air acquires a positive charge when ventilated by an air stream which in turn carries away negative charge released as the trapped bubbles burst. Chalmers¹ suggests that melting is often particularly important and may account for the positive charge sometimes detected near the base of thunderclouds and also for that usually found on continuous rain. Drake³ observed that when small ice particles melt in a wind tunnel the separation of electric charge is highly dependent on convection currents which develop in the meltwater and rapidly transfer bubbles to the surface where they burst. He showed that the onset of strong convection and the most vigorous electrification occurred only when the heat flow rate to the melting particle was above a certain level. According to Iribarne and Mason⁴ this charge is produced by the rupture of the electrical layer at the air/water interface. Our experimental results show a new aspect of this melting electrification.

Ice particles of diameter approximately 3 mm were made from drops of deionized water at conductivity $10^{-4} \text{ ohm}^{-1} \text{ m}^{-1}$. Some (group A) were frozen in still air, others (group B) in an airstream flowing upwards at 11 m s^{-1} ; in both cases the temperature ranged between 0 and -15°C but during the freezing of any one drop it was held constant. Next, each frozen particle in turn was supported on platinum wire of diameter $130 \mu\text{m}$ and the charge was measured as it melted in a special Faraday cage forming part of a small vertical wind tunnel through which air flowed upwards at 7 m s^{-1} , close to the terminal fall speed of the particle. Conditions in the wind tunnel circuit could not be controlled easily, and the inevitably low relative humidity demanded a comparatively high air temperature to yield the minimum heat flow rate for melting. The air temperatures required lay between 11°C and 15°C , and these were chosen so that the heat flow into the particle during melting remained constant. Fig. 1 shows the electrification, in terms of the positive charge appearing on the particle per unit mass melted, plotted against earlier freezing environment temperature. The scatter of the points is not surprising since both charge produced and mass melted are subject to errors of up to 25%.

The curves in Fig. 1 are quite distinct. The upper graph results (group B) confirm those of Drake³ that melting electrification of such particles, frozen in the airstream, will increase steadily as the earlier freezing environment temperature falls. It is now clear from the lower curve (group A) that we have a similar dependence when the drops are frozen in still air. The melting charges, however, are less; group A values are only one sixth those of group B. These combined results suggest that it is the rate of freezing during its formation which mainly controls the charge the ventilated ice particle will acquire on melting.

Fig. 1 Positive charge appearing on the particle for unit mass of ice melted and earlier freezing environment temperature. During melting the rate of heat flow into the particle was held constant. $\times$ (upper curve), Drops frozen in air flowing at 11 m s^{-1} (group B); $\circ$ (lower curve), drops frozen in still air (group A).

Here, as in earlier work, air bubbles in the ice appear to be associated with the melting charges. The ice particles were found to have an opaque core, with many minute bubbles and a few large bubbles exceeding $100 \mu\text{m}$ in diameter. For drops frozen in still air (group A) the core became larger and the number of large bubbles fewer as the freezing environment temperature was reduced. A corresponding trend was observed with drops frozen in the airstream (group B). Moreover for any freezing temperature the core was appreciably larger and the large bubbles were fewer for those frozen in the airstream. These findings agree with the results of related work by Carte⁵ and the association of smaller bubble diameter with larger charge for water of this degree of purity is in general agreement with the theory of Iribarne and Mason⁴.

The water used in these experiments corresponds to rain water in which the main impurity is absorbed carbon dioxide. The CO_2 level in the laboratory was not abnormally high, nor was solid CO_2 used. When, by the addition of NaCl, the conductivity was raised by a factor of 10, to correspond to that of natural cloud water, the group A electrification was only halved. A similar result for the group B electrification may be inferred from the work of Drake³. We would therefore expect these results to apply to the freezing and subsequent melting of precipitation water in the atmosphere. In strongly convective conditions the magnitudes of specific charge shown in Fig. 1 are likely to be reached. At other times the rates of freezing and subsequent melting will be too low for these effects to control the charging of precipitation, and some other mechanism will be dominant. Sometimes, however, successive layers of an ice particle will have been formed at different freezing rates. When they melt they will contribute to the particle's charge to different extents, at times even reversing the sign. The perplexing alternation of signs of charge observed on precipitation particles might be thus explained.

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¹ Chalmers, J. A., *Atmospheric Electricity*, second ed. (Pergamon Press, London, 1967).² Dinger, J. E., and Gunn, R., *Terr. Magn. Atmos. Elect.*, **51**, 477 (1946).³ Drake, J. C., *Quart. J. Roy. Met. Soc.*, **94**, 176 (1968).⁴ Iribarne, J. V., and Mason, B. J., *Trans. Farad. Soc.*, **163**, 2234 (1967).⁵ Carte, A. E., *Proc. Phys. Soc.*, **77**, 753 (1961).