

Electrification Experiments with AgI in the System: Water Vapor, Liquid Water and Ice¹

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ABSTRACT

The charge on an AgI particle, freely suspended in an air current of terminal velocity, has been measured at various temperatures, humidities and cooling rates. The wall of the flow pipe serves as a source or sink for water vapor, simulating neighbor droplets and ice particles in a cloud. The data show that the AgI particle sorbs water below the dew point and thereby acquires negative charge, that the sorbed water freezes at 0°C, and that the electrification is reversible. There are sudden changes in charge at the onset of sorption at the dew point and at freezing and melting, but the largest changes take place in prolonged and extensive sorption or desorption. The electrification follows an exponential rate law, indicating an autocatalytic process. Ice formation at 0°C was also obtained with a dense cloud of AgI and correspondingly small condensate droplets in the expansion chamber. It is concluded that nucleation occurs at 0°C, and that supersaturation pertains to the growth of the ice to detachable size. The data are compared with data in the literature. It is concluded that contradictions among published data are the results of differences in experimental conditions, especially substrate effects in experiments with supported drops. The mechanisms of nucleation, growth and electrification are discussed.

1. Introduction

The freezing of water is accompanied by electrification. The magnitude and the polarity of the charge depend upon the rate of freezing and upon the nature of impurity salts present (Gross, 1968). Freezing drops become electrified only when the precipitated gas, dissolved in the water but insoluble in the ice, bursts out of the drop (Kachurin and Morachevskii, 1966). Under favorable circumstances the charge generated in freezing is very large, and it has been suggested that thunderstorm electricity is generated in this manner.

Freezing is part of the normal precipitation process, and it should therefore be expected to be accompanied by electrification in clouds more generally, including seeded clouds. MacCready and Takeuchi [referred to by MacCready and Baughman (1968)] have measured charges on ice particles in a seeded cloud. They found charges close to the limit of stability by Lord Rayleigh's criterion for charged water drops. Stow and Latham² have reported equally large charges in developing thunderstorm clouds. Berg and Gaukler (1968) observed charges of the same order of magnitude on ice particles in clouds under conditions of natural precipitation, i.e., freezing and growth.

This is not to say that droplets and ice particles in clouds are always strongly charged. There is an abundance of data on weakly charged cloud droplets (e.g., Chalmers, 1967). But the evidence shows that strongly charged ice particles are abundant in clouds under conditions of growth, i.e., when ice particles and liquid droplets are present together in a cloud.

We think that electrification is an integral part of the precipitation process in general, and that it is of particular importance in cloud seeding.

This paper reports charge measurements on a single, freely suspended, AgI particle in a current of air at terminal velocity and at various temperatures and humidities. The particle size was 15–25 μ . Neighbor droplets and ice particles, serving as sources and sinks for water vapor, were simulated by the wall of the vessel, the nature of which, liquid water or ice, was controlled by the rate of cooling or heating. The technique used permits the accurate measurement of a change in charge by 10^2 esu gm^{-1} , corresponding to 5×10^{12} ions gm^{-1} , or approximately 10^{-10} gm of ions gm^{-1} for the particles used. The charge measurement was thus extremely sensitive as compared to mass measurements, and the experiment, therefore, did not require substantial changes in mass. The AgI used was purchased from Deepwater Chemical Co.; it is said to contain less than 0.1% of hygroscopic impurities. Such AgI sorbs water to an appreciable extent only at or above, but not below, saturation (Orr *et al.*, 1958; Corrin *et al.*, 1964).

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² Stow, C. D., and J. Latham, 1967: Airborne studies of the electric properties of convective clouds. Paper presented at Fourth Intern. Conf. Universal Aspects Atmos. Electricity, Tokyo, Sept.

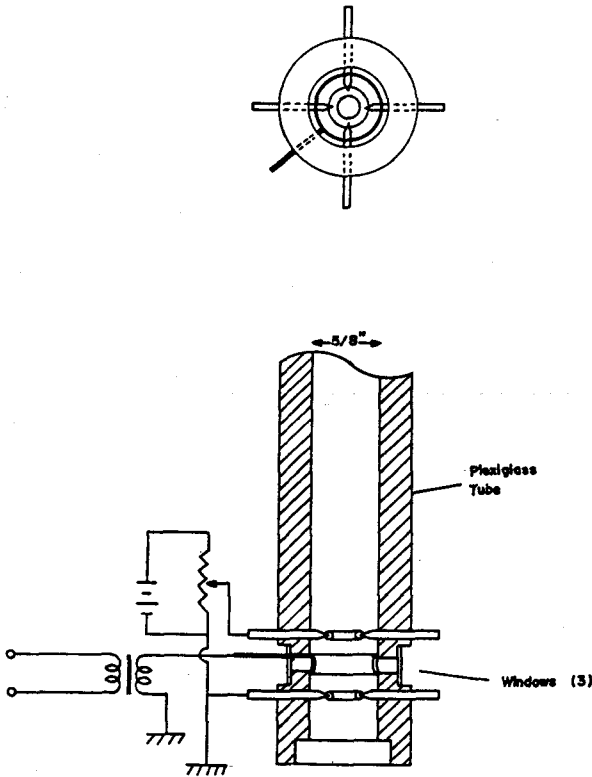


FIG. 1. Suspension chamber.

The results obtained are in general agreement with published information on electrification in freezing. Some additional information was obtained as a result of the high sensitivity of measurement and as a result of the suspension that precludes leakage of charge. On the other hand, the literature contains a few apparent contradictions, e.g., with respect to the freezing of droplets containing Ag particles. The new data seem to resolve some of these contradictions.

2. Experimental technique

A single AgI particle, charged in a corona discharge, was suspended in a vertical current of filtered air at terminal velocity. It was held horizontally by a non-uniform ac field that also provides a restoring vertical force. The particle was held close to the midpoint of the field, where the field strength is close to zero, by adjusting the air velocity. The 60 Hz ac field exerts no net force upon ions or particles $< 5 \mu$. The mass-to-charge ratio m/q was measured at intervals by means of a superimposed, almost uniform, dc field which was applied intermittently only. The applied dc voltage V was adjusted so as to hold the particle at the midpoint of the field with the air current shut off. The value of m/q is related to V by the approximate formula

$$m/q = V \times 10^{-6}, \quad (1)$$

where m is measured in grams, q in esu, and V in volts;

thus, at $V=100$ V the particle has a charge of 10^4 esu gm^{-1} . The techniques for suspension and charge measurement have been described before (Berg and George, 1967). Fig. 1 shows the suspension chamber used in these experiments.

The particle size was determined by taking photographs of the particle. In no case was there any change in size or shape of the particle. The changes observed in m/q were therefore almost entirely due to changes in q .

The suspension chamber was part of a flow system, shown in Fig. 2. Laboratory air was blown through a filter and controlled by a gate valve. The suspension chamber was mounted in a cold box that was cooled by a refrigerator. The air in the cold box was stirred with a fan to give a uniform temperature in the cold box. The air temperature in the flow system was measured with thermistor probes at two locations. They gave nearly the same temperature at the flow rates used. The particle would take the temperature of the air in a few milliseconds. The cold box was made of plywood and insulated by 3 inches of polystyrene foam. The flow system was made of plexiglass tubing. There was no electrostatic shielding.

The cooling rate was controlled by adjusting the expansion valve and by a suitable choice of coolant pressure. Control by switching the refrigerator on and off could not be used at low temperatures because such control affects not only the temperature but also the state of the wall. Thus, in rapid cooling the wall is covered by ice, but when the refrigerator is switched off the ice becomes covered by liquid water.

The optical system used for observation is shown in Fig. 3. It is an ordinary microscope system for Koehler

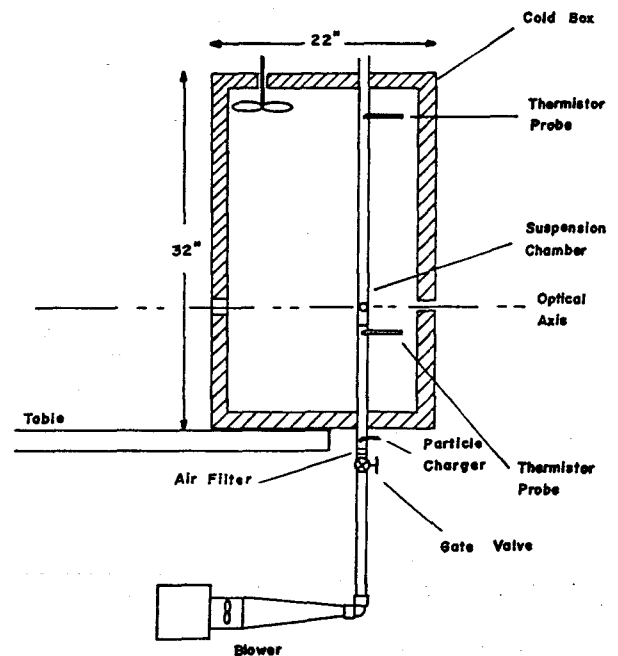


FIG. 2. Diagram of apparatus.

illumination of the particle. The particle was viewed in scattered light at right angle to the optical axis at a magnification of 9.5. This microscope was used for general observations and for charge measurements. Pictures were taken in shadow illumination through another microscope at a magnification of 19.7. We used a Dynafax camera at a framing rate of 500 frames sec^{-1} because the short exposure per frame, 50 μsec , gives a sharp picture even though the particle may vibrate. Fig. 4 is a reproduction of a photograph of a 17 μ particle. Photographs were taken intermittently in connection with the charge measurements.

The device for charging the particles was inserted into the tube just above the air filter. The AgI powder was placed in a small metal bowl, and 5000 V dc was applied between it and a pointed wire a short distance above the powder. The particles were positively charged. The charged particles were carried by an air current to the suspension chamber. They were guided along the axis of the tube and away from the wall by a nonuniform ac field produced by four rods in the wall of the tube. This technique has been described before (Berg and Gaukler, 1968). As a rule, several particles were caught in the field. All but one of them were eliminated by manipulating the field and the air current. When a single particle had become suspended, the charging device was removed, the hole was sealed, and the guiding field was switched off.

As already mentioned, the AgI particle does not sorb water vapor below saturation. However, if water remains in the flow system from a previous experiment, the air might be humidified in the system. In order to check this, charge measurements were conducted at room temperature over a period of time until there was no change in charge with time. When the system was at equilibrium, there was no change in charge over a

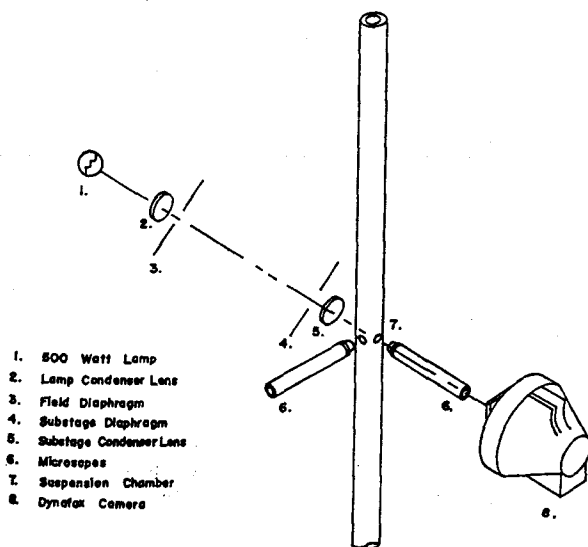


FIG. 3. Optical diagram.

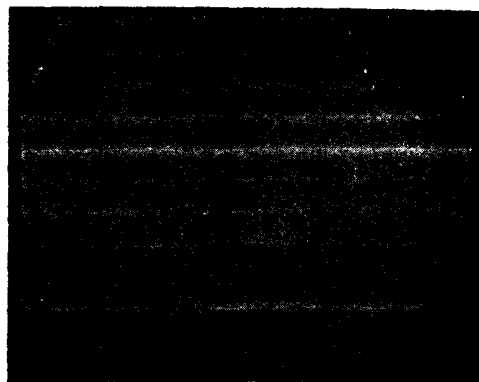


FIG. 4. Reproduction of a photograph of the AgI particle used in taking the data in Fig. 6, particle size 17 μ .

period of several days. It is essential to use filtered air, free of ions and charged dust particles, in this type of experiment. In unfiltered air the particle would lose its charge in a few minutes.

The standard procedure was to suspend a single particle, check the flow system, switch on the refrigerator, bring the air temperature down to some predetermined level, switch off the refrigerator, and make charge measurements intermittently during the temperature cycle. The relative humidity of the air in the laboratory was measured with the Alnor Dew Pointer at intervals.

3. Experimental results

The temperature itself seems to have some influence upon the rates of the processes, but its main effect is to control the relative humidity of the air. At equilibrium in the system, the value of m/q is constant. When the temperature is reduced, m/q remains constant until the dew point is reached; from there on m/q increases. This was verified in numerous experiments. Below the dew point, at saturation with respect to liquid water, m/q increases until the temperature reaches 0C. The water then freezes on the wall and on the AgI particle, the system assumes equilibrium at saturation with respect to ice, and m/q becomes constant. However, if the cooling is very slow, the ice on the wall will be covered with liquid water. In this case m/q increases at an accelerated rate.

When the refrigerator is switched off below 0C and at a constant value of m/q , the change in m/q depends upon the rate of heating of the wall. If the previous cooling was rapid, the temperature in the cold box is low and the heating of the wall is slow. There will then be ice on the wall while the ice on the AgI particle melts. The AgI particle then gives off water to the wall, and m/q decreases. This continues until the ice on the wall melts. The air then becomes saturated with respect to liquid water, and m/q increases until the temperature passes through the dew point, whereupon m/q decreases. If the previous cooling was slow, the temperature in the

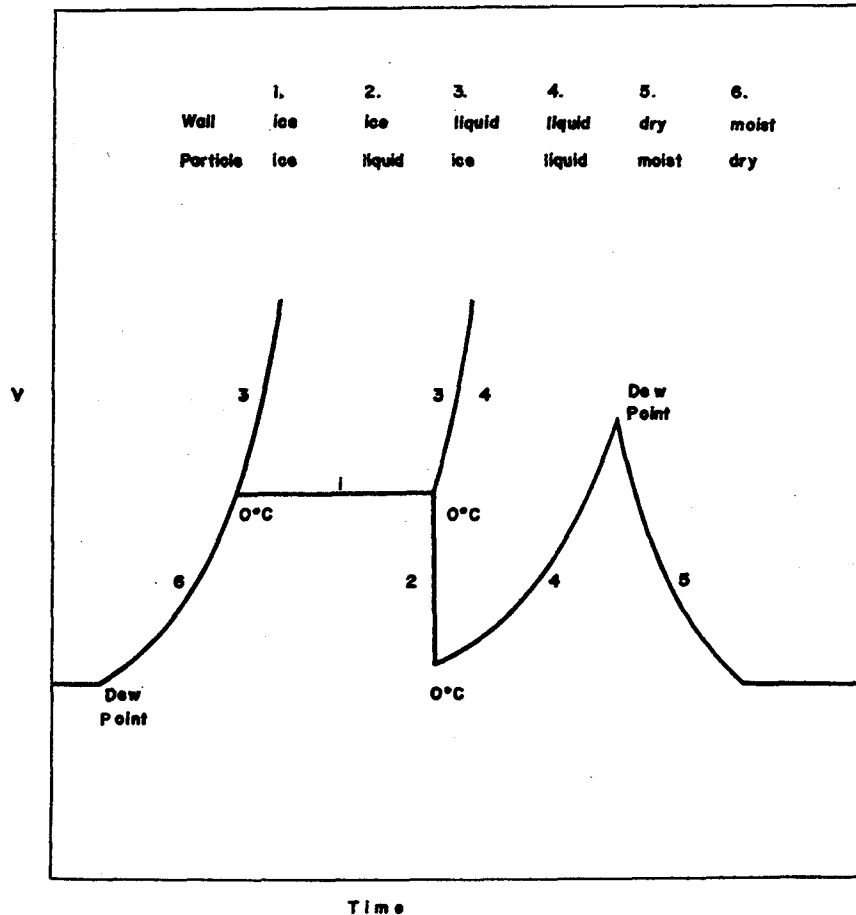


FIG. 5. Schematic representation of $m/q = V$ as a function of time at various rates of cooling and heating for a positive particle.

cold box is comparatively high, and the heating of the wall is rapid. Thus, the ice on the wall as well as that on the AgI particle melts, and the air becomes saturated with respect to liquid water.

These effects are shown schematically in Fig. 5. Typical actual data to illustrate the various cases just described are shown in Figs. 6-9. The plot in Fig. 8 does not bring forth the increase of the rate at 0C. The same data have been replotted in Fig. 9 to show this detail more clearly. The logarithm of the change in m/q plotted vs time gives two straight lines on either side of 0C. The slopes give the time constants 0.033 and 0.051 min^{-1} , respectively.

The plots of the data show that m/q changes rapidly or suddenly at the freezing or melting or at the start of growth or evaporation, but these rapid or sudden changes are not always noticeable or present at all. Nevertheless, when they do occur they are too rapid to be attributable to a change in mass.

A great many photographs were taken of suspended AgI particles for size measurements. In no case could any change in size or shape be detected. The amount of water on the particle must have been very small. The

changes in m/q were large, by a factor of up to 6. They are therefore almost exclusively changes in q . When m/q decreases, q increases, and vice versa.

The largest changes in m/q are those over long periods of growth or evaporation. The value of m/q then changes according to an exponential rate law in most cases, but occasionally a linear rate law was found. These rate laws were found at a constant temperature as well as a gradually changing temperature. The rate of change appeared to be slightly higher at a high constant temperature than at a lower decreasing temperature. The data give some qualitative evidence to this effect, but the variations with the experimental conditions were too large to permit the determination of an activation energy.

The data were otherwise remarkably reproducible, and the effects of the cooling rate could be produced at will by regulating the expansion valve in the cooling system or by switching the refrigerator on and off. There was no noticeable differences between individual particles, except those deriving from the experimental conditions.

The particles used in these experiments were posi-

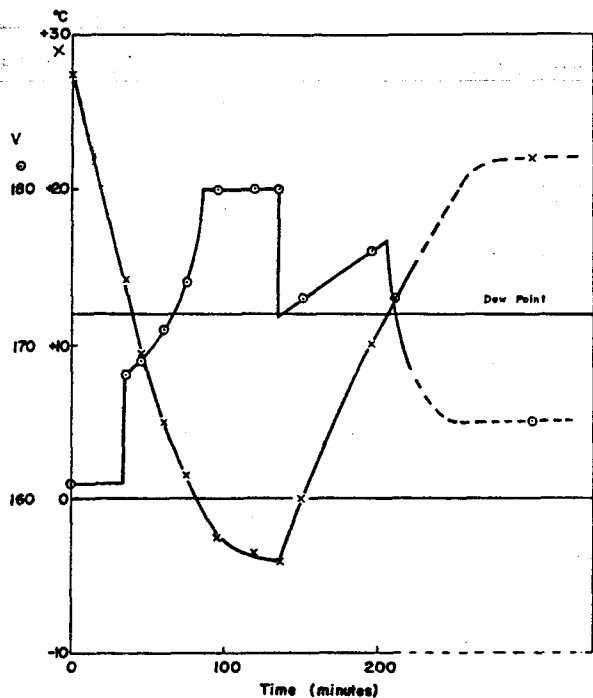


FIG. 6. Plots of $m/q=V$ and temperature vs time for a positive particle.

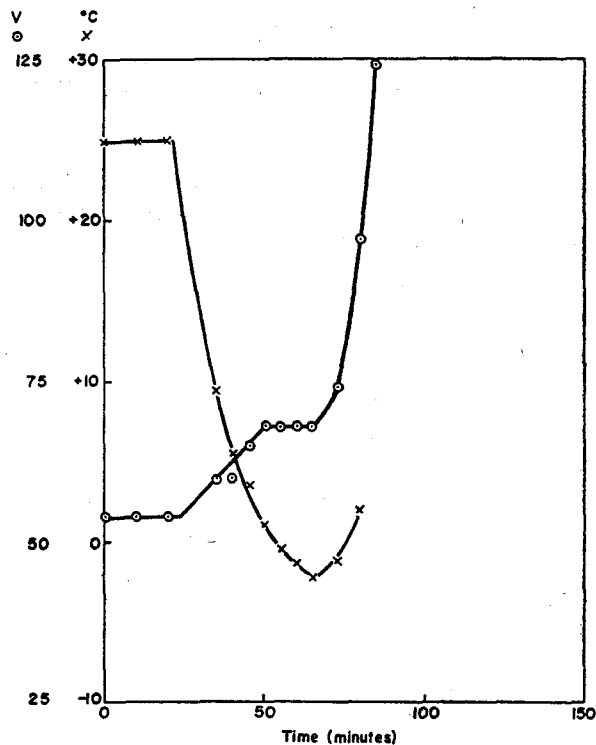


FIG. 7. Plots of $m/q=V$ and temperature vs time for a positive particle

tively charged because the discharge favored this polarity. It is therefore not quite clear from the data whether they pertain to loss and gain of charge or to acquisition of negative and positive charge. Prior to these experiments we conducted some exploratory experiments with negative particles that were charged by friction in a capillary, through which they were blown into the suspension chamber. The suspension chamber was made of aluminum and had a cooling coil wrapped around it in direct contact with the wall. The thermal insulation was poor, and the wall temperature changed rapidly with large temperature differences between wall and particle. The air in the suspension chamber was stagnant. Nevertheless, the data obtained give some qualitative information and resolved the issue of polarity. Fig. 10 shows the data obtained in one such experiment. The value of m/q followed the variations on the wall as the refrigerary was switched on and off. It is noteworthy that these variations have the same polarity as compared to the plots obtained in the later experiments with positive particles. The relative humidity plotted in Fig. 10 was calculated on the assumption of no condensation. Of course, the actual humidity did not exceed saturation. These exploratory experiments were conducted by Mr. Gene E. Carpenter.

4. Discussion

The wall serves as a source or a sink for water vapor and thereby simulates neighbor droplets and ice particles in a cloud. There is no electrification and no

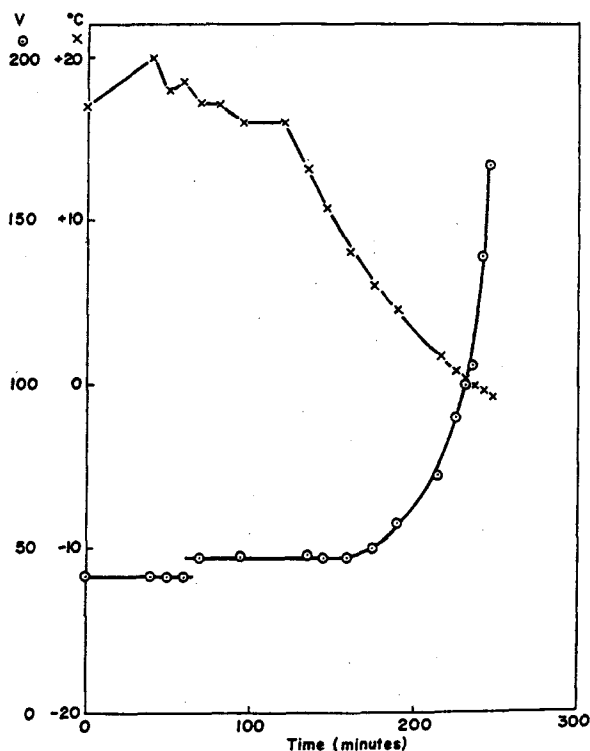


FIG. 8. Plots of $m/q=V$ and temperature vs time for a positive particle.

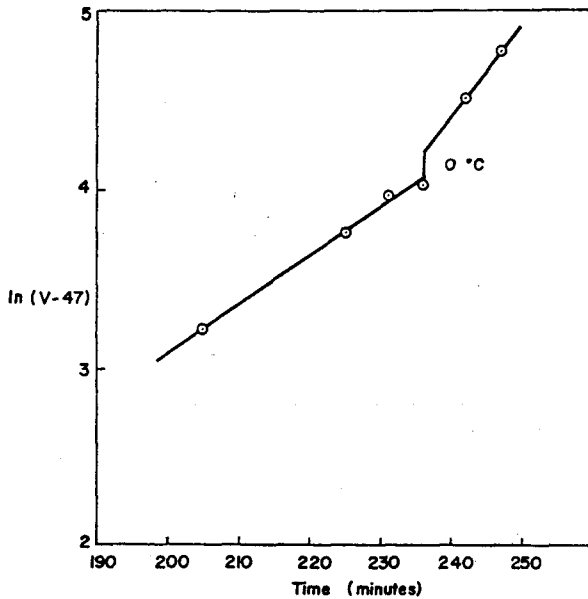


FIG. 9. Plot of logarithm of change in $m/q = V$ vs time on the data in Fig. 8.

sorption of water vapor when there is ice on both wall and particle. An ice particle does not grow and does not acquire charge in a completely glaciated cloud, only in the presence of liquid droplets. However, when the wall and the particle are both liquid, electrification and sorption take place, presumably because the particle is not completely covered by liquid water. Mühleisen (1959) and Dubois (1963) have observed electrification of liquid droplets both when they grow and when they shrink.

In our experiments the AgI particle acquired negative charge in sorption and positive charge in desorption. Mühleisen found electrification to the same polarities for growing and shrinking fog droplets, respectively. Dubois found the opposite polarities for droplets of NaCl solutions.

It seems probable that the polarity would depend upon the presence of impurity salts dissolved in the water. Such an effect has been observed by Workman and Reynolds (1950) in the freezing of bulk water. In our experiments the ice on the AgI acquired negative charge in its growth. This is in agreement with the observations of Workman and Reynolds with halide solutions.

The formation of ice on the AgI particle at, or close to, 0C is in apparent disagreement with the generally accepted threshold at -5C. But Roulleau (1957) and Hoffer and Ogne (1965) have reported ice formation on AgI at -1C and -2C, respectively. It is conceivable that freezing took place at 0C in our experiments because there was so little water, a thin film, or very small droplets, on the AgI particle.

In order to check this, experiments were conducted with a very dense AgI particulate in the expansion

chamber under conditions otherwise identical to those reported (Berg *et al.*, 1968). The condensate droplets were much smaller than previously as a consequence of the greater density of the cloud. Ice was observed as close to 0C as we could measure, certainly above -1C. When the particulate density was reduced, the previous result, a threshold at -5C, was reproduced. The particulate used in these experiments was AgI sublimate prepared from cloud-seeding grade AgI and also from ultrapure AgI (Corrin *et al.*, 1967), that was supplied by Dr. M. L. Corrin, Colorado State University, Fort Collins. The experiments were performed by Mrs. Urte Vaughan.

The evidence shows that nucleation takes place at, or close to, 0C, and that the supercooling pertains to the growth of the ice. Thus, the ice does not grow through a large drop, i.e., indefinitely, unless the water is supercooled to a certain extent, usually -5C.

This result is compatible with those of Edwards *et al.* (1962) and Gokhale (1966), who studied the freezing of water drops containing AgI and supported on paraffin.

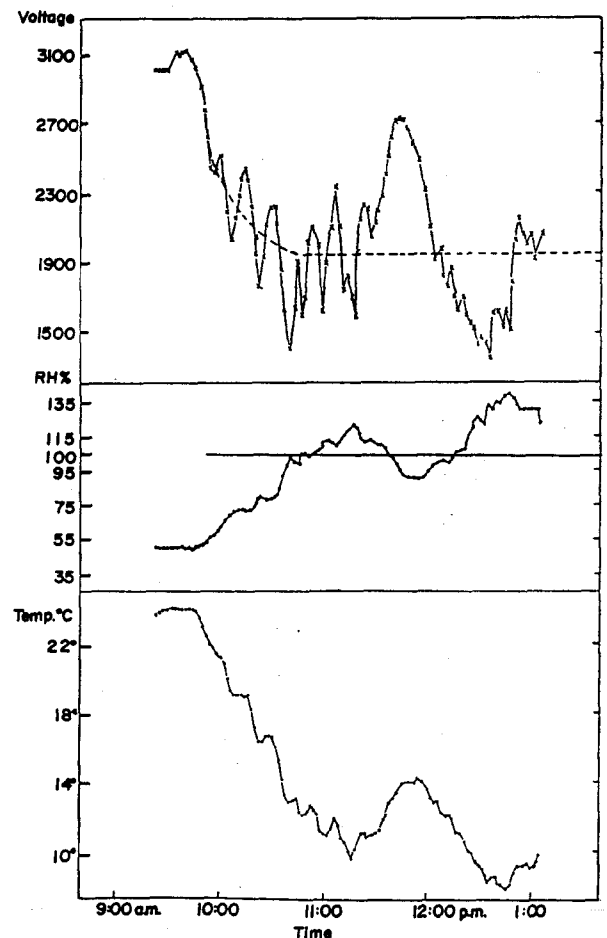


FIG. 10. Plots m/q (voltage), relative humidity (calculated from that at room temperature on the assumption of no condensation), and temperature for a negative particle in still air.

They found that the freezing temperature was a linear function of the logarithm of the surface area or the concentration of the AgI. This indicates a competition between freezing and an opposing process, the freezing temperature being that at which freezing was the faster of the two.

Some information on the opposing process can be gained from a comparison of different experiments reported in the literature. Edwards and Evans (1968), using the same AgI as Edwards *et al.* but another substrate, dimethyl-dichlor-silane, found a threshold at -6°C for the freezing of spray droplets and condensate droplets, and also for the refreezing of melted ice particles. Thus, the opposing process is affected by the substrate, and it is promoted by paraffin but not by dimethyl-dichlor-silane. Hoffer (1961) and Brownscombe and Thorndike (1968) studied the freezing of droplets containing colloidal AgI, embedded in silicone oil and free falling in air, respectively, and found freezing at -16 and -5°C , respectively. Again, there was apparently a substrate effect, the opposing process being promoted by the silicone oil. Substrate effects have also been found by Hosler and Hosler (1955), Mossop (1955), and Yang and Good (1966), who conducted freezing experiments with water in vessels of different wall materials. It appears that the effect of the substrate is related to the gas dissolved in the water and precipitated in freezing. Thus, Mossop found no effect of surface treatment of the glass wall when the water was thoroughly degassed. This matter will be further discussed later on.

Schaefer (1952, 1954) and Berg *et al.* (1968), using unsupported AgI particles, and Edwards and Evans (1968), using AgI particles supported on dimethyl-dichlor-silane, found a threshold for the freezing of condensate droplets at -5 or -6°C . But Hoffer and Ogne (1965), using AgI particles on Millipore filter, observed freezing at -2°C . It is conceivable that the substrate made the difference, but it is noteworthy that Hoeffler and Ogne grew their condensate droplets more slowly, and that they determined freezing by nucleation of an aqueous solution. There are thus several reasons why the results could be different, but we think that the most plausible explanation is that the growth of the droplet was slow, maybe slower than the growth of the ice.

There is evidence that gases dissolved in the water and precipitated in freezing affect both freezing and electrification. A gas bubble in water is charged, gas bubbled through water is ionized and leaves the water charged, and a drop becomes charged in freezing when the precipitated gas bursts out of the drop. In all these cases the electrification is affected by salts dissolved in the water (e.g. Alty, 1926; Pitts, 1927; Harper, 1957; Reiter, 1964). In our charge measurements there was no restraint on the gas, and it escaped as it was precipitated. It was shown by Dorsey (1948) that heating below the boiling point and by Mossop (1955) that

prolonged distillation enhance supercooling, and that subsequent exposure to air raises the freezing temperature. Cavitation produced by shock (Young and van Sicklen, 1913; Goyer *et al.*, 1965) or by ultrasound (Hickling, 1966; Hunt and Jackson, 1966), which occurs only in the presence of gas (Schumb *et al.*, 1955), initiates freezing. It is noteworthy that cavitation is promoted by all gases, including noble gases. In the cases of ultrasound and shock, H_2O_2 and H_2 and their reaction products with solutes are produced, e.g., HNO_2 and HNO_3 , in the presence of air; H and OH radicals are intermediate in these reactions.

The freezing of water involves the rearrangement of intermolecular (hydrogen) bonds. This process is effected, or promoted, by H and OH radicals; also, H and OH radicals, or H^+ and OH^- ions, are instrumental in oxidation-reduction type electrochemical electrification. Thus, freezing causes the precipitation of the gas dissolved in the water, the gas produces a cavity, H and OH are formed in the cavity, and H and OH promote freezing and cause electrification. However, H and OH are minor constituents of the gas in the cavity, and the ice is separated from the water by the gas. The growth of the ice requires the transfer of water from the liquid to the ice through the intermediate gas. Hence, the precipitation of gas both promotes and opposes the growth of the ice.

This chain of evidence links the opposing process in the experiments of Edwards *et al.* and of Gokhale to the precipitated gas and, indirectly, the effect of the substrate to the precipitated gas. The effect of the substrate may be more or less evident, but the oxidizing chromate used in Mossop's experiment has an obvious effect upon H. The effect was promoting, which indicates a promoting effect of OH. Paraffin would act rather as a reducing agent and consume OH and, therefore, oppose freezing. Silicone is known to decompose H_2O_2 vapor, notably into H_2O and O_2 , and should therefore be expected to destroy OH and oppose freezing (Schumb *et al.*, 1955). Silanes would not have this effect, and their stability suggests that they have no appreciable effect.

Further information on the freezing mechanism is provided by data published by Gokhale, who determined the reaction order in freezing (1966) and evaporation (1963). His data on freezing show an equilibrium with a heat of reaction of $0.3 \text{ kcal mole}^{-1}$, i.e., $\frac{1}{3}$ of the heat of fusion of ice. This indicates a 5th-order reaction. His data on evaporation, the rate of surface area change as a function of the relative humidity, give a 3.5th-order reaction, corresponding to a 5th-order reaction for the change in mass. Thus, freezing-melting and evaporation-condensation involve 5 water molecules together. This is in agreement with the thermodynamics of freezing and condensation proposed earlier on the basis of other evidence (Berg, 1964). It is also in agreement with reaction order determinations for the precipitation of salts from aqueous solution. We think

that Gokhale's data on freezing pertain to growth and not to nucleation.

In most of the cases observed in our experiments the change in charge followed an exponential rate law

$$dq/dt = kq, \quad (2)$$

where k is the rate constant. Integration gives the empirical formula

$$q = q_0 e^{kt}. \quad (3)$$

This rate law may be compared with the exponential rate of change of the field strength at ground level during the development of a thunderstorm reported by Reynolds and Neill (1955) and Moore *et al.* (1959). Their observation could pertain either to the charge per ice particle, or to the number of charged ice particles. Their rate constant was eight times that in our experiments, but it may still indicate an exponential increase of the charge per ice particle.

Another related application of the charge measurements in the Facy effect. It has been shown by Rosinski *et al.* (1963) that Stefan's wind does not account for the effect, and also that electrically neutral particles are scavenged by a growing drop. It has been shown by Vittori and Prodi (1967) that a high humidity promotes the scavenging by ice particles as well as by water drops. We think that exposure of the particles to a high humidity electrifies them, as in our experiments with AgI, and that the scavenging effect is a result of electrostatic attraction. It is noteworthy that Vittori and Prodi found preferential deposition of the particulate on the growing part of a snowflake, where charge would be generated. Such a nonuniform charge distribution on growing ice particles has been proposed by Odencrantz *et al.* (1968).

It thus appears that freezing causes electrification, and that electrification promotes growth of the ice. Electrification may therefore be an autocatalytic process as expressed by the rate formula (2). The evidence indicates that the process involves dissolved gases and dissolved salts, and that the electrification is an electrochemical process of the oxidation-reduction type. If this interpretation is correct, it would indicate means for improving the properties of AgI as a seeding agent with respect to the threshold temperature for the formation of ice and with respect to growth of the ice, notably by suitable doping of the AgI.

5. Conclusion

The data and observations reported here confirm and extend those reported by other investigators. Electrification takes place whenever a water droplet or an ice particle grows or shrinks by exchange with its environment. In addition, there is also some electrification in freezing-melting and in condensation-evaporation, but the largest effect is apparently that in prolonged and extensive exchange of mass. The observed changes in

charge, of the order of 10^4 esu gm^{-1} , are as large as those found in natural clouds. The data show that ice nucleation takes place at, or close to 0°C , and that the supercooling usually observed pertains to the growth of the ice to detectable size. The information obtained, combined with that in the literature, throws some light upon the mechanisms of nucleation and growth in freezing. It seems to hold out some hope for improved cloud-seeding agents with respect to threshold temperature.

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