Ice electrification

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I C E E L E C T R I F I C A T I O N

by

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ABSTRACT

When water drops are cooled to $-1^{\circ}C$, nucleated with tiny ice crystals and then allowed to freeze at $-15^{\circ}C$, a thin shell of ice is formed. Further freezing of the water causes internal stresses to be set up and the water escapes through the shell to form spicules. If the pressure becomes too great then some of the ice shell or of the spicule may be ejected, and the residue is found to have a charge of the order of 10^{-3} e.s.u.

A diffusion chamber was designed and constructed. The base was cooled directly being an aluminium heat sink surrounded by solid carbon dioxide. The steady temperature gradient thus set up was measured with horizontal thermo-couples. A 1 mm diameter water drop was suspended in the centre of the chamber on the end of a fine insulating fibre which could be easily raised and lowered. The charge on the drop was measured by raising it into the centre of a Faraday cage which was connected to a sensitive ballistic galvanometer through a Vibrating Reed Electrometer (V.R.E.). The limit of sensitivity was 0.05×10^{-3} e.s.u.

It was found that the sign of the charge on the residue was dependent upon the type of break which had occurred. When more water than ice was given off then the residue had a predominantly negative charge. When no break occurred, no charge was detected.

Of the 633 drops studied, 118 broke in some way. The average charge on the residues was $+0.34 \times 10^{-3}$ e.s.u. and ranged from -17.10 to $+25.30 \times 10^{-3}$ e.s.u.

It was found impossible to account for either the magnitude or the sign of the charge in terms of the Latham and Mason temperature gradient theory but both could be accounted for if the Workman and Reynolds effect was invoked.

It is suggested that in a thundercloud the effect of the turbulence present is to increase the number of breaks which occur while the spicule is still partially liquid, thereby increasing the number of negative residues and giving the cloud the correct polarity, when minute water drops freeze on graupel and break.

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CHAPTER 1

Introduction

The might and power of the thunderstorm has always been written about in literary works but since 1752, when Benjamin Franklin first proved this power to be provided by electric charges, there has been an ever increasing volume of scientific works on it. Among the papers written have been many on the generation of the electric charge. This aspect of the thunderstorm is still under discussion and although several theories have been suggested no conclusions have been reached and it seems possible that a synthesis of several of these theories might be the best solution to the problem.

In view of the many books and papers which have been written on thunderstorms (some of which are tabled at the end of this chapter) it is proposed here to give only a brief account of the results of the work done on the properties of the thunderstorm which have to be accounted for by any theory of charge separation and then to pass on to the work done in connection with some of these theories.

1.1 General Thunderstorm Properties

It has been established that the distribution of electric charge within a thundercloud is approximately as shown in the diagram (Fig. 1) (Simpson and Robinson 1940). The main dipole carries a charge of 30 C separated by 5 Km with the upper positive charge region at a temperature of $-30^{\circ}C$ and the lower negative region at a temperature of $-5^{\circ}C$. The much smaller, positively charged base region carries +5 C and is situated just below the $0^{\circ}C$ isotherm.

The electric moment destroyed by a lightning flash is on average 110 C -Km (Wormell 1939, 1953 and others) and the charge neutralised has been calculated to be from 20 to 30 C (Workman and Holzer 1942). The average interval between flashes is 20 seconds. The rate of recovery of the electric moment after a lightning flash takes an approximately exponential form with a time constant of about seven seconds, which leads to a calculation of the charging current of one amp, taking into account the leakage currents (Mason 1953a).

The connection between precipitation and electrical activity has been established (Kuettner 1950). The central lightning area usually has the highest intensity of precipitation and the lightning originates at the same time as solid precipitation particles appear in the cloud. Further work done by Workman and Reynolds (1950a) shows that the interval between the appearance of precipitation and the first lightning flash is between 10 and 20 seconds. The electrical effect in non-stormy clouds is known to be considerably less than in stormy ones.

Any theory for the generation of charge within a thundercloud must, then, account for these main properties.

1.2 Theories of Charge Generation in Thunderclouds

The main dipole of the thundercloud suggests a gravitational method of charge separation. Two processes occur in this method, firstly a process of separating the charge on to particles of different sizes in a central net neutral region and then, secondly, a process of segregating these particles to form the main dipole.

The only theory yet suggested which does not invoke this double process is the convection theory originally formulated by Grenet (1947) modified by Vonnegut (1955) and later by Vonnegut and Moore (1958).

If a gravitational method is assumed and the relative velocities of the particles is taken to be 11 m sec^{-1} and the recharging current three amp then it has been calculated that the quantity of unsegregated charge of either sign in the central region must be about 1000 C (Mason 1953b, Wormell 1953). Decreasing the value of the relative velocities, that is decreasing the difference in size of the particles, increases the charge to be carried on them. If the particles are much smaller than precipitation particles the calculated charge is too great for them to carry.

Amongst the gravitational theories two main groups may be defined; one postulates that the charges already existing in the atmosphere are preferentially attached to different sized particles and the other that particles which were originally neutral are in some manner broken and charges become attached to the differently sized pieces.

This second group may again be divided into two sections, one in which the theories concern only the liquid form of precipitation and the second which involve the solid form either together with the liquid form or on its own.

From the temperatures existing in a thundercloud this latter subgroup would seem to be the best one to consider when trying to find an acceptable theory, for although supercooled water drops can exist at temperatures down to $-30^{\circ}C$, the natural freezing nuclei in the cloud would tend to freeze some of them and start an avalanche.

Before any general theory on the generation of charge in a thundercloud can be formulated it is obvious that it is desirable to study more closely the electrical effects associated with the phase change from water to ice and vice-versa. Much work has already been done on this topic and it will be briefly reviewed in the next chapter.

General references:-

Byers, H.R. and Braham, R.R.	1949	The Thunderstorm.
Chalmers, J.A.	1957	Atmospheric Electricity.
Kuettner, J.	1950	The Electrical and Meteorological Conditions inside Thunderstorms.
Mason, B.J.	1957	The Physics of Clouds.

CHAPTER 2

Electricity Associated with the Freezing of Water

As noted in the first chapter, the onset of lightning usually occurs at the same time as solid precipitation is first observed in the thundercloud. It seems most probable then that the formation of the precipitation is one of the major processes in the generation of the electric charge.

The main process in the growth of a hail pellet is the accretion of supercooled water droplets on to an ice crystal core grown by sublimation. These supercooled droplets will freeze quickly if the water content and the temperature of the region is low and more slowly if they are high; in wetter or warmer regions the surface of the pellet may become wet if the latent heat cannot be dissipated quickly enough (Mason 1957).

2.1 The Workman and Reynolds Effect

Some of the first work done on the electrical effects associated with the freezing of water was that of Workman, Reynolds and their colleagues in New Mexico. They found that during the freezing of dilute aqueous solutions a potential difference was established across the liquid-solid boundary (Workman and Reynolds 1948, 1950b). The sign and magnitude of the potential difference was dependent upon the contaminants in the water; the ice was usually negative with respect to the water.

The main contaminants in precipitation are calcium carbonate and sodium chloride and when a solution of approximately the same composition

as that found in nature was frozen, the water acquired a charge of about $+10^4$ e.s.u. cc^{-1} of frozen water.

The only exceptions to the negatively charged ice were when solutions of ammonium compounds were used. With a 3×10^{-5} normal solution of ammonium hydroxide the water obtained a charge of -3×10^5 e.s.u. per cc.

They put forward the theory that the negatively charged ions in the solution were preferencially frozen into the ice. The reason for the opposite process in the case of the ammonium salts was thought to be due to the isomorphism of the ammonium ion with the hydroxyl ion. This latter theory was tested by using a solution of ammonium fluoride; the fluoride ion is both negative and similar to the hydroxyl ion in structure; this solution when frozen yielded negative ice, the only ammonium compound to do so.

2.2 Spicule Formation on Freezing Water Surfaces

The formation of spicules on the surface of freezing water was investigated by several people in the 1920 and 1930's and a general summary of the main method of formation was formally stated by Dorsey (1948). The pressure of the enclosed water causes a rupture in the ice surface and a jet of water is ejected, the outside of which freezes immediately, the water continuing to flow along the tube formed until the pressures are equalised or the tube becomes blocked with ice. Blanchard (1951) made further observations of this phenomenon where spicules were formed on freezing water drops. He used 8 mm diameter water drops freely suspended in a vertical wind tunnel. He calculated

that using a temperature of -17.6°C approximately 80% of the spicule should grow within 8 sec of the start of freezing, the remainder of the growth taking place much more slowly. His experiments verified this.

2.3 The Fragmentation of Freezing Water Drops

Further work on the formation of spicules and the breaking of freezing water drops was carried out by Maybank (1960) and Mason and Maybank (1960). Drops ranging from 0.06 to 2 mm in diameter were studied. They were supercooled, nucleated with freezing nuclei and then lowered into a temperature-controlled refrigerated cell. The behaviour of the drops at all stages of freezing were studied. If the pressure differences set up by the expanding water were too great for the ice shell to withstand, then fragmentation and splintering of the drops took place. The percentage of drops breaking was found to be influenced only by the degree of supercooling allowed before nucleation was performed. As the nucleation temperature was raised so was the percentage, the highest being at a temperature of just below the freezing point.

The diameter of the drop and the freezing temperature had no effect on the percentage breaking. There was no noticeable effect when impurities were introduced, except in the case of a 0.2 N solution of sodium chloride, when no spicules were formed. They found that the freezing of such a solution produced only half of the overall increase in volume that was produced by distilled water, so that the excess pressure inside the shell was much less. When de-aerated drops were nucleated and frozen at -13°C four of the twenty broke, whereas with similar but aerated drops none broke. There was no difference between

the behaviour of aerated and de-aerated drops when a nucleation temperature of $-1^{\circ}C$ was used.

Blanchard (1957) showed that the type of freezing which occurs in water drops is controlled by the degree of supercooling achieved before freezing starts. If the drop was supercooled to below $-5^{\circ}C$ then opaque freezing took place, innumerable air bubbles being trapped in the quickly forming outer shell. If the temperature was higher than this when freezing began then a clear outer shell was formed, only a very small quantity of air being trapped. Langham and Mason (1958) suggested that these different patterns of freezing might influence the percentage of drops breaking. Maybank's work verifies this, for as the temperature was raised to $0^{\circ}C$ so the percentage was increased, the more brittle shell giving way under the pressure more readily than the more spongy shell which was formed at nucleation temperatures well below $0^{\circ}C$.

2.4 The Electrification of Breaking Freezing Water Drops - London

After observing the physical breaking of water drops, Mason and Maybank (1960) studied the charge produced when 1 mm diameter water drops broke on freezing. A nucleation temperature of $-1^{\circ}C$ and a freezing environment temperature of $-10^{\circ}C$ were used. Their results showed that no charge was produced until the drop broke and that those which broke and left a major residue were predominantly negative while those which left a minor residue were predominantly positive (Table 4).

The charges detected ranged from +4 to -7×10^{-3} e.s.u. and the average charge for the drops in this temperature class was -0.57×10^{-3} e.s.u. Similar experiments with drops of diameter 0.35 mm yielded

charges smaller by a factor of two.

Latham and Mason (1961a) put forward a quantitative theory to account for this charge separation, which they suggested was due to the migration of protons in ice which has a temperature gradient across it. The concentration of H^+ and OH^- ions in ice rises rapidly with an increase in temperature and the mobility of H^+ ions is about ten times that of OH^- ions. Because of the above properties, when a temperature gradient is maintained across a block of ice the H^+ ions migrate more quickly to the cold end; this sets up an internal electric field which tends to oppose the migration. A steady state is reached when these two currents are equal and opposite. The quantity of charge (q) separated by this process was calculated to be

$q = 4.95 \times 10^{-5}$ (dT/dx) e.s.u. cm^{-2} where dT/dx is the temperature gradient; with the cold end being positively charged.

The experiments performed to verify this theory showed that the above value agreed well with their results if the temperature of the warm end of the ice was below $-7^{\circ}C$, but if it was above this value, a small correction to the theory was required.

Their calculation of the value of q was dependent upon the linear relationship between log k and T^{-1}, where k is the electrical conductivity of ice and T the absolute temperature. Bradley (1957) showed that above $-7^{\circ}C$ this relationship is no longer linear. When Latham and Mason applied the necessary correction, their newly calculated charge separation agreed well with the experimental results

for all temperature differences, except when the warm end was very close to $0^{o}C$.

At warm end temperatures approaching $0^{o}C$ the experimental results were much higher than the calculated values and it was found that as the surface volume ratio was increased so also was the charge separated. These facts suggested that large charges were being produced in the surface layers of the ice.

2.5 The Electrification of Breaking Freezing Water Drops - Durham

The charge produced on the residues of breaking freezing supercooled water drops has also been studied in Durham by Evans (1962) and Evans and Hutchinson (1963). Once again single 1 mm diameter drops were used. They were supercooled to $-2^{o}C$, nucleated and frozen in an environment at $-15^{o}C$. The charges detected ranged from +11.30 to -25.00×10^{-3} e.s.u. and the average charge was -1.1×10^{-3} e.s.u. Using Latham and Mason's value of 4.95×10^{-5} dT/dx e.s.u. cm^{-2} for the charge separation they calculated that the total charge which could be separated in an average drop was 0.30×10^{-3} e.s.u. Of the drops studied 86% had charges greater than this value, 16% being greater by a factor of ten.

If the same corrections as Latham and Mason used are applied to recalculate the charge separation, based on Bradley's 1957 measurements, then the value of Q in the equation

$$q = Q \times 10^{-5} \text{ dT/dx e.s.u. cm}^{-2}$$

is found to be dependent upon the warm end temperature of the ice in a manner shown in figure 2, if the cold end temperature is $-15^{o}C$.

From this it can be seen that if the warm and cold end temperatures are respectively $-0.5^{\circ}C$ and $-15^{\circ}C$ then the charge separated is

$$q = 9.5 \times 10^{-5} \, dT/dx \text{ e.s.u. cm}^{-2}$$

At temperatures nearer to $0^{\circ}C$ the value of Q could be even larger than this because of the surface effect found by Latham and Mason.

Evans and Hutchinson, when calculating the maximum charge separated in a drop, used the value $Q = 5$. As the inner surface of the ice is at $0^{\circ}C$ then the value $Q = 9.5$ or larger ought to have been used. Substituting this new value into their equations gives a value of maximum charge separated in the drop of 0.6×10^{-3} e.s.u. Of the drop residues, 68% had charges greater than this. To account for all the residue charges measured, a value of $Q = 400$ would have to be used.

When other factors such as errors in temperature, thickness of shell, the effect of the spicule and frictional effects were considered by them, the amount of charge separated by the Latham and Mason theory could not be increased sufficiently to account for the electrification of all the drop residues.

From their results it seemed that the amount of freezing which had taken place determined the sign of the charge on the residues. In drops where the break took place across a liquid-solid boundary, 16 out of 18 residues had a negative charge and where it took place across ice alone, 5 out of 6 residues had a positive charge.

Evans and Hutchinson concluded that the charges on the drop residues could not be accounted for by the Latham and Mason theory, but that they

could well be accounted for by the Workman and Reynolds effect (1950), both from the sign and magnitude of the charges.

By the Workman and Reynolds effect (2.1) negative ice and positive water is expected. Therefore if ice alone is given off, positively charged residues are expected, but if ice and water are given off, then either positive or negative residues are expected, dependent upon the quantities of each ejected.

2.6 The Electrification of Breaking Freezing Water Drops - Russia

Very similar work to that done in London and in Durham has been done in Russia by Kachurin and Bekraiev (1960). Drops ranging from 0.2 to 2 mm in diameter were frozen at temperatures ranging from $-3^{\circ}C$ to $-20^{\circ}C$. The charges measured on the residues ranged from +45 to -90×10^{-3} e.s.u. and the average was -3.03×10^{-3} e.s.u.

The charge on the residues was measured, during breaking, on an oscilloscope and the net charge with an electrometer. The oscilloscope showed that these charges were composed of smaller charges formed during the cracking and breaking of the drop. The positive pulses formed always rose steeply and the negative ones gradually. The different signs were associated with the ejection of different particles. The steep positive pulses appeared when comparatively large negative ice particles were ejected and the gradual negative ones when a stream of minute positive water drops was ejected.

This work is again seen to fit closely to Workman and Reynolds effect by which negative ice and positive water are expected.

2.7 The Electrification of Melting Ice

When ice containing air was melted Dinger and Gunn (1946) found that the trapped air was given off at the surface and took with it a negative charge of the order of 1.25 e.s.u. cc^{-1}, if distilled water was used. When contaminated water was used no charging took place.

More recently the experiments of Dinger and Gunn have been repeated by Matthews and Mason (1963); on no occasion did they detect any charge production.

2.8 The Aims of the Work

The present work was undertaken to try to obtain more evidence on the sign and magnitude of the charge left on a residue after a supercooled water drop broke on freezing. To obtain sufficient results for all types of break to be equally represented is no easy matter for usually only 10% of the drops break.

It was hoped to account for the observed charges by one or more of the theories of charge separation; and also to obtain more evidence to decide whether or not the type of break affected the charge separation as was suggested by Evans and Hutchinson (1963).

CHAPTER 3

The Apparatus

To study the freezing process of water drops and the charge produced when they break, the main essentials of apparatus must be an efficient cooling system to produce a steady temperature gradient in an experimental chamber, apparatus for the support and nucleation of the drops, and instruments for the measurement of any charges produced.

In the apparatus designed by Hutchinson (1960) and used by Evans (1962) the essential features were a cooling system using circulating paraffin oil beneath a small perspex chamber. Several disadvantages had been found with this apparatus, including a high temperature gradient, frost formation on the inner windows and a minimum temperature of $-30^{\circ}C$, which is not sufficiently low.

It was decided, therefore, to redesign the apparatus and to try to eliminate these qualities and at the same time to make the apparatus more adaptable for future experiments of a similar nature, by constructing the separate units so as to be independent of each other.

Figure 3 is a photograph of the complete apparatus, while figure 4 shows a complete diagram of the main cooling and diffusion chambers.

3.1 The Cooling System

To reduce the heat gain experienced in Hutchinson's apparatus it was decided to cool the base of the chamber directly by making it the upper surface of a heat sink (Fig. 4).

A six inch cube of aluminium was used as a heat sink, the upper and lower surfaces of which were machine faced and had two nine inch duralumin plates bolted to them, making good thermal contact. Both plates served as mechanical stabilisers and also the upper one presented a uniform temperature surface to the chamber and the lower one acted as a heat catchment area.

The heat sink was housed in a large perspex chamber with false sloping sides, into which the coolant (solid carbon dioxide, "Cardice") could be introduced. The false sloping sides forced the coolant on to the sink. To prevent heat gain from the perspex, the sink was placed on a layer of insulation. The base joints of the chamber were sealed with Apiezon 'Q' Sealing Compound to prevent water, formed on warming the chamber, from seeping into the insulation.

The whole of the perspex chamber was suitably supported with a frame made of steel Handy Angle and then surrounded with insulation three inches thick on the sides and six inches thick on the bottom. The insulators used were "Polyzote" and "Onazote", trade names for expanded polystyrene and expanded ebonite, which have thermal conductivities of 0.8 and 0.7 x 10^{-4} cal. cm^{-1} sec^{-1} $^{o}C^{-1}$ at 10°C respectively. To prevent ice forming in any spaces and causing inconvenience on melting these spaces were filled with cork dust. Finally the insulation joints at the outside were lagged with felt to prevent any air flow, and the whole was bound with strong adhesive tape.

The lid to the cooling system, also made from Onazote, was cut to

fit closely to the diffusion chamber and shaped to fit against the top of the sloping sides. The coolant was replenished merely by removing this lid.

3.2 The Diffusion Chamber

This was constructed of perspex and had double walls (Fig. 4) the inner chamber being the experimental area, the outer one a guard ring area.

The inner chamber dimensions were 15 cm x 15 cm x 30 cm and the chamber reached down to the heat sink but was supported only by the brass guard ring, which rested on the heat sink. The guard ring acted as a base to the guard ring chamber and as a support to the Onazote jacket. (Figs. 5, 7)

The temperature gradient in the experimental chamber was measured by horizontal thermo-couples (3.3), which were also taken through the guard ring chamber horizontally, to reduce heat flow along them.

The guard ring chamber was kept at approximately the same temperature as the inner chamber by the brass guard ring and thus acted as an insulating chamber. The guard ring chamber was made completely air-tight, using Durofix adhesive and Apiezon 'Q' Sealing Compound, except for a small air inlet at the top. Dry calcium chloride was introduced into the chamber and also into a 'U' tube fitted to the air inlet. This was done to prevent the formation of frost in the guard ring chamber.

An Onazote jacket was then fitted around the chambers with windows

suitably placed for viewing and lighting purposes. The windows were double walled and contained a dehydrating agent.

The whole of the diffusion chamber could be moved within the limits of the thermo-couple leads, for maintenance purposes (Fig. 17).

3.3 The Thermo-couples

The temperature gradient in the diffusion chamber was measured by horizontal copper-constantan thermo-couples, the high thermo-electromotive force produced by them being approximately linear with temperature over the small range considered.

Twelve couples were employed in the chamber, the lowest being 2 mm from the base and the others being at 2.5 cm intervals above; two others were also used, one actually in the heat sink, the other in the insulation to check the heat gain; yet another couple was made and left spare.

All the couples were constructed identically, having the same lengths and thus the same resistance. The diffusion chamber junctions were made as small as possible. The common reference junction was kept in a brass block covered in water and maintained at room temperature in a dewar flask.

The complete circuit of the thermo-couples is shown in figure 8. A Scalamp galvanometer was used to indicate the e.m.f. produced. The resistance 'R' was adjusted during the calibration so that for temperatures between $10^{\circ}C$ and $-30^{\circ}C$ a deflection of 1 mm on the Scalamp was equivalent to $1^{\circ}C$ temperature difference, this being the range over which this series of experiments was to be performed. The switch was

used to select the individual thermo-couples in turn. The junction box was filled with cotton wool and was used to prevent stray e.m.f.'s being produced where the connecting wires were soldered to the copper or constantan.

The system was calibrated by using the diffusion chamber junctions as the reference junction and varying the temperature of the dewar flask junction. A calibration curve (Fig. 9) was then plotted.

Under normal use the zero of the Scalamp was set to the reading of the mercury in glass thermometer kept in the dewar flask and temperatures down to -30°C could then be read directly, a small correction being needed below this value.

One side of the thermo-couples was connected to earth, as shown, to prevent pick-up to the electrometer, especially from the operator.

3.4 The Performance

The apparatus so far described worked very well. If a whole block of Cardice was chopped and placed in the cooling chamber in the evening a temperature gradient suitable for use next morning was obtained (Fig. 10); additional Cardice was then added only when necessary to keep the temperature steady.

No frost formed on the inner walls of the chamber, although, due to the operator's breath and to the moisture in the room, condensation did form on the outside of the windows, but this was easily removed.

Ice naturally formed on the thermo-couples during the experiment

but this was melted at three or four day intervals by a hot blast of air from a hair dryer. The chamber was then left overnight to regain its former temperature.

To check the thermo-couples, the position of the $0^{\circ}C$ isotherm could easily be ascertained by raising an ice crystal on the end of a fibre until it melted.

3.5 The Upper Apparatus

To prevent strain on the perspex diffusion and cooling chambers, the apparatus above them was mounted on a steel Handy Angle bridge with a heavy brass plate on it. This plate held the charge detecting equipment, the fibre suspension rod and tube and the nucleating tube (Fig. 4). It was detachable from the bridge and all individual units on it were fixed with tapped holes, so that any unit or the whole plate could be easily removed.

To stop draughts entering the chamber and destroying the temperature gradient a strip of foam rubber was used to seal between the chamber top and the brass plate.

3.6 The Fibre

The fibre used by Hutchinson (1960) and by Evans (1962) had been made from Durofix adhesive. They had found difficulty in controlling it due to its extreme flexibility, although it was only three inches long. In the new chamber a fibre of some ten inches was necessary. Many different materials were experimented with until eventually a combination of stretched P.V.C. string, Durofix adhesive fibre and a fine glass fibre was found to be suitable. Not only did the fibre have to be pliable, to permit moderately rough treatment while a drop

was being placed upon it, but it also had to have a low thermal conductivity, to prevent heat transfer along it to the drop, and a high insulation, to prevent loss of charge, and last, but not least, it had to permit a drop to be suspended from it easily with the minimum possible contact between the drop and the fibre, to prevent the suspension from affecting the drop's behaviour on freezing. This latter difficulty was the hardest to overcome whilst trying to find an acceptable fibre; however it was found that by giving the glass fibre an enlarged end and then coating this with a film of paraffin wax the drop could be suspended as shown in figure 18 (5.9). By coating it with wax it was also found that the spurious charges experienced by Maybank (1960), due to the collision of air borne ice crystals with a glass fibre, were eliminated.

The heat transfer along the fibre was calculated to be sufficient to raise the temperature of a 1 mm diameter drop by only $0.01^{\circ}C$ in two min, assuming no heat loss to the surroundings.

The leakage time constant of the fibre was measured in the usual manner, by allowing a charge to leak away through it to earth for known time intervals. This was done for each fibre used. The time constant never fell below 30 min and the average was 50 min. Charge measurements were usually performed within half a minute, giving 1% loss of charge. If the time lag exceeded half a minute for any reason then the necessary correction was made.

3.7 The Fibre Suspension Rod and Guide Tube

To enable the fibre to be raised and lowered easily into the diffusion chamber it was glued to the end of a rod which moved easily

in a vertical guide tube (Fig. 4). The guide tube could also be easily raised and lowered on a retort stand, which had a special groove cut in it, so that no circular movement could take place and the rod always remained above the small entrance to the chamber. The lower section of the guide tube was made larger than the rod, so that if the fibre became accidentally caught on the guide tube it was not chopped by the rod if this was moved. All the moving parts were lubricated with graphite powder.

3.8 The Nucleation Tube

Nucleation of the drops was achieved by dropping a small quantity of crushed Cardice down a glass tube. This was drawn out of a wide glass tube producing a funnel shaped top (Fig. 4); the lower end was bent so that the Cardice crystals did not hit the fibre and produce spurious effects but the finer ice crystals formed fell in a shower round it. The drop could then be nucleated by lowering it through the shower or by forming a shower round it, depending on the desired nucleation temperature.

The tube was held in place by two rubber stoppers, on a removable plate; thus it could be rotated slightly for final adjustment of the direction of descent of the crystals. If access to the chamber was required while the chamber was cold, it was made by removing this plate and the nucleation tube.

3.9 Charge Measurements

Charges produced by the drop were measured by raising the drop into the centre of a Faraday cage (Fig. 4) connected to a

Vibrating Reed Electrometer (V.R.E.). As two different V.R.E.'s were used (3.10) the Faraday cage was made to fit both.

The Faraday cage (Fig. 5) consisted of two small cylinders, joined together by a short spiral of fuse wire, to enable the drop to be centrally placed in the cage when charge measurements were made. It was connected to the centre of a co-axial socket, the outside of which was connected to a cylindrical earthed shield of wire gauze, to avoid pick-up, especially from the operator. This unit could then be directly connected to a co-axial plug, fitted to the brass plate, and so to a V.R.E. input.

3.10 The Vibrating Reed Electrometer and Fluxmeter

Charges induced on the Faraday cage were to be measured with a V.R.E. but because all readings would have to be taken visually and the period of a V.R.E. is only of the order of 1 sec, a Scalamp fluxmeter functioning as a ballistic galvanometer, of 20 sec period, was connected to the recorder output socket of the V.R.E.

The V.R.E. finally used was an Electronic Instruments Limited Vibron Electrometer, Model 33B. This had connected to it an input resistor of $10^{10}\Omega$ housed in an earthed metal box, containing a heater lamp, to prevent insulation breakdown due to moisture. The lamp was always short circuited during experiments because of pick-up.

Any movement of the input cable caused piezo-electric effects and thus spurious deflections; all input leads were therefore firmly bolted to the upper brass plate.

As the sign of the charge is not known it is desirable to have

a centre-zero meter. Unfortunately the V.R.E. had a side zero meter and no other meter was available. When the meter was set to a central position a current of 0.5 mA flowed through the recorder and because of this no range change could be performed either at the V.R.E. input or at the fluxmeter input. The meter and an equal resistance were therefore fitted to a double-throw, double-pole switch to allow either the meter or the resistance to be in circuit.

Under normal working conditions the resistance was used, the meter being only used occasionally to check if spurious charges were present or not.

The fluxmeter used was connected through a shunt circuit to the recorder output socket of the V.R.E. (Fig. 12), the potentiometer being set during the calibration to give a convenient sensitivity value and to a value such that the limit of sensitivity of 1 mm was approximately that of the random fluctuation of the V.R.E. All range changes were then performed at the V.R.E. input.

Many difficulties were encountered at first, most of which were resolved when it was discovered that the wall mains socket being used was, in fact, not connected to the mains supply but to the constant voltage supply (a difference of 20 v), and also when the co-axial cable being used was replaced by a type with more efficient shielding. Another V.R.E., an E. K. Cole model became available during these difficulties. It was supplied in two separate parts, an input head unit, which it was possible to fit directly on to the

upper brass plate, and an indicating unit. Because of the shorter input cable necessary, the input capacity would have been lower, and so the new model would have been more sensitive. However, movement of the suspension rod caused large spurious deflections, even when the very short input cable was replaced by a wire in a solid metal shield to eliminate the solid insulation. It was decided that the movement must have been causing spurious charges in the head unit despite its anti-vibrational mountings.

The less sensitive Vibron Electrometer was therefore used.

3.11 Calibration of the Vibrating Reed Electrometer (V.R.E.) and Fluxmeter

The V.R.E. and fluxmeter were calibrated connected together as they were to be used. The Faraday cage was replaced by a variable boxed condenser. A known voltage was then applied to one side of the condenser and the other side was connected to the V.R.E. input (Fig. 13). The known charge produced was then correlated with the deflections on the fluxmeter. This was performed for the 10 mV, 30 mV and the 100 mV ranges of the V.R.E. input.

The voltage was obtained from a potential dividing circuit. If the current is constant the voltage applied is proportional to 'R'. The current was kept constant by measuring the voltage 'V' across a 5000 Ω resistance and keeping this constant by varying 'P' if necessary. The voltage 'V' was measured with a direct reading potentiometer. The switch used was an ordinary morse key which

was found to produce less spurious charge than any other tried, this charge producing less than 1 mm deflection on the fluxmeter. All components were completely screened by using earthed metal boxes and co-axial cables.

To obtain the correct value of the capacity used, the stray capacities were found by varying the capacity 'C' and keeping the voltage constant. From the graphs obtained the stray capacity was found to be only very small and to be in series with the main capacity.

Thereafter the capacity 'C' was kept constant and the resistance 'R' was used to vary the charge applied to the V.R.E. A graph of the resistance against the fluxmeter deflections was plotted for the three input ranges (Fig. 14). The formula used to evaluate the charge then was:-

$$Q = \frac{R \times C \times C^{o} \times V}{(C + C^{o}) \quad 5000} \times 3 \times 10^{-3} \text{ e.s.u.}$$

where:- 'Q' Charge in e.s.u.,

'V' Voltage measured by the direct reading potentiometer,

'R' Resistance across which the condenser was connected,

'C' Box capacity in μF,

'C^{o}' Stray capacity in μF.

The percentage of the charge lost by the Faraday cage, due to it having a broken surface, was found by lowering a fibre carrying a charge first into the Faraday cage and then into a Faraday cage having a continuous surface, except for the small hole where the fibre was inserted. The fluxmeter deflections for the two cages were then plotted on a graph and it was found that only 1% of the charge was lost.

The calibration curves (Fig. 14) gave values of sensitivities of:-

10 mV range $(0.050 \pm 0.002) \times 10^{-3}$ e.s.u./mm;

30 mV range $(0.148 \pm 0.007) \times 10^{-3}$ e.s.u./mm;

100 mV range $(0.71 \pm 0.03) \times 10^{-3}$ e.s.u./mm;

when the stray capacity and the percentage lost by the Faraday cage where taken into account.

3.12 Visual Observations

The drops were originally viewed through an ordinary telescope, the magnification of which was kept constant, focusing being achieved by horizontal movement. It was held in a special fitting made to slide easily on a retort stand. The height of the object from the base of the chamber could be read directly from a cut metre ruler, which was also clamped to the retort stand.

The chamber was illuminated from the back and a square water bath was used as a heat filter. This method of viewing worked fairly well except for the rather inferior definition and the occasional difficulty of moving the telescope accurately.

In May 1962 a more powerful stereoscopic telescope became available and a special stand was constructed for it, for horizontal viewing. Vertical movement was achieved by two side screws and horizontal movement by its own ratchet. The magnification could be altered at the turn of a knob without altering the focus.

Using an eyepiece of x 20 and an objective of x 0.5 the working clearance was 20 cm and magnifications of x 5, x 8, x 14,

x 25 and x 40 could be obtained. For the higher magnifications, however, a brighter light was required. The limit of measurement was 10^{-3} mm corresponding to half a division on the highest magnification.

A 35 mm camera attached to one eyepiece of the telescope was used to photograph the drops. Great difficulty was however experienced in focusing and the photographs produced were with few exceptions of poor quality.

3.13 The Tape Recorder

In order that the drop might be viewed continuously a tape recorder was used to record comments on the growth, size and appearance of the drop. This proved a highly successful arrangement for recording as on the play-back the exact sequence and times of events could be written down more fully than would otherwise have been possible. It also proved useful for recording the comments of any visitor while viewing a drop, as the similes invented by a person who is viewing a phenomenon for the first time are sometimes entirely different from those of a person who has seen the identical phenomenon some hundreds of times.

Photographs of the apparatus can be seen in figures 3, 15 and 16 at various stages of being dismantled, while figure 17 was taken before the perspex windows were added.

CHAPTER 4

Experimental Procedure

In order that the results from the experiments on the freezing and breaking of water droplets could be compared with the results of Evans and Hutchinson and with Maybank and Mason an experimental procedure as similar as possible to theirs was adopted. All the drops in the series were also delt with as uniformly as possible.

In essence the procedure for each drop was to suspend it on the end of a fibre, lower it to the nucleation isotherm of $-1^{\circ}C$, nucleate it with small ice crystals and lower it to the freezing environment where the process of freezing and splintering was watched.

When the chamber had attained steady conditions the positions of the desired nucleation and freezing isotherms were located, using the thermo-couples. The telescope was then set to the freezing isotherm level.

The details of the procedure were as follows. The suspension tube and rod were raised above the upper brass plate (Fig. 15) and a drop was placed on the end of the fibre with a piece of stranded wire. Any spurious charge was then removed by placing a small radio-active source near to the entrance of the chamber while the drop was lowered past it. The suspension rod was gently lowered until the drop was near the nucleation isotherm and then the suspension tube was lowered so that it formed a complete earthed shield around the fibre. Finally the radio-active source was removed and the drop

was raised into the Faraday cage to make sure that all the charge had been removed and then lowered to the nucleation isotherm where it was left for about a minute for it to take up the temperature of its surroundings.

A small piece of cardice was crushed into fine crystals and then a few of these were dropped down the nucleation tube. The suspension rod was then quickly lowered so that the drop passed through the shower of ice crystals formed by the cardice in the chamber, and reached the freezing isotherm. The suspension rod was then clamped in position so that the drop could not move vertically again. After some practice the whole process of nucleation took less than one second.

The drop was then watched whilst the freezing and any subsequent breaking took place.

If no break occurred after the drop appeared to have frozen completely then it was raised into the Faraday cage to see if there was any charge on it, melted and used again.

If the drop broke it was allowed to settle after swinging and the type of break which had occurred was recorded. The amount of charge on the remnant was measured by raising the drop into the Faraday cage. Three readings of the charge were taken and the average value calculated from them. The drop was then melted and the remnant diameter was measured. When the charge had been removed with the radio-active source the drop was used again, a record being

kept as to whether the drop was a new or an old one.

The following information was obtained for all drops studied:- nucleation temperature; freezing temperature; diameter; freezing appearance; size of spicules and their rate of growth. If the drop broke then the following information was also noted:- time and type of break; amount of swing of remnant; remnant charge and diameter.

4.1 Precautions taken

As the drops which broke were to be classified later into those which broke when the spicule was partially liquid and those which broke when the spicule had frozen completely, this factor was determined before the charge measurements were taken so that the results would not be influenced by the former results of other workers.

It was realised that there could be several causes of spurious charges becoming attached to the drop and so in the first drops which were studied these were checked.

As the temperature at the level of the Faraday cage was controlled by the room temperature, it was sometimes above the $0^{\circ}C$ isotherm and thus any charge produced by the melting of the drop by the Dinger and Gunn process would have been detected also. Therefore when the Faraday cage was below the $0^{\circ}C$ isotherm, a drop which had frozen and not broken and had no charge on it was raised above the Faraday cage and allowed to melt. The drop was then lowered back into the Faraday cage. No charge was detected on these drops. If the charge associated with the melting of ice reported to occur by Dinger and Gunn had been produced then for the

average drop of 1.5 mm diameter a charge of 1.7×10^{-3} e.s.u. should have been detected.

Spare ice crystals, produced by the cardice crystals, bombarded the freezing water drop and the fibre but these were found to produce no spurious charge. Maybank had found them to produce spurious charge when he used a glass fibre but the coating of paraffin wax which was given to the glass in this case seemed to eliminate them.

To check that the charge was not due to the movement of the fibre and the ice at any time, the fibre was swung violently from the top on several occasions but no charge was ever detected. This was done when there was no drop on the fibre, when there was a newly discharged drop on it and when there was a discharged ice sphere on it. If the fibre hit the Faraday cage then charge was detected near the top of the fibre but this was easily separated from the charge on the drop.

During the start of the experiments some of the drops fell off the fibre accidentally. When a water drop fell off only small charges of the order of 10^{-4} e.s.u. were recorded, but when an ice sphere fell off, the charges were of the order of 10^{-2} e.s.u. In later experiments therefore no drop was accepted if the break in the drop took place near to the fibre.

CHAPTER 5

Freezing Appearance

Several photographs of the drops at various stages during freezing were taken and these accompany this chapter. Under normal circumstances the drops were nucleated at a temperature of $-1^{\circ}C$ and frozen in an environment at $-15^{\circ}C$. Occasionally however the drop was not nucleated by the ice crystals and remained liquid at temperatures of $-15^{\circ}C$ for about twenty seconds after it had been lowered. Such drops were regarded as having been nucleated at $-15^{\circ}C$ and were not included in the main classification; they are considered at the end of this chapter.

The first sign that freezing was taking place was the spreading of a thin ice film over the surface of the drop. The ice shell was then seen to grow thicker and small irregularities to appear on the surface. These surface irregularities were presumably formed by water seeping out through cracks and freezing on the surface.

After this initial formation of the shell further freezing took place in two distinct ways but before considering them several general comments on the appearance of the drops which held for both types of freezing can be made.

In all the drops studied some form of bulge or spicule was formed. The external feature always had an oval cross section which is consistent with its being formed by the water being forced out of a crack or fissure. The spicule or bulge was always formed

in the lower hemisphere of the drop. Quite frequently two or three spicules started to grow but only one of these grew very large, the position of the other smaller spicules seemed to govern the method of cracking of the drop if any occurred (5.4).

5.1 Smooth Spicule Growth

Most frequently once the spicule had started to grow it did so in a smooth even way, like that on the drop in figure 18. The water on expanding in the centre of the drop forced out a small bulge, the outside of this froze immediately and a small ice tube was formed along which further water was pushed when more of the liquid interior froze. The spicule grew in this way for some seconds and then small bubbles appeared in the spicule. These bubbles were formed at the ice-water boundary and were then liberated into the liquid core where they travelled either back up into the drop or to the end of the spicule, where they forced out small extensions at the end of the spicule. Two such extensions can be seen in the figure. During the formation of the spicule, thickening of the ice shell had been taking place. When the spicule had finished growing and the expanding water had no longer this easy method of escape, a series of cracks spread through the drop and the central water froze quickly. Any break which occurred in the drop did so either shortly after the formation of the spicule or while the cracks were spreading through the drop centre. The former were usually the spicule type of break and the latter the central type.

5.2 Bubbly Spicule Growth

The bubbly type of spicule grew initially in a very similar way to the smooth spicule but at the same time that the spicule started to grow a number of small bubbles was released at the ice-water boundary in the main drop (Fig. 19a). These bubbles were forced inwards by the advancing ice until they met in the centre. At the same time bubbles were being liberated in the growing spicule. The formation of this type of spicule seemed to be both by water from the centre pushing out into the spicule and by the bubbles formed in the centre and in the spicule pushing the spicule out. The outline of the spicule was noticeably much more irregular than in the smooth type of growth. After the growth of the spicule very little of the drop remained liquid and no cracking of the drop took place. This was probably because the air bubbles could be compressed to allow the little remaining liquid to freeze; this also probably accounted for the fact that very few of the drops which grew in this way broke.

The growth of this type of spicule started much later than that of those which grew in a smooth way, and the process of spicule formation also took longer.

Those drops which formed only a large bulge were also included in this category (Fig. 20), for the bulge appeared to be pushed out purely by the bubbles. The inward movement of the bubbles can be seen quite clearly in this series of photographs, although for the bubbles to be confined to one sector, as in the figure, was unusual.

The movement of the bubbles usually took place along one radial plane of the drop, advancing to the centre and so forming a daisy flower shaped pattern (Fig. 21a). When viewed parallel to the plane the bubbles could be seen to be confined to the one diameter. In figure 21b two planes of bubble movement are seen, one towards the main bulge and the other to a smaller subsidiary bulge. Figure 21c shows both the flower pattern and the bubbles in the spicule which started at the side of the spicule and then proceded inwards to the centre of the spicule and out towards the end of it.

5.3 Classification of Breaks

The types of break which occurred in the drops are shown diagrammatically in figure 22. To make comparison with Evans and Hutchinson's results easier, they have been classified into the same groups. On breaking, the drop remnant was usually displaced, sometimes so violently that it passed out of the field of view of the telescope. The violent nature of breaking made it difficult to see very clearly anything which actually occurred at the time. The type of break could be seen as soon as the remnant settled down again and the distance of the swing could be roughly estimated. Sometimes the swing was straight but occasionally the force of the breaking tended to twist the drop, indicating that the piece given off had been projected tangentially. Sometimes the swing of the remnant was the only indication that any part of the drop had broken and it was only by turning the remnant round that the cause of the

movement was found. The frequency of occurrence of the different types of break are given in table 2.

5.4 Central Breaks

The more violent swings of the fibre were associated with drops which had broken through the centre, during the final stages of freezing of the drop, when a series of cracks was spreading through the centre. No two drops ever broke in the same way. The break was usually along the weakest line in the drop, the position of this line governing the exact nature of the break (Fig. 22).

In the first stages of the growth of the ice shell it was normal for at least two spicules to start growing, the one which continued to grow being that at the weakest point in the shell. When the major spicule had grown at this point however the shell was strengthened by the spicule and the weakest point in the thickening shell was then at one of the subsidiary spicules. When a break occurred through the centre of the drop it usually took place along the axis of the smaller spicules. The position of the small spicules, therefore, tended to determine the line of break. Most frequently the part ejected contained the major spicule. If the weakest point in the ice shell was still at the major spicule then the drop tended to split in two with part of the spicule being given off and part remaining on the remnant. The break usually went near to the central core of the drop.

The amount of the drop given off in this type of break varied between twenty and eighty per cent by weight. This class could not

therefore be considered to be composed of major residues only.

5.5 Spicule Breaks

Breaks which occurred across the spicule only were twice as numerous as the central type of breaks. Although the break did sometimes cause the fibre to move violently, usually the amount of movement was not such as to cause the drop to go out of the field of view of the telescope.

The breaks which involved only the spicule fell into two classes, those which occurred while the interior of the spicule was still liquid and those which occurred when complete freezing of the spicule had taken place. Two factors were taken into account when judging whether the spicule was only partially or completely frozen; firstly its appearance and, secondly, the time interval between the completion of the growth of the spicule and the time of the break.

The break was most frequently perpendicular to the long axis of the spicule, but sometimes it was parallel to it (Fig. 22). When the break was perpendicular to the axis it was usually either near to the base of the spicule or at one of the small extensions mentioned earlier (5.1).

Photographs showing some typical spicule breaks are given in figure 23, where b) had a solid spicule on breaking and a), c) and d) a partially liquid spicule.

5.6 Bubble Breaks

When the bubbles formed in the spicule during its growth were

pushed out to the end of the spicule then the force was sometimes great enough to thrust them out of the end of the spicule, leaving the end with the characteristic shape shown in figure 24. There was little or no movement associated with this type of break and the statistics for them are therefore far from complete, for if the drop was in a position perpendicular to that shown in the photograph then the break was never noticed.

5.7 Cracked Drops

When the final cracks were spreading through the centre of the drops it often occurred that one of these cracks was more prominent than the others and it opened out. It seemed probable that if the drop had been freely suspended then it would have been forced into two, for the crack frequently went up to the fibre. Two photographs of this type of drop are given in figure 25. The second of these also shows clearly the method in which some of the central type of breaks occurred through the minor of the two spicules.

5.8 Discussion of the Types of Freezing which Occurred

A total of 633 drops were studied where the nucleation and the freezing temperatures were $-1^{\circ}C$ and $-15^{\circ}C$ respectively. The distribution of these within the different types of freezing described is given in table 1, while table 2 gives the frequency of occurrence of the different classes of breaks, as shown diagrammatically in figure 22.

The most notable feature in table 1 is the frequency with which the different types of freezing did occur and the frequency with which

the different types of growth produced breaks. It is evident that if all the drops had grown in a smooth even way then the number of breaks which occurred would have been greatly increased.

All the drops were treated as nearly identically as possible at all stages in nucleating and freezing them, and the reason for the different behaviour of the drops on freezing was finally attributed to idiosyncracies of the operator. There were several sessions of experimenting when the drops nearly always grew in a bubbly manner. These included such times as when photographs of the drops were being taken and when other people were observing the drops whilst the author was operating the lowering mechanisms. It was also noticed that the proportion of drops which froze in the two ways varied from day to day and also that the proportion would vary from the morning to the afternoon. This pointed to the possibility that as all the drops were treated in as nearly similarly a way as possible, the governing factor was the facility of the operator for each drop. It is obvious that at such times as mentioned above, the process of lowering the drop from the nucleating position to the freezing position would take longer, and it is reasonable to assume that for varying reasons the reactions of the operator either at different times on the same day or on different days would change.

Other factors which might have caused the different types of growth appeared to have no effect. A check was kept on whether the drop being studied had been used previously and if so the type of freezing which had occurred at each successive refreezing. The

former behaviour of the drop did not however affect the freezing of the drop. It was quite common for the same drop to freeze in any of the ways on successive freezing. This ruled out the possibility of such factors as impurities, temperature and suspension, affecting the drop.

The only other possible difference in the freezing of the drops lies in the nucleation process. It is conceivable that the drops would freeze differently if the quantity of nucleating ice crystals colliding with them varied greatly.

The freezing of supercooled water was investigated by Blanchard (1957) who described the type of freezing for nucleation temperatures of above and below $-5^{\circ}C$. If the drops were nucleated at temperatures above $-5^{\circ}C$ then a clear type of freezing was experienced as the air, released on freezing, had time to escape, below $-5^{\circ}C$ opaque freezing took place.

When a supercooled water drop is nucleated at a temperature of $-1^{\circ}C$ immediately a shell of ice is formed which is transparent, for the air, which is released due to its lower solubility in ice compared to that in water, escapes to the atmosphere. If the drop is lowered quickly to an environment at $-15^{\circ}C$ further freezing of the drop will take place at a rate dependent upon the rate at which the latent heat can be dissipated. If, however, the speed with which the drop is lowered is altered, the rate of freezing will be non-uniform in the second stages of the formation of the shell. The speed of lowering

the partially frozen drop could then govern the behaviour of the air released by the further freezing.

The variations in the frequency with which the different types of growth produced breaks has already been mentioned. When bubbly types of growth occurred, the stresses set up by the expansion of the water on freezing could be accommodated by the compression of the air bubbles, but when a clear type of freezing took place part of the ice shell or spicule was forced off. It follows also that with the increase of bubble activity in the bubbly types of growth there will be an increase in the number of bubble breaks occurring.

5.9 Nucleation at -15°C

When the drop remained liquid for several seconds before freezing at the -15°C isotherm, it was regarded as having been nucleated at this temperature. The type of freezing which took place was entirely different from that which occurred if the drop was nucleated at -1°C. The drop remained liquid for some twenty seconds and then suddenly turned opaque. Further freezing of the ice shell took place slowly and small bulges appeared on the surface. One of these grew slowly, then broke, and water was seen to spread over the surface. Another bulge then started to grow either at the same place or very close to it. The whole process could be repeated several times before the drop was frozen through. Because of the opacity of the ice it was impossible to see any of the interior of the drop. As freezing of the main drop took place, the whole of it went gradually darker. No extensive cracking of the drop took place but on several occasions a small crack

grew gradually larger until it opened out. Figure 26a shows a drop which was nucleated and frozen at $-15^{\circ}C$. The bulge is situated in the upper hemisphere of the drop which was usual in drops nucleated at this temperature.

Only rarely did drops nucleated at $-15^{\circ}C$ form spicules of any size but when they did, the spicule grew from the same place as the initial bulge, which was formed as above. They then grew in a smooth, even way, turning downwards under the influence of gravity. These spicules, like those which grew on drops nucleated at $-1^{\circ}C$, always had an oval cross section. The drop in figure 26b clearly shows this, for the second of these two photographs of the same drop was taken after the drop had been rotated through ninety degrees.

Only one of the drops frozen in this way broke and this did so through the centre. The charge left on the drop residue was $+1.95 \times 10^{-3}$ e.s.u. but all that can be said is that this is well within the range for the drops nucleated at $-1^{\circ}C$ (Chapter 6).

Thirty-one drops were nucleated at $-15^{\circ}C$ and of these twenty-six formed bulges only, four grew long spicules and one broke.

The different behaviour of the drops when nucleated at $-15^{\circ}C$ can be seen to agree with the work of Blanchard (1957) who found that when supercooled water drops were nucleated at temperatures above $-5^{\circ}C$ a clear form of ice was produced but that at temperatures below this an opaque type of ice, in which innumerable air bubbles were trapped, was formed. The very much lower frequency of breaking at these temperatures can be attributed to the spongy nature of the ice shell, in which the

air bubbles will be compressed by the expanding interior rather than the shell being ruptured.

Incidentally the drops used to illustrate this section also illustrate the difference between a good and a bad type of suspension. The first drop (Fig. 26a) is situated on the very tip of the fibre, whereas the second one (Fig. 26b) is situated around the end of the fibre. The first type of suspension was always used where possible and on only a few occasions was the drop found to be suspended in the second manner. Bad suspension had two consequences; firstly it tended to bind the drop together so that any cracks which might have caused a break to occur were not effective and secondly if any break did occur it was likely to be near to the fibre and so affect the charge (4.1).

CHAPTER 6

Charging of the Drop Residues

The charges left on the drop residues have been classified in two distinct ways, firstly in the manner adopted by Mason and Maybank (1960) and secondly after Evans and Hutchinson (1963). The drops considered in this chapter all belong to the main class of drops studied, which had diameters from 0.9 to 1.7 mm with an average diameter of 1.3 mm, and which were nucleated at $-1^{\circ}C$ and frozen in an environment at $-15^{\circ}C$. The charge on the only drop which broke after being nucleated at $-15^{\circ}C$ has already been discussed. (5.9)

6.1 Major-Minor Drop Residue Classification

When the drop residues were separated into major and minor ones by mass, as was done by Mason and Maybank, then the charges on them were found to be distributed as shown in table 3. Of the major residues, 47 had a positive charge, 32 a negative charge and 27 had no detectable charge. These last 27 all belonged to the bubble break class. The average for the 106 residues was $+0.45 \times 10^{-3}$ e.s.u.

The charges on the twelve minor residues were equally divided between positive and negative and the charges themselves were of a comparable magnitude, the average being -0.59×10^{-3} e.s.u.

The average for the 118 residues was $+0.34 \times 10^{-3}$ e.s.u. The discrepancies with the results of Mason and Maybank are shown in table 4, where the results obtained by them and by the author are tabulated in a comparable form.

6.2 Type of Break Classification

When the drops were classified into those which broke in the

different ways described in chapter 5 then the charges on the residues were distributed as in table 5.

When a central break had occurred, the residues were found to be equally divided into positive and negative charged residues, with averages for both of the same magnitude.

When the drop had broken across the spicule whilst it was still partially liquid, the majority of the residues had a negative charge, the average of which was -3.15×10^{-3} e.s.u. The average value for those which had a positive charge was only $+1.35 \times 10^{-3}$ e.s.u.

Conversely when the drop broke across the spicule when it had completely frozen, the residues were predominantly positively charged, and the respective averages were +5.10 and -1.27×10^{-3} e.s.u.

On those occasions when the drop break was of a bubbly form, the charges on the residues were noticeably less than in the other classes, and this class of drops had been found previously to have no detectable charge on them. Nineteen of the drops had a positive charge left on the residue with an average of $+0.12 \times 10^{-3}$ e.s.u. and seven had a negative charge with an average value of -0.09×10^{-3} e.s.u. Twenty-seven drops in this class had no detectable charge but were included when the average of $+0.03 \times 10^{-3}$ e.s.u. for the group was being calculated.

The average for the 118 drop residues was $+0.34 \times 10^{-3}$ e.s.u.

These results compare very closely with the results of Evans and Hutchinson, who also found that central breaks left residues of both signs, that liquid spicule breaks left predominantly negatively

charged residues, and that solid spicule breaks left residues which had a predominantly positive charge.

All the drops which broke in some way produced some charge, except for some of the bubble type of break. None of the drops which remained whole had any charge on them, nor was any ever detected on those drops which cracked but did not separate. It is possible that some form of minute splintering of the shell sometimes took place, however, without being noticed, and that a charge of less than 0.05×10^{-3} e.s.u., the limit of sensitivity of the Vibrating Reed Electrometer, was present on the 515 drops which did not break. The average for the 633 drops which were studied then becomes $+0.065 \times 10^{-3}$ e.s.u. if an average value of zero is taken for the 515 drops which did not break: $+0.08 \times 10^{-3}$ e.s.u. if an average of $+0.02 \times 10^{-3}$ e.s.u. is taken: and $+0.05 \times 10^{-3}$ e.s.u. if an average of -0.02×10^{-3} e.s.u. is taken for those drops not noticeably breaking.

Discussion

6.3 Kachurin and Bekraiev

When similar work to that described was done in Russia and the charge on the drop residues was recorded on an oscilloscope, a small charge of the order of $+1.5 \times 10^{-3}$ e.s.u. was recorded at the start of the break, corresponding to negative particles of ice being given off. Later both negative and positive pulses were recorded when water or ice respectively were given off. The residue charge was the algebraic sum.

It seemed probable then that at the time of the formation of the

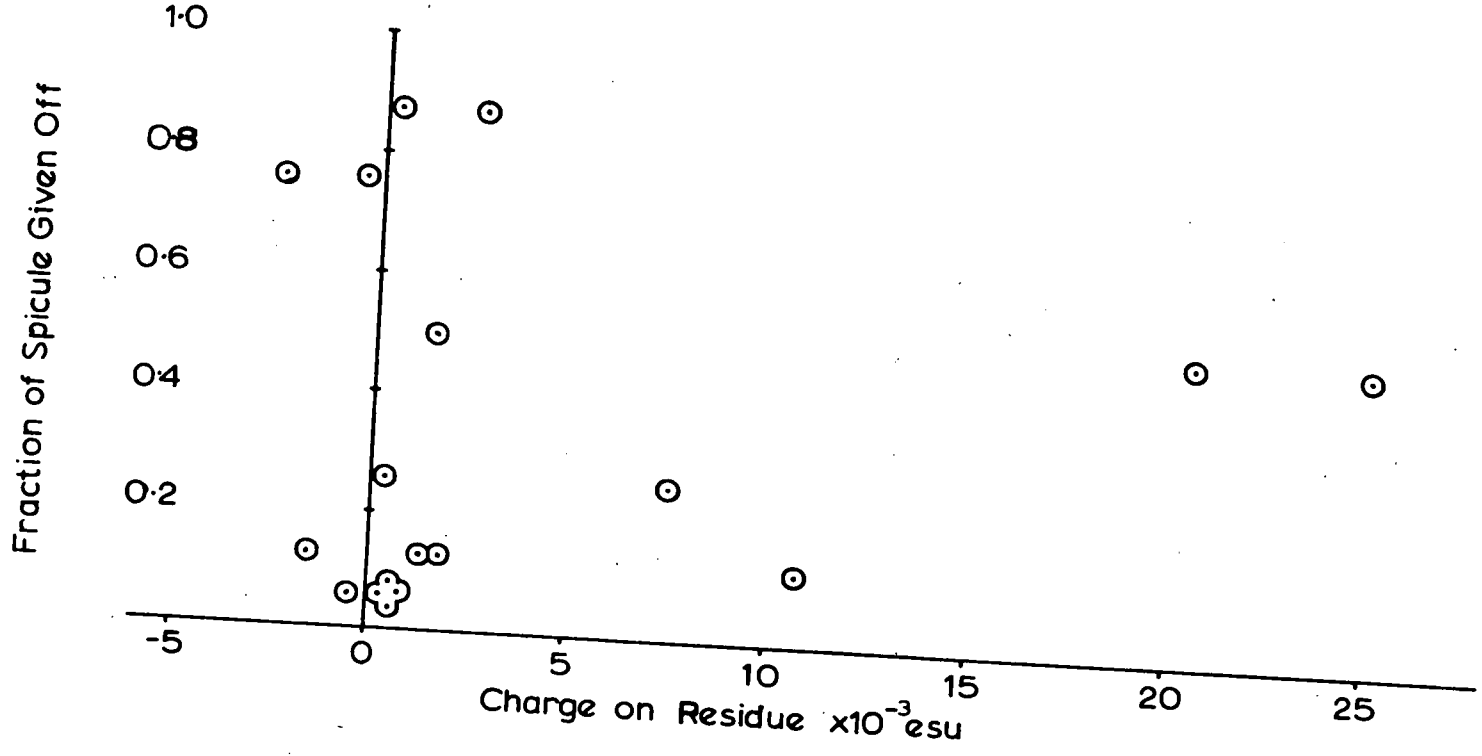

Fig 27 Residue Charge - Fraction of Solid Spicule Given Off

open cracks in the drops studied here (5.7), a positive charge should have been left on the drop. However no charge was ever recorded on them and any charge was certainly less than the limit of sensitivity of the V.R.E.

It is essential to note here however that despite this difference in the results, the Russian work does suggest that the water in the drop acquires a positive charge and the ice a negative charge, as also does the present work (6.4).

6.4 Workman and Reynolds

The potential difference set up across the liquid-solid boundary during the freezing of dilute aqueous solutions was investigated initially by Workman and Reynolds in America. They found that the water usually acquired a positive charge and the ice a negative one. A value of 10^{+4} e.s.u. per cc of water frozen can be taken for the charge separated (2.1).

In the present work, when solid spicule breaks occurred, as described (5.5), the part ejected would be composed almost entirely of ice, thus leaving the drop residue with a positive charge, if the Workman and Reynolds effect is in operation. This was found to be the case in fourteen of the nineteen drops so breaking. Those drops which were left with a negative charge had a noticeably smaller value than the positive ones.

Because of the nature of the freezing, the base of the spicule is more likely to be connected to the liquid core of the main drop by small channels and cracks, and therefore the nearer the break is to

the base of the spicule, the more likely is water to be given off with the solid ice ejected. In this class then the magnitude of the charge on the residue will be governed by the different quantities of ice and water given off. The amount of ice ejected will increase as more of the spicule is ejected thus greater positive charging is expected. As the break approaches the base of the spicule more water is also expected to be expelled and so the positive charging will be reduced and if sufficient water is expelled the charge could be reversed.

As any exact measurements of the particles ejected were impossible, the amount of ice given off was estimated from the position of the break along the spicule, all the spicules having approximately the same cross section and length. The graph of the charge left on the residues against the fraction of the spicule ejected (Fig. 27) shows that the charge did attain its maximum positive value when approximately half of the spicule had been ejected. The graph is however far from conclusive both because of the errors involved and because of the limited number of results for the class.

If Workman and Reynolds value of charge separated across a liquid-solid boundary of 10^4 e.s.u. per cc of water frozen is assumed, then some calculations on the expected charge separation in a drop can be made. The dimensions used in the following calculations have been taken from the drop photographs which accompany Chapter 5.

Volume of shell $V = \frac{4}{3} \pi (R^3 - r^3)$ cc

Charge situated in shell $Q = V \times 10^4$ e.s.u.

Volume of spicule $Vs = \pi^a \times {}^b \times {}^h$ cc

Charge situated in spicule $q = Vs \times 10^4$ e.s.u.

When the following values are used:-

		Average	Minimum
R	Radius of drop	0.065 cm	0.045 cm
r	Internal radius of shell	0.025 cm	0.035 cm
a	Major cross sectional semi-axis of spicule	0.008 cm	0.006 cm
b	Minor cross sectional semi-axis of spicule	0.004 cm	0.003 cm
h	Length of spicule	0.15 cm	0.06 cm

then

Average	Minimum
$Q = 10$ e.s.u.	$Q = 2.0$ e.s.u.
$q = 150 \times 10^{-3}$ e.s.u.	$q = 34 \times 10^{-3}$ e.s.u.

If, therefore, only half of a solid spicule is ejected, a maximum charge of 75×10^{-3} e.s.u. can be expected to be left on the residue. It can be seen that all the charges on the solid spicule break residues fall well within this range. If the charge values are calculated with the minimum values for the parameters then the charge separated is still sufficient to account for all but the two highest charges recorded.

When the break occurs across a spicule which is still partially liquid, both water and ice are given off, the quantities of each depending on the exact constitution of the drop at the time of the break. The amount of ice will be the thin sheath of ice, whilst the

amount of water will depend on the excess pressure in the interior. It is therefore difficult to make any calculations on the charge which might be produced under such circumstances. However, the amount of ice given off cannot exceed that of the sheath and the quantity of positive charge produced cannot be greater than the charge situated within it. If a shell thickness of only 0.001 cm is taken and the average spicule dimensions already given are used then the charge situated in the sheath can be calculated to be 50×10^{-3} e.s.u. The quantity of water given off is not controlled and it is expected that in most cases, because of the high pressures built up, more water than ice will be ejected, and the net charge on the drop residue is then expected to be negative.

The results for this class did in fact show that of the twenty-nine drops 65% had a negative charge, and that the average positive value was substantially less than the negative one.

When the break occurs across the centre of the drop, it is even more difficult to make any calculations of the expected charge. In such cases the freezing had usually proceeded further than in the spicule type of break, but the fracture usually went near to or through the liquid core of the drop. The charges are therefore expected to be approximately equally divided between positive and negative. The magnitude of the charge is only limited by the total charge separated, which is 10 e.s.u. for average and 2 e.s.u. for minimum values.

From the results it can be seen that these expectations are not exceeded.

In the case of the bubble type of break the only substances which

appeared to be ejected were the thin ice cap and the air bubbles themselves. The charge left on the drop residue will then, if the ice is assumed to be negative, be predominantly positive, but if, however, a small quantity of water escapes, the charge sign will be reversed. In either case the magnitude of the charge will be very small.

The volume of ice in the cap is only of the order of 10^{-7} cc, taking the dimensions of the drop in figure 24 and the corresponding charge separated is then 10^{-3} e.s.u.

The drop residue charges for this class are predominantly positive and all have a very small magnitude.

The main difficulty in trying to apply the Workman and Reynolds effect quantitively to the results lies in finding the correct value of charge separation to be used, as the dissolved impurities in the water they used had a great effect on the charge separated. If a value of charge separated as low as 2×10^{3} e.s.u. were used instead of the value 10^{4} e.s.u. then all the observed charges could still be accounted for. From a qualitative point of view the only impurities which affected the sign of the separated chargewereammonium ones.

Both from considerations of the sign and magnitude of the charges attached to the drop residues in the various classes the results concur with considerable accuracy with the Workman and Reynolds effect.

When the results as a whole are considered, the average value for all the drops of $+0.34 \times 10^{-3}$ e.s.u. is of both the correct sign and magnitude for this effect also, for in the majority of cases ice only or mainly ice will be ejected from the drops.

6.5 Latham and Mason

The Latham and Mason theory of charge separation within ice having a temperature gradient across it, whereby the cold end of the ice attains a positive charge of $5 \times 10^{-5} \frac{dT}{dX}$ e.s.u. cm^{-2} has already been mentioned (2.4). This charge separation value is inapplicable to the charge produced when supercooled water drops freeze and break for two major reasons. The first has been mentioned previously (2.5) and is due to the increase of charge separated when the warm end of the ice nears $0^{\circ}C$, the second factor which tends to increase the charge separated is the non-uniform nature of the ice when it is grown in this manner.

When the warm end of the ice is at a temperature of $-0.5^{\circ}C$ and the cold end is at $-15^{\circ}C$ then the charge separated becomes 9.5×10^{-5} $\frac{dT}{dX}$ e.s.u. cm^{-2} (2.5 Figure 2).

Brook (1958) found that when the ice had irregularities in it then the charge separation could be increased by a further factor of two. The surface charge density then becomes approximately 2×10^{-4} $\frac{dT}{dX}$ e.s.u. cm^{-2}.

Taking this value for the charge separated and using typical values for the dimensions of the drops then the surface charge on a drop can be calculated, in a similar way to that of Evans and Hutchinson (1963). Because Evans and Hutchinson found that the Latham and Mason value for the charge separated could not account for the charge left on their residues, in the following calculations the values tending to give the highest charge separation have been used when estimating the average

values and the values used for the maximum values have been increased to within the limits of being only just probable. Taking, for instance, the values used for the thickness of the ice shell, figure 18a shows that when the spicule starts to grow the ice shell of the main part of the drop has a thickness of 0.03 cm; this value has been taken for the thickness at the time of break, although breaks never occurred so soon in the growth of the drop. The maximum value of 0.01 cm for the thickness is obviously much smaller than met with in practice.

Charge density	$q = 2 \times 10^{-4} \frac{dT}{dX} A$ e.s.u.
Area half way through drop shell	$A_d = 4\pi \frac{(R + r)}{2}^2$ cm^2
Area half way through spicule shell	$A_s = 2\pi \frac{(a + b - t)}{2} h$ cm^2
Charge separated in main drop	$q_d = \frac{2\ dT}{R - r} A_d 10^{-4}$ e.s.u.
Charge separated in spicule	$q_s = \frac{2\ dT}{t} A_s 10^{-4}$ e.s.u.

When the following values are used:-

		Average	Maximum
R	Radius of drop	0.065 cm	0.085 cm
r	Internal radius of shell	0.035 cm	0.075 cm
a	A Major cross sectional semi-axis of spicule	0.008 cm	0.010 cm
b	Minor cross sectional semi-axis of spicule	0.004 cm	0.006 cm
t	Thickness of spicule shell	0.003 cm	0.002 cm
h	Length of spicule	0.15 cm	0.20 cm
dT	Temperature difference across shell	14.5°C	14.5°C

then:-

Average	Maximum
$q_d = 3.04 \times 10^{-3}$ e.s.u.	$q_d = 23.2 \times 10^{-3}$ e.s.u.
$q_s = 4.10 \times 10^{-3}$ e.s.u.	$q_s = 12.0 \times 10^{-3}$ e.s.u.

The total charge situated on the surface of the drops has then an average calculated value of $+7 \times 10^{-3}$ e.s.u. and a maximum calculated value of $+35 \times 10^{-3}$ e.s.u.; and the drop residue could only have this quantity of negative charge on it if the whole of the outer surface of the drop were to be stripped off at the time of the break. The charges measured on the residues were all less than the maximum calculated value but not less than the average calculated value.

Of the 17 central breaks which occurred, 7 exceeded the predicted total charge separation of 3×10^{-3} e.s.u., and of the 48 spicule breaks, 10 exceeded the predicted total charge separation within the spicule.

For the Latham and Mason theory to be accepted as the sole charging mechanism in the drops, their surface charge density (q) has to be increased to at least $q = 5 \times 10^{-4} \frac{dT}{dX}$ e.s.u. cm^{-2}. Only then could all but four of the charges on the drop residues be accounted for.

The two major factors which tend to increase the charge separation, have already been included in the calculations. A further correction due to the impurities which might be present in the water can also be added. The impurity which was found to increase the charge separation most increased it by a factor of two.

If all these corrections to the original Latham and Mason value of $q = 5 \times 10^{-5} \frac{dT}{dX}$ e.s.u. cm^{-2} for the charge separation are made and all the predicted charge on the surface of a drop is separated when a break occurs, then the magnitude of the majority of the charges can be accounted for by their theory.

When the results are examined in terms of the type of break, the charges observed on the residues and the charges expected on them by the Latham and Mason temperature gradient theory, several discrepancies appear, even if only the sign of the charge is considered.

Under the Latham and Mason theory, the major residues are predicted to have a negative charge associated with them because the exterior of the ice shell, which carries the positive charge, is being predominantly ejected. The minor residues are expected to carry charges of either sign as both the exterior and interior surfaces of the shell are being given off. Although as can be seen from table 3 the latter is true, the majority of the major residues carried a positive charge. The average for all the major residues was $+0.45 \times 10^{-3}$ e.s.u. If those drops which had only bubble breaks are excluded from the major residue class because it is conceivable that the charges there originate from some other mechanism, then there are only 28 positive residues and 25 negative ones but the average for the class then becomes $+0.85 \times 10^{-3}$ e.s.u., which is still not in complete accord with the Latham and Mason theory.

The results of Mason and Maybank's work on the charging of drop residues after shattering had taken place, are given in table 4, where

the results of the present work have also been summarised. Although the two sets of results are not strictly comparable because of the differences in temperature and diameters, the results when classified in a similar way should show some similarities. The different average values suggests that the results must be classified in some other manner.

When central breaks occur then charges of either sign are expected, dependent upon the proportions of inner and outer surfaces of the shell ejected. This is seen to be the case (Table 5).

When a liquid spicule break occurs then once again neither of the signs is expected to predominate, for again approximately equal quantities of inner and outer surfaces will be ejected, although a slight bias towards negative charging could be expected. The results once again concur with the Latham and Mason theory.

When a solid spicule breaks, part of the outer surface of the main shell is, in effect, ejected and the drop residue is therefore expected to have a negative charge associated with it. Table 5 shows clearly that in this class the drop residues were predominantly positively charged, in direct opposition to the Latham and Mason theory. Only five of the nineteen drops carried a negative charge and these were substantially smaller than the positive ones.

Again when bubble breaks occur, the charges are expected by the Latham and Mason theory to be equally distributed about zero, for similar quantities of the inner and outer surfaces of the ice are ejected, however 75% of them were positively charged.

The presence of the temperature gradient method of charge separation in ice as formulated qualitatively by Latham and Mason is well established, but it would appear from the foregoing calculations and discussion that the theory must be adjusted considerably from that given (Latham and Mason 1961a) if the charging of breaking, freezing, supercooled water drops is to be explained by it. Latham and Mason themselves mentioned the existence of a surface layer effect when the temperature of the warm end of the ice was near to $0^{o}C$ and it is probable that at such temperatures the temperature gradient effect is completely masked. When considering the electrical properties of freezing supercooled water drops it is essential to consider not only the outer ice shell, but also the inner water and the electrical effects associated with the boundary and the change of phase.

6.6 Dinger and Gunn

Although the Dinger and Gunn effect cannot be invoked to explain the charges left on all the drop residues, because it was formulated for the melting and not the freezing of ice, it is possible that the charges left on the residues after a bubble break could be associated with it, for the effect concerns the release of trapped air bubbles.

Dinger and Gunn found that the charge acquired by ice when it melted and the trapped air bubbles were released was +1.25 e.s.u. per cc. When a drop of 1.00 mm diameter is melted a charge of $+0.6 \times 10^{-3}$ e.s.u. can be expected to remain on the drop. No charge of this size was ever detected when the frozen drops were melted (4.1) but the trapped air bubbles might have been reabsorbed by the drop.

For the charges in the bubble break class to be explained in terms of the Dinger and Gunn effect it is desirable that only one tenth of the air in the drop be released; for the average charge in the class was only $+0.03 \times 10^{-3}$ e.s.u. and the maximum was $+0.35 \times 10^{-3}$ e.s.u. The charge sign and magnitude are therefore of the correct order, however Matthews and Mason (1963) found no charging of the same magnitude as Dinger and Gunn (1946).

6.7 Further Methods of Charge Separation

(a) Friction

When two parts of a drop are separated some frictional forces must be encountered. The charge thus produced can be expected to be a function of the area of the surfaces separated.

When a central break occurred the area of separation was of the order of 10^{-2} cm^2 whilst when a spicule break occurred the area was only of the order of 10^{-4} cm^2. However no marked difference in the magnitude of the charges for the different classes was evident (Table 5).

(b) Energy

The energy involved in the separation of the two parts of a drop can be estimated from the distance of swing and the mass of a drop remnant if such factors as air resistance, irregularity of shape of the remnant, and flexibility of the fibre are ignored.

The graph of the energy involved against the charge on the residue (Fig. 28) shows that the departure from zero charge tends

to be greater for the higher energies of swing. The differences of energy of swing were found to be mainly dependent upon the distance of swing, because the mass of the remnant varied little comparatively from drop to drop. The drops included in this graph were taken from all classes of breaks.

It is impossible here to state anything but the general trend, for the results are far from conclusive. The accuracy of measuring the energy involved was low, because the speed with which the breaks took place allowed only a rough estimate of the distance of swing to be made.

6.8 Conclusions

The charge left on the residues of supercooled drops which on freezing had broken was on average $+0.34 \times 10^{-3}$ e.s.u. The charges on the residues were distributed in the same way as those on the residues of Evans and Hutchinson (1963). In those cases where the break had taken place across a liquid-solid boundary the charges were mainly negative and in those where only ice was involved the charges were mainly positive.

The Latham and Mason temperature gradient theory does not seem to account for the charges on the drop residues completely, but the charges do appear to be explicable in terms of the Workman and Reynolds effect.

CHAPTER 7

Meteorological Significance

The main property of a thundercloud which must first be accounted for by any theory is its polarity. There exists an upper positive charge region and a lower negative one. Only when this general condition has been satisfied can the other properties be discussed.

Electrical effects within a thundercloud seem to be associated with the formation of soft hail or graupel pellets. These are formed by the accretion of small water droplets on to a central core of ice, which is initially formed by the sublimation of water on to a freezing nucleus (Mason 1957). If then it is assumed that the water drops caught on the surface freeze and break in a similar manner to the individual drops studied, the net charge gained by the pellet will be the algebraic sum of all those on the charged residues.

The average charge of all the drops studied was positive. Thus if the pellets are formed as above, the average charge on them will be positive. When the two differently charged particles are then separated by gravity, a thundercloud of inverted polarity would be formed.

Latham and Mason (1961b) found that when a hail pellet was grown artificially by the accretion of water droplets the hail did in fact acquire a negative charge, which was in agreement with the work of Mason and Maybank and would give the thundercloud the correct polarity.

The most obvious difference between drops used in the individual experiments described and those found in clouds is the size. The

drops under experiment had diameters ranging from 0.9 to 1.7 mm but there was no correlation between the diameter and the charge on the residue. However, with the much smaller drops met with in a thundercloud it is most likely that smaller charges will be associated with them, especially if the Workman and Reynolds effect applies. The size of the drops had little effect on the frequency of breaking. Mason and Maybank also found that the frequency of breaking was not affected by the size but that the charge on the residues of smaller drops tended to be smaller.

In a thundercloud then, although the charges on the individual droplet residues are probably less than those found experimentally, there is no evidence to suggest that the sign will be different.

Another great difference between the conditions in the experimental chamber and the thundercloud lies in the stability of the air. In a thundercloud many updraughts exist and the air is in a turbulent state. In the experimental chamber the drops are not subject to the same conditions as prevail in the cloud and the behaviour of the droplets in the cloud are not truly represented by the experimental conditions.

In stable conditions the heat liberated by the drop when it freezes is not removed by the surrounding air as quickly as in turbulent conditions. The drop will therefore cool more slowly and small thermal currents will be established. Mason and Maybank showed that the rate of cooling of the drops had no effect on their fragmentation. The thermal currents could however have some effect on the charge production. When the drops fracture, tiny ice crystals are given off and if these

are ejected with only a small velocity then they could be caught in the thermal currents and later recollide with the drop. In the turbulent conditions of a cloud, the particles have more chance of escaping, especially as the pellets are falling all the time. The effect of the turbulence might then be to increase the charge separated, and could in a few cases reverse the sign.

A more obvious effect of the turbulence in the thundercloud would be to increase the number of drops breaking. The structure of the spicules formed on the drops is extremely delicate, as can be seen from the photographs in chapter 5, and with the smaller droplets in a cloud it will be even more so. It would be expected then that in the thundercloud many of the spicules would be broken by outside interference before the excess pressures built up are sufficient to break the shell. No such breaks occurred in the experimental chamber, as neither strong air currents or other particles were present. If this type of external breaking does occur then the percentage of drops breaking while the spicule is still partially liquid will be greatly increased, for the spicules are much weaker before freezing has been completed.

Of the 118 breaks studied, 29 were of a liquid spicule type, the average for these being -1.29×10^{-3} e.s.u. as compared with the overall average of $+0.34 \times 10^{-3}$ e.s.u. If the number of liquid spicule breaks occurring is doubled, then the overall average becomes negative.

The effect of external breaking of the water drops by the turbulent conditions and the other hail pellets could be to change the sign of the

net charge left on the hail pellets from positive to negative, which would yield a cloud of the correct polarity.

It is impossible here to make any theoretical calculations on the magnitude or rate at which charge could be separated in the thundercloud by this method, because of the great uncertainty of the extent of the effect of external breaking.

Latham and Mason (1961b) found that as the impact velocity of the water drops on to the artificial hail pellet was increased from 0 to approximately 10 m sec^{-1} the charge production increased in a negative direction but that above this velocity the charge magnitude decreased again. The latter decrease they attributed to the splashing of the drops on impact. The former increase they attributed to the water drops in a slow air stream freezing from the hail pellet surface upwards and therefore not tending to break as frequently. It is possible however that this increase could also be due in some part to the external breaking of the freezing droplets by the stronger air currents.

CHAPTER 8

Summary and Future Work

Experiments on the freezing and breaking of individual supercooled water drops have now been carried out by four independent workers and the results have in three cases proved to be substantially in agreement.

The Russian and Durham work had tended to show that only by making large assumptions could the electrification of the drop residues be explained in terms of the Latham and Mason theory (1961a) but that this electrification could be explained by the Workman and Reynolds effect (1950b). The work described has clearly confirmed this.

Because of the greater number of drops studied, the dependence of the sign of the charge on the drop residues upon the type of break which had occurred, first suggested by Evans (1962), has also been more clearly demonstrated.

The average charge on the drops which broke was $+0.34 \times 10^{-3}$ e.s.u. whilst the charges ranged from -17.10 to $+25.30 \times 10^{-3}$ e.s.u. Those drops which broke through the centre yielded almost equal numbers of positive and negative residues, those which broke across a partially frozen spicule yielded 65% negative residues whilst those which broke when the spicule was completely frozen yielded 75% positive residues. Finally, those drops from which there was ejected only the thin ice cap on the end of the spicule together with a small quantity of air yielded 75% positive residues. The charges on the first three classes of breaks were of the same order of magnitude, while those in the last

class were noticeably smaller.

If the electrification of thunderstorms is due to the freezing of many supercooled water drops on to the surface of hail pellets, and these break in a similar manner to the individual drops studied, then the net charge on the hail pellets would be positive and a cloud of the wrong polarity would result. However it is suggested that when the turbulent conditions in a thundercloud are considered more partially frozen spicule breaks are likely to occur which will increase the number of negative residues and make the net charge on the hail pellets negative.

8.1 Suggestions for Future Work

From the preceeding paragraph it seems that any future study of the freezing and breaking of supercooled droplets must be undertaken under conditions more like those prevailing within a thundercloud, the significance of work done in stable conditions being very difficult to assess in relation to turbulent conditions. Further investigation is required into the extent to which turbulence does cause external breaking of the drops to occur and the charge associated with the breaking of drops due to collisions.

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REFERENCES

BLANCHARD, D. C. 1951 A verification of the Balley-Dorsey theory of spicule formation on sleet pellets. J. Met. 8 p. 268.

BLANCHARD, D. C. 1957 Artificial Stimulation of Rain. Pergamon Press, London, p. 233.

BRADLEY, R. S. 1957 The Electrical Conductivity of Ice. Trans. Faraday Soc. 53 p, 687.

BROOK, M. 1958 Recent Advances in Thunderstorm Electricity. Pergamon Press, New York, p. 383.

BYERS, H. R. and BRAHAM, R. R. 1949 The Thunderstorm. Washington D.C. U.S. Weather Bureau.

CHALMERS, J. A. 1957 Atmospheric Electricity. Pergamon Press, London.

DINGER, J. E. and GUNN, R. 1946 Electrical effects associated with a change of state of water. Terr. Mag. Atmos. Elect. 51 p. 477.

DORSEY, N. E. 1948 The Freezing of Supercooled Water. Trans. Amer. Phillos. Soc. 38 p. 247.

EIGEN, M. and DE MAEYER, L. 1958 Self Dissociation and protonic charge transport in water and ice. Proc. Roy. Soc. A. 247 p. 505.

EVANS, D. G. 1962 The Electrification of Ice and Supercooled Water Drops. M. Sc. Thesis, Durham University.

EVANS, D. G. and HUTCHINSON, W. C. A. 1963 The Electrification of Freezing Water Drops and of Colliding Ice Particles. Quart. J. R. Met. Soc. 89 p. 370.

GRENET, G. 1947 Essai d'explicationde la charge electrique de nuages d'orages. Ann. Geophys. 3 p. 306.

HUTCHINSON, W. C. A. 1960 Ice crystal contact electrification. Quart. J. R. Met. Soc. 86 p. 406

KACHURIN, L. G. and BEKRAIEV, V. I. 1960 Investigation of the electrification of crystallising water. Dolk. Akad. Nauk. U.S.S.R. 130 p. 57.

KUETTNER, J. 1950 The electrical and meteorological conditions inside thunderclouds. J. Met. 7 p. 322

LANGHAM, E. J. and MASON, B. J. 1958 Heterogeneous and homogeneous nucleation of supercooled water. Proc. Roy. Soc. A. 247 p. 493.

LATHAM, J. 1961 Electric charge transfer in ice and its application to thunderstorm electricity. Ph. D. Thesis, London University.

LATHAM, J. and MASON, B. J. 1961a Electric charge transfer associated with temperature gradients in ice. Proc. Roy. Soc. A. 260 p. 523.

LATHAM, J. and MASON, B. J. 1961b Generation of electric charge associated with the formation of soft hail in thunderclouds. Proc. Roy. Soc. A. 260 p. 537.

MASON, B. J. 1953a A critical examination of theories of charge generation in thunderstorms. Tellus. 5 p. 446.

MASON, B. J. 1953b On the generation of charge associated with graupel formation in thunderclouds. Quart. J. R. Met. Soc. 79 p. 501.

MASON, B. J. 1957 The Physics of Clouds. Clarendon Press, Oxford.

MASON, B. J. and MAYBANK, J. 1960 The fragmentation and electrification of freezing water drops. Quart. J. R. Met. Soc. 86 p. 176.

MATTHEWS, J. B. and MASON, B. J. 1963 Electrification accompanying melting of ice and snow. Quart. J. R. Met. Soc. 89 p. 376.

MAYBANK, J. 1960 Ph. D. Thesis, London University.

SIMPSON, G. C. and ROBINSON, G. D. 1940 The distribution of electricity in thunderclouds, II. Proc. Roy. Soc. A. 177 p. 281.

WORKMAN, E.J. and HOLZER, R. E. 1942 The electrical structure of thunderstorms. N.A.C.A. Tech. Notes, No. 850.

WORKMAN, E. J. and REYNOLDS, S.E. 1948 A suggested mechanism for the generation of thunderstorm electricity. Phys. Rev. 74 p. 709.

WORKMAN, E. J. and REYNOLDS, S. E. 1950a Thunderstorm research programme at New Mexico School of Mines. Proc. Conf. Thund. Elect. Chicago, p.29.

WORKMAN, E. J. and REYNOLDS, S. E. 1950b Electrical phenomena occurring during the freezing of dilute aqueous solutions and their possible relationship to thunderstorm electricity. Phys. Rev. 78 p. 254.

WORMELL, T. W. 1939 The effects of thunderstorms and lightning discharges on the earth's electric field. Phil. Trans. Roy. Soc. A. 238 p. 249.

WORMELL, T. W. 1953 Atmospheric Electricity: some recent trends and problems. Quart. J. R. Met. Soc. 79 p.3.

VONNEGUT, B. 1955 Possible mechanism for the formation of thunderstorm electricity. Wentworth Conf. Report p. 169.

VONNEGUT, B. and MOORE, C. B. 1958 Recent Advances in Thunderstorm Electricity. Pergamon Press, New York, p. 317.