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"The Melting of Hail and Snow"

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### ABSTRACT

This thesis sets out to investigate two separate aspects of the atmospheric melting of small ice particles; namely the rates of melting and the associated electrification by the bursting of gas bubbles during the melting. The first part describes experimental tests of the heat transfer equations used to describe the melting of small ice spheres, and finds that the theory based on the assumption of spherical symmetry is in good agreement with the observed times of complete melting, over a wide range of environmental conditions. The results are then applied to the melting of small ice spheres and cones in the atmosphere, and simple equations are derived to predict the distance of fall for complete melting and the associated depth of the 'radar melting band'.

The first part concludes with an investigation into the equivalence of an evaporating ice sphere and an ice-coated thermojunction, by examining the conditions under which the surface just begins to melt in airstreams of different humidity.

The second part of the thesis investigates the generation of electric charge associated with the bursting of gas bubbles during the melting of ice. It

is found that the experimental results cannot be interpreted simply in terms of the rupture of an electric double layer during bubble bursting. In particular, the presence of  $\text{CO}_2$  in the environment leads to an unexpected increase in the charge generation. The difference in the behaviour is attributed to the presence of strong internal convective motions in the melting specimens which may control the dynamics of the bubble bursting process.

The results of the investigations are used to provide a reasonable interpretation of the several features of the pocket of positive charge found in the base of a thunderstorm.

Finally, in the Appendix, some unexplained charging effects, that do not come directly within the scope of this work, are examined; and some lines of further research are indicated.

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PUBLICATIONS

Part of the work described in this thesis will appear in the October 1966 issue of the Q.J.R.Met.Soc. under the title,

"The Melting of Small Ice Spheres and Cones."



## CHAPTER ONE

### Introduction and Review

#### 1.1 Introduction

In middle latitudes, including the British Isles,, the greater proportion of precipitation reaching the ground has existed in an ice-phase at some stage of its development in the atmosphere. Although Findeisen (1939) believed that all rain in middle latitudes composed of large drops originated as snow or hail, there have been recorded cases of rain falling from layer clouds with their tops warmer than  $0^{\circ}\text{C}$ . (Marchand 1903, Kinshott 1950, Mason and Howorth 1952). In summer cumulus clouds there is also evidence, from radar observations, of the coalescence mechanism alone operating to produce rain in the early stages of cloud growth; (Battan 1953, Feteris and Mason 1956), but in many of the cases reported by these observers the clouds subsequently reached proportions that indicated the presence of an ice-phase.

Because of the importance of the ice-phase in the initiation of precipitation by the Bergeron process, and in the development of the electrical structure of the thunderstorm (Mason 1957), much theoretical and experimental attention has been given to the formation and growth of ice crystals, hailstones and snowflakes in the atmosphere. By comparison, relatively little attention has been given to the subsequent melting of these solid hydrometeors in their passage to the ground.

The interpretation of radar echoes from precipitation, in particular the so-called "melting band", has long been based on theories of melting that have not been tested in real atmospheric conditions. Various assumptions concerning the radar reflectivities of melting ice particles have been made in calculations on the intensity distribution of the echoes from the melting band and in calculations on its depth. In particular, spherical or axial symmetry has been assumed for the melting of small, solid ice particles. That is not usually the case, as is demonstrated by the experiments performed by Mason (1956) which are mentioned below.

Since radar observations have become such powerful tools in the investigation of the atmosphere, it was initially hoped that this thesis might examine the melting of ice particles and snowflakes both visually and with radar. However, the radar facilities did not become available in time, but the visual observations and the formulation of the equations of melting actually undertaken in this work should assist in providing a more accurate basis for future interpretation of radar patterns in the melting region.

In setting up the experimental system to study melting under conditions approximating to those likely to be found in the real atmosphere, it was also found

that the apparatus was very suitable for examining a further important aspect of melting: namely the associated electrification. It has been proposed by various workers that generation of charge during melting, by the bursting of air bubbles trapped in the ice, might lead to an explanation of

i) the presence of a small pocket of positive charge below the main centre of negative charge, and below the 0°C level in thunderstorms.

ii) the presence of a negative space charge found over melting snow on the ground.

iii) the vertical, predominantly positive, electric current carried by continuous rain from layer clouds.

Whilst considerable attention has been given to the electrification associated with melting by a number of workers mentioned below, there has been little attempt to examine the phenomena under conditions approximating to those likely to be encountered in the atmosphere. Most of the experiments have been conducted with bulk ice specimens, and very low wind speeds of order a few cm per second. The apparatus constructed to examine the melting of ice particles visually and to test the heat transfer equations, was found to be suited, with slight modification, to an examination of the charging during melting under more realistic conditions.

## 1.2 The Melting Equations

In relation to the radar interpretation problem mentioned above, and in order to estimate the chances of hailstones reaching the ground, Mason (1956) calculated the rates at which small ice spheres would melt in falling through atmospheres of specified lapse rate and humidity, whilst retaining their melt water. Mason tested the equations by suspending small spheres at the interface of carbon tetrachloride and liquid paraffin at room temperature, and found results in good agreement with the theory after certain assumptions were made regarding the heat and mass transfer coefficients applicable to this case of "zero" Reynolds Number. A small degree of internal convection in the drops was reforded; and during the later stages of melting the ice core floated to the top of the drop. Since both these effects are likely to cause a significant departure from the symmetrical melting assumed for non-zero Reynolds Number it seems essential that the equations derived by Mason should be put to the test under conditions more closely resembling those likely to be found in a real atmosphere.

An attempt to relate the melting to more realistic conditions was made by Macklin (1963), who reported some measurements on large spheroids which were partly melted by exposure to a stream of warm air for 1-2 minutes in a wind tunnel, the quantity of melt water

being determined by weighing. The heat transfer as inferred from the degree of melting of spheres was in fair agreement with that given by the heat transfer equations established for the evaporation of water drops, (Ranz and Marshall, 1952), with the assumption that all the melt water was shed in the airstream. When the water is retained, as will be the case for particles whose melted diameter is less than about 5mm, the theory and experimental behaviour is more complicated.

Apart from the two experiments mentioned there seems to be little published work on the heat transfer equations associated with the melting of ice particles.

### 1.3 Electrical Effects

#### 1.3.1 Charge produced by bursting bubbles.

Interest in the electrical effects of melting ice has increased ever since Dinger and Gunn (1946) first carried out a series of laboratory experiments which suggested that charge separation took place when air bubbles trapped in the ice burst at the surface after being released from the interior during melting.

In one experiment, Dinger and Gunn measured the charge acquired by an airstream passed over melting ice contained in a nickel-plated copper tube. The system was electrically shielded and the air deionised by passage through earthed copper turnings. During the melting process, maximum charges of order 1 esu per cm<sup>3</sup>

of melt water were detected, and it was found that the air acquired a negative charge. The magnitude of the charging varied with the rate of melting and concentration of impurity in the sample. Tap water, and distilled water to which slight amounts of impurity had been added, gave no detectable electrification.

In a further experiment the charge acquired by the melt water itself was recorded. In this case the ice samples were frozen in a platinum dish by a sealed cooling coil, in an apparatus which allowed various gases to be admitted to the environment while freezing took place. After freezing, the dish was transferred to an enclosure in which the temperature was controlled and the ice allowed to melt at a known rate. The charge on the dish was recorded by an electrometer. The results were similar to those found in the case where the charge given to the airstream was recorded, except that the charge acquired by the dish was always positive. No charging was recorded when the samples were melted in an environment of  $\text{CO}_2$ . No charging was recorded for ice containing few or no bubbles. The charge generation was found to be a maximum for water of pH value about 6, and increased with decreasing conductivity.

The experiments were repeated by Matthews and Mason (1963), but they failed to detect any charges greater than  $10^{-2}$  esu per  $\text{cm}^{-3}$ , though this was probably

due to the poisoning of the specimens by the presence of strong concentrations of  $\text{CO}_2$ , (Correspondence 1964). Similar experiments have been conducted by Magono and Kikuchi (1963), McCready and Proudfit (1963) and Dinger (1965). The highest level of charging was found by Dinger (1965) and was about  $+6.0 \text{ esu/gm}$  of melt water for triply distilled water.

In all cases it has been recognised that the rate of melting, bubble size and impurity concentration in the ice are the controlling factors on the magnitude of the charge separation. However, the experimenters have concentrated on trying to establish a mean charging rate for bulk ice specimens prepared in different ways without more than a qualitative reference to the controlling factors mentioned above.

Iribarne (1966a) has carried out an extensive laboratory investigation into the bursting of nitrogen bubbles at a water surface, both at room temperature and at  $+2^\circ\text{C}$ , for a wide range of bubble sizes and low concentrations of impurity. A detailed discussion of his results will be given later in this work, but it is pointed out that they show a critical dependence on both bubble size and impurity concentration.

In a paper awaiting publication, Iribarne (1966b), discusses the question of whether the charging process is wholly able to account for the presence of the lower

subsidiary pocket of positive charge in a thunderstorm. He finds that the salt concentration taken from rain analysis data for continental and maritime air are just those which give the main variation in his experiments with bubbles bursting in water. This is a complicating factor in any extrapolation of his results to real hydrometeors, but he concludes that in most cases the concentration of  $\text{CO}_2$  is likely to be the dominating factor in determining the magnitude of the charge separation. In the light of further complicating factors, such as the effect of shape of the hydrometeors and the dynamics of the bubble bursting at high fall speeds, he declines to calculate possible charge separation rates and concludes that there is insufficient evidence at present to decide whether the mechanism is able to account for the subsidiary pocket of positive charge in thunderstorms. However, he does demonstrate that it can explain the magnitudes both of the negative space charge over melting snow and of the vertical electric current found in steady rain.

### 1.3.2 Creation of subsidiary charge.

Of other known methods of atmospheric charge generation there are at least two that operate under the right conditions to produce charges of positive sign below the  $0^\circ\text{C}$  level. These are the drop breaking mechanism proposed originally by Simpson (1909), and

the action of point discharge currents from the ground, (Grenet 1947, Vonnegut 1953). Before discussing them it is worth recording the known features of the positive charge pocket. From the analysis given by Mason (1957), in which he reviews the whole subject of electrification of clouds, the following points emerge.

The pocket of positive charge is situated at or below the  $0^{\circ}\text{C}$  isotherm, and is usually located in the region of most intense precipitation in the centre of the storm. It may be represented approximately by a charge of about 4 Coulombs in a sphere of radius 0.5 Km, i.e. a charge density of  $8\text{C/Km}^3$ . The most intense electric fields in the thunderstorms are often found near the  $0^{\circ}\text{C}$  level in the region between the main negative charge and the lower positive charge, and in many cases these fields are probably instrumental in initiating discharge from cloud to ground. The main centre of negative charge is situated around the  $-5^{\circ}\text{C}$  isotherm.

In an investigation on the charging by the break-up of water drops in a strong vertical jet of air, Simpson found that the heavier fragments from 8mm drops carried away a net positive charge of order  $10^{-2}$  esu per  $\text{cm}^3$  of water, and that ions of both signs, but carrying a net negative charge, were left behind in the airstream. Although this rate of charge generation is too small to account for the observed magnitude

of the charges associated with thunderstorms, it was suggested by Mason (1957) that there could be an appreciable increase in the charge separation rate for drops breaking in an intense electric field such as are found in the bases of thunderstorms. This possibility was explored by Matthews (1963).

#### Matthews' Mechanism

Matthews allowed water drops, which had an equivalent spherical diameter of 15 mm, to rupture in free fall in the presence of a vertical electric field. He found that on average, the heavier fragments carried a charge of one sign ( dependent on the direction of the field), and that the smaller fragments carried the opposite sign and were left in the air. The mean magnitudes of the charges separated in fields of 30, 300 and 1500 V/cm were 0.1, 0.9 and 2.8 esu/cm<sup>3</sup> of water respectively. With the assumption that the charge separation varies inversely as the radius, this would give charges of 0.25, 2.0 and 5.5 esu/cm<sup>3</sup> for drops of 6 mm diameter, this being the natural limit in size for typical raindrops.

Matthews found that the charge separation took place in such a way as to tend to destroy the inducing field by the gravitational separation of the heavy and lighter fragments. This would mean that water drops formed below the melting level and below the main

centre of negative charge in a thunderstorm would carry negative charge on the heavier fragments.

In his conclusions on the relevance of the mechanism to the production of the positive pocket of charge below the main centre of negative charge he says, "This mechanism operating in the lower part of a thundercloud could produce heavier positive fragments in the downward field between the two main thunderstorm charging centres. In this way the appearance of the small region of positive charge at the bases of most thunderstorms may be explained."

Now it has been established beyond all reasonable doubt that the main centre of negative charge in a thunderstorm is situated at temperatures below  $0^{\circ}\text{C}$ , a mean level being  $-5^{\circ}\text{C}$  (Mason 1957), the main centre of positive charge being some several kilometres higher up. It has also been established that, except for a few reported cases of lightning from clouds wholly warmer than  $0^{\circ}\text{C}$ , the occurrence of solid precipitation elements is a prerequisite of intense electrical activity. It therefore seems most unlikely that large drops of water will be present in regions of the thunderstorm well above the  $-5^{\circ}\text{C}$  isotherm. In an examination of thunderstorms on the Zugspitze, Kuettner (1950), found that even in cases where the cloud bases were warmer than  $0^{\circ}\text{C}$ , rain was present at cloud base

on only 21 per cent of occasions.

Even if large raindrops were present in the region between the main centres of charge, and electrification by rupturing took place, it is difficult to see how Matthews visualises the positive fragments falling through the main centre of negative charge, and emerging as a separate collection of positive drops some distance below it.

### Vonnegut's Mechanism

VC An alternative model is proposed by Vonnegut (1953), who considers that the convective motion of the air in a thunderstorm, together with point discharge from the ground, could act in a manner akin to a giant Wimshurst machine, and might separate sufficient charge to account for the total generation of the thunderstorm. In this model, positive charge is transported by the updraughts to the top of the cloud from ions generated near the ground by point discharge. The lower negative charge is similarly supplied by the conduction charge from the upper atmosphere and transported downwards in downdraught regions. With this model, the presence of the lower pocket of positive charge in the downdraught region would be difficult to explain without there being some generating mechanism at work near the 0°C level.

In correspondence with Latham (1965), Vonnegut

says that there is insufficient evidence at present to decide whether point discharge is likely to be a major mechanism in thunderstorm electrification but believes it to be important as a possible secondary generator. In this way it might account for the lower positive charge by point discharge in the field set up between the main negative centre and the ground, the positive ion current being captured by falling drops. Measurements of the point discharge current at the ground give a value of about  $0.02 \text{ amp/Km}^3$  at the time of most intense vertical field (Simpson 1949). If this current were spread over the entire cross-section area of the lower negative charge, a mean value being about  $3\text{Km}^2$ , then it could provide positive charge to the atmosphere at a rate of about  $3.6 \text{ C/min}$ .

Although this is of more than sufficient magnitude to explain a small region of charge of  $4 \text{ C}$ , it does not explain why the charge is localised near the melting level and in an area approximately one quarter the horizontal extent of the main negative charge. Nor does it take into account the fact that the positive current carried to the ground by the precipitation elements in this region of the storm is of the same order of magnitude as the upward ion current (Mason 1957).

Measurements made on individual precipitation elements in the region of most intense vertical electric field by Smith (1955) and Gunn (1950), have shown that

the charges carried by these elements are often greatly in excess of that which can be explained by the simple Wilson capture of ions, unless it is assumed that the vertical field increases considerably with height above the ground. However, the presence of a positive space charge would actually reduce the field in this region between itself and the ground.

The above arguments do not preclude the possibility of the final value of about 4C being a dynamic equilibrium condition after build up over a long period. We will therefore examine this possibility in more detail.

During the growth of the thunderstorm, and before the onset of precipitation, the integrated point discharge current would provide a positive space charge distributed in the lower regions of the storm. If we assume that the point discharge current increases linearly with time from a value of about zero to a value of  $0.02 \text{ amp/km}^2$  during a mean period of 20 minutes before precipitation falls from cloud base, then the total charge separated in a cylinder of cross-section  $1 \text{ km}^2$  will be about 12C. The vertical distribution of the charge will depend on both the updraughts and field strengths present in this region. Using a value of  $1.5 \text{ cm/sec/V/cm}$  for the mobility of typical atmospheric ions, and assuming a mean vertical field over the interval of order 300 V/cm (Mason 1957), then the vertical drift velocity of the ions due to the field

*No. 1*  
*was in cloud drops*  
*2 cc*  
will be of order 4.5 m/sec. We may combine this with the updraught to give a figure of order 10 m/sec. for the total upward movement of the ions. Over the 20 minute period of development of the storm then, a charge of 12 C may be distributed in a cylinder of cross-section  $1 \text{ Km}^2$  and height of order 12,000 m: i.e. a mean charge density of order  $1.5 \text{ C/Km}^3$ .

With the onset of precipitation, the distribution of this charge will be affected by capture of the ions by the falling raindrops below the  $0^\circ\text{C}$  level. However, in order to enhance the mean charge density to a value of order  $8 \text{ C/Km}^2$  typical of the positive charge pocket, it would be necessary for the drops to fall a distance of order 5 Km even if they swept up all the ions in the intervening space, and assuming the drops to have zero initial charge. In practice, the precipitation elements will, on balance, have a strong negative charge associated with their fall from the main negative charge centre, and are in any case unlikely to capture the entire ion current. If these considerations were to increase the fall distance necessary to acquire a charge density of  $8 \text{ C/Km}^3$  by a factor of two, then the precipitation elements would have reached the ground as the melting level is usually of order only 2 - 5 Km above ground. It thus appears that although the mechanism of point discharge is able to supply charge in sufficient quantity, there is difficulty

in visualising a method whereby it is concentrated in a small region near the  $0^{\circ}\text{C}$  level in a thunderstorm. On the other hand it does seem more likely that it will explain the positive current to earth carried by precipitation in the centre of the thunderstorm, which is of the same order of magnitude as the ion current.

In summary therefore, it is concluded that although several mechanisms have been proposed to explain the subsidiary positive charge in a thunderstorm, none of them has been clearly demonstrated to be capable of wholly accounting for its sign, magnitude and distribution.

#### 1.4 Division of Present Work

Because of the two separate aspects of the melting problem investigated in this thesis, the heat transfer and electrification, it has been decided to present the work in two separate sections.

The first part deals with the melting equations of small ice spheres and cones under a wide range of meteorologically significant conditions. The final equations are then used to compute the distances of fall for complete melting to take place under typical atmospheric conditions. The section concludes with an investigation into the equivalence of an ice-bulb hygrometer and an evaporating ice sphere in the same airstream, by determining the conditions under which surface melting takes place.

The second half of the thesis deals with the

electrification of melting ice and snow, and particularly attention is paid to the analysis in terms of the controlling factors previously mentioned. Finally, the results are discussed in relation to the various atmospheric charging phenomena which it has been suggested that they might explain.

PART ONE

"The Melting Equations"

## CHAPTER TWO

### The Melting Equations.

#### 2.1 Theory of a Melting Ice Sphere

Consider a sphere of melting ice in a moving air stream of constant velocity, temperature and humidity. If, at any instant, the ice core of radius  $a$  is surrounded by a uniform symmetrical layer of water of radius  $(b-a)$ ,  $b$  being the overall radius of the particle, then the basic equations governing the transfer of heat between it and the air are given by Mason (1956) as

$$\begin{aligned}
 L_f \rho_i 4\pi a^2 da/dt &= -4\pi ab K_w T_s / (b-a) & (2.1) \\
 \text{(Latent heat of melting)} & \quad \text{(transfer through water)} \\
 &= -4\pi b [C_h K_a (T - T_s) + C_m L_v D (\rho(T) - \rho(T_s))] \\
 & \quad \text{(conduction thru' air)} \quad \text{(condensation on particle)}
 \end{aligned}$$

where  $\rho_i$  is the density of the ice sphere,  $T_s$  its surface temperature,  $T$  the temperature of the remote environment,  $K_a$ ,  $K_w$  the thermal conductivities of air and water,  $L_v$ ,  $L_f$  the latent heats of condensation and fusion,  $D$  the coefficient of diffusion of water vapour in air,  $\rho(T)$ ,  $\rho(T_s)$  are respectively the concentrations of water vapour in the environment and very near the surface of the sphere.  $C_h$  and  $C_m$  are coefficients that allow for the enhanced exchange of heat and water vapour between the sphere and a moving air stream.

If the relative humidity of the air is  $H$ , and the air in contact with the sphere is assumed to be saturated

at the surface temperature  $T_s$ , we may write

$$q(T) - q(T_s) = H(q_{\text{sat}}(T) - q_{\text{sat}}(T_s)) \quad (2.2)$$

and empirically it is found that

$$q_{\text{sat}}(T) = 4.84 + 0.30T + 0.016T^2 \text{ g.m}^{-2} \quad (2.3)$$

for  $0^\circ\text{C} < T < 15^\circ\text{C}$  to within  $\frac{1}{2}\%$ .

According to the careful measurements of Ranz and Marshall (1952) on evaporating water drops, the transfer coefficients take the form

$$C_h = 1.0 + 0.27(Re)^{\frac{1}{2}} \quad (2.4)$$

$$C_m = 1.0 + 0.26(Re)^{\frac{1}{2}} \quad (2.5)$$

for  $10 < Re < 500$

where  $Re$  is the Reynolds Number of the sphere. Thorpe and Mason (1966) give slightly different formulae for the case of evaporating ice spheres. However, an ice sphere with a layer of water on its surface will behave essentially as a water sphere, and therefore the above values of the transfer coefficients are used in all the calculations on melting. Ranz and Marshall also found that the value of the diffusion coefficient,  $D$ , for water vapour in air given in the International Critical Tables as  $D = 0.22 \text{ cm}^2 \text{ sec}^{-1}$  appeared to be too large and suggested a value of  $0.20 \text{ cm}^2 \text{ sec}^{-1}$  for its value at  $0^\circ\text{C}$ . Thorpe and Mason, working with evaporating ice spheres, suggest a value of  $D = 0.206 \text{ cm}^2 \text{ sec}^{-1}$

with an overall uncertainty of  $\pm 3\%$ . Further evidence that the ICT value may be too high is put forward by work of Hilpert (1932) on evaporating wet plates, and by Powell (1940) on spheres. In the calculations made in this work, a value of  $D = 0.20 \text{ cm}^2 \text{ sec}^{-1}$  is adopted.

With the substitution for the vapour densities in terms of temperature given by equations (2.2) and (2.3), equation (2.1) lends itself to solution by numerical integration. The solution is much simplified if  $b$  is assumed to remain constant during the melting process. This will be nearly true in the case of solid ice spheres of density close to  $0.9 \text{ g cm}^{-3}$ , particularly at high humidities where condensation on the sphere will partly compensate for the change due to an increase in density, as demonstrated in the calculation below.

The rate of increase in mass at any instant for a sphere due to condensation is given by

$$dm/dt = 4\pi bDC_m \{ \rho(T) - \rho_{\text{sat}}(T_s) \} \quad (2.6)$$

If the sphere is melting in a saturated air stream of temperature  $T^\circ\text{C}$ , then a mean value for its surface temperature over the melting period will be of order  $T/2^\circ\text{C}$ . For a sphere of radius 1mm in a saturated air stream of velocity  $4 \text{ m sec}^{-1}$  and temperature  $10^\circ\text{C}$ , equation (2.6), combined with equation (2.3), gives

$$(dm/dt)_{\text{mean}} \approx 3.5 \times 10^{-6} \text{ g. sec}^{-1}$$

Experimentally, it is found that the melting time for a 1mm radius sphere under these conditions is of order 50 seconds. Hence, the fractional change in mass,  $f_m$ , over the whole melting period is approximately

$$f_m = 50 \times (dm/dt)_{\text{mean}} / \frac{4}{3} \pi b^3 \approx 0.05$$

The corresponding per cent change in radius is of order 1.7%, compared with that of order 3% due to the density change in melting.

Turning again to equation (2.1), then given the values of  $b$ ,  $T$ ,  $H$  and the wind speed (hence  $Re$ ), and assuming  $\rho_i = 0.9 \text{ gcm}^{-3}$ ,  $T_s$  can be calculated as a function of ' $a$ ', and the time taken for the sphere to melt completely with these constant environment conditions is then given by

$$t_m = \int_0^t dt = - \frac{L_f \rho_i}{k_w b} \int_b^0 \frac{a(b-a)}{T_s(a)} da \quad (2.7)$$

Values of  $t_m$  were calculated for a wide range of initial parameters using an IBM 7090 computer. The integral was evaluated using Simpson's Rule with ten radius steps. The values of constants used were those appropriate to  $0^\circ\text{C}$ . To avoid an expression of the form  $0/0$  arising at one of the limits, the integral was evaluated over the range  $b=0$  to  $b=0.0001 \text{ mm}$ .

The computation was tested for sensitivity to

changes in the values of the constants used, and for doubling the number of intervals in the numerical evaluation of the integral by Simpson's Rule. The results can be expressed in terms of % change in the melting time for a typical case and are given in Table (2.1) below.

Table (2.1)

| Alteration                          | % Change in $t_m$ |
|-------------------------------------|-------------------|
| $D \pm 5\%$                         | $\pm 2\%$         |
| $K_a \pm 5\%$                       | $\pm 2\%$         |
| $C_h/C_m \pm 5\%$                   | $\pm 1\%$         |
| $K_w + 2\%$                         | $- 2\%$           |
| Simpson's Rule with<br>20 intervals | $< 0.1\%$         |

During melting, the surface temperature of the drop varies between  $0^\circ\text{C}$  and the wet-bulb temperature of the environment, the maximum value being  $10^\circ\text{C}$ . Since both  $D$  and  $K_a$  vary in the same sense as the temperature changes, the net effect of using the values of these constants at  $0^\circ\text{C}$  is less than 1% when the temperature varies between  $0^\circ\text{C}$  and  $10^\circ\text{C}$ . The variation of  $K_w$  in this range is of order 2%, but when the time integral is taken the mean effect will be of order 1%. Hence,

the maximum error in the calculation of  $t_m$  is estimated to be of order 2%.

An analysis of the results showed that for a given air temperature, the computed values of  $t_m$  could be represented empirically by the relationship

$$b^2/t_m C_m^{0.82} = B(H-E) \quad (2.8)$$

where  $H$  is the relative humidity, and  $B$  and  $E$  are constants which are nearly independent of wind speed and humidity. Varying the particle radius,  $b$ , between 0.5 mm and 3 mm for a fixed wind speed produces a spread of  $\frac{1}{2}\%$  in the empirically derived values of  $b^2/t_m C_m^{0.82}$ , while varying the wind speed from 0.5 to 4.0 m sec<sup>-1</sup> for constant  $b$  produces a variation which is  $\sim 1\%$ . Equation (2.8) is plotted in Figs. (2.5) and (2.6) for four values of air temperature and for relative humidities ranging from 20 to 100%.

## 2.2 Experimental Arrangement

In order to study the melting of ice and snow particles under a wide range of temperature and humidity conditions, a closed circuit wind tunnel system was constructed inside a cold room. A schematic layout of the system is shown in Fig. (2.1). The pump used was a B.V.C. vacuum cleaner with a maximum capacity of 2000 l/min., controlled by a voltage regulator which allowed continuous variations of flow in the range from

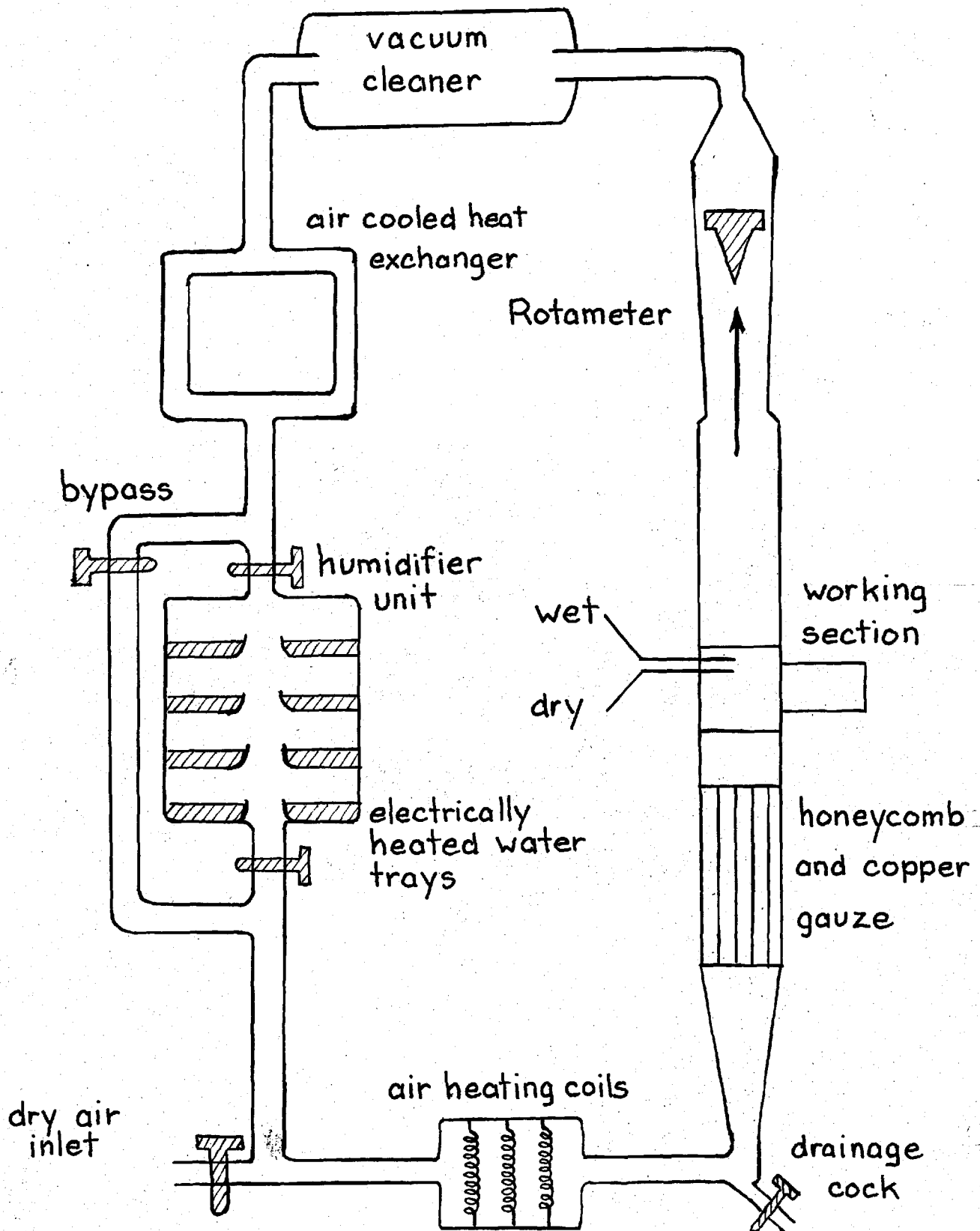


Fig. 2.1 Schematic layout of closed-circuit wind tunnel system.

zero to 2000 l/min. At high air flow rates it was found that the pump heated the air stream by as much as  $10^{\circ}\text{C}$  over the ambient temperature of the cold room. For this reason a heat exchanger with a large surface area was included in the circuit. This consisted of 32 thin walled copper tubes mounted between two shallow brass cylinders 18 inches in diameter and 1 inch deep. The tubes were equally spaced round the circumference of the cylinders. Details of the construction are shown in Fig. (2.2). A small coil-wound heater capable of 200 watt dissipation was mounted just before entry to the wind tunnel, and was controlled by a Variac voltage regulator.

The wind tunnel itself consisted of a vertical Perspex tube 10 cms in diameter and approximately 2 m long, surmounted by a Rotameter flow device of nearly the same diameter. The working section of the tunnel was removable. The air flow in the tunnel was stabilised by use of honeycomb sections, and a series of copper gauzes gave a uniform temperature profile across the tunnel. A nylon micromesh filter was used to remove particulate matter from the air stream. The components were connected together with  $1\frac{1}{2}$  inch bore flexible hose, and the system was provided with a drainage cock at its lowest point.

Experiments were carried out with a number of

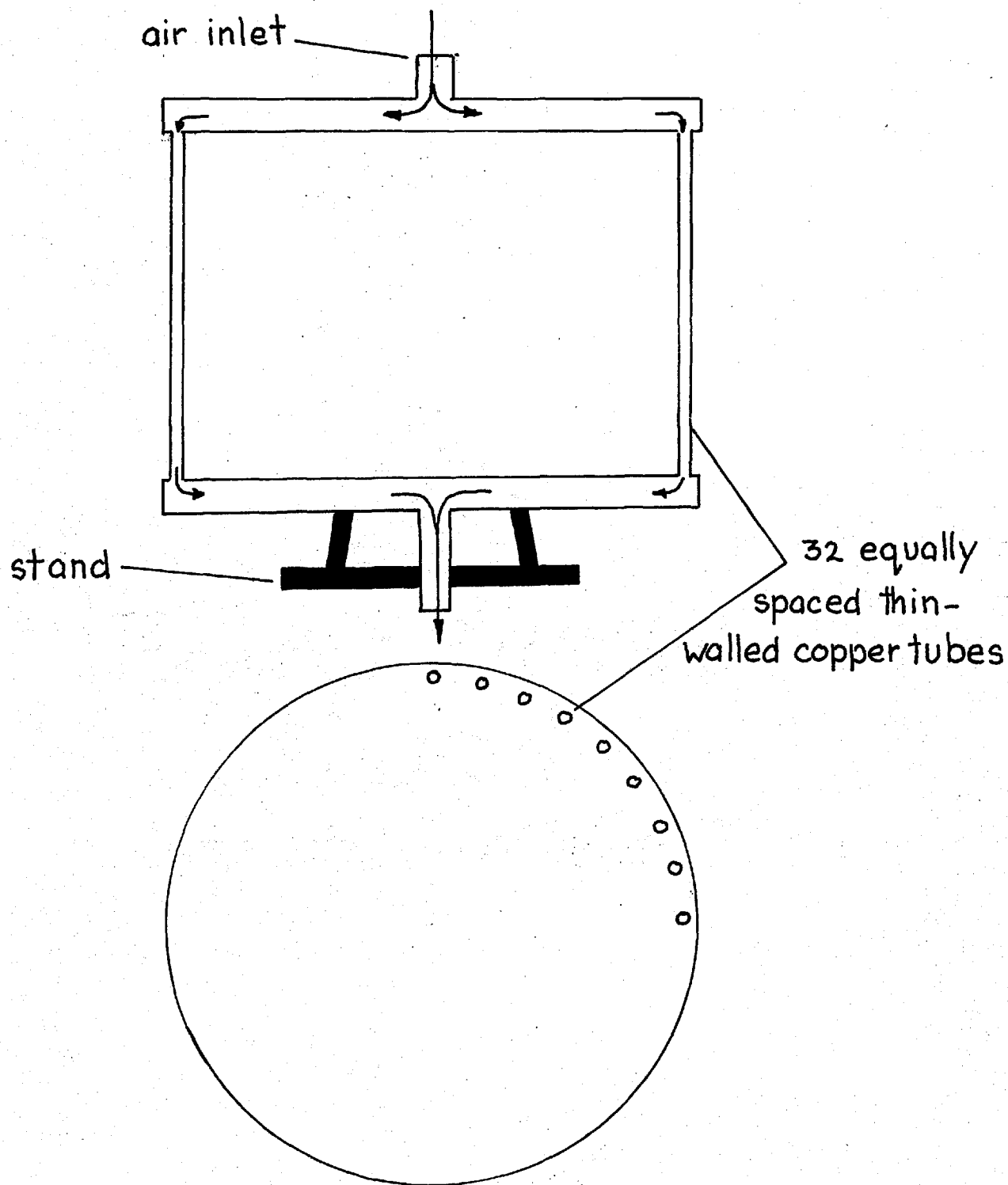


Fig. 2.2 Detail of heat exchanger.

humidifying units, but none proved to be entirely satisfactory in permitting independent control of both humidity and temperature. The different methods used were based on the following ideas:-

- i) Passing heated air through wet muslin gauze.
- ii) Injecting steam from a small boiler.
- iii) Mixing a saturated air stream with a dry air stream.
- iv) Passing the air stream over heated water trays.

The method finally adopted, as a compromise, was a combination of (iii) and (iv). The air stream was first saturated at the working temperature by passage over electrically heated water trays. The humidifying unit was then by-passed, and the air stream allowed to become progressively drier over a long period of time by slight mixing with the cold- room air. In this way a whole range of different humidities could be obtained, the rate of change being as little as 5% per hour.

The air temperature of the working section was determined using a 46 s.w.g. Manganin/contra thermocouple. The humidity was determined using a 46 s.w.g. Manganin/Contra wet-bulb thermojunction. The junction was wound with a single layer of clean cotton thread for a distance of 3 cm on either side and distilled water was applied to the junction with a small paint brush. The vapour pressure of the air stream was calculated from the

psychometric equation

$$e_s(T') - e(T) = A_w(T - T') \quad (2.9)$$

where  $e(T)$  is the vapour pressure of the air stream at temperature  $T$ , and  $e_s(T')$  is the saturation vapour pressure at the wet-bulb temperature  $T'$ , and  $A_w = 0.62 \text{ mb}^\circ\text{K}^{-1}$ . The adoption of this value for  $A_w$  is explained in the next chapter. The relative humidity was then calculated using Meteorological Tables Met. O. 265 part III, 2nd edition, assuming normal atmospheric pressure.

At all wind speeds and humidities, the temperature profile in the tunnel was flat over the central portion of the tunnel, the variation being less than  $0.05^\circ\text{C}$  out to a radius of 4 cm. The velocity profile and turbulence level in the tunnel were investigated using a hot-wire anemometer. The turbulence level, expressed as r.m.s. fluctuation/mean velocity, was in all cases  $< 2\%$  over the central tunnel section. The major part of the fluctuation was due to variation in volume flow rather than eddies; a stream line test using steam injection detected no eddies. The hot wire was found to be unsuitable for absolute velocity determination. It was suspected that the temperature corrections applied to the calibration for the cold room temperature of order  $-10^\circ\text{C}$  were greatly in error, as the integrated volume flow was approximately

20% lower than indicated by the Rotameter. In spite of this, it was found that the velocity dropped by only 10% from that at the centre when approximately 2 mm from the walls for velocities  $> 0.5 \text{ m sec}^{-1}$ .

In all calculations, the tunnel velocity has been taken as the mean velocity indicated by the Rotameter. The maximum error introduced because of this is estimated to be about 3% in the wind speed, which will produce an error in the calculation of  $C_h$  and  $C_m$  of about 1% for Reynold's numbers in the range 10 to 500.

### 2.3 Experimental Procedure

Ice spheres with diameters between 0.2 and 5.0 mm were made by freezing drops of distilled water from a hypodermic syringe. The drops were suspended on a mesh of  $10\mu$  diameter fibres, made by drawing fine threads of Evostik glue, and were nucleated and frozen in a small chamber attached to the working section of the wind tunnel.

The technique of manipulating the spheres is indicated in Fig. (2.3). A small hole in the preparation chamber exposed the drops to the air from the cold room and allowed them to become supercooled and nucleated with small ice crystals. Once the drops had frozen they were transferred on the sliding carrier into the wind tunnel, partially melted there, and then returned to the chamber and refrozen. In this way it was possible

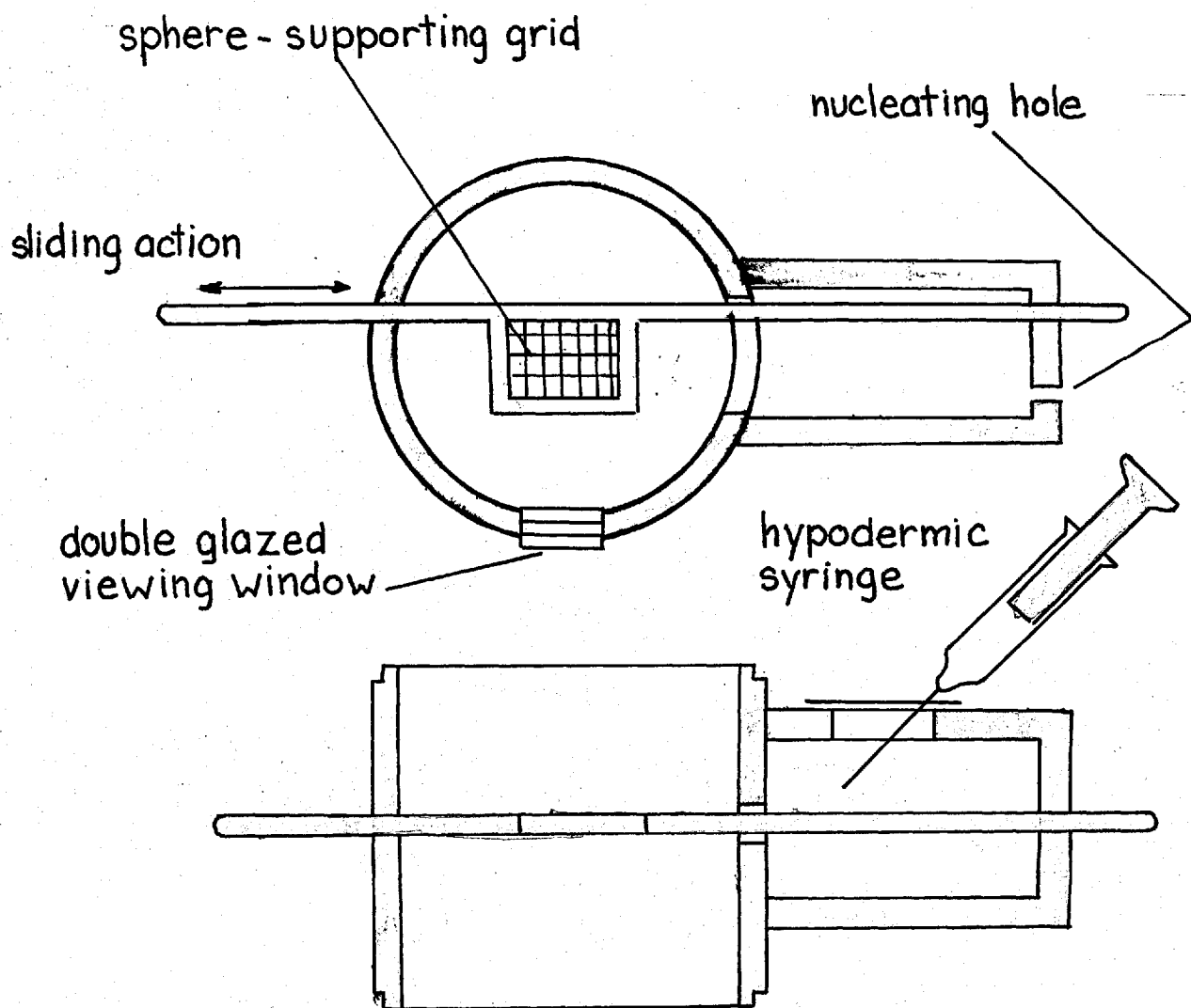


Fig. 2.3 Detail of transfer mechanism.

to produce good, smooth ice spheres that contained few air bubbles and had densities close to  $0.9 \text{ g cm}^{-3}$ . Such spheres were re-introduced into the wind tunnel and allowed to melt in an air stream of constant velocity, temperature and humidity. Their melting was observed with intermittent, filtered illumination through a low power microscope and the times until the last traces of ice vanished determined with an accurate stop-watch.

During the melting period the ice core was observed to float to the top of the drop and considerable convective motions were seen in the water portion. The convective currents, which sometimes rotated the ice core in unpredictable fashion, could be studied by the motion of small air bubbles released by the melting ice. The velocity of these currents, whose general pattern is indicated in Fig. (2.4), increased when the temperature and humidity gradients and the wind speed were increased, but never exceeded a few cm/sec. The larger air bubbles ( $d > \frac{1}{10} \text{ mm}$ ) released during melting tended to rise to the top of the drop, while smaller bubbles were carried down by the convection currents and tended to burst near the stagnation point at the base of the drop. In the final stages of melting the ice core usually collected a layer of bubbles which persisted for up to a minute after the last traces of ice had disappeared.

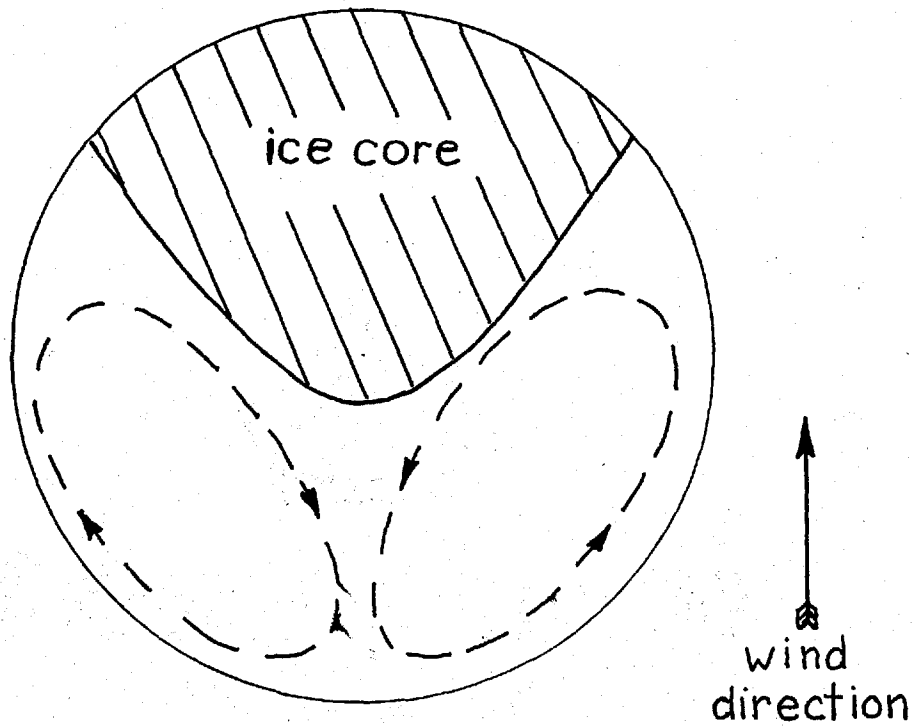


Fig. 2.4 Schematic diagram showing convection cells and conical ice core in melting sphere.

Experiments were conducted at four different temperatures between  $0^{\circ}\text{C}$  and  $10^{\circ}\text{C}$ , for several different wind speeds in the range  $0.5$  to  $4.0 \text{ m sec}^{-1}$ , and with humidities ranging from  $20$  to near  $100$  per cent. The actual range of humidity employed with any given temperature and sphere size was limited by the consideration that the sphere diameter should not change by more than  $5\%$  during the melting process.

The experimental results, plotted in the form of  $b^2/t_m C_m^{0.82}$  against the relative humidity of the air stream, are shown in Figs. (2.5) and (2.6), where they may be compared with the corresponding relationships computed from equation (2.8).

#### 2.4 Discussion of Results

Random errors in the determination of the complete melting time ( $\pm 1\%$ ), external radius,  $b$ , ( $\pm 3\%$ ), and  $C_m$  ( $\pm 3\%$ ) combine to produce a probable error of about  $\pm 7\%$  in the experimentally determined values of  $b^2/t_m C_m^{0.82}$ , the error in the measured value of the relative humidity being estimated at  $\pm 3\%$ . In addition to some scatter, Figs. (2.5) and (2.6) shows that, especially at higher air temperatures, the experimental points show a systematic deviation from the computed relationships in a sense that indicates that the ice melts more slowly than predicted by theory, i.e. by equation (2.8). The systematic errors that could arise are listed below

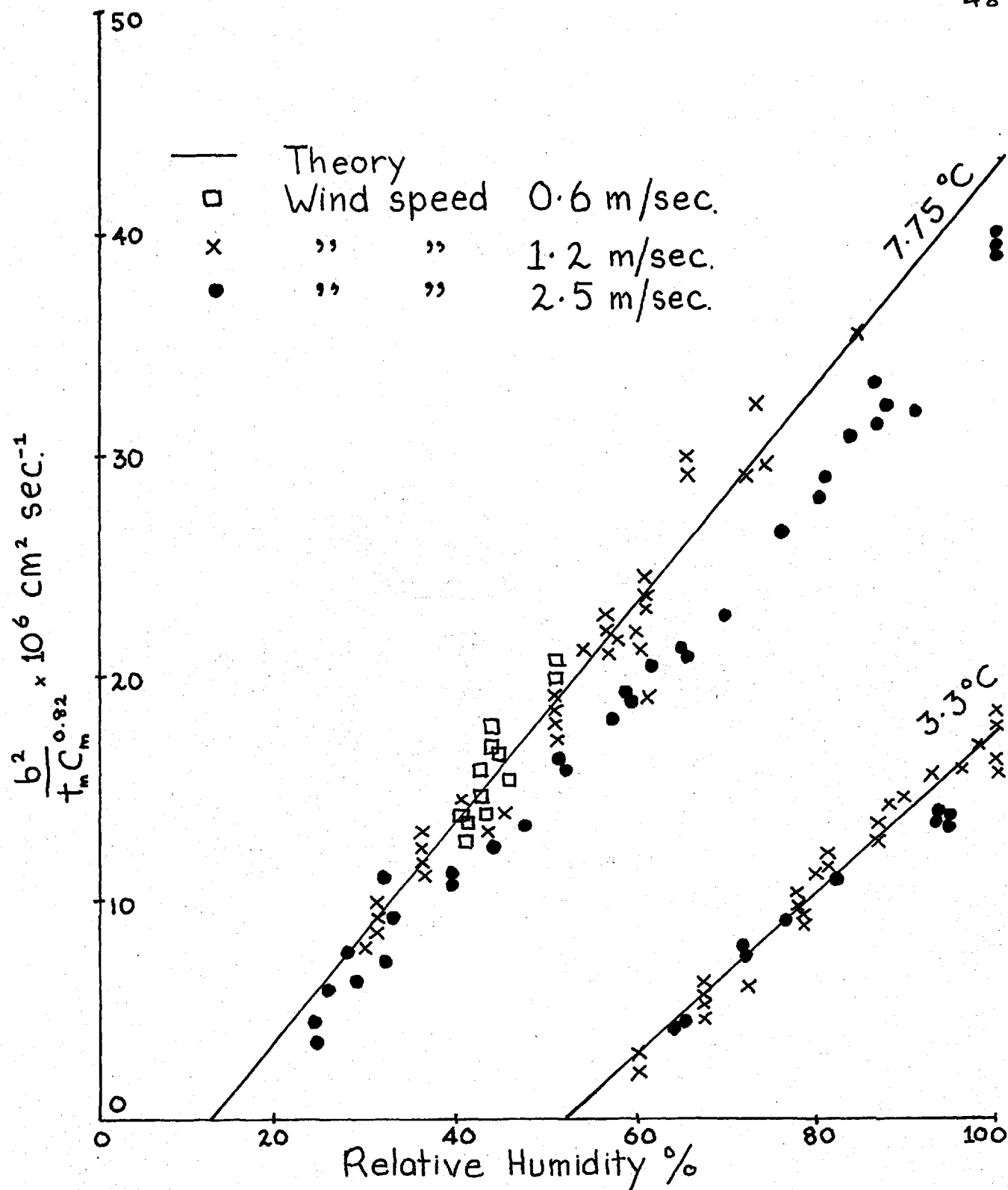


Fig. 2.5 Empirical function  $b^2/t_m C_m^{0.82}$  plotted against Relative Humidity for ice spheres. Air temps. 3.3°C and 7.75°C.

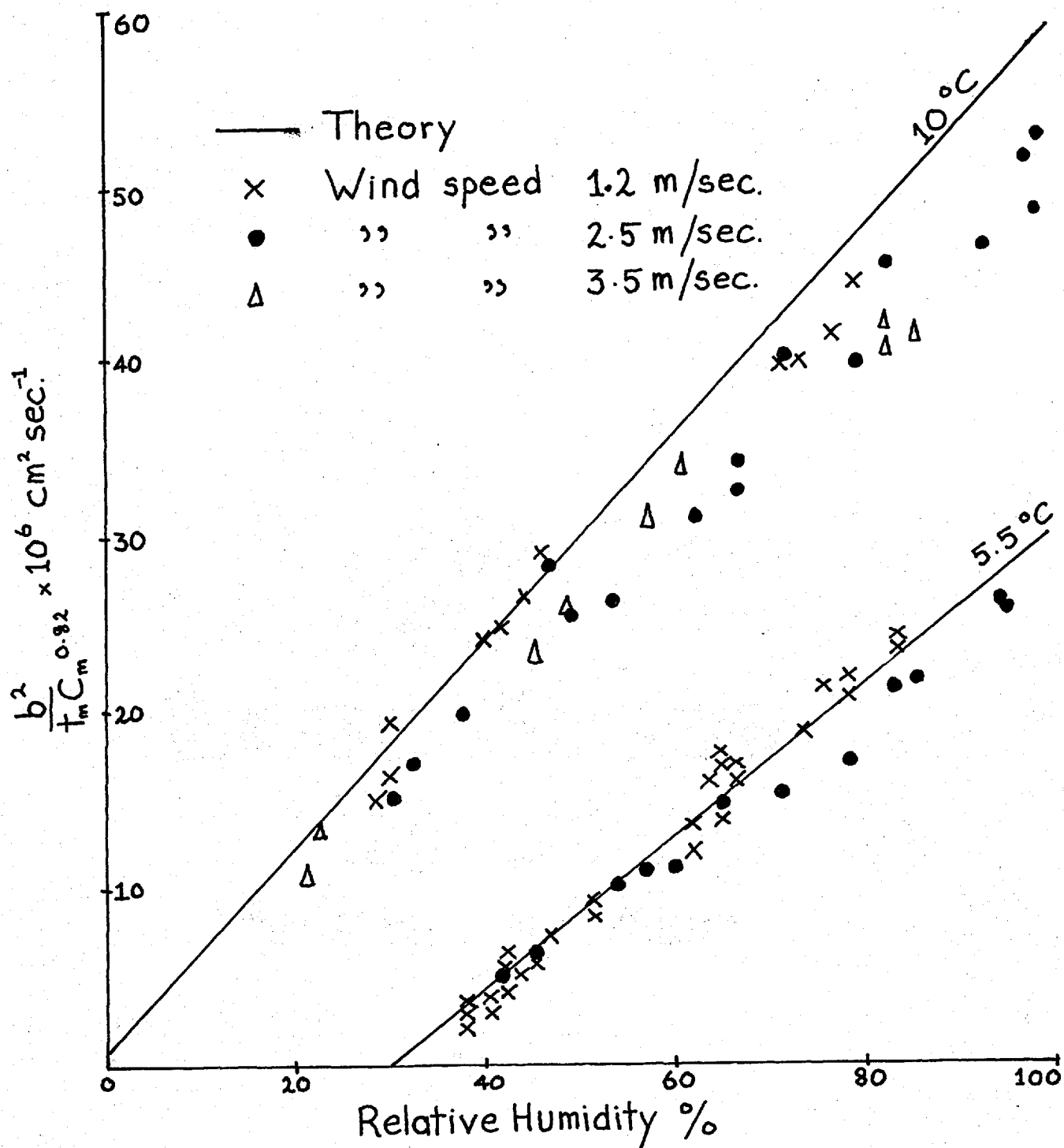


Fig.2.6 Empirical function  $b^2/t_m C_m^{0.82}$  plotted against Relative Humidity for ice spheres. Air temps. 5.5°C and 10.0°C.

and considered below separately.

- a) Radiation received from the warm air stream.
- b) Conduction of heat along the supporting fibres.
- c) Modification of the diffusion field by the supporting fibres.
- d) Presence of air bubbles in the ice inhibiting heat transfer.
- e) Finite time for melting to begin after introduction of the spheres into the wind tunnel.
- f) Absence of term involving the heat required to raise the temperature of the water shell in equation (2.1).

(a) The ratio of heat received by radiation to heat received by conduction to the sphere from the warm air stream, is given approximately by

$$\frac{4\pi b^2 \alpha \sigma (T^4 - T_s^4)}{4\pi b C_h K_a (T - T_s)} \quad (2.10)$$

where  $\alpha$  = absorbtivity of ice  $\approx 1.0$

$\sigma$  = Stefan's Constant =  $1.35 \times 10^{-12}$  watt  $\text{cm}^{-2}$  deg.  $\text{K}^{-4}$ .

Using  $b = 1 \text{ mm}$ ,  $C_h = 5.0$  and assuming the extreme condition of the air being at  $+10^\circ\text{C}$  while the surface temperature of the sphere is at  $0^\circ\text{C}$ , then this ratio is less than 0.01. In general, the temperature difference between the air stream and the sphere surface will be considerably less than  $10^\circ\text{C}$ , and the sphere will not

be composed entirely of ice with high absorbtivity.

The integrated effect will then lead to an error of much less than 1% in the melting time, and has consequently been neglected.

(b) The ratio of heat conduction from the air to heat conduction along the fibres to the sphere is given approximately by

$$\frac{4 \pi b C_h K_a (T - T_s)}{8 K_f \left(\frac{\pi d^2}{4}\right) (T - T_s) / \alpha} \quad (2.11)$$

where  $K_f$  is the fibre conductivity,  $d$  the fibre diameter and  $\alpha$  the boundary layer thickness for the sphere, given approximately by

$$\frac{\alpha}{b} \approx \left(\frac{\pi D}{v b}\right)^{1/2} \quad (2.12)$$

where  $v$  is the wind speed and  $D$  the coefficient of water vapour diffusion in air. This gives a mean value of 0.1 for  $\frac{\alpha}{b}$  for the range of wind speeds and sphere diameters used in this experiment. The factor 8 appearing in the denominator of Equation (2.11) takes into account the possibility that the sphere might be situated at the intersection of two pairs of fibres at right angles.

Using the values

$$K_f = 3.5 \times 10^{-4} \text{ cal. cm}^{-1} \text{ sec}^{-1} \text{ } ^\circ\text{C}^{-1}$$

$$C_h = 5.0$$

$$d = 10^{-3} \text{ cm}$$

$$b = 10^{-1} \text{ cm}$$

$$K_a = 6.0 \times 10^{-5} \text{ cal. cm}^{-1} \text{ sec}^{-1} \text{ } ^\circ\text{C}^{-1}$$

in equation (2.11), then the ratio of heat conduction from the air to heat conduction along the fibres is of order  $10^3:1$ , and the conduction along the fibres is negligible.

(c) The fibres were drawn as fine as possible in order to minimise both heat transfer and any modification of the diffusion field for the spheres. In order to test the effects of the method of support, a variety of positions were used, the spheres being supported on different numbers of fibres either from above, below or passing through the sphere. No significant variation in the complete melting time could be detected for any of the variations used.

(d) The presence of air bubbles trapped in the ice during freezing might modify the heat transfer. However, no significant difference in overall melting time could be found between spheres with few bubbles and those prepared with large concentrations of bubbles.

(e) With the simplifying assumption that

$$C_h = C_m = C \quad (2.13)$$

and

$$\rho_{\text{sat}}(T_2) - \rho_{\text{sat}}(T_1) = \beta(T_2 - T_1)$$

Mason (1956) calculated the time,  $\tau$ , taken for an ice sphere to warm from temperature  $T_1$  to  $T_2$  in an air stream saturated with water vapour at temperature  $T$  to be

$$\tau = \frac{\rho_i c_i b^2}{3C(K_a + L_v D/\beta)} \log_e \left( \frac{T - T_1}{T - T_2} \right)$$

where  $\rho_i$ ,  $C_i$  are respectively the density and specific heat of ice. When the air temperature in the wind tunnel was  $+10^\circ\text{C}$  the cold room was held at about  $-10^\circ\text{C}$ , so putting  $T = 10^\circ\text{C}$ ,  $T_1 = -10^\circ\text{C}$ ,  $T_2 = 0^\circ\text{C}$ , and  $\beta = 3 \times 10^{-7} \text{ g cm}^{-3} \text{ } ^\circ\text{K}^{-1}$  we have

$$\tau = 1100 \frac{b^2}{C} \text{ secs.} \quad (2.14)$$

Some typical values of  $\tau$  are compared with the complete melting times,  $t_m$ , of spheres in an air stream of velocity  $3.5 \text{ m sec}^{-1}$  and temperature  $10^\circ\text{C}$  in Table (2.2).

TABLE (2.2) VALUES OF  $\tau$  AND  $t_m$  FOR SPHERES OF VARIOUS SIZES

| Radius, $b(\text{cm})$ | $C$ | $\tau(\text{sec})$ | $t_m(\text{sec})$ | $\tau/t_m$ |
|------------------------|-----|--------------------|-------------------|------------|
| 0.5                    | 5.0 | 0.55               | 11                | .050       |
| 1.0                    | 7.0 | 1.57               | 35                | .045       |
| 2.0                    | 9.5 | 4.63               | 105               | .044       |

Since it takes a few seconds to position the ice sphere in the wind tunnel and to focus the telescope upon it, the onset of melting cannot be determined exactly and timing the melting period from the instant the sphere enters the tunnel may lead to an overestimate of  $t_m$  by about 5% in the worst cases.

(f) The calculation of the time of complete melting,  $t_m$ , is considerably complicated if accurate account is taken of the warming of melt water. However the magnitude of the effect may be readily estimated in the case where the air stream is saturated with water

vapour and the final surface temperature of the drop  $T_s = T$ . The average temperature of the well-stirred melt water will then be approximately  $T/2^\circ\text{C}$  and its heat content  $C_w T/2$  has to be compared with the latent heat  $L_f$ . If  $T = 10^\circ\text{C}$ ,  $C_w T/2 = 5 \text{ cal gm}^{-1}$  corresponding to an increase in the melting time of about  $5/80$  i.e. 6%. Thus it seems that the heating of the melt water and the finite time for the sphere to reach  $0^\circ\text{C}$  after introduction to the tunnel, taken together, may account for the observed discrepancy of about 10 per cent between the observed and calculated melting times at the higher air temperatures and humidities, and also for the smaller discrepancies at lower temperatures and humidities.

### 2.5 The Melting of Small Ice Cones

Small, artificial, conical hailstones were prepared by freezing distilled water in conical brass moulds. These were supported on a mesh of fibres in the same manner as the spheres, and allowed to melt in the wind tunnel under constant environmental conditions, the time for complete melting being recorded as before. The dimensions of the types of cone used are given in Table 2.3 below. Types A and C were supported with their points facing away from the wind, while type B faced with its point into the wind. These are the aspects that would occur in natural fall, as predicted by the criteria due to Jayaweera (1965). Cone A had a hemispherical cap.

Table 2.3 Dimensions of Cones

| Type | Body Ht.<br>mm | Min. Dia.<br>mm | Max. Dia.<br>mm | Cap Ht.<br>mm | Eq. Sphere Rad.<br>mm |
|------|----------------|-----------------|-----------------|---------------|-----------------------|
| A    | 2.38           | 1.13            | 3.57            | 1.78          | 1.70                  |
| B    | 2.38           | 1.13            | 3.57            | -             | 1.38                  |
| C    | 4.50           | 0.53            | 3.57            | -             | 1.61                  |

In each case it was found that the profile of the melting cone assumed a nearly spherical shape within one third to one half of the melting time. Because of the difficulty of formulating the theory of heat and vapour transfer under these changing boundary conditions, the problem has been examined by comparison with spheres of equal volume. The ratio  $t_c/t_s$ , the ratio of the melting time of a cone to that of a sphere of equal volume, has been plotted as a function of the relative humidity of the environment for the three types of cone and for two values of air temperature in Figs. 2.7 and 2.8.

From the graphs it can be seen that at low relative humidities the cones melt faster than spheres of equal volume, the discrepancy being greater the more the cones deviate from the spherical shape. However, with increasing humidity and more rapid condensation on the melting particles, this tendency is reduced and finally reversed. Even so, wide variations in the humidity do

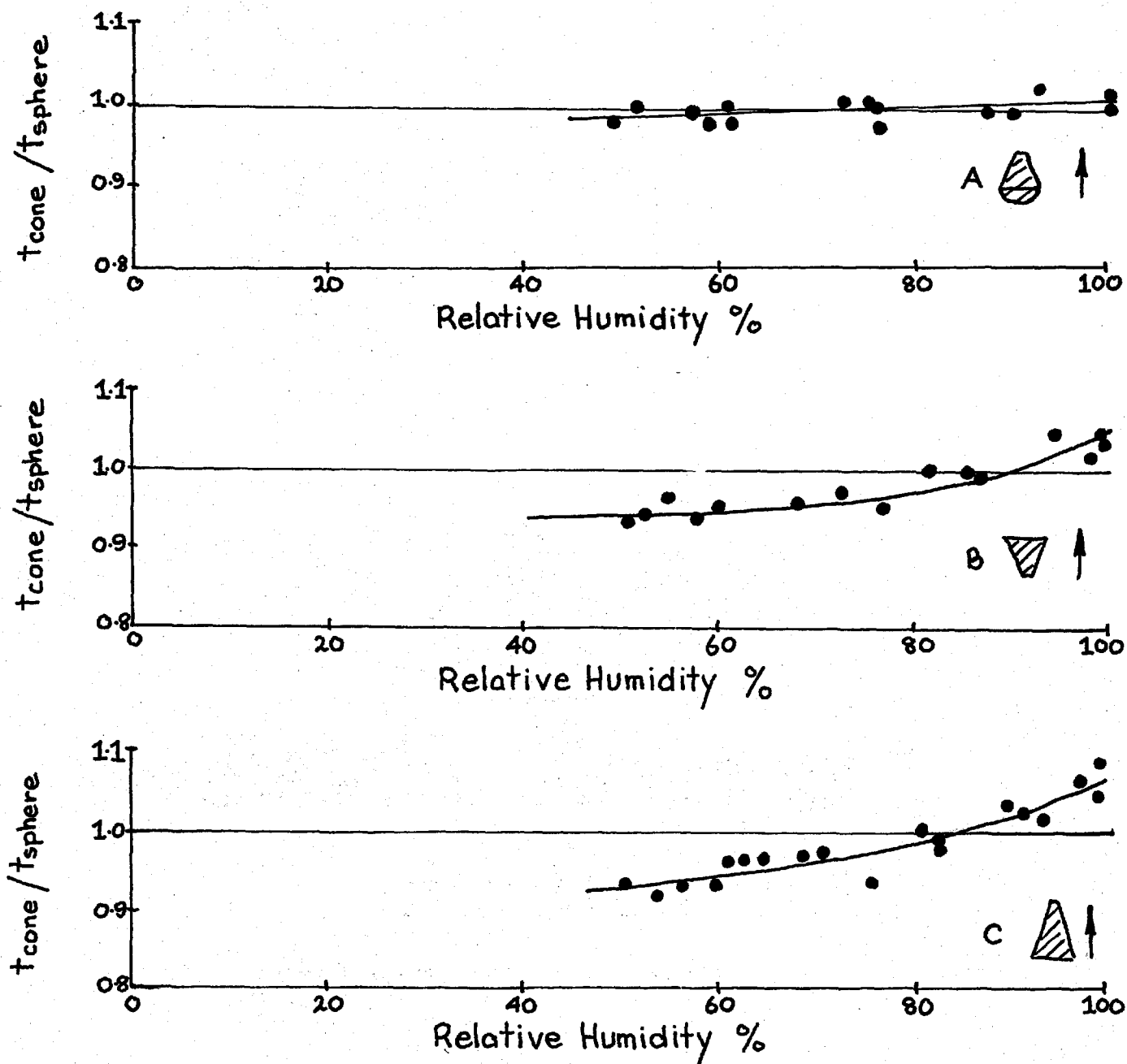


Fig. 2.7 Ratio of complete melting time for cones to that of spheres of equal volume as a function of Relative Humidity. Density  $0.9 \text{ g cm}^{-3}$ ; Wind speed  $3.5 \text{ m sec}^{-1}$ ; Air temperature  $3.3^\circ\text{C}$ .

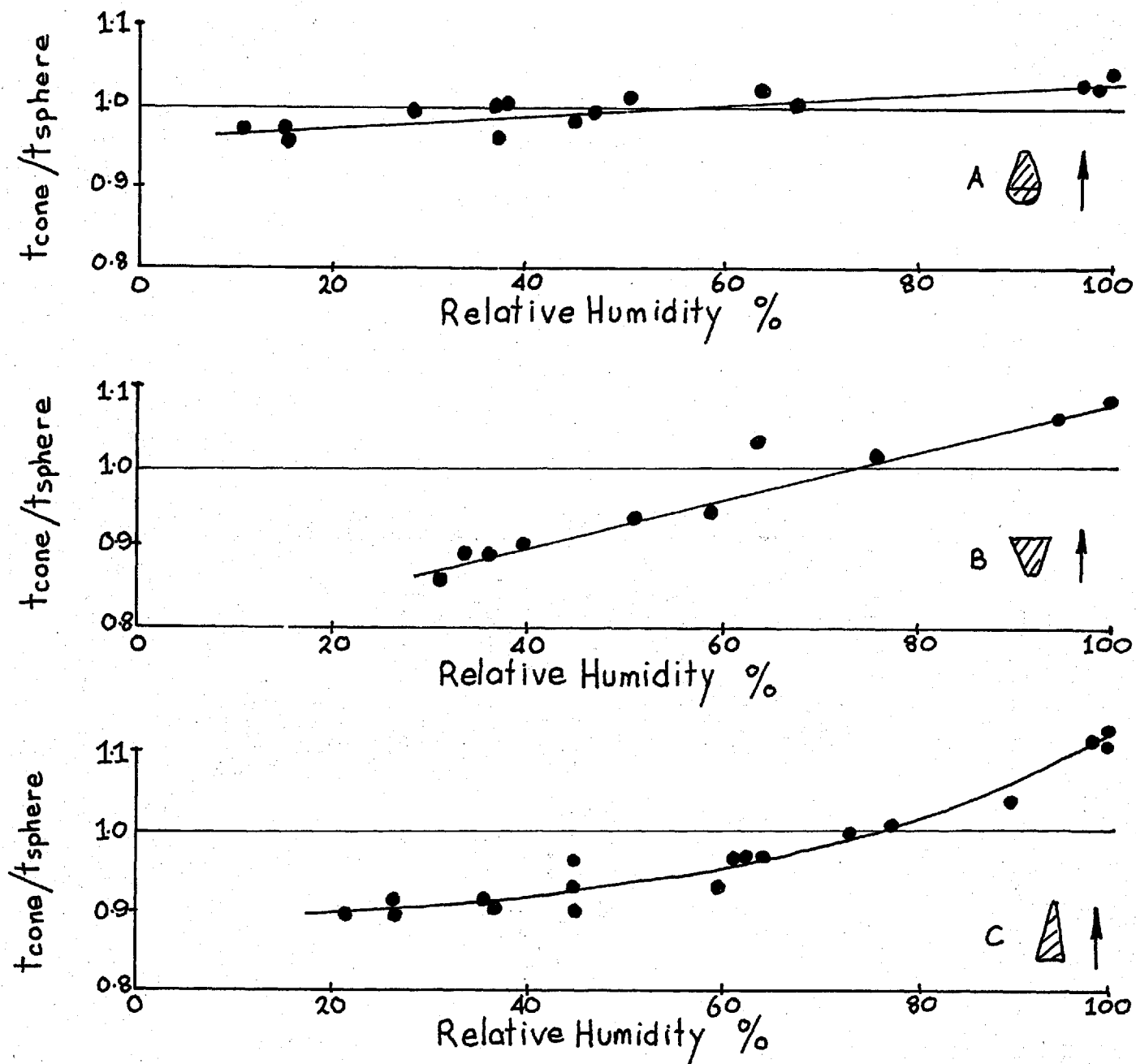


Fig. 2.8 Ratio of complete melting time for cones to that of spheres of equal volume as a function of Relative Humidity. Density  $0.9 \text{ g cm}^{-3}$ ; Wind speed  $3.5 \text{ m sec}^{-1}$ ; Air temperature  $7.75^\circ \text{C}$ .

not cause the complete melting times of the cones to differ by more than about 10% from the corresponding values of spheres of equal volume.

Equation (2.1) could be adapted to describe the melting of cones with the inclusion of "shape factors" that modify the diffusion terms of heat and water vapour. Because the boundary conditions are changing in shape during the melting it would not be possible to refer these factors to the initial cone profile in more than a qualitative way. However, the results indicate that the vapour diffusion term is reduced in comparison with the heat diffusion term. At low humidities this gives a smaller rate of cooling by evaporation, hence faster melting than the equivalent sphere. At high humidities the condensation rate is similarly decreased, leading to a longer melting time than for the equivalent sphere. At median humidities where the mean vapour density gradient is nearly zero, the melting time is approximately the same as for the equivalent sphere. These observations suggest that Equation (2.1) could be written with parameters based on the "spheres of equivalent volume" hypothesis, and need only be modified by an empirical factor reducing the condensation term. It can be seen from Figs. (2.7) and (2.8) that the effect is greater the more the cone departs from the spherical.

However, empirical factors of this nature do not

give much insight into the physical reasons for the behaviour during melting, and their calculation is more academic than useful. For meteorological purposes, the fact that even over the widest range of humidities likely to be found in the atmosphere, the melting times of cones do not differ by more than 10% from that of spheres of equal volume, means that the "equivalent sphere" approximation is sufficiently accurate for calculating rates of melting of small ice particles under atmospheric conditions.

## 2.6 The Melting of Snow

Some samples of natural snow were collected in the winter of 1963-4 and melted in the wind tunnel. However, the effort was concentrated on the techniques of collection, storage and handling, and only a few preliminary observations were made in readiness for the following winters. It is unfortunate that in the 1964-5 and 1965-6 winters there has been only one heavy snow fall and a few light showers. Consequently little experimental work has been done on the melting of snow. The few qualitative observations that have been made are recorded below.

i) The melting time of flakes collected from the same snow fall appears to be independent of flake size. This is probably an indication that the individual crystals comprising the flakes, melt as more or less individual units. Visual observation supports this

view. The original crystal positions in the melting flake do not change until melting is very nearly complete, but in the final stages the wet crystals are drawn into a roughly spherical droplet and the last traces of the ice rapidly disappear.

ii) Small fragments occasionally broke away from the snow flakes in the wind stream before they began to melt. Once melting had begun, however, this was observed only rarely. The effect occurred predominantly with flakes formed from clusters of dendritic crystals.

iii) The break-up during melting of large, conglomerate snow flakes indicates a consistent pattern. These conglomerate masses usually consisted of a number of individual and identifiable flakes that had probably stuck together when wet. This type of flake was typically found when the air temperature at the ground was slightly above  $0^{\circ}\text{C}$ , and occasionally had cross-sections of over  $10\text{ cm}^2$  in the horizontal plane. Where the individual flakes had stuck together, regions of volume overlap acted as the nuclei for formation of water drops. The final number of water drops formed appeared to be approximately the same as the number of these overlapping regions. With four different samples collected on different occasions, it was observed that the water drops formed had diameters between 0.5 and 1.5 mm; the mean diameter of all the drops being 0.9 mm..

Since the flakes were melted on a mesh of  $10\mu$  fibres spaced to form squares of 3 mm side, it is quite likely that the results were affected by the support, and they may be criticised on these grounds.

## 2.7 Conclusions

### 2.7.1 Spheres

When allowance is made for heating of the water shell, and the finite time for melting to begin after introduction of the spheres to the wind tunnel, the experimental results for spheres of diameter 0.2 mm to 5.0 mm show agreement with theory to within  $\pm 3\%$  in the time of complete melting, over the whole range of each of the parameters air temperature,  $0^{\circ}\text{C}$  to  $10^{\circ}\text{C}$ , humidity, 20% to 100%, and wind speed, 0.5 to 4 m sec<sup>-1</sup>. This range of wind speed includes the fall speed of the smaller spheres. This suggests that the equations may be used to predict the melting times of spheres falling in an atmosphere in which the temperature and humidity profiles vary with height. Although the heat transfer equations, cast in the form of Equation (2.1), are based on the steady-state theory, Mason (1956) showed that the thermal relaxation time for drops composed entirely of either ice or water, defined as the time for the drops to reach  $(1 - 1/e)$  of their equilibrium temperature difference, when suddenly introduced into a new, water saturated environment, is only of order

a few seconds. Experiments by Gunn (1951) with water drops showed that for a drop of 1.35 mm radius falling in air at  $28.8^{\circ}\text{C}$  and having a relative humidity of 22% the thermal relaxation time was 4.35 secs. In the case of melting ice particles, the internal convective motions in the water layer will decrease this relaxation time once the melting is well established. Since these times are small, compared with the time for complete melting that will be calculated, no serious error will be introduced by using the steady-state equations.

Following Mason (1956), two types of atmosphere are considered that probably represent the extremes of cases likely to be found in practice. The first deals with the case in which the air is water saturated at all levels of descent, corresponding to melting entirely within the cloud but neglecting collection of liquid water. The second considers the case when there is neither evaporation from nor condensation on the particle at all levels of descent; this corresponds to the case where melting begins just at cloud base and the particle encounters an increasingly dry atmosphere below it. Any real case is likely to fall between these extremes except under rare circumstances when the humidity immediately below cloud base is very low.

For the saturated case, Equation (2.1) gives a pair of simultaneous, linear, differential equations that

cannot be solved analytically. Using the approximation of equation (2.3), and the substitution  $T = \alpha vt$  where  $\alpha$  is the lapse rate and  $v$  the fall speed of the particle, the melting times were computed using an IBM 7090 computer. A fourth order Runge-Kutta programme was used, and the ice-core radius and surface temperature evaluated at every 2 second interval. To avoid difficulty at the end point, the computation was terminated when the core radius was just less than 1% of the initial radius. This is estimated to introduce an error in the computed melting time of less than 0.01%. The changes in fall speed and the drop diameter during melting were neglected. The values of fall speed used were the terminal velocities for freely falling water drops as determined by Gunn and Kinzer (1949). These are given in Table (2.4) below, together with the values of  $C_h$  and  $C_m$  calculated from equations (2.4) and (2.5).

Table 2.4 Terminal Velocities of Falling Water Drops

| Drop Radius<br>mm | Fall Speed<br>cm/sec | $C_h$ | $C_m$ |
|-------------------|----------------------|-------|-------|
| 0.05              | 403                  | 5.45  | 5.26  |
| 0.10              | 650                  | 8.95  | 8.65  |
| 0.15              | 806                  | 11.85 | 11.40 |
| 0.20              | 883                  | 14.10 | 13.60 |
| 0.25              | 909                  | 15.85 | 15.32 |

Although the range of Reynold's Numbers involved, 0 to 2000, is greater than that for which equations (2.4) and (2.5) have been quoted, 0 to 500, they probably give a good estimate of  $C_h$  and  $C_m$ . The computed melting time is found to be approximately proportional to  $(C_h + C_m)^{-\frac{1}{2}}$ , so any error introduced from this source will be halved in the estimation of the melting time.

A further source of error lies in the use of equation (2.3) to compute the saturation vapour pressure as a function of temperature. This equation is only true for a pressure of 1000 mb. If the melting level is assumed to be at a height of order 700 mb pressure, then the error in the computation for a sphere of initial radius 1 mm is estimated to be about 1%, and has been neglected.

The warming of the melt water has been allowed for by adding 6% to the computed times, based on the calculation of section 2.4(f) for constant environment conditions. Although this is only an approximation it is not likely to lead to an error greater than 1% in the computed melting time.

The total error from all sources is estimated to be not greater than 3%. Since the purpose of the calculation is to determine the variation in melting time under two extreme atmospheric conditions, this level of error is not important.

The results of these calculations for the case of an atmosphere with lapse rate  $6^{\circ}\text{C Km}^{-1}$ , and which is water saturated at all levels are shown in Figs. (2.9) and (2.10), where they are compared with the results of Mason (1956), adjusted for a lapse rate of  $6^{\circ}\text{C Km}^{-1}$ .

For an atmosphere in which there is neither condensation on, nor evaporation from the melting spheres, equation (2.1) is capable of analytical solution. We then have

$$L_f \rho_i a \, da/dt = - \frac{b K_w}{(b-a)} T_s \quad (2.15)$$

and

$$T_s = \frac{C_h K_a (b-a)}{a K_w + C_h K_a a(b-a)} T \quad (2.16)$$

and using the substitution

$$T = \alpha v t \quad (2.17)$$

then the time,  $t_f$ , for the ice core to reach a fraction,  $f$ , of its original radius is given by

$$\int_0^{t_f} t \, dt = - \frac{L_f \rho_i}{b K_w K_a C_h \alpha v} \int_b^{f b} a \{a K_w + C_h K_a (b-a)\} da \quad (2.18)$$

The times for complete melting ( $f = 0$ ), and the corresponding distance of fall are shown in Figs. (2.9) and (2.10), where they are compared with those of Mason.

An examination of the results shows that the difference between the fall distances for the two

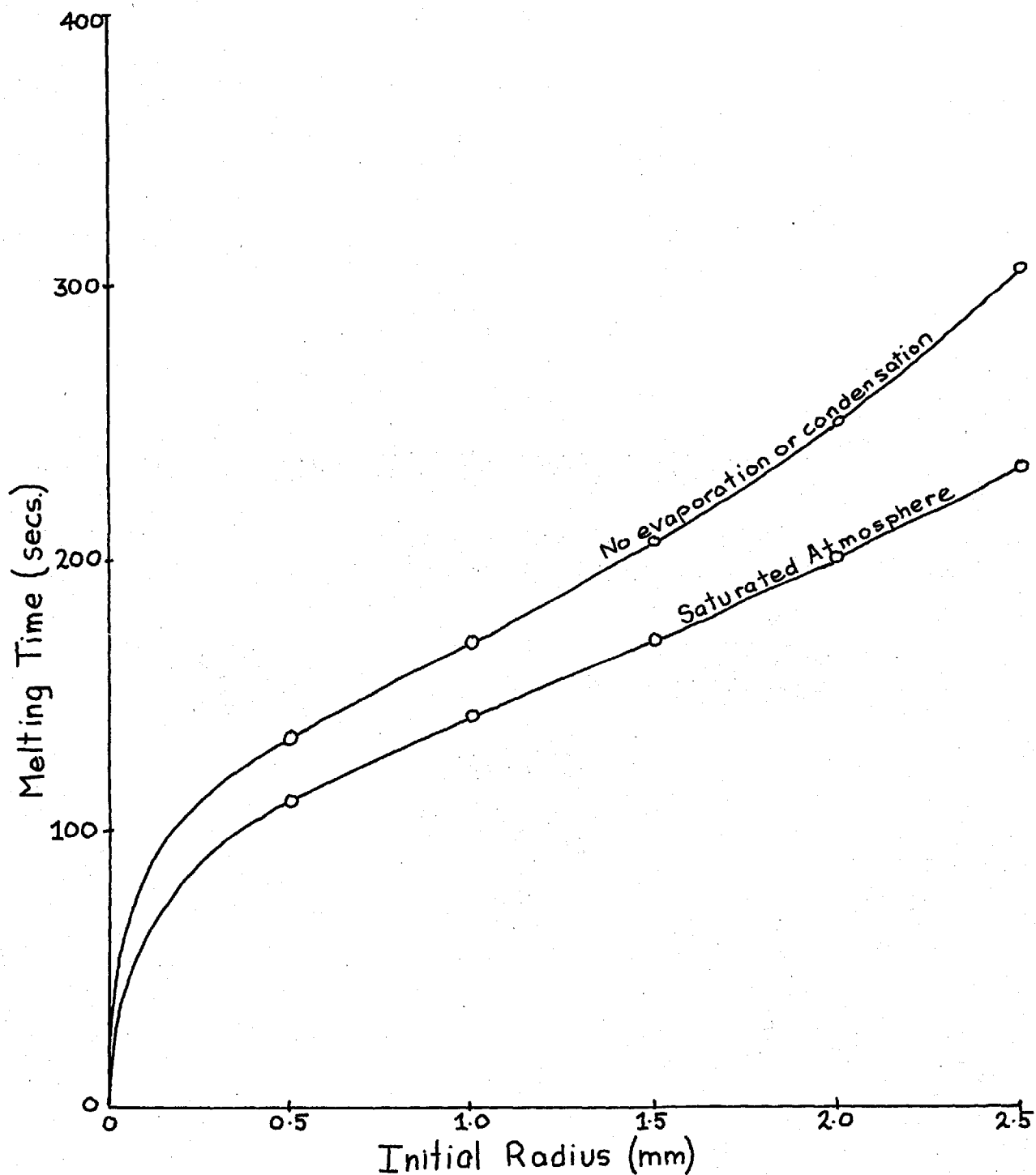


Fig. 2.9 Complete melting time for solid ice spheres falling from  $0^{\circ}\text{C}$  level as a function of their initial radius.  $\alpha = 6.0^{\circ}\text{C Km}^{-1}$ .

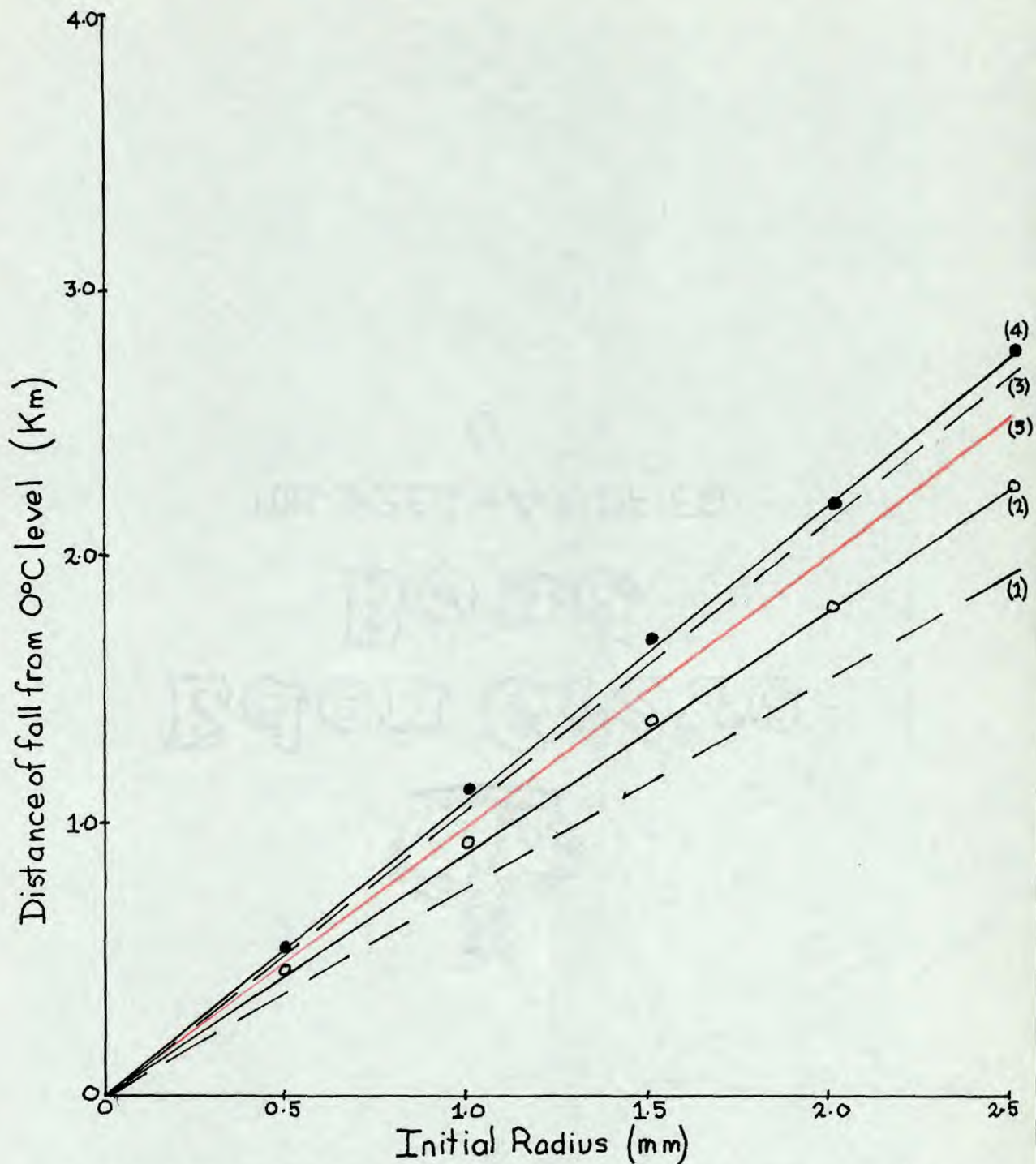


Fig. 2.10 Distance of fall of solid ice spheres from 0°C level for complete melting.  $\alpha = 6.0^\circ\text{C Km}^{-1}$

— — — Mason (1956).

Curves (1) and (2) Saturated Atmosphere.

Curves (3) and (4) No evaporation or condensation.

Curve (5)  $Z = (2.55a/\alpha^4) \times 10^6$

atmospheres is much less than that found by Mason. The slight difference in curves (3) and (4) in Fig. 2.10 are due to a difference in the form of the heat transfer coefficient,  $C_h$ , assumed by Mason. The curves show that in both types of atmosphere considered, the distance of fall is very nearly directly proportional to the initial radius of the spheres.

Since the results for the two extreme types of atmosphere do not differ by more than 20% over the whole radius range, the distance of fall for complete melting to take place under all normal atmospheric conditions may be calculated from a single, simple formula with an accuracy of 10%. This level of accuracy will usually be sufficient for most meteorological purposes. Since the distance of fall is also very nearly inversely proportional to the lapse rate,  $\alpha$ , to the power one half, we have

$$Z = (2.55 a / \alpha^{1/2}) \times 10^6 \quad (2.19)$$

where  $Z$  is the distance of fall from the  $0^\circ\text{C}$  level for complete melting to take place,  $a$  is the initial particle radius and  $\alpha$  the lapse rate in  $^\circ\text{C Km}^{-1}$ . This relationship is shown as the red curve (5) in Fig. (2.10).

In the case of the saturated atmosphere, the calculation neglects the possible accretion of cloud

droplets which might contribute to the rate of melting of the ice spheres. If we consider an ice sphere of 5 mm diameter melting completely during a fall of 3 Km, then the volume swept out by the sphere is approximately  $6 \times 10^{-3} \text{ m}^3$ . Assuming a liquid water content of  $1 \text{ g.m}^{-3}$  in the lower regions of the cloud, and that the ice sphere collects all the cloud drops in its path, then the increase in mass of the sphere will be  $6 \times 10^{-3} \text{ g}$ . The mass of the ice sphere itself is approximately  $450 \times 10^{-3} \text{ g}$ , so the collection of cloud droplets will lead to an increase in mass of the ice sphere of order 1%, and the contribution of this to the melting can be neglected within the accuracy of Eq. (2.19).

In order to show how much of the sphere remains as ice at each stage of its descent, Figs. (2.11) and (2.12) have been plotted to show the distance of fall from the  $0^\circ\text{C}$  level as a function of the fractional water shell thickness of the spheres. This is important in assessing the radar reflectivities, as Kerker, Langleben and Gunn (1951), have shown that an ice sphere with a water film thickness greater than one fifth of its original radius will scatter as effectively as a sphere of the same mass composed entirely of water. It was observed in the wind tunnel that for this fraction of melting the ice core was still approximately

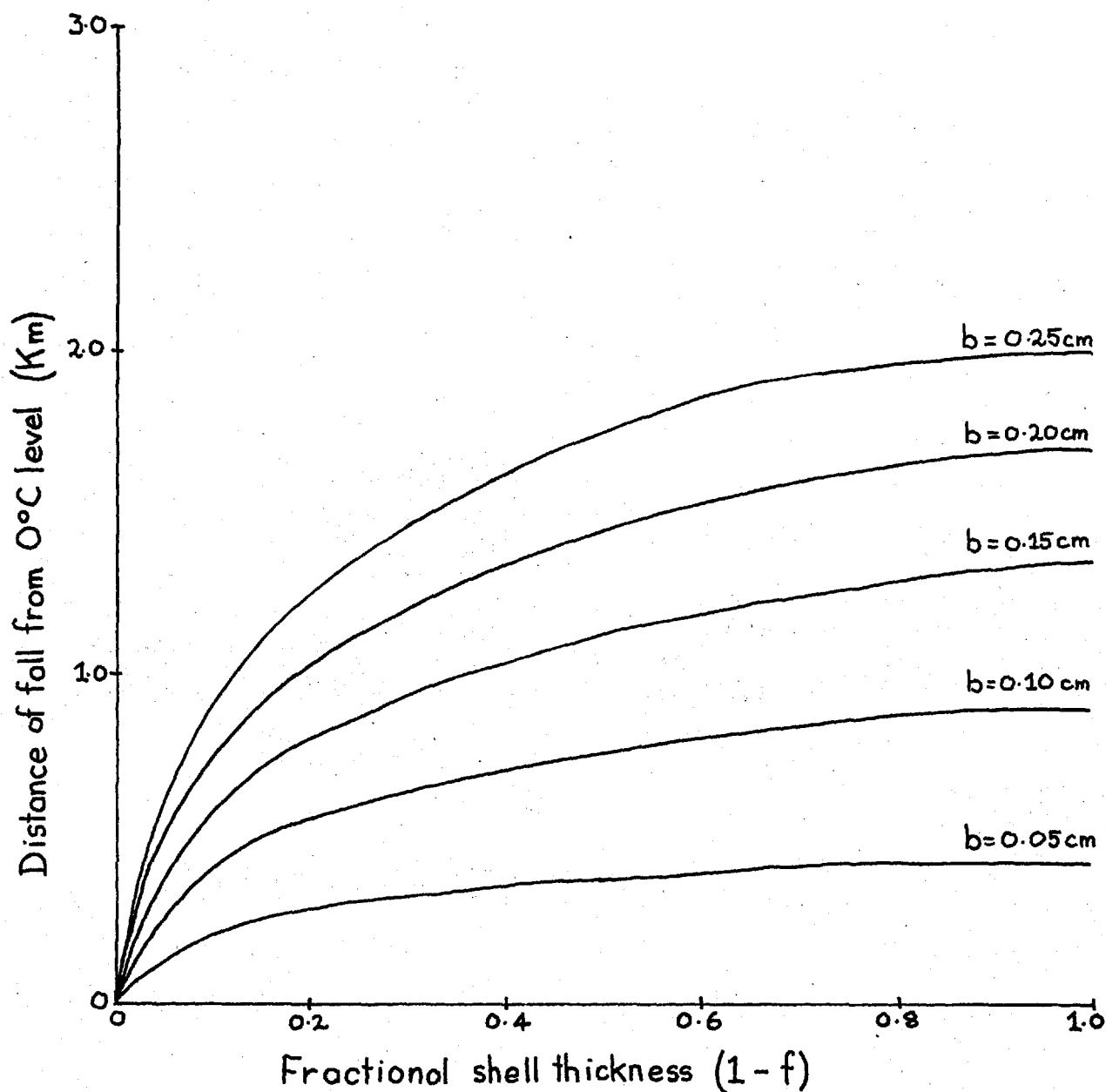


Fig. 2.11 Graph showing fractional water shell thickness as a function of distance of fall from  $0^\circ\text{C}$  level for solid ice spheres of different initial radii. Atmosphere saturated at all levels.  $\alpha = 6.0^\circ\text{C Km}^{-1}$

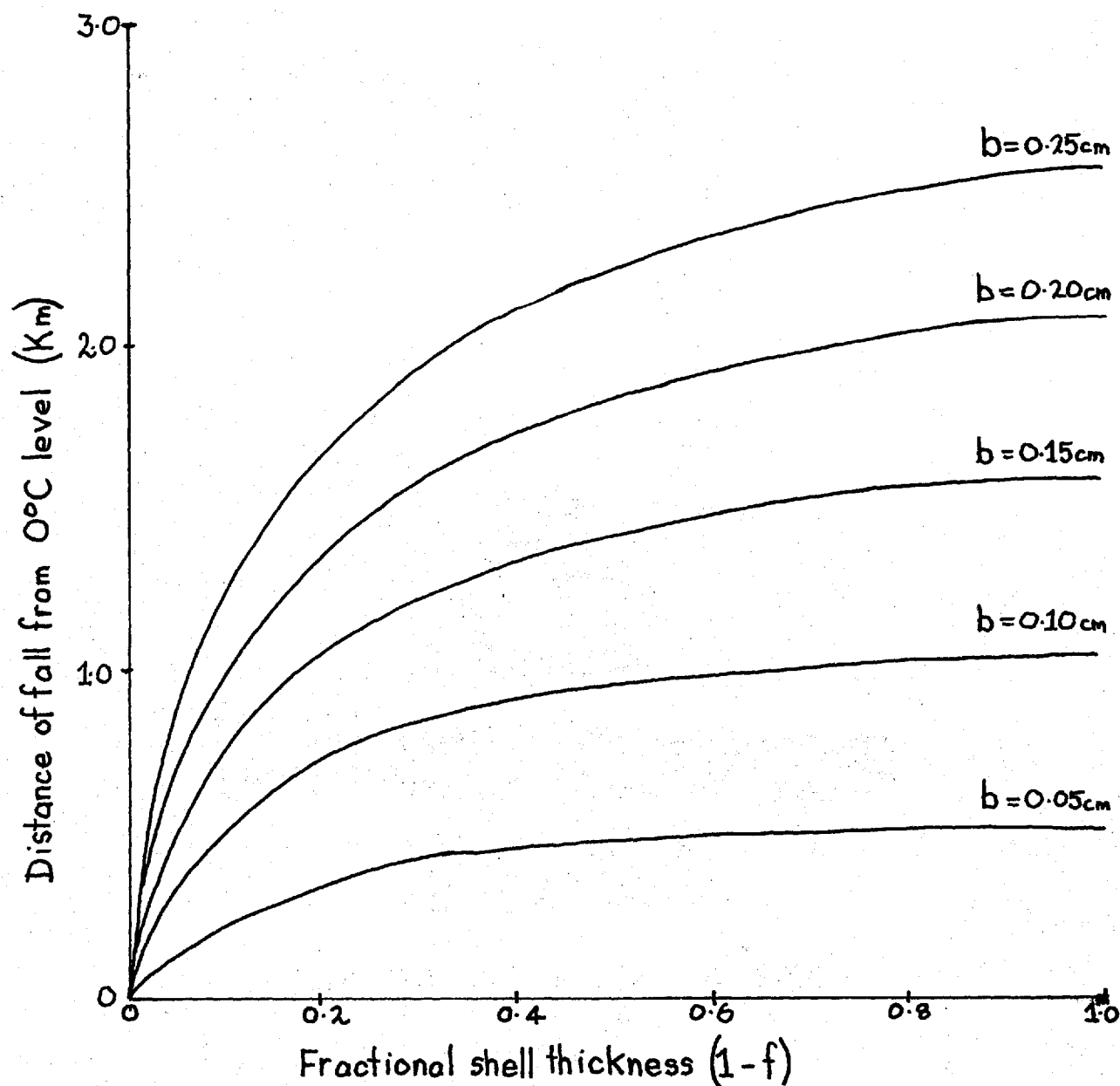


Fig. 2-12 Graph showing fractional water shell thickness as a function of distance of fall from 0°C level for solid ice spheres of different initial radius. No evaporation or condensation on sphere.  $\alpha = 6.0 \text{ }^{\circ}\text{C Km}^{-1}$ .

symmetric with respect to the water shell, the pronounced conical shape and associated convection in the water shell beginning when the volume of ice had decreased to approximately one half. The display of Figs. (2.11) and (2.12) is also useful in assessing the chances of the particles reaching the ground, as this statement implies that the particle still contains a large fraction of its volume in the form of ice.

An examination of Figs. (2.11) and (2.12) shows that the melting proceeds very rapidly in the latter stages; on average, the fraction is increased from 0.5 to 1.0 in only the last 10% of fall. If we assume that the particle may be classified as reaching the ground if it has a volume of ice greater than 50% of its original volume, then the distance of fall is approximately 70% of that for complete melting to take place, and can be written

$$Z_{0.5} = (1.8 a / \alpha^{1/2}) \times 10^6 \quad (2.20)$$

where  $Z_{0.5}$  is the distance of fall from the  $0^{\circ}\text{C}$  level for half the volume of ice to have melted and 'a' and  $\alpha$  are as previously defined. At this stage of melting the water shell thickness is approximately one fifth of the original radius, so Eq. (2.20) also gives the fall distance from the  $0^{\circ}\text{C}$  level for the reflectivity to become equal to that for spheres of the same mass composed entirely of water.

### 2.7.2 Cones

The melting times of solid ice cones in an environment of constant humidity, temperature and wind speed, show differences of up to 10% when compared with spheres of equal volume. In a saturated atmosphere, cones of all shapes considered take longer to melt than the corresponding spheres. Hence, the distance of fall in the atmosphere saturated with water vapour at all levels is likely to lie nearer to the value predicted by Eq. 2.19. In an atmosphere in which there is no condensation or evaporation from the cones, the region of humidity is such that the melting times of the cones are approximately equal to that of spheres of equal volume. Hence the melting time in this type of atmosphere will approximate to that of the spheres of equal volume, and Eq. 2.19 may be used once again to predict the fall distance to within  $\pm 10\%$ . Also, it is reasonable to assume that on this basis Eq. 2.20 will predict the fall distance for the volume of ice to decrease to one half of the original volume.

### 2.7.3 Snow

The preliminary observations do not really justify making definite conclusions, as an insufficient number of samples were examined, and the method of support may well have interfered with the experimental behaviour.

#### 2.7.4 General Conclusions.

Small, solid ice hydrometeors found in the atmosphere, will, in general, have a shape intermediate between that of the sharpest pointed cones used in these experiments and that of perfect spheres. Eq. (2.19) and (2.20) have been found to be applicable to describe the atmospheric melting for both extremes of shape to an accuracy of about 10%, and are thus likely to be applicable to all shapes of solid hydrometeor found in practice.

Hence we may restate that for all natural solid hydrometeors having an equivalent spherical diameter less than about 5 mm, the fall distance for complete melting to take place is given by

$$Z = (2.55 a / \alpha^{1/2}) \times 10^6 \quad (2.19)$$

to within 10%. Also, the ice particles will reach the ground with half their volume remaining as ice if the melting level is at or below a height given by

$$Z_{0.5} = (1.8 a / \alpha^{1/2}) \times 10^6 \quad (2.20)$$

to within 10%. The thickness of the melting band as seen by radar will also be given by Eq. (2.20) with the same level of accuracy, when the particles are of solid form.

## CHAPTER THREE

### An Evaporating Ice Sphere As A Wet-Bulb Thermometer

#### 3.1 Introduction

In spite of the intensive effort in the twentieth century to develop new and accurate methods of measuring humidities, the wet-bulb hygrometer in its many forms is probably still the most versatile and widely used of all instruments. In all the types of wet-bulb hygrometric instrument the calculation of humidity is made by recording the temperature difference between two thermometric devices, one of which is kept dry and the other moistened by water. A psychrometric equation is then applied in order to relate the temperature difference to a difference in vapour pressure. Unfortunately, there is no unique equation that applies to all types of hygrometer, as both the geometry of the system and the way in which it is used enter into the calculations. In practice, it is customary to design and use hygrometers to certain standards laid down by various Meteorological Authorities, and to read vapour pressures and humidities from tables constructed by these authorities on the basis of these standards. Using the wrong tables with such instruments can lead to large errors in the computation of humidities and vapour pressures.

In Britain, it is customary to expose hygrometric instruments in Stevenson Screens, thus restricting the

ventilation to "light wind" conditions. In the United States, the standard instrument is the Assman, or Sling Psychrometer, which utilises the experimental fact that there is a limiting "wet-bulb depression" when the instrument is well ventilated.

When the size of the instrument is reduced by using thermocouples, many of the errors and troubles of the larger instruments using mercury-in-glass type thermometers become more easily controlled. With the smaller geometry it is possible to design instruments that do not suffer appreciably from radiation and conduction errors, and the wind speed at which the limiting wet-bulb depression is reached falls from a few metres per second to some tens of centimetres a second. For these reasons the thermocouple wet-bulb hygrometer has been an important research instrument in the laboratory, and it has become possible to realise a single, practical, psychrometric equation for many different varieties of this instrument which need only conform to a few basic design standards.

To all intents and purposes, an isolated, evaporating water or ice sphere should act as a wet- or ice-bulb thermometer, and their surfaces should attain the wet- or ice-bulb temperature of the air stream, subject to the limits of water purity and radiation errors. For this reason

they may act as the standard devices for determining the quality of given hygrometric instruments, providing the surface temperature of the sphere can be accurately determined. In much of the previous work with water and ice spheres, the assumption that the surface temperature is the same as that which would be recorded by a well designed wet- or ice-bulb thermometer in the same air stream has been the basis of calculations on the rates of evaporation. It seems desirable that this assumption should be put to direct experimental test. For this reason, a direct-comparison experiment was undertaken with the wind tunnel described previously. The hypothesis was tested by examining the conditions under which an ice-bulb and an ice sphere will just begin to melt when placed in the same air stream at temperatures between 0°C and 10°C.

### 3.2 Theory

The transfer of water vapour and heat between a stationary ice sphere and a moving air stream of constant velocity, temperature and humidity may be expressed by the following equations

$$dm/dt = 4\pi r D C_m \{ \rho(T) - \rho(T_s) \} \quad (3.1)$$

and

$$L_s dm/dt = 4\pi r K_a C_h (T_s - T) \quad (3.2)$$

where  $dm/dt$  is the rate of change of mass of the sphere of radius  $r$ ,  $D$  is the diffusion coefficient of water vapour in air,  $K_a$  the thermal conductivity of air,  $L_s$  the latent heat of sublimation of ice,  $\rho(T)$ ,  $\rho(T_s)$  are the concentrations of water vapour in the remote environment and at the surface of the sphere respectively,  $T$ ,  $T_s$  are the temperature of the environment and of the surface of the sphere respectively, and  $C_h$ ,  $C_m$  the coefficients of vapour and heat transfer to a sphere in a moving air stream. In the steady state, equations (3.1) and (3.2) may be combined to give

$$\rho(T) - \rho(T_s) = \frac{K_a}{L_s D} \frac{C_h}{C_m} (T_s - T) \quad (3.3)$$

and, if the air in the immediate vicinity of the sphere is assumed to be saturated at the surface temperature  $T_s$ , we have

$$\frac{e_{sat}(T_s)}{T_s} - \frac{e}{T} = \frac{K_a}{L_s D} \frac{C_h}{C_m} \frac{R}{M_w} (T - T_s) \quad (3.4)$$

where  $e$  denotes vapour pressure,  $R$  is the Universal Gas Constant and  $M_w$  the molecular weight of water. If  $-5^\circ\text{C} < T_s < 10^\circ\text{C}$  and  $(T - T_s) < 10^\circ\text{C}$ , equation (3.4) may be expressed in the normal form of the psychrometric equation

$$\begin{aligned} e_{sat}(T_s) - e(T) &= \frac{K_a C_h R}{L_s C_m D M_w} \left( \frac{T + T_s}{2} \right) (T - T_s) \\ &= \frac{C_p}{L_s} \frac{p}{\epsilon} \frac{K}{D} \frac{C_h}{C_m} (T - T_s) \\ &= A_i (T - T_s) \end{aligned} \quad (3.5)$$

to within an accuracy of  $\pm 2\%$ , where  $p$  is the atmospheric pressure,  $\xi$  the density of water vapour relative to air,  $K$  is the thermal diffusivity of air, and  $C_p$  the specific heat of air at constant pressure.

Evaluation of the Psychrometric Constant,  $A_i$ , requires numerical values for  $C_h$ ,  $C_m$ , and  $D$ . None of these quantities has been determined with great accuracy, but recent careful measurements of rates of evaporation of ventilated water drops by Ranz and Marshall (1952) and of ice spheres by Thorpe and Mason (1966) are consistent with  $C_h/C_m = (5.0 \pm 0.2) \text{ cm}^{-2} \text{ sec}$  at  $0^\circ\text{C}$  and 1000 mb over the range of Reynold's Numbers  $10 < \text{Re} < 500$ . If this figure and appropriate values of the other parameters are substituted in equation (3.5), we find that for  $-5^\circ\text{C} < T_s < 0^\circ\text{C}$ ,  $p = 1000 \text{ mb}$

$$A_i = (0.54 \pm 0.03) \text{ mb } ^\circ\text{K}^{-1} \quad (3.6)$$

The corresponding value for a water sphere is obtained by replacing  $L_s$  by the latent heat of condensation to give

$$A_w = (0.61 \pm 0.03) \text{ mb } ^\circ\text{K}^{-1} \quad (3.7)$$

Turning now to the thermocouple psychrometer, errors may arise due to radiative heat transfer between the wet junction and its warmer surroundings which

are not allowed for in the theory, and by conduction of heat down the wires. Pasquill (1949) has shown that the conduction error may be eliminated by ensuring that the 'wet-bulb depression' in a given air stream is independent of both the aspiration rate and the length of the wick. Monteith (1953) discusses the inherent radiation error and how this depends on the geometry and the aspiration rate. He shows that for a 40 swg copper/contra junction covered with a single layer of cotton thread over a length of more than about 2 cm, 99% of the theoretical limiting wet-bulb depression will be obtained when placed at right angles to an air stream flowing at more than  $1 \text{ m sec}^{-1}$ . Under these conditions Monteith calculates the constant to be

$$A_w = 0.624 \text{ mb } ^\circ\text{K}^{-1} \text{ at } p = 1000 \text{ mb}$$

which, although based on early measurements of heat transfer to long cylinders, is very close to the value derived for an evaporating water drop.

### 3.3 Experimental

The behaviour of ice spheres and ice coated thermojunctions under the same conditions was compared by determining the temperature and humidity conditions under which their surfaces attained  $0^\circ\text{C}$  and just began to melt in a moving air stream. The thermocouple psychrometer consisted of two welded 40 swg copper/contra

junctions, one of which was wound with a single layer of clean cotton thread up to a distance of 3 cm on either side of the junction. Distilled water was applied to this wick with a small paint brush and the resulting ice-coated junction was placed at right angles to the air stream in the wind tunnel described in Chapter Two. The second junction, acting as a reference, was placed in a stirred ice/distilled-water mixture, whose temperature as recorded by a standard mercury-in-glass thermometer was always  $0^{\circ}\text{C} \pm 0.02^{\circ}\text{C}$ . The air temperature was recorded by a similar pair of thermocouple junctions without wicks, one of which was placed about 1 cm below the ice covered junction and the other in the ice/distilled-water mixture.

Ice spheres of diameter 0.5 mm to 2.0 mm were formed from distilled water and suspended in the tunnel at the same height as, and about 2 cm from the ice coated junction, on a fibre of  $10\mu$  diameter. Both the ice sphere and the ice-coated junction were illuminated by a microscope lamp through a glass heat filter and a 10 inch column of 5% cupric chloride solution. This gave a diffuse blue light that was ideal for observing changes of surface detail on the ice specimens through a low-power telescope.

That changes of  $\pm 1.0 \text{ m sec}^{-1}$  in air speed about the usual value of  $3.6 \text{ m sec}^{-1}$ , and changes of  $\pm 2 \text{ cm}$  in the wick length produced no observable change in the

temperature of the ice-coated junction indicated that the conduction error was negligible. Moreover, calculations suggest that heat transfer by conduction along the supports would have been less than 0.1% of that conducted to the junction through the air. The ice sphere and ice junction were in the same radiation field and, in any case, the geometry and aspiration rate were such that, according to Monteith, the radiation error to the thermocouple was less than 1%.

Having placed the ice specimen in position, the air stream was allowed to attain a constant velocity while being heated through  $0^{\circ}\text{C}$  at a rate of about  $0.05^{\circ}\text{C}$  per minute. By starting with nearly saturated air at  $0^{\circ}\text{C}$  and allowing some exchange between this air and the drier air of the cold room between each run, the ice surfaces could be made to melt with air temperatures ranging from  $0^{\circ}\text{C}$  to  $10^{\circ}\text{C}$ . The onset of melting was judged when microscopic details on the ice surfaces began to disappear, and the reflected highlights became smooth and bright. At this stage the temperature of the air stream was held constant.

Under all conditions the onset of melting on the surfaces of the ice spheres and of the ice-coated junction were judged to coincide to within  $0.05^{\circ}\text{C}$ , and melting began over the whole surface of the sphere simultaneously. In all cases the onset of melting

coincided with a wet junction reading of  $-0.05 \pm 0.05^{\circ}\text{C}$ , this reading becoming  $0^{\circ}\text{C} \pm 0.05^{\circ}\text{C}$  within a few seconds of the air temperature becoming constant.

### 3.4 Conclusions

Providing that a thermocouple hygrometer is designed to eliminate radiation and conduction errors, by using the principles suggested by Pasquill (1949) and Monteith (1953), it will record a temperature of  $0^{\circ}\text{C}$  at the same instant as the surface of an ice sphere just becomes wet in the same air stream, over the range of air temperature  $0^{\circ}\text{C}$  to  $10^{\circ}\text{C}$  and associated humidity between 100% and 0%. Although this only demonstrates the equivalence of an ice sphere and an ice-coated thermojunction at a particular surface temperature, i.e.  $0^{\circ}\text{C}$ , it is reasonable to assume that the surface temperature will be the same over the complete humidity range 0% to 100% at temperatures between  $0^{\circ}\text{C}$  and  $10^{\circ}\text{C}$ .

The results also imply that equation (3.3) is applicable to shapes other than spheres, providing their radii of curvature are sufficiently large so that there is no reduction of vapour pressure at the surface due to curvature of the surface. That is, any modification of the diffusion coefficients  $C_h$  and  $C_m$  by shape factors  $F_h$  and  $F_m$  has to retain the property that  $F_h/F_m \approx 1.0$ .

PART TWO

"Electrification of Melting Ice and Snow"

## CHAPTER FOUR

### Introduction to the Electrification Associated with the Melting of Ice.

#### 4.1 The electric double layer

It was seen in Chapter One that the charging associated with the melting of ice is thought to be due to the bursting of air bubbles, that are released from the ice during melting, at the air/water interface. The separation of charge is considered to be an effect of stripping off a portion of the electric double layer at the air/water surface and the bubble surface, by the droplets formed by the bursting bubbles. The mathematics of this electric double layer has been treated in detail by Debye and Huckel (1923), Chapman (1913) and Gouy (1917) and involves a complicated analysis of the molecular properties of surfaces. A summary of the simpler concepts is well presented by Adamson (1960), and this summary forms the basis of the treatment presented here.

If, for any reason, a plane surface of a liquid containing ions of both sign carries a uniform charge density,  $\sigma$ , then the surface may be characterised by a potential  $\psi_0$  which decreases as one proceeds into the liquid where a state of dynamical electrical neutrality exists. If  $\psi$  is the potential at any depth  $x$  from the surface, we may write three equations which determine the rate of decrease of this potential as a function of

the distance,  $x$ , from the surface.

The first equation determines the probability of finding ions of either sign at a position characterised by the potential,  $\psi$ .

$$\begin{aligned} n^- &= n_0 e^{+ze\psi/\kappa T} \\ n^+ &= n_0 e^{-ze\psi/\kappa T} \end{aligned} \quad (4.1)$$

where  $e^{ze\psi/\kappa T}$  is the Boltzman factor for ions of charge  $ze$ ,  $e$  being the electronic charge and  $z$  the valence of the ions.  $n_0$ ,  $n^+$  and  $n^-$  are the concentrations of ions far from the surface, and at a depth characterised by the potential  $\psi$  respectively.

The second is formed from the knowledge that the integral of the net charge density,  $\rho$ , at all points in the liquid must be equal and opposite to the surface charge. Hence for a plane surface we may write for unit cross-section

$$\begin{aligned} \sigma &= - \int_0^\infty \rho dx = - \int_0^\infty ze(n^+ + n^-) dx \\ \text{or } \sigma &= -2n_0ze \sinh(ze\psi/\kappa T) \end{aligned} \quad (4.2)$$

Finally, the charge density at any point is related to the potential gradient at that point by Poisson's equation,

$$\nabla^2 \psi = -4\pi \rho / D \quad (4.3)$$

where  $D$  is the dielectric constant of the medium.

Combining equation (4.2) and (4.3) for the case of an infinite plane surface we have,

$$\frac{d^2\psi}{dx^2} = \frac{8\pi n_0 z e}{D} \sinh(ze\psi/kT) \quad (4.4)$$

If we assume  $ze\psi \ll kT$ , then the exponential terms of the hyperbolic function may be expanded in a series, and only the first few terms need be considered. On doing this, we have

$$\frac{d^2\psi}{dx^2} = 8\pi n_0 z^2 e^2 \psi / DkT = \kappa_g^2 \psi \quad (4.5)$$

When a mixture of ion types is involved, then

$$\kappa_g^2 = (4\pi e^2 / DkT) \sum_i n_i z_i^2 \quad (4.6)$$

The solution of equation (4.5) is exponential in form, and for the case of  $ze\psi \ll kT$ , (singly charged ions at room temperature) it reduces to the simple form

$$\psi = \psi_0 e^{-\kappa_g x} \quad (4.7)$$

The quantity  $1/\kappa_g$  is hence the distance at which the potential falls to  $1/e$  of its value at the surface, and determines the effective thickness of the double layer.

Since the double layer thickness is determined by the quantity  $1/\kappa_g$ , it can be seen from equation (4.6) that increased concentrations of ions in solution will lead to a decrease in the layer thickness. For monovalent

ions at room temperature, the layer thickness is typically of order  $100 \text{ \AA}$  for ionic impurity concentrations of  $10^{-2}$  to  $10^{-3}$  moles/litre.

Although this simple treatment of the theory of the electric double layer is useful in giving typical values to some of the quantities involved, in order to give a more complete description which takes into account the finite size of ions and the interionic actions, it is necessary to develop a much more complex mathematical treatment that is not justified for the purposes of this work. However, some of the features of the more complex analysis are important, and the main conclusions of the arguments are presented below.

Rather than the single diffuse layer of charge extending into the liquid suggested by the simple theory, Stern (1924) considers two separate regions. One, close to the surface, forms a compact double layer a few molecular diameters thick and is rigidly attached to the surface. The second is similar to the diffuse region previously described and is mobile in the sense that it is not resistant to shear movements in the liquid. This aspect of mobility of the double layer is obviously important when we come to consider the motion of bubbles rising through the liquid.

In the case of 'pure' water, from which all ionic impurities have been removed, the surface charge associated with the double layer is supposed to arise

from the statistical arrangement of hydroxyl groups at the surface. At room temperature there is a statistical excess of about 4% of hydroxyl groups with their oxygen atoms directed away from the surface, thus giving it a negative charge layer. The charge density as computed from the solution of equation (4.2) is about  $60 \text{ esu/cm}^2$  for 'pure' water at room temperature.

When ionic impurities are added to the water, the double layer thickness is reduced as previously indicated. At some critical concentration the sign of the surface charge layer can be reversed. From the discussion, it would appear that the exact chemical nature of the ionic impurity is at least only of secondary importance (perhaps in modifying the surface properties of the water) and one might expect essentially similar electrical effects for the same concentrations of ionic impurity independent of their chemical nature.

In this treatment, the finite time of the electric double layer to form after the creation of a new surface has not been dealt with; nor can any reference be found to a mathematical treatment of the phenomenon. Iribarne (1966a) estimates that for laboratory prepared conductivity water from an ion exchange column, the time constant is of order  $10^{-1}$  second.

#### 4.2 Bubbles bursting at a water/air interface

The dynamics of the bubble bursting process has

been studied with the aid of high speed photography by Woodcock et al (1953), Kienzler et al (1954) and Knelman et al (1954). This has revealed that the bubble rises into the surface and forms a thin hemispherical cap. As the liquid drains from this cap, a point is reached when the film ruptures. The water then rushes into the sides of the cavity so formed, and a thin jet rises in the centre and breaks up into a few small drops of order  $1/10$  th the bubble diameter. These drops are ejected vertically away from the surface at very high velocity. (See Fig (4.1))

Besides the drops formed from the break up of the central jet, Mason (1954) and Blanchard (1963) have found that for bubbles with diameter greater than about 0.5 mm, a few small drops of order 10 microns diameter are formed from the rupture of the cap. These are usually ejected at low angles to the horizon. Mason (1954), working with a small expansion cloud chamber, found that besides the drops of order 10 microns diameter formed from the rupture of the cap, a few hundreds of sub-micron size drops were also formed close to the surface.

It is essential to distinguish between the two sources of drops formed during the bursting process. Henceforward, the drops formed from the rupture of the water film will be called "film drops" and those from break up of the jet "jet drops".

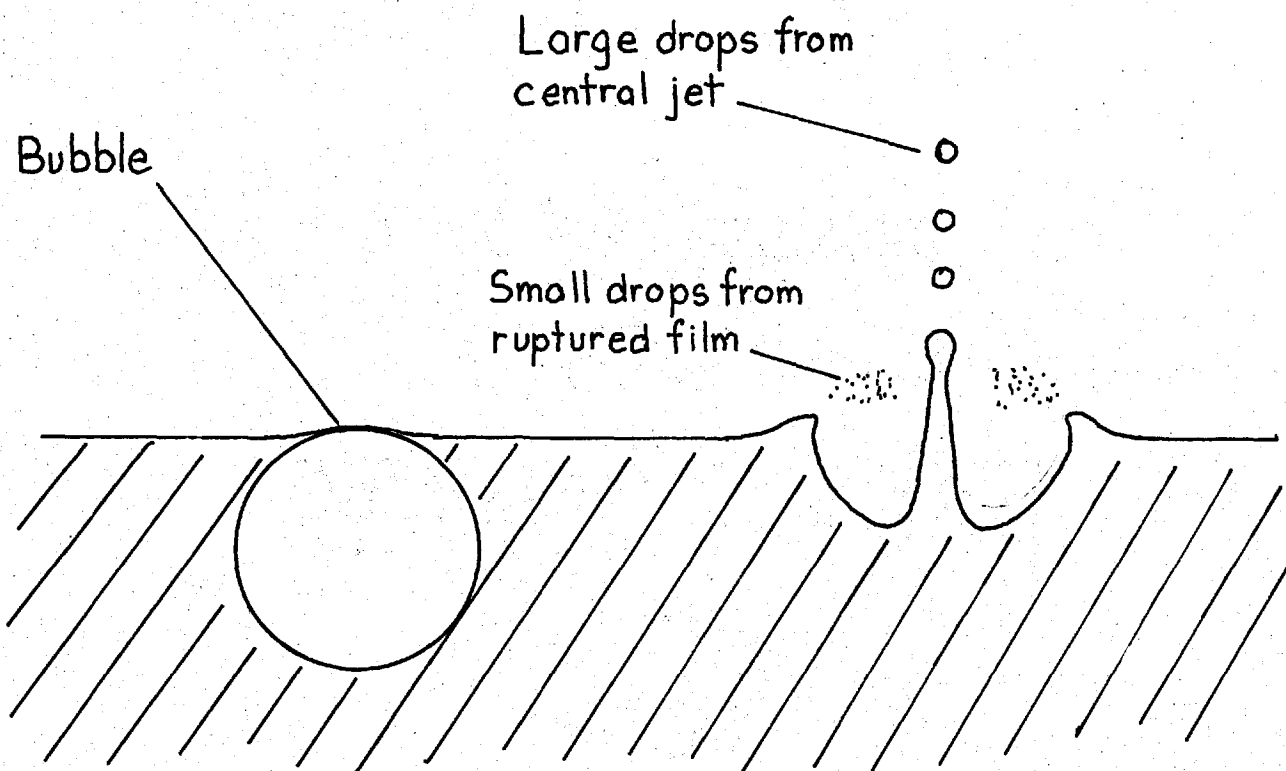


Fig. 4.1 Drop production from bursting bubble.

Blanchard (1963), in an extensive investigation into the bursting of air bubbles in the size range 0.1 mm to 5 mm diameter at a sea water surface and a distilled water surface, found a number of effects which are relevant to the interpretation of how charge is separated by this process. With these bubbles, a mean of about 10 film drops were formed from the larger bubbles, and only 1 or 2 for bubbles of less than 0.5 mm diameter, when bursting in sea water. With his apparatus the sub-micron size drops noted by Mason would probably have escaped detection. The effects he discovered are listed below.

i) Small traces of surface active agents, (organic oils), introduced to the salt water surface entirely eliminated the production of detectable film drops. This was considered to be due to stabilisation of the film by duplex layers of the oils.

ii) With bubbles of less than 0.5 mm diameter, none of the organic oil film could be detected in the jet drops. Blanchard shows that for these smaller bubbles, the process of jet ejection after rupture of the surface film takes place so rapidly that the surface does not have sufficient time to spread into the central cavity. *how is it formed*

iii) Bubbles rising into a surface in salt water had a much longer life at the surface before bursting than they did in distilled water. In some cases with salt

water the bubbles rose to a maximum height then partially sank back into the liquid before bursting. In distilled water the bubbles did not appear to show any deceleration in their upward motion before bursting. Since bubbles which sink back into their liquid before bursting will have a much smaller cap area, the number of film drops produced in salt water is likely to be smaller than that for distilled water.

iv) Organic oils present on the surface did not prevent the appearance of film drops if the air bubbles rose to the surface in clusters, or at the same point in rapid succession. In this case the effect of bursting in rapid succession was to renew the water surface between each event and thus prevent the surface film from becoming stabilised in the region where the bubbles were bursting.

v) By "ageing" the bubbles in the bulk solutions before allowing them to burst at the surface, the number of film drops was reduced. This was thought to be due to the absorption of long-chain organic molecules into the bubble surface during the long life of the bubble in the water.

Before proceeding to discuss these results in relation to experiments on charge separation, it is essential to try to determine where the water is drawn from that goes into the two types of drop. Harper (1957) suggests that the limit of film thickness for the bubble cap before bursting is of order  $100 \text{ \AA}$ . Since this film

has two sides, it may be expected that providing the double layer is of greater thickness than about  $50 \text{ \AA}$ , a separation of charge might occur on the film drops. However, the situation is complicated if one considers the formation of the bubble film as due to the squeezing of the water from the surface region by the rising bubble. See Fig. (4.2a). In this case, the cap will contain electric double layers from two different sources: one from the bulk liquid surface, the second from the bubble surface in the interior of the liquid. The final state of the film before rupture would be difficult to predict since the motion of the bubble through the liquid may have caused it to shed all but the non-mobile, Stern portion of the electric double layer discussed in section (4.1). This is typically a few molecular diameters thick. On the other hand, the bulk water surface will in all probability contain surface active agents of one sort or another in any practical case, even under the cleanest laboratory conditions available.

Although Harper (1957) considers the bursting of the bubble cap to be the agent for charge separation, the experiments of Blanchard (1953) on the electrification of bubbles bursting at a sea-water surface and a distilled water surface, showed that the charge on the jet drops was approximately 100 times the charge on the film drops. However, the experiments of Mason (1954) suggest that about 100 drops of sub-micron size may well have

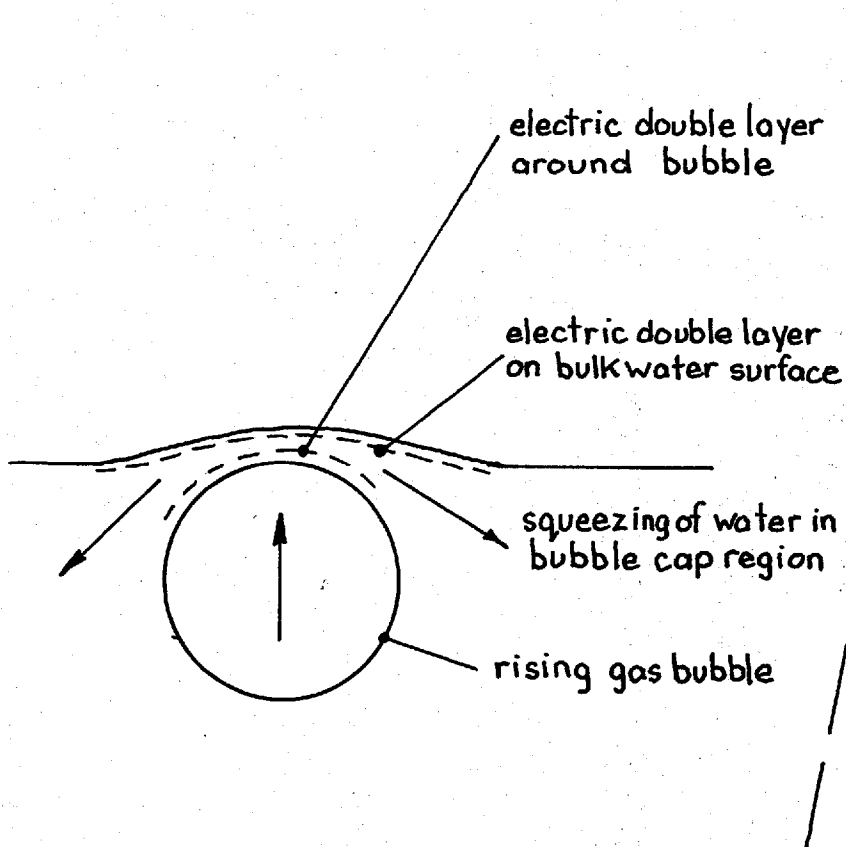


Fig. 4-2(a) Squeezing of water to form cap by bubble rising into surface.

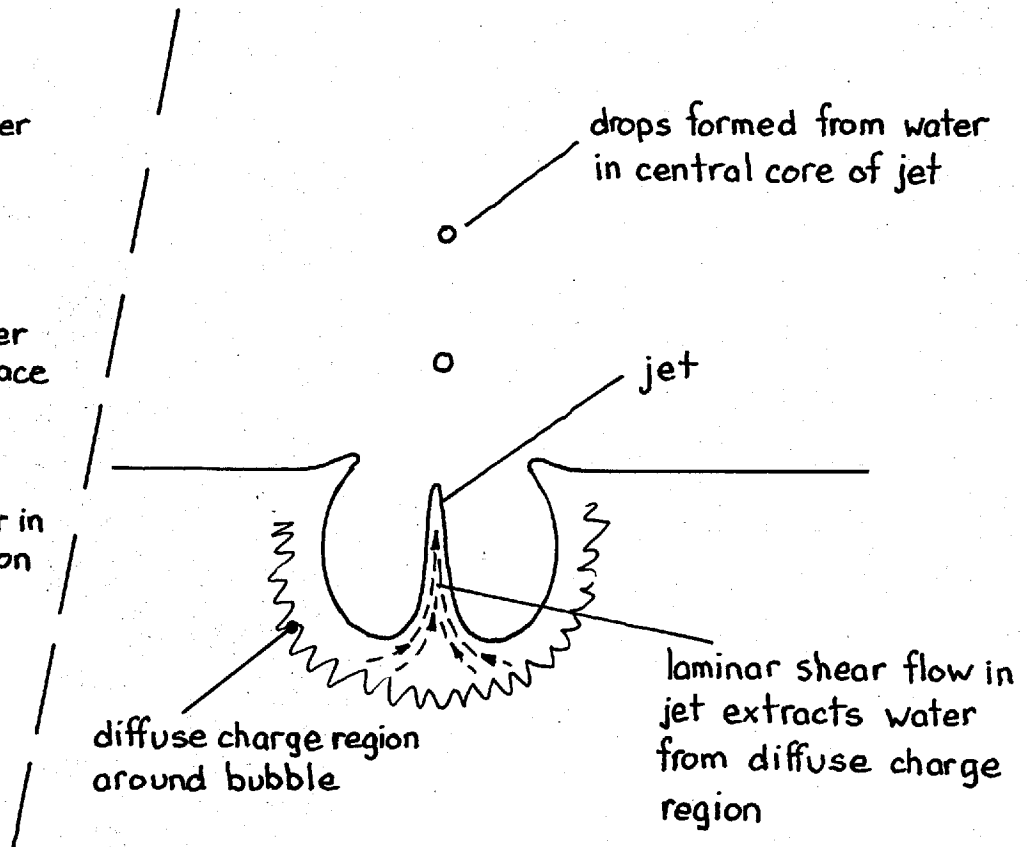


Fig. 4-2(b) Blanchard's conception of water flow in rising jet.

escaped detection in Blanchard's work, and if these drops were to carry only 1% each of the charge on the jet drops the contribution from both sources would be of the same order of magnitude, and possibly the relative importance of the two sources reversed.

Turning to consideration of the water source for the jet drops, Blanchard suggests that laminar shear flow occurs in the rising central jet, the water going to form the drops coming from the central core of this jet. See Fig. (4.2b). In this way, the water from the diffuse region of the electric double layer at the base of the bubble predominates in the drops formed by the break up of this jet, rather than water which is drawn from the interior bubble surface. In the case of large bubbles (diameter greater than 1 mm) the evidence conflicts with this suggestion, as the organic oil films placed on the bulk liquid surface manifested themselves in the jet drops. This shows that at least part of the water going into these drops must have come from the layers close to the surface. It will also be seen that the concept is at variance with the sign of the charge found by Blanchard on the jet drops, discussed in the next section.

#### 4.4 Charging experiments with bursting bubbles

Iribarne (1966a) has carried out a series of experiments on the bursting of nitrogen bubbles at a water surface. The bubbles were formed from a capillary

tube and rose through a sufficient depth of water to give them a lifetime of more than one second before reaching the surface and bursting. This gave a reasonable assurance that the double layer would be well established round the bubbles. The water surface at which the bubbles burst was continuously renewed by allowing it to overflow its container. The debris from the bursting was collected by blowing a slow stream of nitrogen over the surface, and into a copper filter. The charge on the debris was recorded by measuring the current through a Vibron Electrometer connected to this copper filter. The nitrogen stream also served to keep the water surface in slow motion in the bursting region, thus preventing 'rafting' of the bubbles at high bubbling rates, at their point of emergence on the surface. The results of his experiments are shown in Figs (4.3) and (4.4) for water samples at 25°C containing impurities due to different concentrations of NaCl and CO<sub>2</sub>. The effects due to CO<sub>2</sub> are interpreted as the formation of Carbonic acid in solution, H<sub>2</sub>CO<sub>3</sub>, and its subsequent dissociation into H<sup>+</sup> and HCO<sub>3</sub><sup>-</sup> ions. Experiments were also conducted at +2°C, the form of the curves in the two cases were essentially similar.

It can be seen from the graphs that the charge is critically dependent on the size of the bubbles in the range 50 to 200 microns radius. The experiments with 'pure' water (conductivity better than 10<sup>-7</sup> mho cm<sup>-1</sup>)

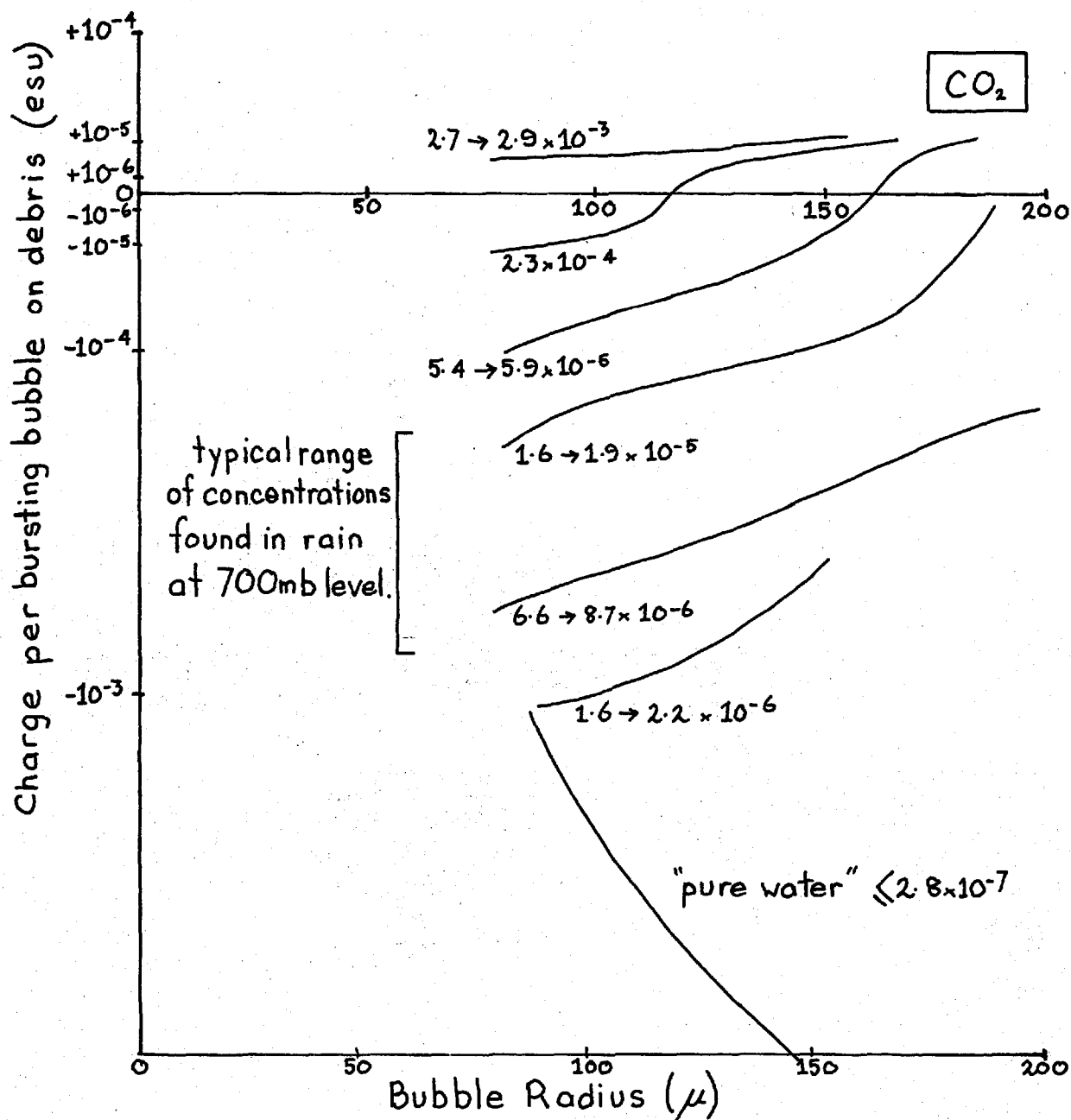


Fig. 4.3

Variation of net charge on the debris per bubble bursting event as a function of radius, and for a range of  $CO_2$  concentrations in moles/l. Water temperature  $\sim 25^\circ C$ . Nitrogen bubbles. After Iribarne (1966 a).

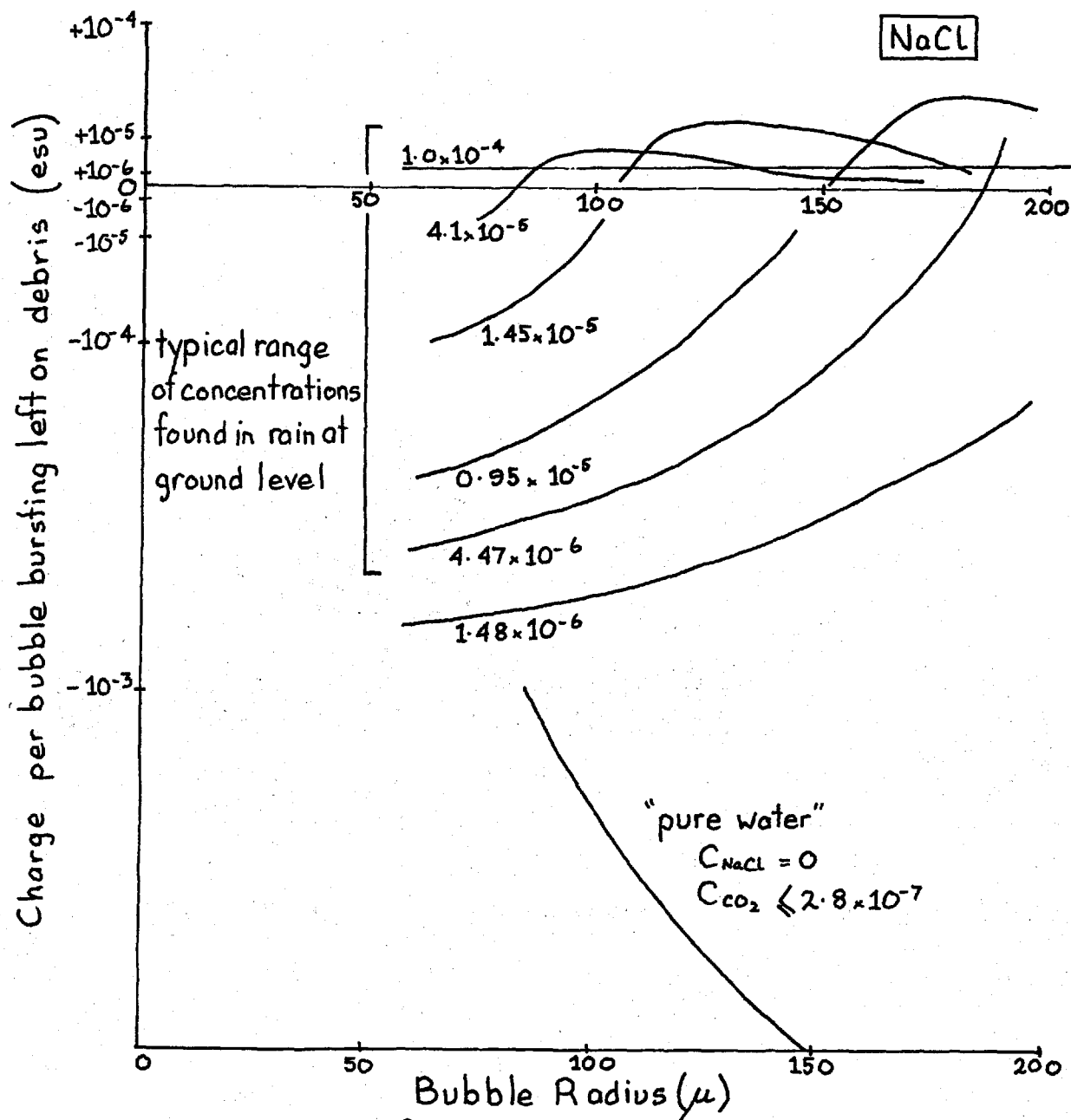


Fig. 4.4 Variation of net charge on the debris per bubble bursting event as a function of radius, and for a range of NaCl concentrations in moles/l. Water temperature  $\sim 25^\circ\text{C}$ . Nitrogen bubbles. After Iribarne (1966a).

indicate an increasing charge per bubble left on the debris with increasing bubble size in this range, whilst all the water samples with impurity concentrations in the range  $10^{-7}$  to  $10^{-4}$  moles/litre show a decreasing negative charge per bubble left on the debris with increasing bubble size, and eventual reversal of sign of the charge to small, positive values. This type of behaviour was typical of all the other types of ionic impurities used by Iribarne besides NaCl and  $\text{CO}_2$ , as might be expected from the arguments of section 4.1.. For concentrations greater than about  $10^{-4}$  moles/litre, the charge per bubble was found to be nearly independent of the bubble size and had small positive values of order  $10^{-6}$  esu per bubble. Typical ranges of the concentrations of NaCl and  $\text{CO}_2$  found in rain water are indicated on the graphs.

Blanchard (1963) found that the jet drops and film drops in his experiments previously described always had a positive charge of order  $10^{-7}$  esu (jet) and  $10^{-8}$  esu (film): the total charge per bubble was then of order a few times  $10^{-7}$  esu. This is consistent with the results found by Iribarne for NaCl concentrations greater than  $10^{-4}$  moles/litre and at higher radii.

Although Iribarne follows Blanchard's argument in supposing the main charge to be carried by the drops from the central jet, his experiments were unable to

decide the relative importance, or even the signs, of the jet and film drops, but recorded only the net charge separated on all the debris of the bursting. If the jet drops at high impurity concentrations carry positive charge as he supposes, then it is difficult to reconcile this with the water having come from the diffuse layer of charge at the base of the bubbles, for at these high impurity concentrations a reversal of sign of the double layer effect will have taken place. That this is the case is indicated by Iribarne's results which show predominantly negative charge on the drops for ionic concentrations of less than about  $10^{-4}$  moles/litre. In fact, it would seem that his experiments indicate that the charge comes predominantly from the surface layers of either the bubbles and/or the bulk water surface. This is also indicated by the extreme sensitivity of the charge magnitude to small traces of surface active agents on the bulk water surface noticed by Iribarne, and also by Dinger and Gunn (1946) in their experiments with melting ice. If the jet drop water came from below the under-surface of the bursting bubble, then the surface active agents which have to spread into the cavity and up the sides of the jet would not have sufficient time to be incorporated into the jet drops. However, it must be pointed out that if the bubble film drops contribute significantly to the charge separation, then the surface

active agents might well have their effect in reducing the charge carried by the film drops, thus reducing the net separation of charge.

Calculations by Iribarne show that the magnitudes of the charges separated by bursting bubbles are consistent with it being a double layer effect. The variation of the magnitude of the charge with bubble size may be qualitatively interpreted as due to the extraction of different depths of water from the double layers in going to form the drops of both types on bursting.

In conclusion it may be stated that experiments with bursting bubbles at water surfaces containing ionic impurities show a qualitative agreement with the charge separation being primarily due to disruption of electric double layers. The exact mechanism by which this occurs is still very much in doubt.

There seems to be no immediate reason why the results of an experiment with bubbles bursting at a water surface after release from ice during melting, should differ materially from the results found by Iribarne and Blanchard, for similar bubble sizes and impurity concentrations. However, if the nature of the melting of solid ice hydrometeors in the real atmosphere is considered, one might expect differences due to the dynamics both of the internal convective motions in the melting particles described in Chapter Two, and the drag

forces at the air/water surface due to the high wind velocity. Before going on to outline the experiments designed to investigate the self charging of melting ice, it will be useful to review the bubble structure and impurity concentrations of atmospheric hydrometeors..

#### 4.5 Properties of Atmospheric Ice

The size and distribution of bubbles in ice depends on the freezing rate as well as the volume of dissolved gases in the water prior to freezing. Both these are linked to the temperature of the environment in which the freezing takes place. Because of the variability of these conditions found in the atmosphere, the sizes of bubbles which occur in particles found in the atmosphere range from about 1 micron to a few millimetres in diameter. For instance, in falling snow, the bubbles are typically a few microns in diameter and in solid hydrometeors of a few millimetres diameter the bubbles are typically from tens to hundreds of microns diameter. In large hailstones the upper limit is extended to millimetre size due to the trapping of air spaces during coagulation of individual particles.

An analysis by Junge (1963) of the salt concentrations in rain finds that in Continental rain the concentration of NaCl is typically in the range  $10^{-5}$  to  $10^{-6}$  moles/litre, and in Maritime rain the upper limit is of order  $10^{-4}$

moles/litre. Concentrations of other salts such as  $(\text{NH}_4)_2\text{SO}_4$  and  $\text{MgSO}_4$  showed similar concentrations. The value for  $\text{NaCl}$  can be much higher in coastal regions, reaching a figure of  $10^{-3}$  moles/litre in some cases. If the concentration of  $\text{CO}_2$  is calculated by assuming the water to be in equilibrium with the atmospheric concentration of  $\text{CO}_2$  at any level, then the range is  $10^{-5}$  to  $10^{-6}$  moles/litre. The analysis of rain at the ground will not necessarily be typical of that found at higher levels in the atmosphere, as scavenging will take place in the lower regions; however, since it appears from Iribarne's work that the charge per bubble will decrease with increasing concentration of these impurities, by using these ranges in the following experiments on melting, a lower limit may be fixed for the charging rate likely to occur in the atmosphere.

The results of Iribarne's work suggests that there is an upper limit (of order a few hundred microns radius) above which the charge separation becomes unimportant for typical concentrations of  $\text{CO}_2$  and  $\text{NaCl}$ . It should be noted that the main variation in his results occurs in precisely the range of impurities typical of the atmosphere, and for bubble sizes typical of atmospheric ice particles.

#### 4.6 Previous experiments on charging of melting ice

The results of previous work in the field of

electrification associated with the bursting of bubbles in melting ice has been reviewed in Chapter One. However, it is worth recording the possible variables that have been found, or suspected, of controlling the magnitude of these charges. These are listed below.

- i) Ionic impurity concentration in the ice.
- ii) Size and distribution of bubbles in the ice.
- iii) Nature of gas used to form bubbles.
- iv) Nature of gas in melting environment.
- v) Rate of melting of the ice.
- vi) Dynamics of the bursting process. i.e. coalescence, rafting, frictional drag, convection in the water.
- vii) Surface contamination.

Because many of these factors are interrelated in a complex way, it was decided to split the investigation into two parts. The first would look at the melting of large specimens of ice of up to about a few  $\text{cm}^3$ , in a small horizontal wind tunnel, and concentrate on examining the effects of bubble size, purity of the sample and nature of the environment, to see if the results of Iribarne's work could be applied to the generation of charge during melting of ice. The second part would use a modified form of the vertical wind tunnel used to study the heat transfer problem, and concentrate on seeing if the dynamics and melting rates were likely to have any effect on the rate of charge production under conditions likely to be applicable to the atmosphere.

## CHAPTER FIVE

### Development of apparatus and preliminary results.

#### 5.1 Introduction

This chapter deals with the development of apparatus and techniques for the manufacture of bulk ice specimens of controlled purity and bubble structure. Preliminary tests with ice samples grown in bulk, and either melted in bulk or divided into samples of about 1 to 5 mm diameter, yielded unsatisfactory results for reasons which will be discussed below. The techniques for melting the specimens were also explored, as the charging levels were found to depend critically on the manner in which the specimens were handled and melted. The system finally adopted was one in which the ice specimens were made and melted in a completely closed environment, and were not handled at any stage of the experiment. In order to do this it was necessary to abandon that part of the experiment designed to examine the charge per bubble, and to concentrate on relating the charge to a mean distribution of bubble sizes formed by freezing the ice specimens at different rates, and melting under conditions resembling those likely to be found in the atmosphere.

##### 5.2.1 Water production

In order to obtain ice specimens of controlled purity and bubble size, density and gas content, it was decided that the best starting point would be to construct

apparatus to produce de-gassed, de-ionised water. This 'pure' water could then be doped with known concentrations of impurity, and bubbles introduced into it via a capillary connected to a gas cylinder, whilst the freezing process took place. The method of doing this is shown in Fig. (5.1). All connections between sections of apparatus and glass tubing were either standard ground glass cone or ball joints. Before use, all glass components were cleaned by immersion in hot chromic acid for thirty minutes, followed by washing in distilled water.

De-ionised water which had a conductivity close to the intrinsic value was obtained from an Elgastat de-ionising column. This water was stored under nitrogen in a 10 litre glass reservoir. By opening a tap, a sample of the water could be forced into a flask through Pyrex tubing. The opening from this flask to the atmosphere was fitted with a cotton wool filter and 'U' tubes containing Sodium Hydroxide, to absorb particulate matter and  $\text{CO}_2$  entering the flask.

The water in the flask was then boiled for a period of about ten minutes to drive off dissolved gases. The 'U' tubes could be by-passed during boiling to prevent them from filling with water. After boiling, the water was allowed to flow through a counter-current heat exchanger made of Pyrex and then through a mixed resin de-ionising column and sintered glass filter. The conductivity-

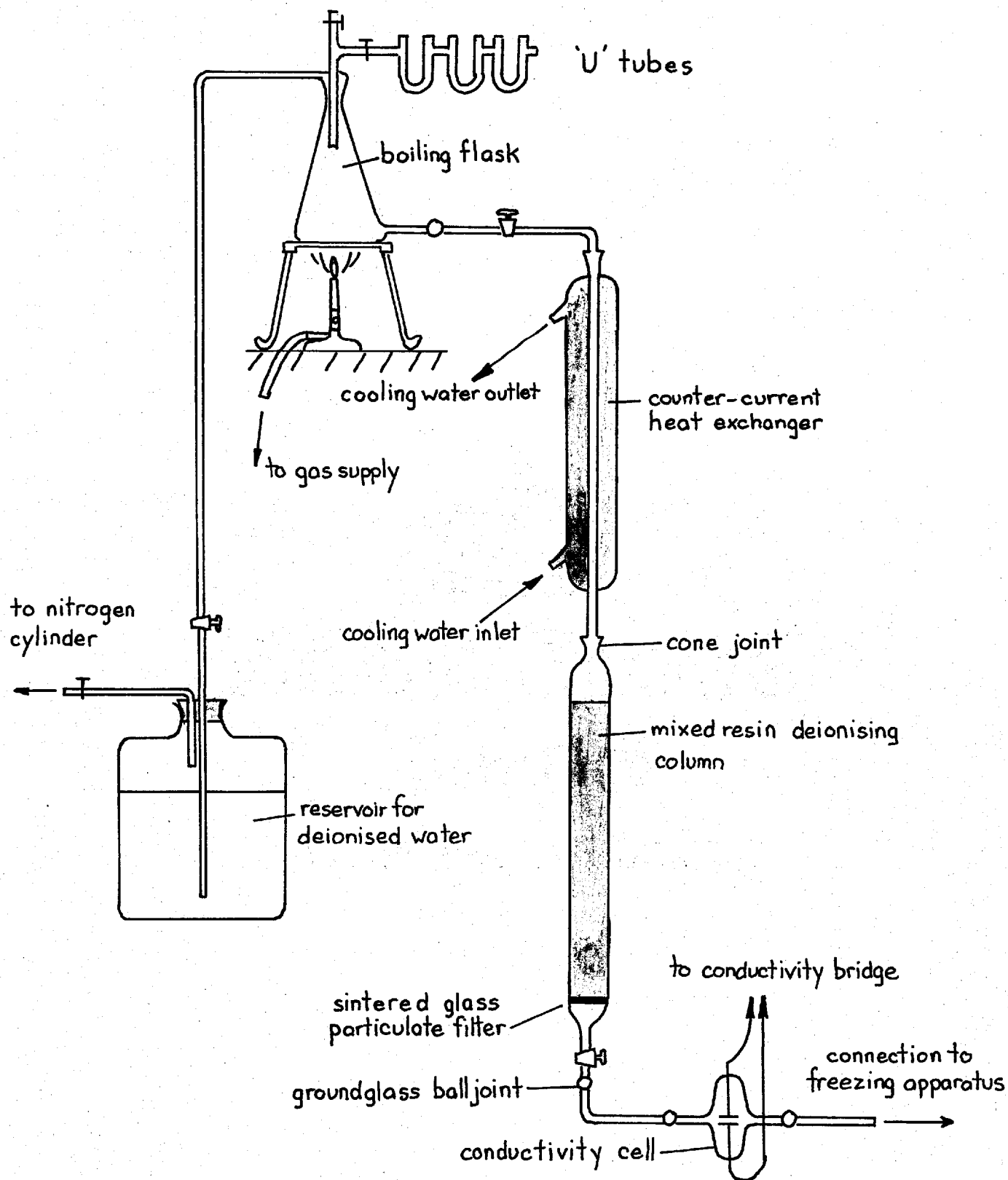


Fig. 5.1 Layout of system for production of degassed and deionised water.

of the water emerging from the column was monitored by a conductivity cell with bright platinum electrodes. After flushing the column with about 10 litres of de-gassed water, it was found that the water had a constant conductivity reading of  $0.045 \mu \text{ mho cm}^{-1}$  at  $20^\circ\text{C}$ . The cell was then disconnected and used in the next section of the apparatus, which was used with the two types of freezing techniques described in sections 5.2.2 and 5.2.3.

#### 5.2.2 Bulk specimen production. Method (i)

In order to produce ice specimens of controlled bubble size and density from the pure water the apparatus shown in Fig. (5.2) was constructed. This consisted of a vertical Pyrex tube about 1 metre long with a standard cone joint at each end. This tube stood in a capillary tube housing and was held firmly in position by springs. Water from the de-ionising system described previously could be admitted through a connection in the capillary tube housing. The capillary tube itself was held in position by a tight fitting rubber tube. In order to prevent impurities from the rubber getting into the water, the tapered section of the capillary tube seated hard against a P.T.F.E. sealing ring. The top section of the Pyrex tube could be connected, via a cone joint, to the conductivity cell.

The freezing head consisted of a thermally insulated reservoir filled with a mixture of solid  $\text{CO}_2$  and silicone

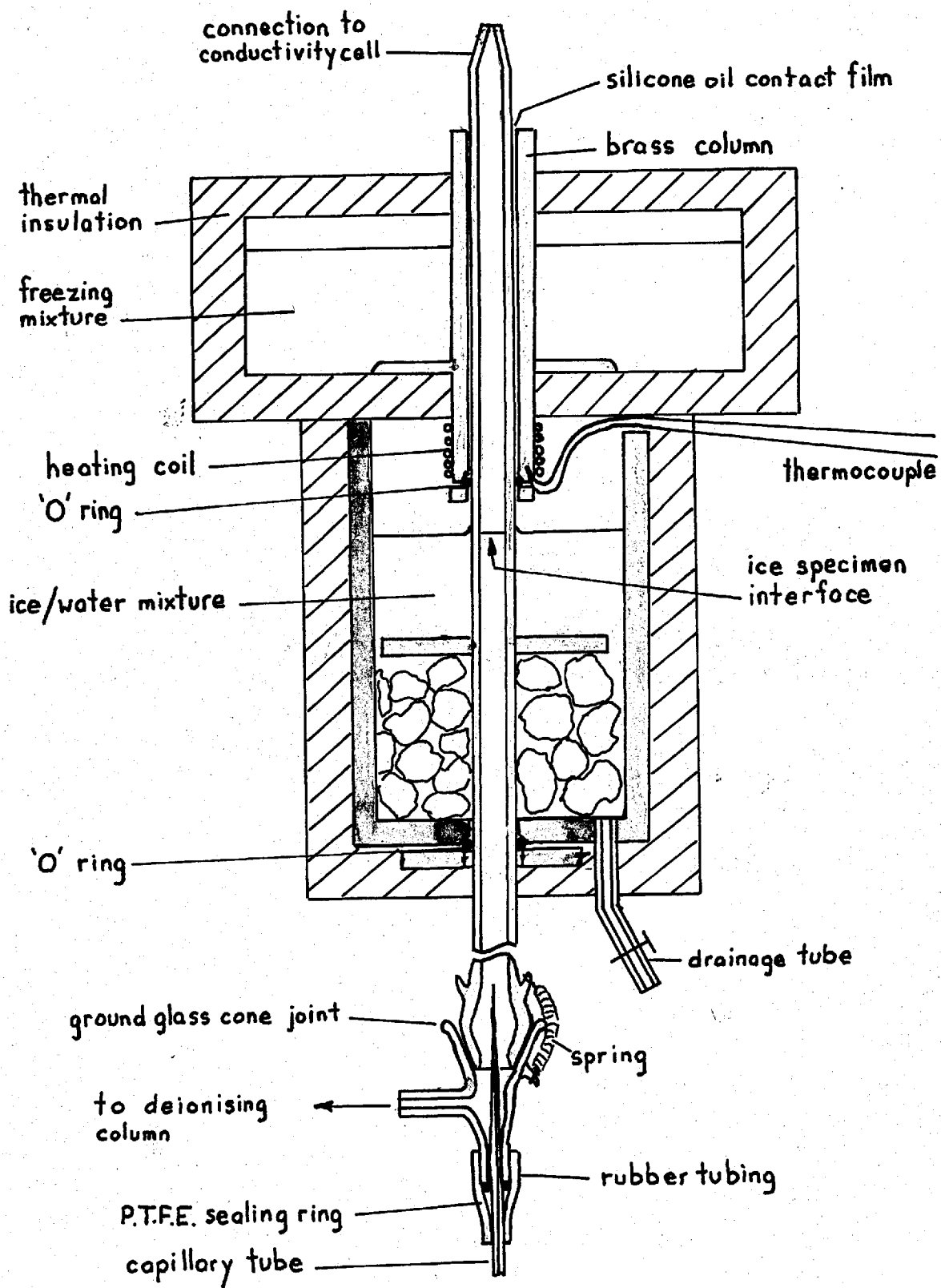


Fig. 5.2 Detail of freezing apparatus for controlled bubble ice specimen production. Half size.

oil, which fitted coaxially over the Pyrex tube. A central brass column enabled the reservoir to be removed from the system without draining the contents. It was held in position against an 'O' ring seal which supported the entire weight of the reservoir and its contents. Good thermal contact was ensured by filling the gap between the brass column and the central Pyrex tube with a thin film of silicone oil. The lowest part of the brass column, which projected below the reservoir, was wound with a small heating coil and also carried a thermocouple.

Below the freezing head, The Pyrex tube carried a coaxial, thermally insulated chamber supported and sealed by an 'O' ring at its lowest point. This chamber could be filled with a mixture of ice and water, and was equipped with a run-off valve in the base. The ice in the mixture was kept below the water level by a weighted grid. The principal of operation of the system is as follows.

Water from the de-ionising column was allowed to flow through the vertical Pyrex tube until the conductivity reading reached a minimum. Although provision was made for doping the flow with a known impurity concentration, this facility was never used (for reasons which are discussed below) and will not be described. When the minimum conductivity had been reached the freezing mixture was poured into the reservoir, and the water in the Pyrex tube allowed to freeze inwards from the walls

of the Pyrex tube, the conductivity being monitored the whole time. In this way one could be reasonably certain of not introducing impurities in making ice from 'pure' water.

After a few minutes the Pyrex tube passing through the reservoir became sealed with ice and the freezing level projected gradually downwards into the coaxial chamber below the reservoir. By controlling the current in the heating coil and by lowering the water level in the lower chamber at a constant rate, the ice/water interface could be kept exactly horizontal while moving with a constant speed down the tube. Gas bubbles were then introduced via the capillary tube, and rose to the ice/water interface where they were incorporated into the ice column. In this way, using a constant rate of bubble production, it was theoretically possible to produce ice with bubbles of constant size and a given density.

In practice the system had a number of drawbacks.

- i) The rate of growth of the ice/water interface could not be maintained constant for more than about 0.5 cm of specimen length.
- ii) The water in the lower chamber tended to freeze around the Pyrex tube at its upper surface, obscuring the interface from view.
- iii) The gas bubbles introduced by the capillary tended to rise up the middle of the tube and collect at the centre of the ice/water interface. In order to prevent

them from coalescing at this point it was necessary to work with very slow rates of bubble production. This problem was partially overcome by passing a small current through a few turns of heating wire wound around the Pyrex tube. This caused sufficient convection to stir up the bubbles and prevent coalescence. However, it also caused the ice/water interface to become slightly concave towards the water, so that if the current was a little too large the bubbles again tended to collect and coalesce in the centre.

iv) The maximum length of specimen was restricted to about 2 cm. Although this would have been sufficient, it was found in practice that the first  $1\frac{1}{2}$  cm were used to adjust all the different controls, and the useful length was reduced to about 5 mm.

v) Once they were made, the specimens were removed by dismantling the conductivity cell, the freezing reservoir and the lower chamber. The Pyrex tube was then removed from its seating in the capillary tube housing and taken into a cold room where it was allowed to warm up to  $0^{\circ}\text{C}$ . The specimen was then slightly melted until it slipped out of the bottom of the Pyrex tube into a clean container. The ice was then refrozen in an atmosphere of nitrogen. Unfortunately this operation further reduced the useful volume of the specimen.

In order to overcome these problems, it was decided to modify the apparatus in an attempt to produce much

larger and more uniform specimens.

### 5.2.3 Bulk specimen production. Method (ii)

Fig. (5.3) shows the modification to the apparatus used to produce larger specimens of ice. The capillary housing, vertical Pyrex tube and conductivity cell were used as before. The freezing head was modified in such a way that the lower chamber made an air-tight seal against the reservoir. Instead of using water in this chamber, it was filled with silicone oil. In this way the ice was prevented from forming on the outer walls of the Pyrex tube, and the ice/water interface could be easily seen. The reservoir also had anchor points for three springs to be fixed to its under surface.

The freezing head assembly was suspended from a lever and pulley system, and could slide up and down the vertical Pyrex tube. The assembly was pulled down by the three springs on the lower section, and the rate of descent controlled by the lever operating on an oil filled piston with a small bleed valve. The lever was used at small angles to the horizontal, thus giving a nearly uniform rate of descent, the fine adjustment to the rate of movement of the freezing level being controlled by the level of silicone oil in the lower chamber.

This apparatus was used in a similar manner to method (i), a small heating coil being used to distribute

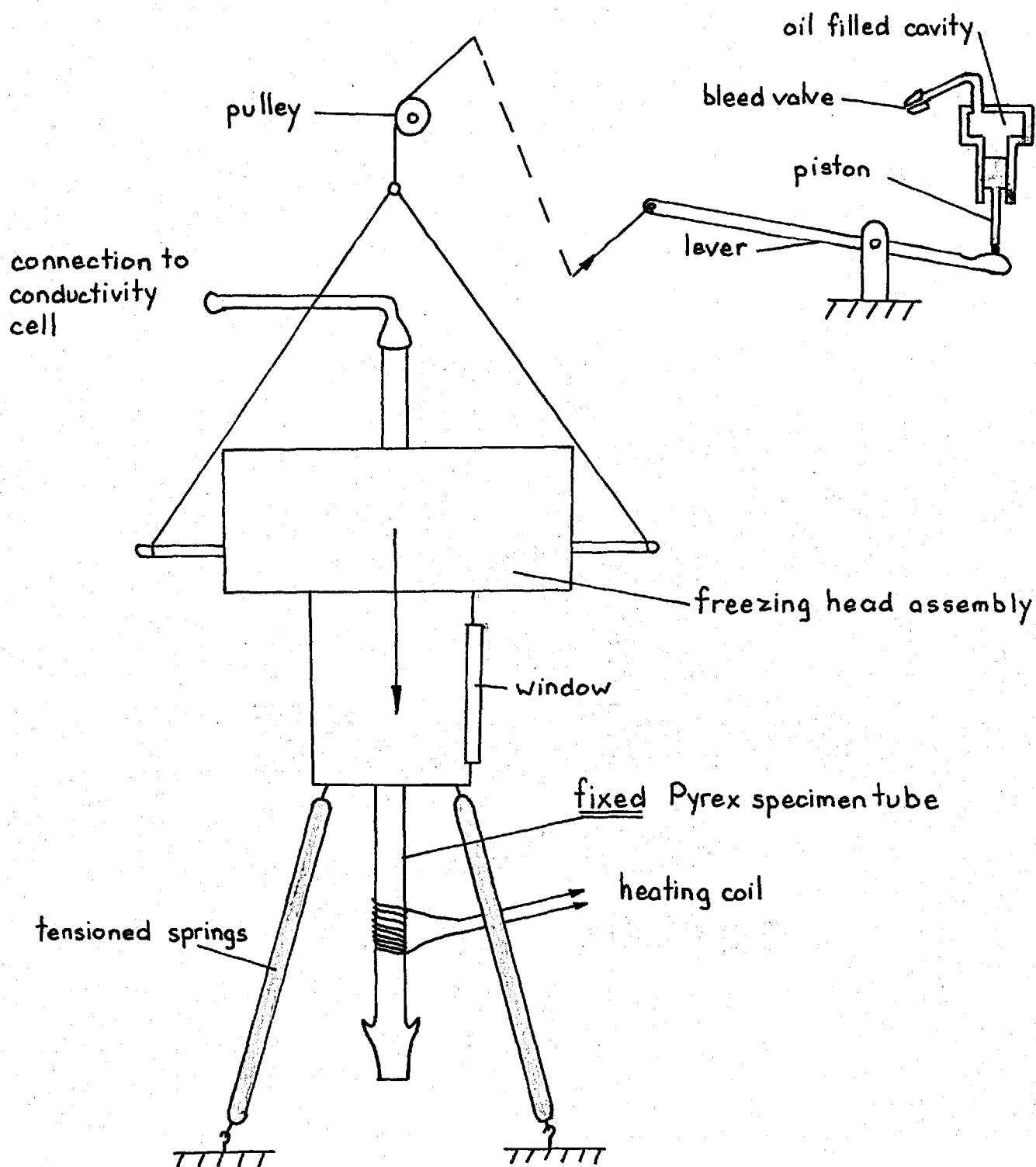


Fig. 5-3 Modification to freezing apparatus for controlled bubble ice specimen production. Schematic.

the bubbles over the whole area of the interface. With this arrangement, the ice/water interface could be kept close to the freezing reservoir, and nearly horizontal. However, in order to avoid coalescence it was still necessary to use fairly slow bubble rates of order 30 per minute. The ice specimens were removed from the apparatus and stored in the manner previously described.

Although this apparatus was a considerable improvement on the first, it was found that the strong temperature gradients induced in the ice specimens tended to make them crack. However, specimens with fairly uniform bubble density were obtained having volumes up to  $5 \text{ cm}^3$ . Results using these specimens will be discussed in section (5.4).

#### 5.3.1 Wind tunnel development

The system used to study the charge separation from the melting of large ice specimens is shown in Fig. (5.4). The wind tunnel consisted of a horizontal brass tube with removeable end section. The bulk ice specimens were placed in a nickel dish and mounted on the centre pin of a coaxial plug. The plug fitted tightly in the outer wall of the brass tube, and acted as the earthing connection for the Faraday Cage system. The gas in which the specimen was to be melted was passed through an electric heater, a particulate filter made of cotton wool and a chamber containing tightly packed

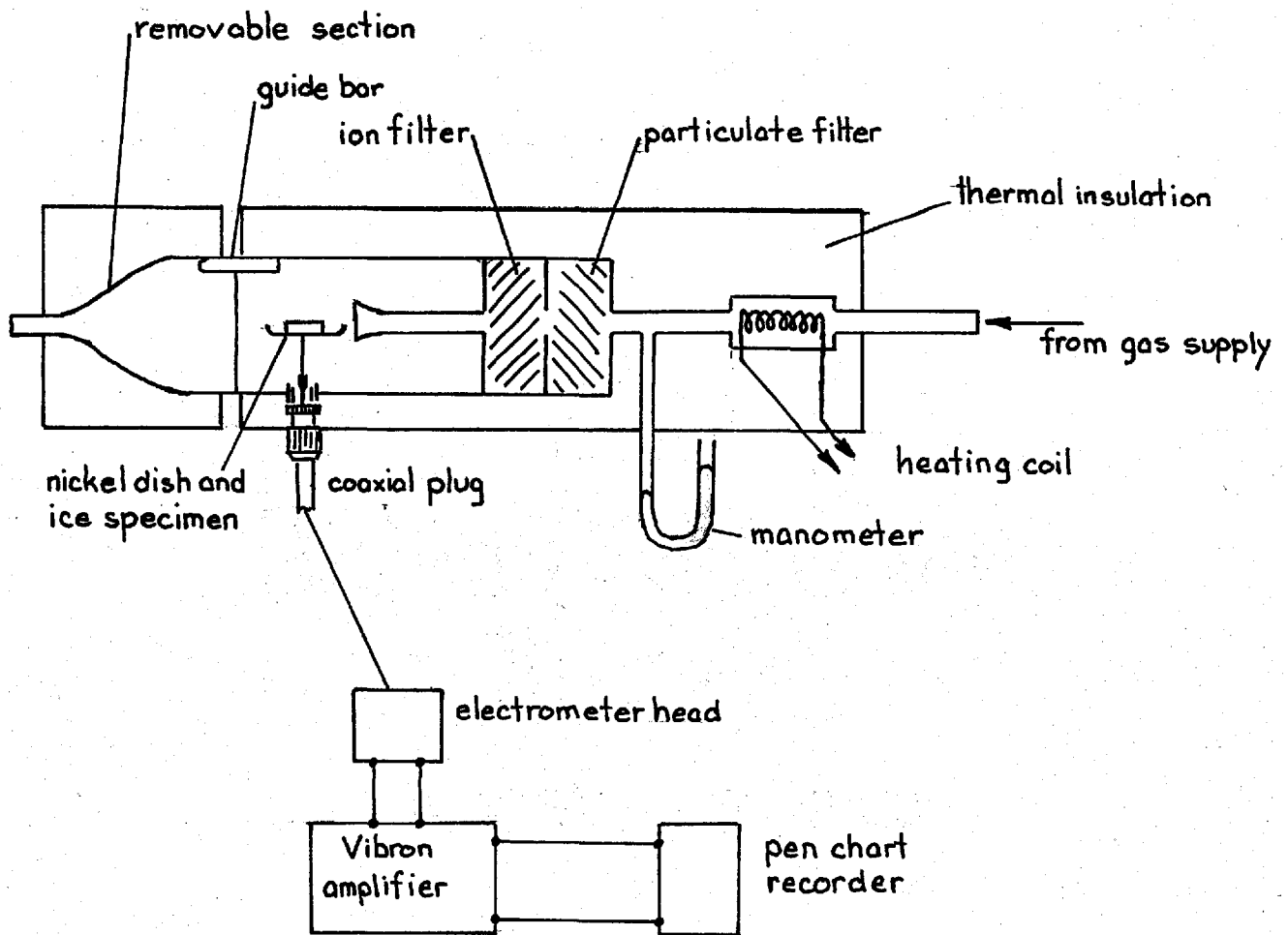


Fig. 5.4 Schematic layout of horizontal wind tunnel.

copper gauze which acted as an ion filter. The gas then passed through a metal tube with flared end and over the nickel dish containing the ice specimen. The gas escaped through a second tube into the atmosphere. A thermocouple measured the temperature of the gas just before passage over the ice specimen. The tunnel had two small windows in the same section as the coaxial plug, thus allowing illumination and viewing. The gas flow was observed by reading a small differential manometer placed in the inlet tube. Before using the tunnel, the system was set up without a specimen in the nickel dish, and the absence of any charge checked under all conditions of wind speed and temperature likely to be used during the experiment. No charging could be detected above the background drift of the Vibron electrometer used to record the charge. The sensitivity of this instrument and associated circuitry was of order  $10^{-4}$  esu..

Using this apparatus, visual studies of the melting process of bulk ice specimens immediately indicated that the results of charge measurements would not easily be interpreted in terms of the initial bubble structure of the ice. Even with specimens containing only some few tens of bubbles per  $\text{cm}^3$ , of diameter in the hundred micron range, rafting and coalescence took place during the melting. After melting had proceeded for a short while, the ice began to float on the film of water formed.

Bubbles released at the under surface of the ice tended to remain trapped there for some time, and considerable coalescence took place before their escape. Also, rafting occurred at the air/water surface, and the bubbles sometimes persisted for periods of up to a minute before bursting, coalescence again taking place. This effect could be reduced by increasing the wind speed and directing it at a small angle towards the surface, thus keeping the surface in slow motion. However, this is likely to influence the escape of the smaller drops from the bursting bubbles and so possibly reduce the charge separation.

It was also observed that when melting had only just begun, so that the film of water on the ice was very thin, the bubbles did not burst in the conventional manner. The bubble cap tended to rupture before the bubbles were fully released from the ice, and the water did not rush into the cavity to form jet drops, but drew away from the edges leaving a small, dry, indented area on the surface. This gradually filled with water as the melting proceeded.

These effects must have been present in the experiments performed by previous workers in the field of electrification during melting, and it will be shown that the aspect of the 'dynamics' of the bursting process plays an important part in determining the level of

charge recorded with any given specimen. Attempts to eliminate the effects mentioned above proved fruitless, and it was decided to abandon these tests with large ice specimens and to proceed to examine the charging with the specimens supported in a vertical wind tunnel. It was suspected, and found to be true, that much greater levels of charge were recorded, due to the possibility of a change in the dynamics of the melting process and the absence of a container for the specimens..

### 5.3.2 Large vertical wind tunnel

By this stage of the investigation it had become obvious that a great deal of attention must be given to designing the apparatus to investigate the charging during melting, as the nature of the support and the dynamics of the melting process were having a profound effect on the quantity of charge generated. For this reason it was decided to use a modified version of the vertical wind tunnel used in the first half of this work on the heat transfer equations, to design apparatus that would be capable of examining the charge generation under conditions likely to be representative of those occurring in natural free fall in the atmosphere. The modified working section of the wind tunnel system used for this investigation is shown in Fig. (5.5).

The lower half of the Perspex tunnel was replaced by a metal section completely filled with close-packed

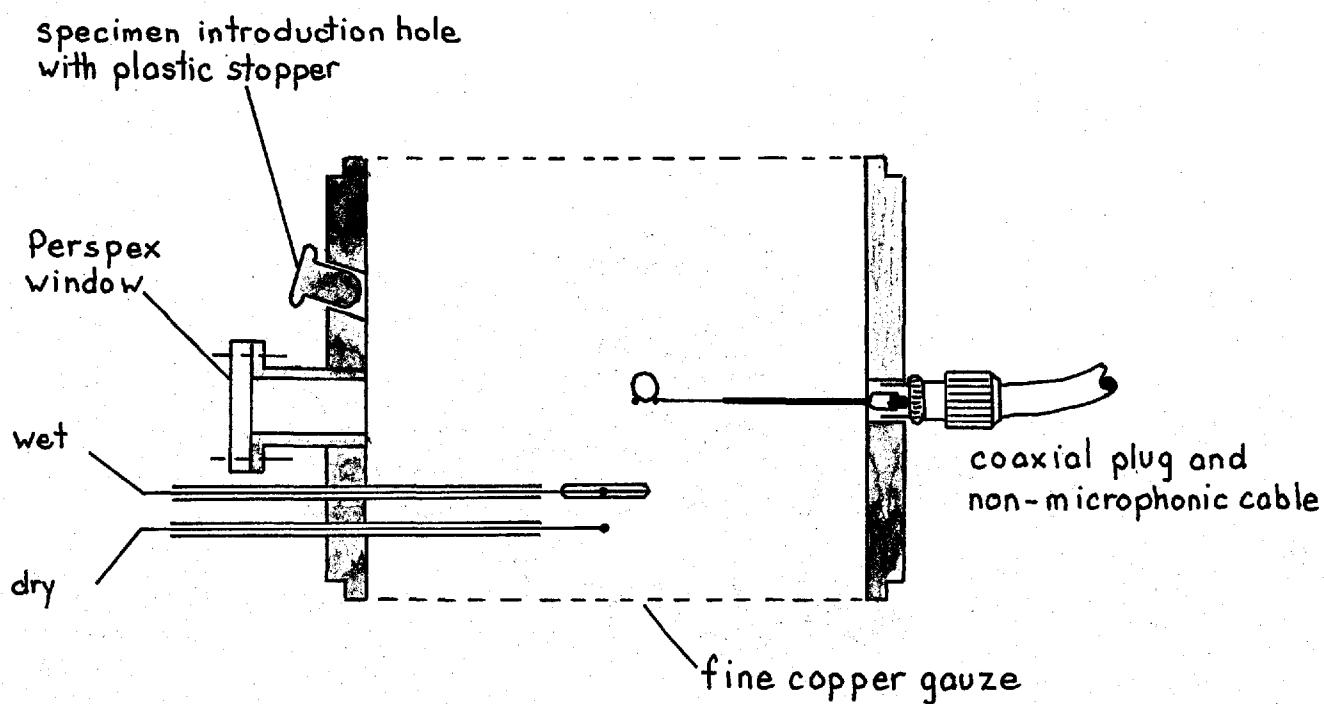


Fig. 5.5 Detail of working section of vertical wind tunnel.

copper gauze to act as an ion filter, and surrounded by thermal insulation. The working section, shown in detail in Fig. (5.5) was made from aluminium tube closed at each end by fine copper gauzes. It had two Perspex windows for illumination and viewing, each screened with copper gauze on the inside. The specimens were supported on a small loop of 46 swg contra wire attached to the central pin of a coaxial plug and thence to a Vibron electrometer and recorder via 7 feet of non-microphonic coaxial cable. The coaxial plug was sealed into the side of the working section with a layer of natural Ceresin wax. This had an insulation resistance greater than  $10^{15}$  ohm even at humidities as high as 95%. A small hole in the working section above the level of the coaxial plug was used for introduction of the specimens to the support. The system was earthed through the outer sheath of the coaxial cable, and formed a Faraday Cage.

#### 5.4 Preliminary results

##### 5.4.1 Bulk specimens in horizontal tunnel

Long cylinders of ice up to about  $10 \text{ cm}^3$  were obtained from the apparatus described in section (5.2.3). These were cut with a stainless steel blade to yield useful volumes of up to about  $5 \text{ cm}^3$ , the residue being melted in a dip type conductivity cell in an inert atmosphere of nitrogen. The blade was heated to red heat before use, and the cutting operation performed at a

temperature of about  $-5^{\circ}\text{C}$  in a slow stream of nitrogen. When cut, the specimens were placed in a nickel dish using clean forceps which had been heated to red heat to remove volatile contaminants.

The mean bubble size in the ice specimens were measured by examination of selected specimens with a microscope. Due to shrinking of the bubbles during their passage from the capillary tube to the freezing interface, there was a spread about the mean value of about  $\pm 10\%$ . The bubble density was inferred from the rate of bubbling of the capillary tube and the growth rate of the ice/water interface. The bubble sizes in the ice were assumed to be the same as in the melt water. The nickel dish was then transferred to the small horizontal wind tunnel described in section (5.3.1) and the specimens melted under a variety of conditions.

The Vibron electrometer used to record the charge had an internal resistance of order  $10^{16}$  ohm, and the total capacity of the system was 240 pf. Since the insulation resistance of the system was of order  $10^{15}$  ohm at the highest humidities likely to be used, the shortest time constant of the system was given by

$$\tau = CR = 240 \times 10^3 \text{ secs.}$$

which is effectively infinite compared with the time for each experiment which is of the order of a few minutes. The voltage output from the Vibron was amplified and fed

to a pen chart recorder.

It was found that it was essential to avoid changes of temperature and movement or vibration of the coaxial cable, as either of these effects manifested itself as a charge recorded by the Vibron. For this reason it was essential to use only steady state conditions for melting. A drift rate check on the electrometer was taken between each experimental run.

The results of the experiment are broadly summarised in Table (5.1) below for melting in nitrogen. The charges recorded are expressed in terms of esu per bubble. The conductivities referred to are those measured by the dip cell at a temperature of  $20^{\circ}\text{C}$ . The increase in conductivity over the intrinsic value of about  $0.05 \mu\text{mho cm}^{-1}$  is thought to be due to the absorption of  $\text{CO}_2$  during the melting, and corresponds to a concentration of less than  $10^{-6}$  moles/litre. The results for melting in air were essentially similar to those for melting in nitrogen and have not been shown.

From these results, a number of factors were immediately striking.

- i) In no instance was any charge recorded for specimens containing no bubbles.
- ii) No great difference could be found for the results of melting in air or nitrogen.
- iii) The column in Table (5.1) giving the charge per

TABLE 5.1  
Charges from bulk specimens

X indicates no charge detected

✓ indicates charge just detected

| wind speed<br>cm/sec | Melting temp.<br>°C | Bubbles       |        | mean conductivity<br>at 20°C<br>$\mu$ mhos<br>cm <sup>-1</sup> | no.<br>of<br>samples | charge/bubble<br>esu |                      |                      |
|----------------------|---------------------|---------------|--------|----------------------------------------------------------------|----------------------|----------------------|----------------------|----------------------|
|                      |                     | dia.<br>$\mu$ | no./cc |                                                                |                      | max.                 | min.                 | mean                 |
| 10                   | 10                  | 147           | 51     | .11                                                            | 7                    | X                    | X                    | $<10^{-5}$           |
|                      |                     | 147           | 94     | .17                                                            | 5                    | ✓                    | X                    | $<10^{-6}$           |
|                      |                     | 147           | 232    | .09                                                            | 4                    | $2 \times 10^{-4}$   | ✓                    | $0.6 \times 10^{-5}$ |
|                      |                     | 253           | 45     | .20                                                            | 3                    | X                    | X                    | $<10^{-5}$           |
|                      |                     | 253           | 87     | .21                                                            | 3                    | X                    | X                    | $<10^{-6}$           |
|                      |                     | 253           | 270    | .13                                                            | 2                    | ✓                    | X                    | $\sim 10^{-6}$       |
|                      |                     | —             | 0      | .15                                                            | 3                    | X                    | X                    | X                    |
|                      | 20                  | 147           | 41     | .08                                                            | 4                    | X                    | X                    | $<10^{-5}$           |
|                      |                     | 147           | 110    | .13                                                            | 7                    | $1.3 \times 10^{-5}$ | $1.0 \times 10^{-6}$ | $8 \times 10^{-6}$   |
|                      |                     | 147           | 273    | .13                                                            | 3                    | $7.3 \times 10^{-6}$ | $4.0 \times 10^{-6}$ | $6 \times 10^{-6}$   |
|                      |                     | 253           | 45     | .19                                                            | 7                    | X                    | X                    | $<10^{-5}$           |
|                      |                     | 253           | 105    | .13                                                            | 6                    | $7 \times 10^{-6}$   | X                    | $3 \times 10^{-6}$   |
|                      |                     | 253           | 241    | .20                                                            | 4                    | ✓                    | ✓                    | $\sim 10^{-6}$       |
|                      |                     | —             | 0      | .25                                                            | 3                    | X                    | X                    | X                    |
| 100                  | 10                  | 147           | 63     | .20                                                            | 9                    | $3 \times 10^{-3}$   | $5 \times 10^{-6}$   | $2 \times 10^{-4}$   |
|                      |                     | 147           | 132    | .14                                                            | 7                    | $9 \times 10^{-5}$   | $4 \times 10^{-6}$   | $5 \times 10^{-5}$   |
|                      |                     | 147           | 247    | .11                                                            | 2                    | $5 \times 10^{-5}$   | $1 \times 10^{-5}$   | $3 \times 10^{-5}$   |
|                      |                     | 253           | 52     | .15                                                            | 3                    | ✓                    | ✓                    | $<10^{-5}$           |
|                      |                     | 253           | 127    | .12                                                            | 2                    | $1 \times 10^{-5}$   | ✓                    | $<10^{-5}$           |
|                      |                     | 253           | 222    | .18                                                            | 4                    | $9 \times 10^{-6}$   | $1.3 \times 10^{-6}$ | $3.5 \times 10^{-6}$ |
|                      |                     | —             | 0      | .11                                                            | 4                    | X                    | X                    | X                    |
|                      | 20                  | 147           | 50     | .11                                                            | 3                    | $1.3 \times 10^{-4}$ | $1.0 \times 10^{-5}$ | $6.5 \times 10^{-5}$ |
|                      |                     | 147           | 99     | .21                                                            | 3                    | $1.7 \times 10^{-4}$ | $7.1 \times 10^{-5}$ | $9 \times 10^{-5}$   |
|                      |                     | 147           | 217    | .10                                                            | 4                    | $4.0 \times 10^{-5}$ | $1.3 \times 10^{-6}$ | $7.3 \times 10^{-6}$ |
|                      |                     | 253           | 43     | .13                                                            | 3                    | $1.1 \times 10^{-5}$ | $0.9 \times 10^{-5}$ | $1 \times 10^{-5}$   |
|                      |                     | 253           | 107    | .09                                                            | 3                    | $7.0 \times 10^{-5}$ | $1.4 \times 10^{-5}$ | $3.5 \times 10^{-5}$ |
|                      |                     | 253           | 248    | .11                                                            | 4                    | $2.0 \times 10^{-5}$ | ✓                    | $4.7 \times 10^{-6}$ |
|                      |                     | —             | 0      | .18                                                            | 3                    | X                    | X                    | X                    |

bubble shows large fluctuations in the mean values for specimens having different bubble densities even though melted under similar conditions. Although the records were examined to see if there was any possibility of the specimen volume being involved, no greater consistency was found in the values.

iv) The magnitude of the mean charge per bubble is much smaller than expected by comparison with the work of Iribarne (1966a). (Order  $10^{-3}$  esu per bubble).

v) The charges remaining on the melt water were always either positive, or were too small to be detected.

As discussed previously, it was felt that the rafting and coalescence of bubbles during melting partly accounts for the great variation in the results. Although it was not definitely established, it was suspected that the maximum charges per bubble occurred on chance occasions when the ice core was set in motion in its liquid.

Some samples of natural snow collected at ground level from a country area were also melted in the horizontal tunnel. The charges recorded using these specimens were always less than  $10^{-4}$  esu per  $\text{cm}^3$  of melt water, and erratic in sign. A test of the salt content of the samples showed an NaCl concentration of order  $10^{-5}$  moles/litre. With these samples the problems of coalescence and rafting were much more acute due to

the higher bubble density. The experiments were not prolonged as it was felt that little useful information could be gained by their study..

The problems brought to light by the results of this investigation were pursued further by proceeding to use the vertical wind tunnel described in section (5.. 3.2), the main intention being to use the results to design a further system once a fuller understanding of the 'ideal' melting conditions were obtained.

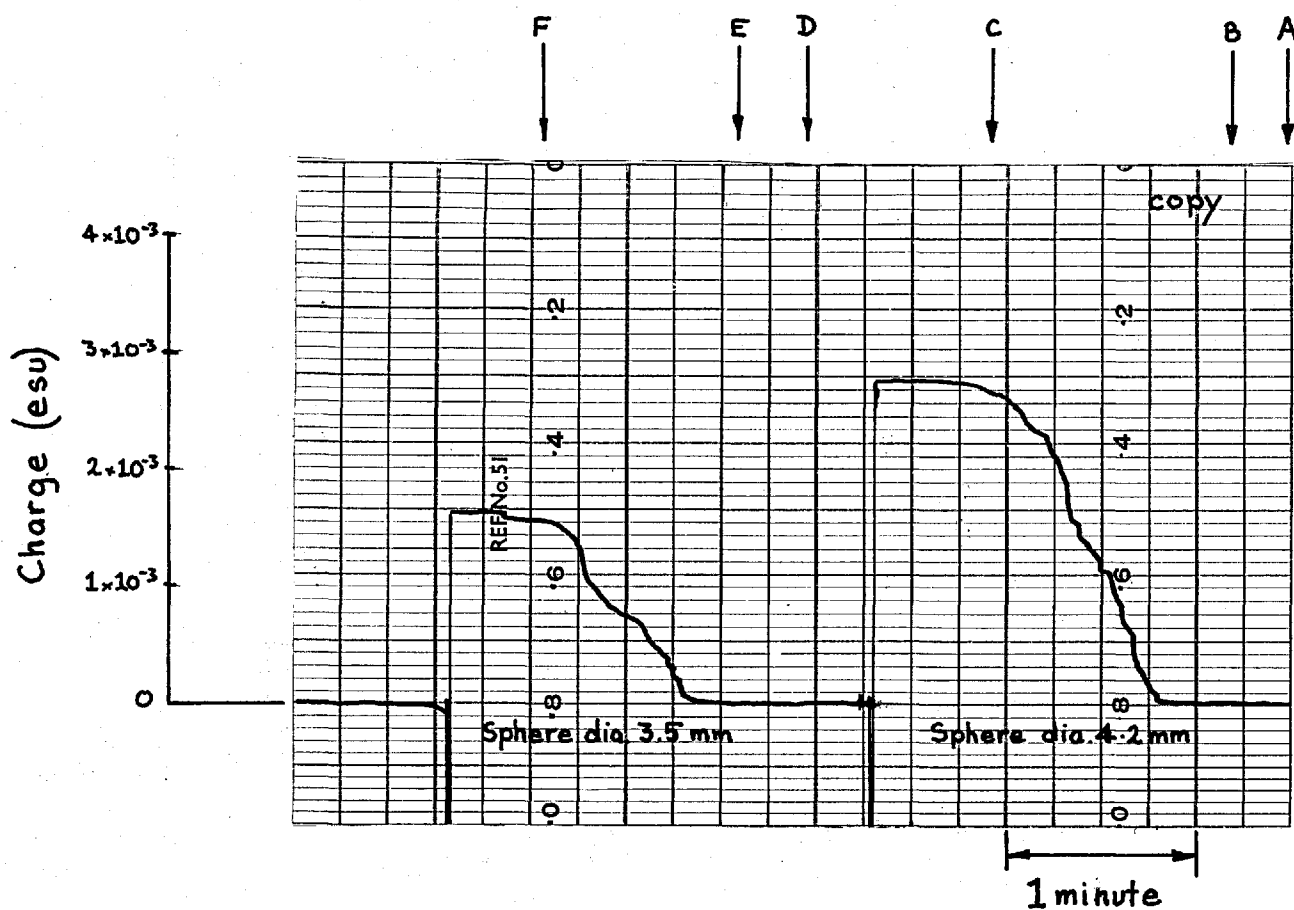
#### 5.4.2 Melting in large vertical wind tunnel

Bulk specimens of ice as used in the horizontal tunnel were divided into elements of approximate equivalent spherical diameter 3 to 5 mm for use in the wind tunnel. The elements were stored in an atmosphere of nitrogen until required.

A small drop of 'pure' water from a hypodermic syringe was then squirted onto the tip of a stainless steel needle. This was allowed to freeze while in contact with one of the small specimens of ice, and in this way the specimens could be picked up and transferred to the wind tunnel. They were quickly inserted through the port-hole on the downstream side of the working section and rested gently on the wire loop support, which was frequently cleaned by boiling in distilled water. The warm air stream melted the surface of the specimen, and the needle could then be removed leaving the specimen held

in position by surface tension forces so that it sat on top of the loop. The port-hole was then sealed with a tight fitting plastic stopper, and the specimen allowed to melt under conditions of constant temperature, humidity and wind speed, the charge being recorded by the Vibron electrometer. Some typical records from the pen chart recorder are shown in Fig. (5.6).

It is seen from the records that the charging begins a short while after the melting process begins, and continues a short while after the melting process ends. The continuation after the melting is complete is probably due to the lag of the recorder, but could also be partly caused by the continued bursting of bubbles which collect at the downstream side of the drop after the last traces of ice have disappeared. Since it will be seen that the range of charging levels are greatly increased over those recorded in the bulk melting case, (this is interpreted mainly as an effect of the dynamics of the melting process), it is thought that the initial delay and small slope of the characteristics are due to the absence of convective motions in the water portion of the melting drop. A further possibility is that surface impurities become gradually diluted during melting and allow greater separation of charge per bursting bubble event. On the traces, some step changes of charge can be seen, although the damping of the instrument



A, D spheres positioned in wind tunnel  
 B, E charging record begun  
 C, F melting finished

Air temperature + 8°C; Humidity ~ 75%  
 Nitrogen bubble dia. 147  $\mu$ ; Approx 490 per cc.

Fig. 5.6 Typical records accompanying Table 5.2.

TABLE 5.2

Charges from divided specimens

X indicates no charge detected

✓ indicates charge just detected

| wind speed<br>cm/sec | Air temperature<br>°C | Bubbles       |       | Approx Humidity<br>% | no of samples | charge/bubble<br>esu |                      |                      |
|----------------------|-----------------------|---------------|-------|----------------------|---------------|----------------------|----------------------|----------------------|
|                      |                       | dia.<br>$\mu$ | no/cc |                      |               | max.                 | min.                 | mean                 |
| 100                  | +6                    | 82            | 490   | 50                   | 7             | X                    | X                    | $<10^{-5}$           |
|                      |                       | 147           | 320   | 50                   | 9             | ✓                    | X                    | $<10^{-5}$           |
|                      |                       | 82            | 490   | 80                   | 10            | ✓                    | ✓                    | $\sim 10^{-5}$       |
|                      |                       | 147           | 320   | 85                   | 10            | ✓                    | ✓                    | $\sim 10^{-5}$       |
|                      | +8                    | 82            | 490   | 45                   | 6             | $3.3 \times 10^{-4}$ | $2 \times 10^{-5}$   | $7 \times 10^{-5}$   |
|                      |                       | 147           | 320   | 50                   | 9             | $2 \times 10^{-4}$   | ✓                    | $5 \times 10^{-5}$   |
|                      |                       | 82            | 490   | 80                   | 9             | $5.2 \times 10^{-4}$ | $1.2 \times 10^{-4}$ | $4.1 \times 10^{-4}$ |
|                      |                       | 147           | 320   | 80                   | 10            | $2.7 \times 10^{-4}$ | ✓                    | $6.3 \times 10^{-5}$ |
| 200                  | +6                    | 82            | 490   | 50                   | 5             | X                    | X                    | $<10^{-5}$           |
|                      |                       | 147           | 320   | 45                   | 6             | ✓                    | ✓                    | $\sim 10^{-5}$       |
|                      |                       | 81            | 490   | 80                   | 7             | $1.7 \times 10^{-4}$ | ✓                    | $5 \times 10^{-5}$   |
|                      |                       | 147           | 320   | 80                   | 9             | $8 \times 10^{-4}$   | $5 \times 10^{-5}$   | $1.4 \times 10^{-4}$ |
|                      | +8                    | 82            | 490   | 55                   | 10            | $1 \times 10^{-4}$   | $4.2 \times 10^{-6}$ | $3.6 \times 10^{-5}$ |
|                      |                       | 147           | 320   | 50                   | 10            | $6.1 \times 10^{-4}$ | $2.2 \times 10^{-4}$ | $4.2 \times 10^{-4}$ |
|                      |                       | 82            | 490   | 75                   | 9             | $9.1 \times 10^{-4}$ | $3.1 \times 10^{-4}$ | $5.9 \times 10^{-4}$ |
|                      |                       | 147           | 320   | 75                   | 10            | $4.2 \times 10^{-4}$ | $1.5 \times 10^{-4}$ | $8.3 \times 10^{-5}$ |

mean conductivity of melt water  $0.32 \mu \text{ mho cm}^{-1}$

tended to smooth out all but the largest ones. A summary of the results obtained with specimens of this nature is given in Table (5.2). The results have been expressed in terms of the mean charge per bubble.

It is seen from the table that although the range of charge per bubble over all conditions is approximately an order of magnitude greater than that obtained from the melting of bulk specimens in the small horizontal tunnel, the mean charge per bubble is not greatly increased. This reflects the great variability in the individual results which could not be tied down to any particular change in the melting conditions or the sample. A list of possible causes is given below.

- a) Chance contamination of the specimens during the many handling, cutting and transfer operations.
- b) Chance absorption of impurities from the air stream at any stage of melting.
- c) Contamination of the support before positioning the specimen.
- d) Absence or presence of strong convective motions in the melting specimens (possibly affected by the support)..
- e) Chance coalescence of bubbles during melting, probably associated with (d) above.

It was decided that in order to investigate these possibilities in detail, a more abundant supply of specimens was necessary without having to go through

the many stages of preparation and possible contamination.. The following technique of manufacture was adopted.

Various samples of water were stored in a thoroughly cleaned, all glass hypodermic syringe. To prepare a specimen, a drop of diameter in the mm range was squirted onto the tip of a stainless steel needle held above a flask containing liquid nitrogen. As soon as the drop was nucleated by stray ice crystals, it was lowered beneath the surface of the liquid nitrogen and allowed to freeze completely. The drops were then transferred rapidly to the wind tunnel as previously described. Sections of spheres frozen in this way revealed that the bubble structure was remarkably uniform throughout the sphere, and the bubble sizes were typically in the range 10 to 50 microns radius, with a density of order  $10^6$  per  $\text{cm}^3$  of ice. Three types of water sample were used: water of the lowest conductivity available from an Elgastat de-ionising column, old distilled water and tap water. The results of the melting experiments with these samples are discussed below, in terms of the mean charge per  $\text{cm}^3$  of melt water.

- i) Tap water gave erratic charging of both signs, but never exceeded  $10^{-3}$  esu per  $\text{cm}^3$  of melt water.
- ii) Except in the case of tap water, the melted specimen always became positively charged.
- III) No significant difference was found between the

results for distilled water and those for the 'pure' water. The mean charge per  $\text{cm}^3$  under all melting conditions using specimens made from distilled water was 0.38 esu per  $\text{cm}^3$ . The mean charge for the same range of conditions for the specimens made from 'pure' water was 0.41 esu per  $\text{cm}^3$ .

iv) No significant variation was found with the wind speed in the range 0.5 to 2.5  $\text{m sec}^{-1}$ .

v) No significant variation was found with humidity in the range 20 to 95%.

vi) No significant variation was found with air temperature in the range  $0^{\circ}\text{C}$  to  $10^{\circ}\text{C}$ , providing that the temperature was more than  $1^{\circ}\text{C}$  above the value at which melting would only just begin. (Below this value, evaporation prevents the water shell from developing and convective activity never sets in).

vii) The artificial introduction of increased concentrations of  $\text{CO}_2$  into the air stream produced no significant variation in the mean levels of charge recorded.

viii) No variation in the mean charge was found with the specimen volume.

A histogram of results for the combined cases of distilled water and 'pure' water is shown in Fig. (5.7)..

The results may be summarised by saying that the mean charge per  $\text{cm}^3$  could not be altered by varying any of the parameters upon which it was suspected that it might depend. From 700 specimens used, the value of this

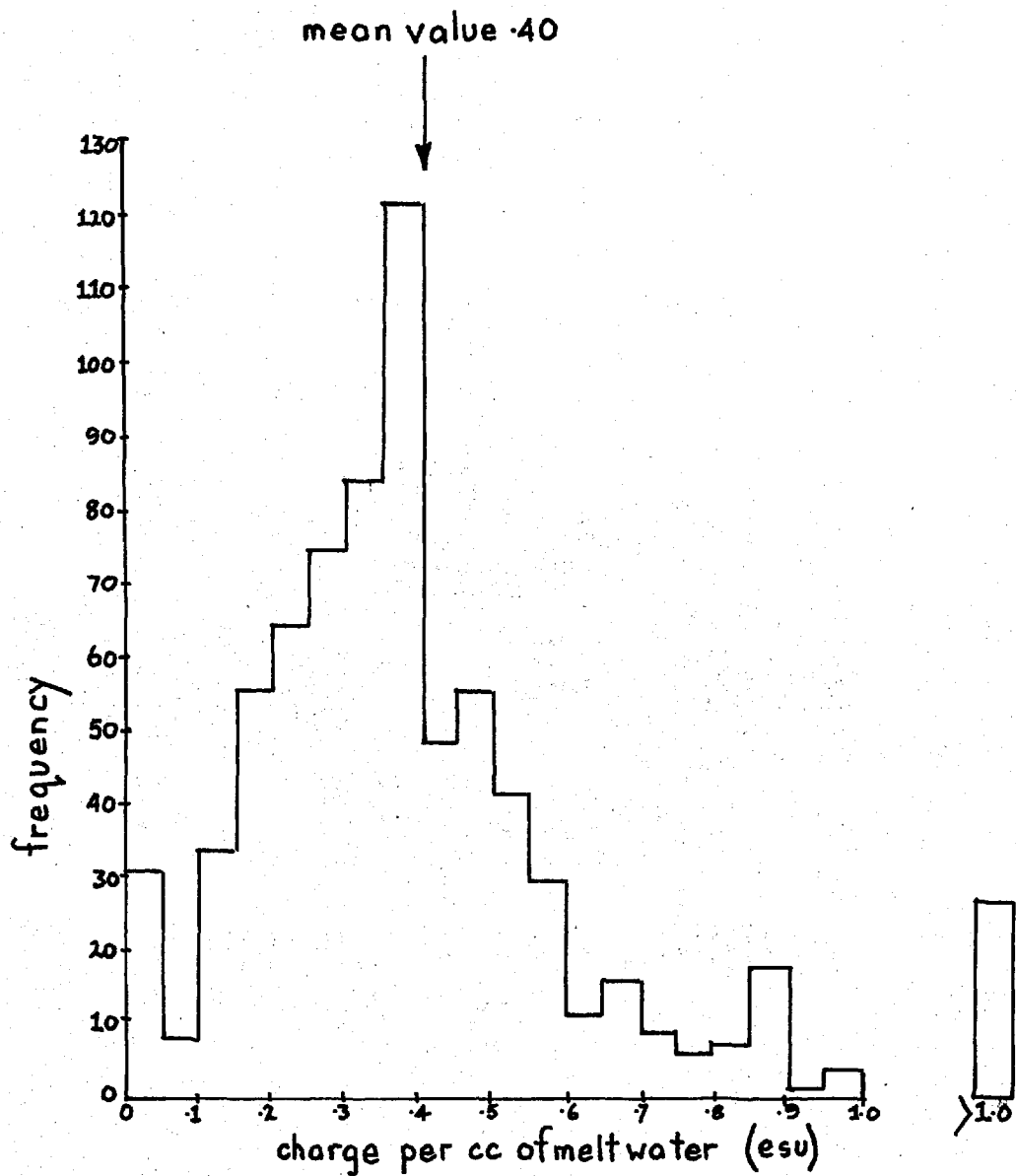


Fig. 5.7 Histogram of results for spheres prepared in liquid nitrogen. Bubble sizes 10 to 50  $\mu$  radius. Melted in air at wind speeds 0.5 to 2.5 m/sec. Temperature 0°C to 10°C.

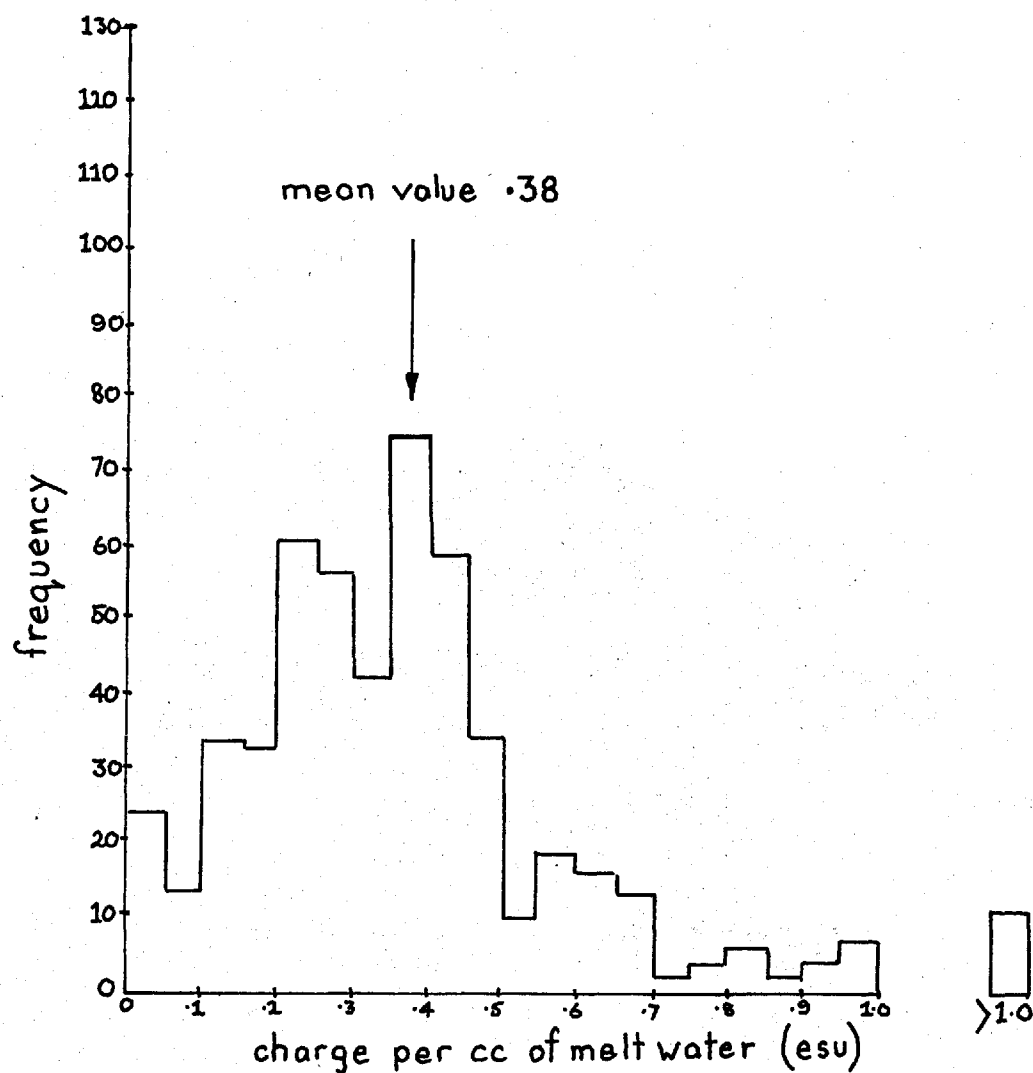
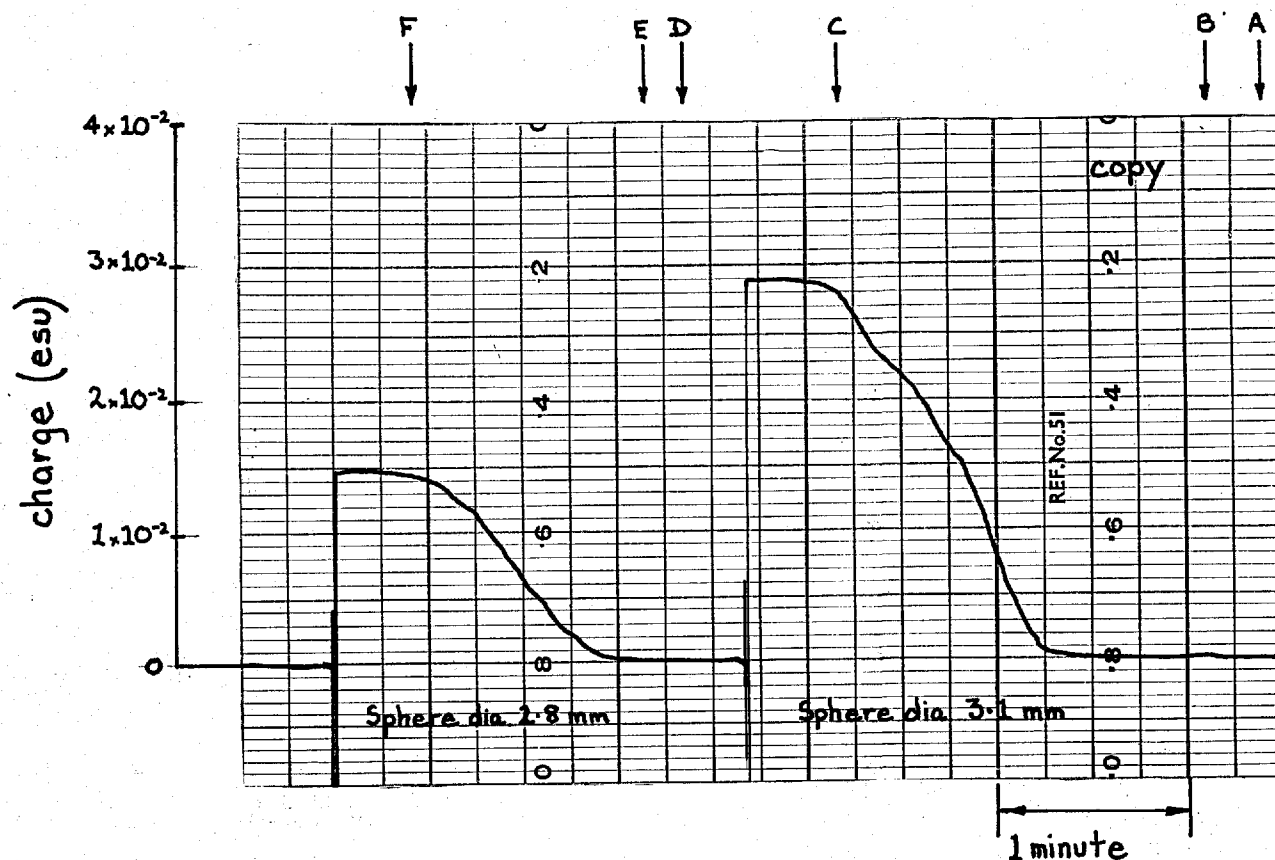


Fig. 5.8 Histogram of results for spheres prepared over liquid nitrogen. Bubble sizes 30 to 200  $\mu$  radius. Melted in air at wind speeds 0.5 to 2.5 m/sec. Temperature 0°C to 10°C.



A, D sphere positioned in wind tunnel  
 B, E charging record begun  
 C, F melting finished

Air temperature  $+5^{\circ}\text{C}$ ; Humidity  $\sim 80\%$   
 Bubble range 30 to  $200\mu$ .

Fig. 5.9 Charging records for spheres in vertical tunnel.

mean charge was  $+ 0.40$  esu per  $\text{cm}^3$ .

Some final experiments in which the ice spheres were made by suspension over boiling liquid nitrogen, and had much fewer bubbles in the range 30 to 200 microns radius in concentrations of order  $10^4$  per  $\text{cm}^3$ , gave a mean charge for the same range of melting conditions of  $+ 0.38$  esu per  $\text{cm}^3$  from 500 specimens. A histogram of the results is shown in Fig. (5.8). Some typical records of the charge generation are shown in Fig. (5.9). Their interpretation is essentially similar to that of Fig. (5.6).

### 5.5 Conclusions

(1) The results with large specimens of ice melted in the horizontal wind tunnel gave unsatisfactory results due to rafting and coalescence of the bubbles. The melting of specimens in this fashion is unlikely to give results that can be applied to melting in the atmosphere, as the dynamics of the melting process seems to play an important part in determining the level of charge generated with any given specimen. In particular, the convective activity in the water portion of a melting ice particle appears to be important.

(2) The melting of small volumes of ice, of uniform bubble size and density, in the vertical wind tunnel, showed a greater range of charge per bubble, although the mean charge per bubble was not significantly increased. In view of the greater consistency of the results found

with the ice sphere prepared in or over liquid nitrogen, it is felt that the variation in the individual results was due to contamination of the specimens during the handling, cutting and transfer to the wind tunnel. The increase in the magnitude of the maximum charge per bubble over the horizontal tunnel case is considered to be due to a change in the dynamics of the melting process.

(3) The results with small spheres prepared in or above liquid nitrogen gave fairly consistent charging results, with an occasional specimen, more than might be expected from the statistical distribution, showing a charge of up to five times the mean charge per  $\text{cm}^3$  of melt water. The mean charge per  $\text{cm}^3$  showed little dependence on any of the following factors.

- a) Temperature of air stream in range  $0^\circ\text{C}$  to  $10^\circ\text{C}$ .
- b) Humidity of air stream in range 20 to 100%.
- c) Wind speed in range 0.5 to  $2.5 \text{ m sec}^{-1}$ .
- d) Bubble structure of the ice.
- e) Difference in conductivity between old distilled water and 'pure' water.
- f) Increase in  $\text{CO}_2$  content of the air stream.

(4) From over 1000 individual results, the ice specimens had always acquired positive charge, or the charge was too small to be detected; except in the case of tap water when small negative charges of order  $10^{-4} \text{ esu cm}^{-3}$  of melt water were detected.

(5) The mean charge per  $\text{cm}^3$  of melt water obtained with spheres made from distilled water and conductivity water was about  $+ 0.40$  esu per  $\text{cm}^3$ . This is an order of magnitude too small to explain the presence of the subsidiary pocket of positive charge in a thunderstorm..

It was felt at this stage that some dominating factor had been overlooked in the experiments, as even the presence of increased concentrations of  $\text{CO}_2$ , so strongly felt by previous workers to have a poisonous effect on the charge generation, had failed to alter the mean charge per  $\text{cm}^3$  of melt water. There was also the few isolated results showing charges much greater than the mean level that needed explanation. In some cases these appeared to correspond to a new water sample, suggesting a contaminating effect on the water stored in the glass syringe used to make the ice specimens. The renewal of the water sample in the syringe also coincided with cleaning of the support in the wind tunnel.

Further experiments with water samples that had been stored for up to 24 hours in the glass syringe showed no significant alteration of the mean charge, and still gave isolated results much greater than the mean. The same behaviour was true when extended experiments were conducted without cleaning the support. However, by touching the support lightly with a finger before use,

the charging levels were reduced by approximately an order of magnitude. Also, by introducing a small piece of cotton wool which had been dampened with paraffin into a section of the wind tunnel remote from the working section, the charging levels were similarly reduced after the first few results. Since previous workers in the cold room had used open baths of paraffin in their experiments, it was felt that this might be having an effect on the results.

For this reason, together with other considerations of design obtained by experience, such as the need to have more flexibility of control of the melting conditions in the environment, a new wind tunnel was designed on a much smaller scale that would operate outside the cold room, but still at temperatures below  $0^{\circ}\text{C}$ . In this system, the ice spheres could be frozen and melted under controlled conditions in situ, thus avoiding possible contamination of their surfaces.

With this new system, it was hoped to examine the charge generation in terms of atmospheric conditions of freezing and melting, rather than to interpret the results in terms of a quantitative theory of the electric double layer. A much greater level of success was obtained, and the wind tunnel, which was used for the remainder of the experiments performed, is described in detail in the next chapter.

## CHAPTER SIX

### Charge Separation for 'Pure Ice' Melting in Nitrogen

#### 6.1 Experimental Arrangement

##### 6.1.1 Wind tunnel

The final design of the wind tunnel system used in the remainder of the experiments with melting ice particles is shown in Fig. (6.1) and (6.2). A photograph of the layout is shown in Fig. (6.3).

The hot and cold gas inlets could be connected either to cylinders of gas, or to separate vacuum cleaner pumps. The cold line contained an 80 foot length of thin-walled copper tube mounted inside a cold room to act as a heat exchanger. After passing through the wind tunnel, the flow of gas could be measured by a Rotameter flow device before exhausting to the atmosphere. The velocity of the gas in the vicinity of the mounted specimens in the wind tunnel was assumed to be given by the mean volume flow divided by the area of the inlet orifice. The flow of the hot and cold gas streams could be controlled by separate valves before entry to the wind tunnel where, after mixing, the gas passed through a plug of cotton wool and a tightly packed volume of copper gauze. The cotton wool acted as a particulate filter and was frequently renewed, and could be examined for evidence of particles in the gas flow. The copper gauze served as an ion filter for the gases.

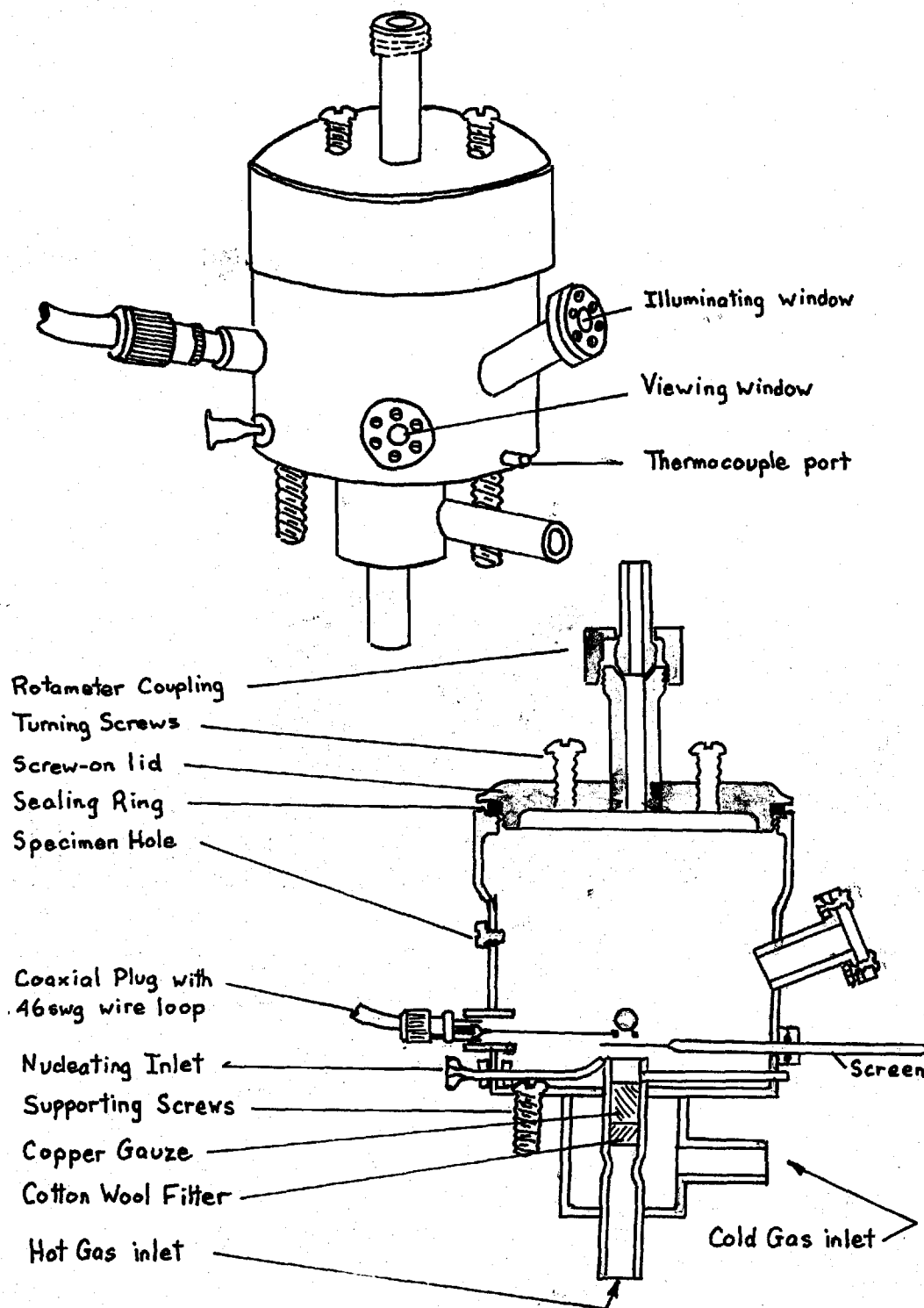


Fig. 6.1 Detail of small vertical wind tunnel.  
Approx half scale.

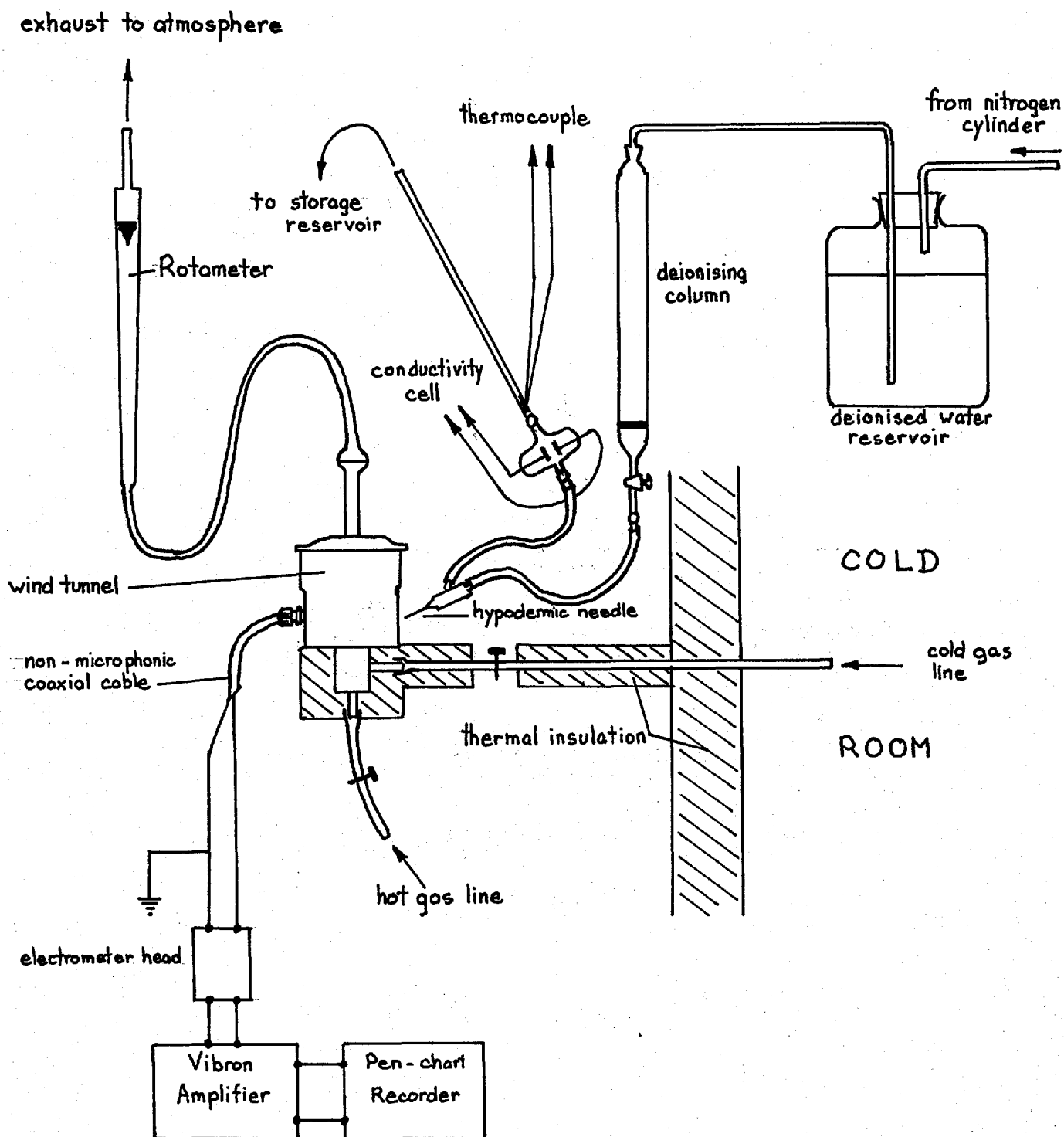


Fig. 6.2 Schematic layout of wind tunnel system showing gas flow, water circuit and electrical connections.

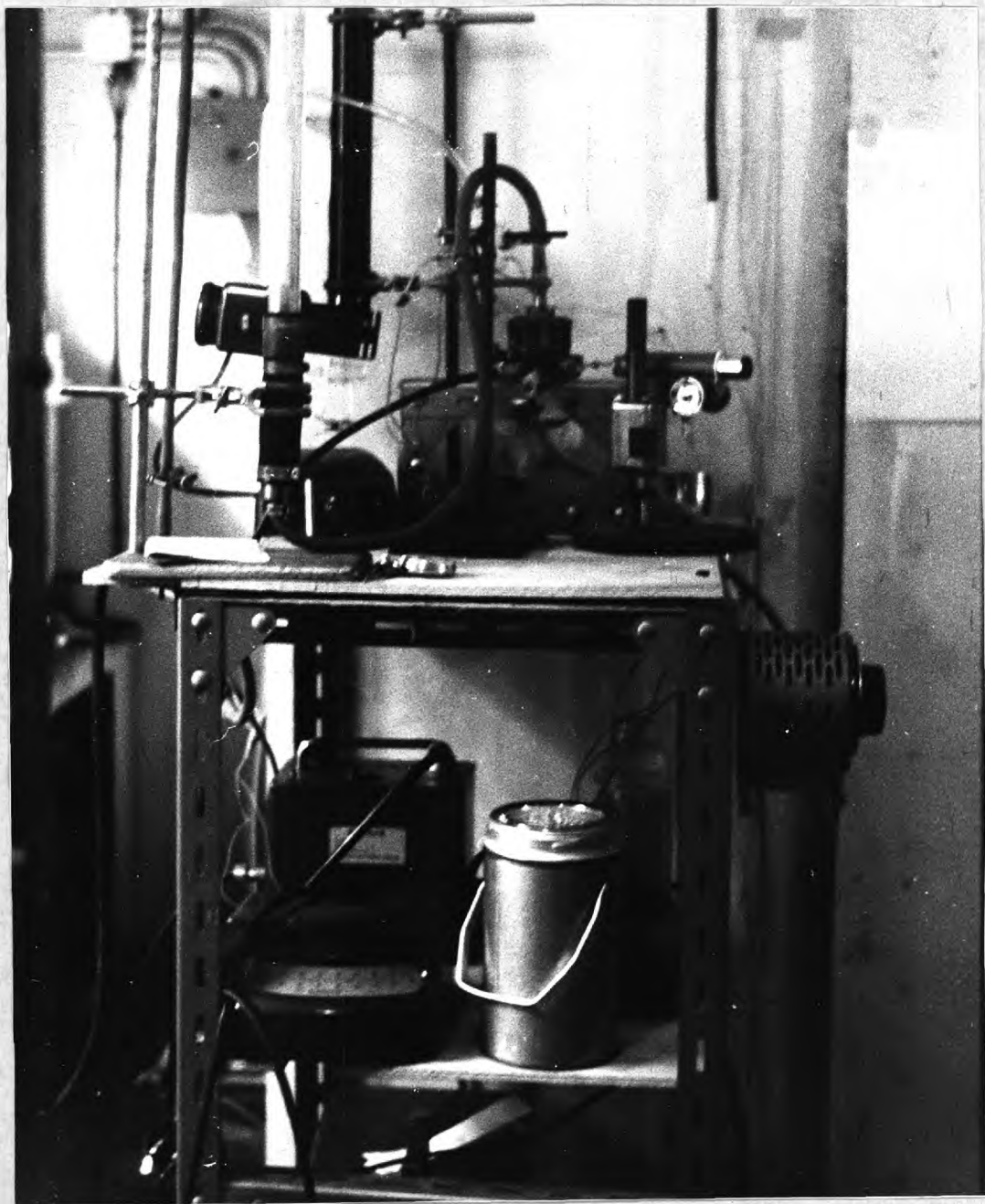


Fig. 6.3 Photograph of wind tunnel system.

The wind tunnel consisted of a thin-walled brass chamber with a screw-on lid, and had two electrically screened windows for illumination and viewing of the melting specimens. The specimens were mounted on a 46 swg contra loop attached to the central pin of a coaxial plug, which was sealed into the tunnel wall with Araldite glue. The specimens sat approximately 0.5 cm above the mouth of the gas inlet tube. To prevent debris from falling into the gas inlet tube, a small screen could be moved to cover the entrance when either positioning or removing specimens from the wireloop. The temperature of the inflowing gas was recorded by a thermocouple in the inlet tube which was sealed into the side with Araldite glue. A fine brass tube, with a nozzle directed towards the specimen mounting, served as a means of introducing small ice crystals into the gas stream. This tube had a Luer cone fitting on the end external to the tunnel for attachment of a hypodermic syringe. The specimens could be introduced either through a small hole in the tunnel wall, normally left sealed by a screw and 'O' ring, or by removing the lid. The tunnel was mounted on a stand via two screws welded to the base.

The electrical connections and recording device were similar to that used in the preliminary experiments, except that a Vibron electrometer having a maximum internal resistance of  $10^{12}$  ohms replaced the one having

a resistance of order  $10^{16}$  ohms.. The time constant of the recording system was then reduced to about 4 minutes, which was comparable with the length of each melting experiment, and it was necessary to correct all records of charging for this effect. The system was checked for electrometer drift under all conditions of temperature, humidity and wind speed likely to be used in the experiments. No charging greater than  $10^{-4}$  esu was recorded for any of these conditions for periods of up to 10 minutes..

#### 6.1.2 'Pure water' system

The continuous flow system for obtaining 'pure' water is shown in Fig. (6.2). De-ionised water was stored in a 10 litre glass reservoir in an atmosphere of nitrogen. The nitrogen pressure forced the water to flow through the system at a given rate.. After passing through the de-ionising column described in Chapter Four, the water flowed into a small brass chamber to which was soldered a stainless steel hypodermic needle. Fig. (6.4).. When not in use, the needle was stored in 'pure' water to avoid contamination. The main flow passed out of this chamber and into a conductivity cell. From the cell the water overflowed into a second glass reservoir where it was stored for re-cycling. The temperature of the water was recorded by a thermocouple mounted close to the conductivity cell.

All connections in the water system were made of

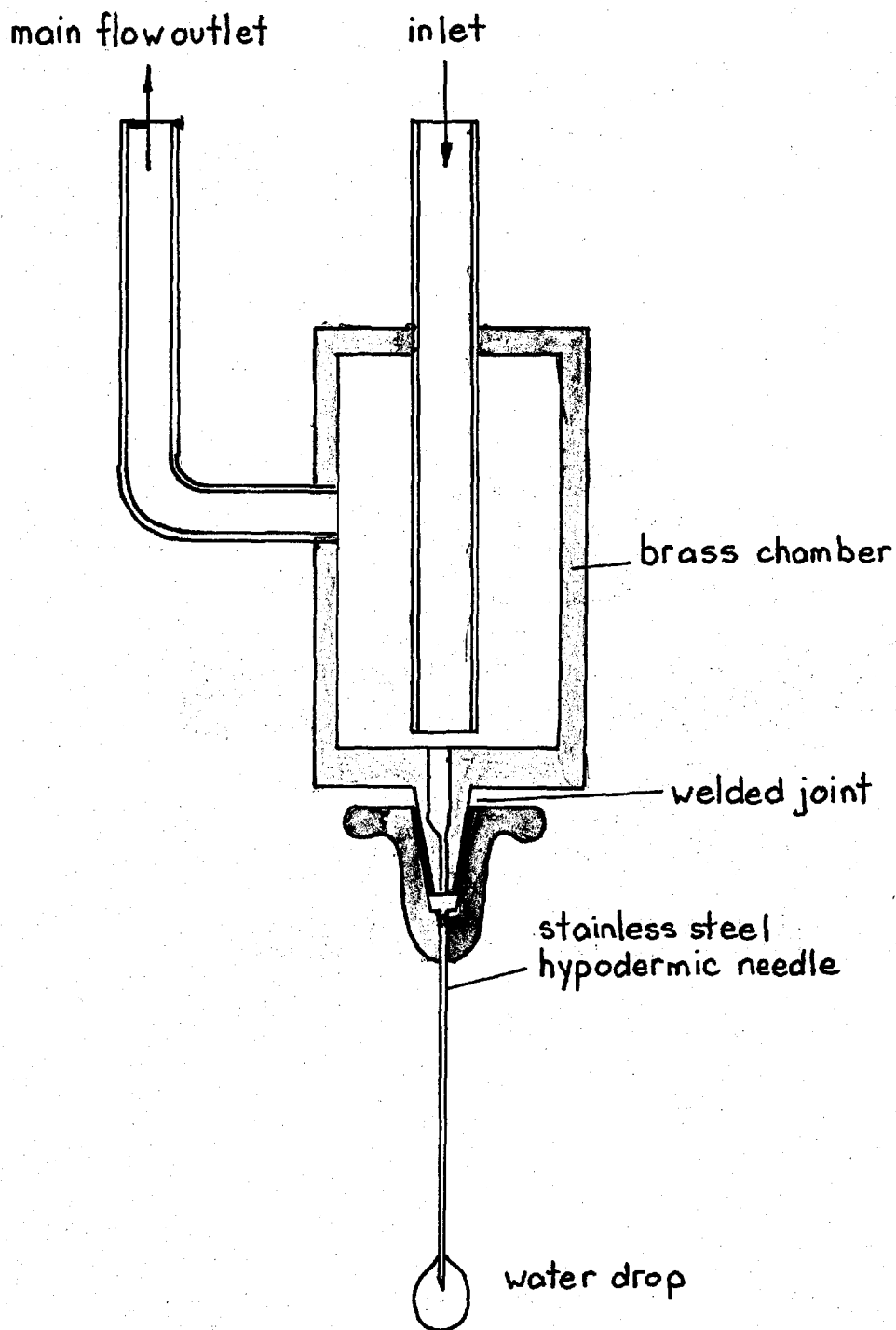


Fig. 6. 4 Detail of 'pure' water drop production chamber.

glass, except for two short tubes attached to the small brass chamber made of polythene, thus making the chamber and needle moveable. All glass components were thoroughly cleaned in hot Chromic acid before use, and washed out with distilled water. After flushing the system with about 10 litres of water it was found that the conductivity reading of the water entering the cell was extremely close to the intrinsic value for 'pure' water.. ( $0.045 \mu\text{mho cm}^{-1}$  at  $20^{\circ}\text{C}$ ).. It was assumed that the small flow of water issuing from the hypodermic needle was at least as pure as that indicated by the conductivity reading..

It was found that after leaving the water system for some days, it was essential to flush it through with at least 5 litres of water before use, as a slight soapiness appeared in the water, probably from the walls of the polythene tubes. Although this did not affect the conductivity reading, it significantly reduced the charging effects, and it was necessary to dispose of this 5 litres used for flushing the column, as the de-ionising column did not remove the soapiness on re-cycling. The presence of the soapiness was detected by vigorously shaking a cleaned glass container full of water from the system. The slightest persistence of bubbles at the surface was regarded as suspect, and the system flushed until this persistence was reduced to less than about 0.1 seconds..

## 6.2 Experimental procedure

It was decided to begin the examination by taking the most scrupulous care to avoid contamination or impurities at any stage of preparation or melting. To this end, only 'pure' water was used directly from the de-ionising system, with exposure only to an inert atmosphere of nitrogen. The results after introduction of known amounts of impurities could then be compared with these 'clean' results. The charge generation was to be examined in the light of the following factors.

- a) Rate of freezing
  - b) Rate of melting
- } (Temperature, humidity, wind speed)

### 6.2.1 'Pure' water in nitrogen

Before use, the tunnel system was thoroughly cleaned and all loose particulate matter removed by allowing a complete cylinder of nitrogen to exhaust through the apparatus at high pressure. The particulate filter was then replaced, and a periodic examination of it showed no visible sign of particulate matter in the system. After this procedure, the wire loop support for the specimens was boiled in distilled water for a few minutes. This was repeated frequently during the experiment.

The nitrogen flow rate and inlet temperature were then set by adjusting the separate valves on the hot and cold gas inlets. When conditions were steady, the small

hole for introduction of the specimens in the side of the wind tunnel was opened and the hypodermic needle inserted. The screen covered the gas inlet tube during this operation. The tip of the needle was held near the entrance for a short while. This caused the nitrogen stream from the wind tunnel to force the water drops issuing from the needle to be blown back along the needle, thus cleaning it and ensuring the removal of any water contaminated with  $\text{CO}_2$  during transfer from the storage bath to the wind tunnel. The needle was then inserted further into the wind tunnel and held just above the wire loop support until a water drop of the required size was formed on the end. The presence of the inlet screen prevented the drops from being blown along the needle. The water drop was then transferred to the wire loop, the needle withdrawn from the tunnel and the opening sealed. The inlet screen was then withdrawn, and the drop allowed to sit in the air stream for a few seconds in order to attain temperature equilibrium..

A hypodermic syringe without needle was then dipped in liquid nitrogen. After a few seconds it had reached a very low temperature, and it was withdrawn from the liquid nitrogen to a position just above the surface. Myriads of tiny ice crystals from just above the surface were then sucked into the hypodermic. This was then quickly attached to the Luer fitting on the wind tunnel

nucleating hole, and the ice crystals discharged into the inlet gas stream. The water drop in the wind tunnel was immediately nucleated, and was allowed to freeze completely in the nitrogen stream. After freezing, the dimensions of the ice specimens were recorded using a low power telescope. The ice specimens were approximately spherical, with occasional bulges and spikes.. In all further discussion the specimens are classified in terms of the equivalent spherical dimensions of the corresponding water sphere.

It is worth recording that the freezing process took place in an asymmetric manner. Fig. (6.5). On nucleation, a thin transparent shell of ice formed over the entire surface. The freezing then proceeded from the base of the drop upwards, forcing the ice shell to bulge and sometimes crack at the top of the drop. Occasionally long spikes were formed at the upper part of the drops having lengths of up to twice the sphere diameter. The longer spikes tended to curve in such a way that their points turned back towards the sphere. This is probably a consequence of the gas flow in the wake of the freezing specimens. Sometimes large cracks were formed in the specimens at the latter stages of freezing, and occasionally the ice sphere blew completely to pieces. This usually occurred in the very last few seconds of freezing.

Gas bubbles were released continuously during the

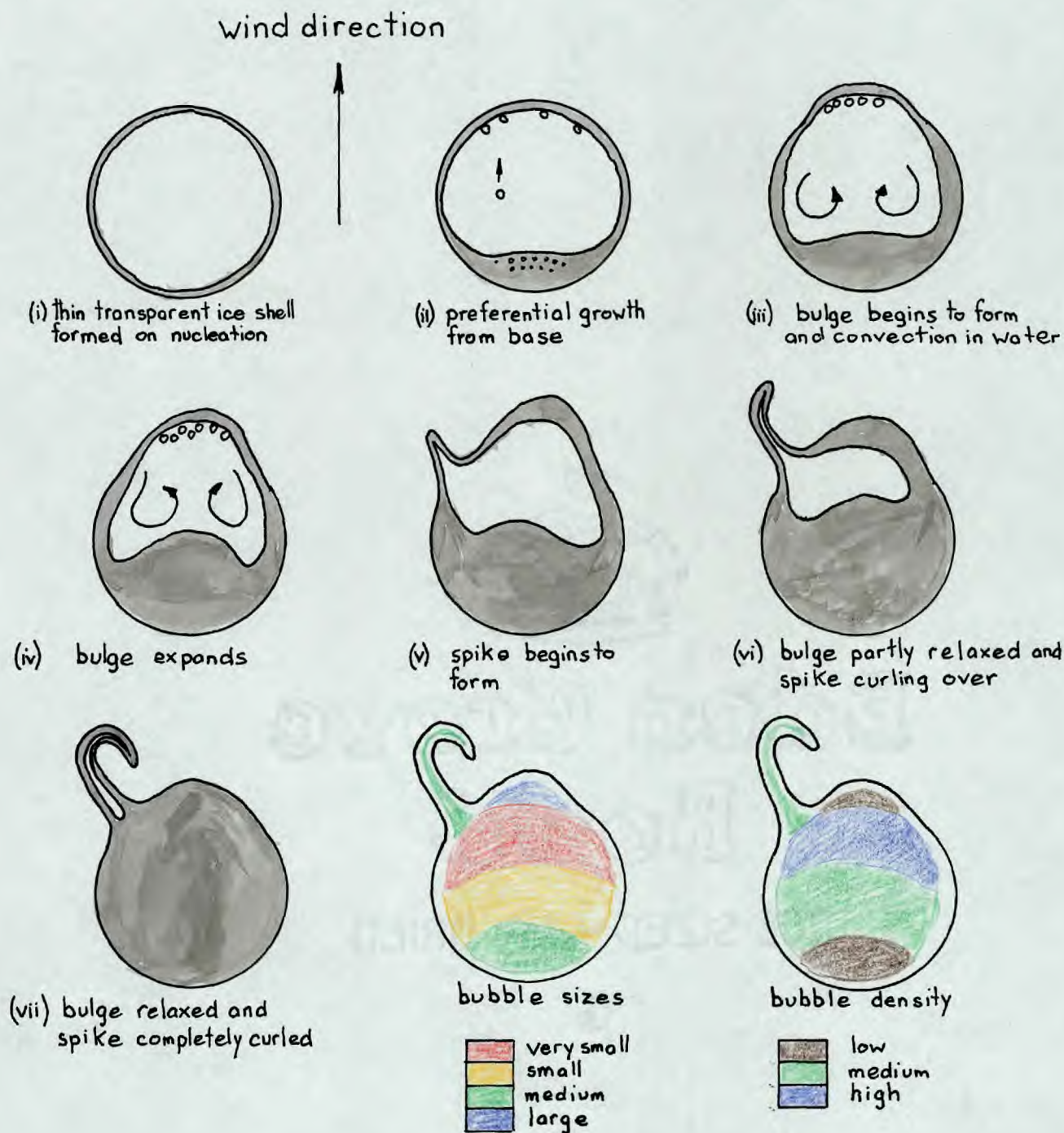


Fig. 6.5 Typical stages in freezing of water drops in wind tunnel with associated bubble structure.

freezing process and were incorporated into the ice structure. Sections of some of the specimens made in this way revealed that there was a pronounced layer structure of bubbles that indicated the contours of the freezing surface at different stages. The water in the bulges and spikes was usually the last to freeze, and this was accompanied by the release of a number of bubbles of very uniform size in the range 50 to 100 microns diameter. These could be observed as long spiral chains in the spikes.

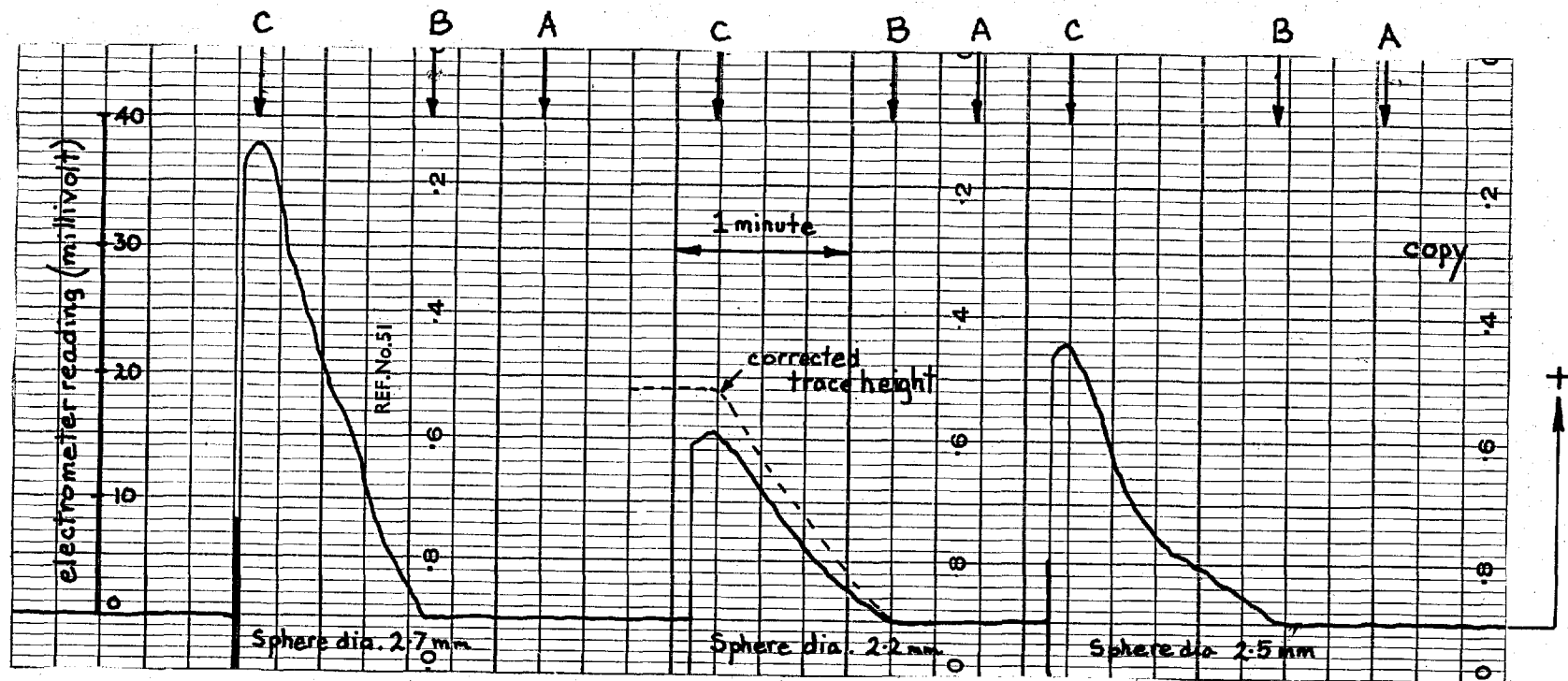
The time constant for absorption of nitrogen into the specimens prior to nucleation was observed by preparing a sample of boiled water in a hypodermic syringe, and using this to make the specimens while still hot. No difference in bubble structure could be found in the two cases even when only a few seconds elapsed between positioning the specimen and its subsequent nucleation. It will thus be assumed that in all cases the gas content of the water drops used to make the ice specimens is in equilibrium with the gas flow in the wind tunnel system before nucleation.

When the specimens were completely frozen, the Vibron electrometer and recorder were switched on, and the absence of any charging effect tested. By operating the inlet valves, a new wind speed and air temperature for melting could be selected. This operation typically

took 10 seconds to reach the selected values, which could then be maintained by continual adjustment of the controls. The ice specimens were allowed to melt completely in the new environment. The time for complete melting and the charge generation were recorded whilst visual observations were made of the melting process. After melting, the water drops were removed by suction with a small pipette.

### 6.3 Results for 'pure' water specimens in nitrogen

Some typical records of the charge generation are shown in Fig. (6.6). It is seen that in all cases the generation of charge begins sharply some time after the specimen has begun to melt. This period is greater than the 10 seconds required to adjust the temperature and wind speed of the new environment, and is interpreted as due to the melting of the thin transparent surface shell formed on nucleation of the water drop, which contains no gas bubbles, and the fact that convection in the water portion does not begin until approximately  $\frac{1}{3}$  to  $\frac{1}{2}$  of the time for complete melting has elapsed. The charging finishes at the same instant as the melting finishes, and visual observation showed that the maximum persistence of bubbles after the last traces of ice had disappeared was of order 0.1 seconds. The decay in the charge after the melting finishes is due to the finite time constant



Capacitance of recording system = 262 p.f.

Spheres frozen at  $-5^{\circ}\text{C}$ . Melted at  $+12^{\circ}\text{C}$ . Wind speed  $8. \text{m. sec}^{-1}$

- A Melting begins
- B Convective activity begins
- C Melting ends

Fig. 6.6 Typical charging records for spheres melting in nitrogen.

of the electrometer system, and the corrected maximum charge is of order 20% greater than that indicated, for the cases shown. A corrected record is shown for comparison. It was found that for specimens prepared and melted under identical conditions, the generated charge expressed in terms of esu per  $\text{cm}^3$  of melt water varied by as much as 50% in some cases, though the spread about the mean value was typically of order  $\pm 20\%$ . Possible errors lay in the measurement of diameter and in slight variations in wind speed and temperature of the environment which can have a major effect. No significant variation could be found in the mean charge with size of the specimen in the range 1.0 to 5.0 mm diameter: consequently the experiments were later restricted to spheres having a mean diameter of 2.5 mm, as this was found to be experimentally convenient. The graphs discussed below were constructed by taking the mean charge per  $\text{cm}^3$  from 10 specimens to plot each point.

Fig. (6.7) shows the effect of gas temperature on the charge, for specimens frozen under identical conditions and melted at a constant wind speed. Because of the extremely low humidity of the nitrogen, melting only just began at a gas temperature of  $+9^\circ\text{C}$ . In this case the spheres took longer than 15 minutes to melt, and any charging was not detectable. A rapid increase of the charge per  $\text{cm}^3$  of melt water then takes place as the gas

temperature increases, followed by a slow decrease for temperatures above about  $+14^{\circ}\text{C}$ . The rapid increase in charge was observed to coincide with the development of strong internal convection in the melting ice specimens. The convective activity was not always equally strong for apparently identical specimens and conditions, and this probably accounts in part for the large variation in charge per  $\text{cm}^3$  of melt water between individual results. The highest charges from any group of ten results used to plot one point on the graphs was definitely correlated with the greatest convective activity as observed visually.

Essentially the same behaviour as with the variation with gas temperature is found when an examination of Fig.(6.8) is made, where the effect of wind speed in the melting environment for given freezing conditions, and gas temperature used for melting, is shown. This indicates that the charge generation is probably associated with the rate of melting rather than gas temperature and wind speed individually.

The results of Figs.(6.7) and (6.8) have been replotted in terms of the approximate mean heat transfer factor, in Fig.(6.9). This factor has been calculated on the assumption that the nitrogen stream has a humidity of 0%, and that the mean temperature difference between the melting sphere and the environment is equal to  $T_w/2$ ,

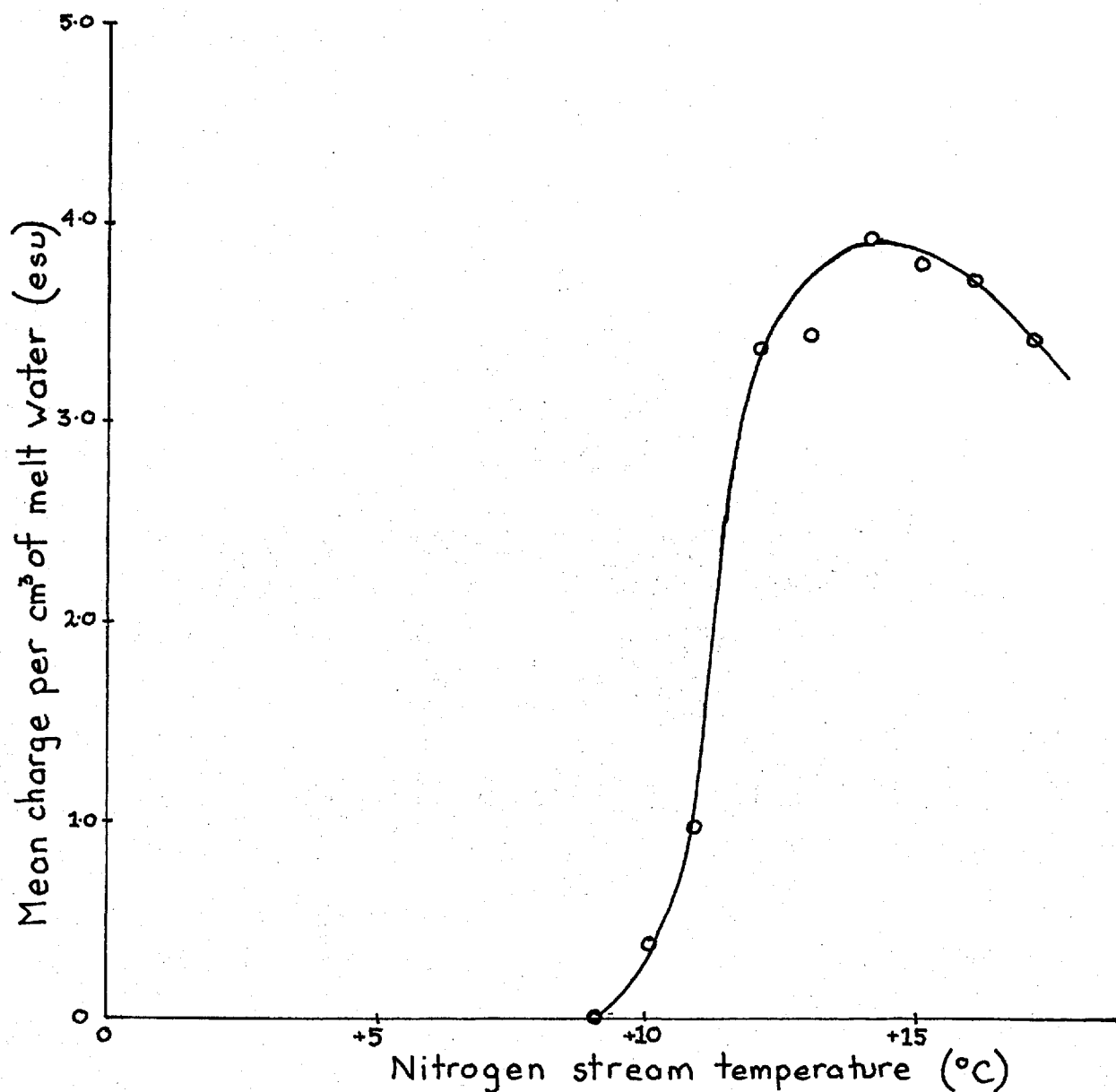


Fig. 6.7 The mean charge per cm<sup>3</sup> for melting ice specimens made from 'pure' water, as a function of melting environment temperature.  
Freezing temperature  $-5^{\circ}\text{C}$ . Windspeed  $8 \text{ m. sec.}^{-1}$

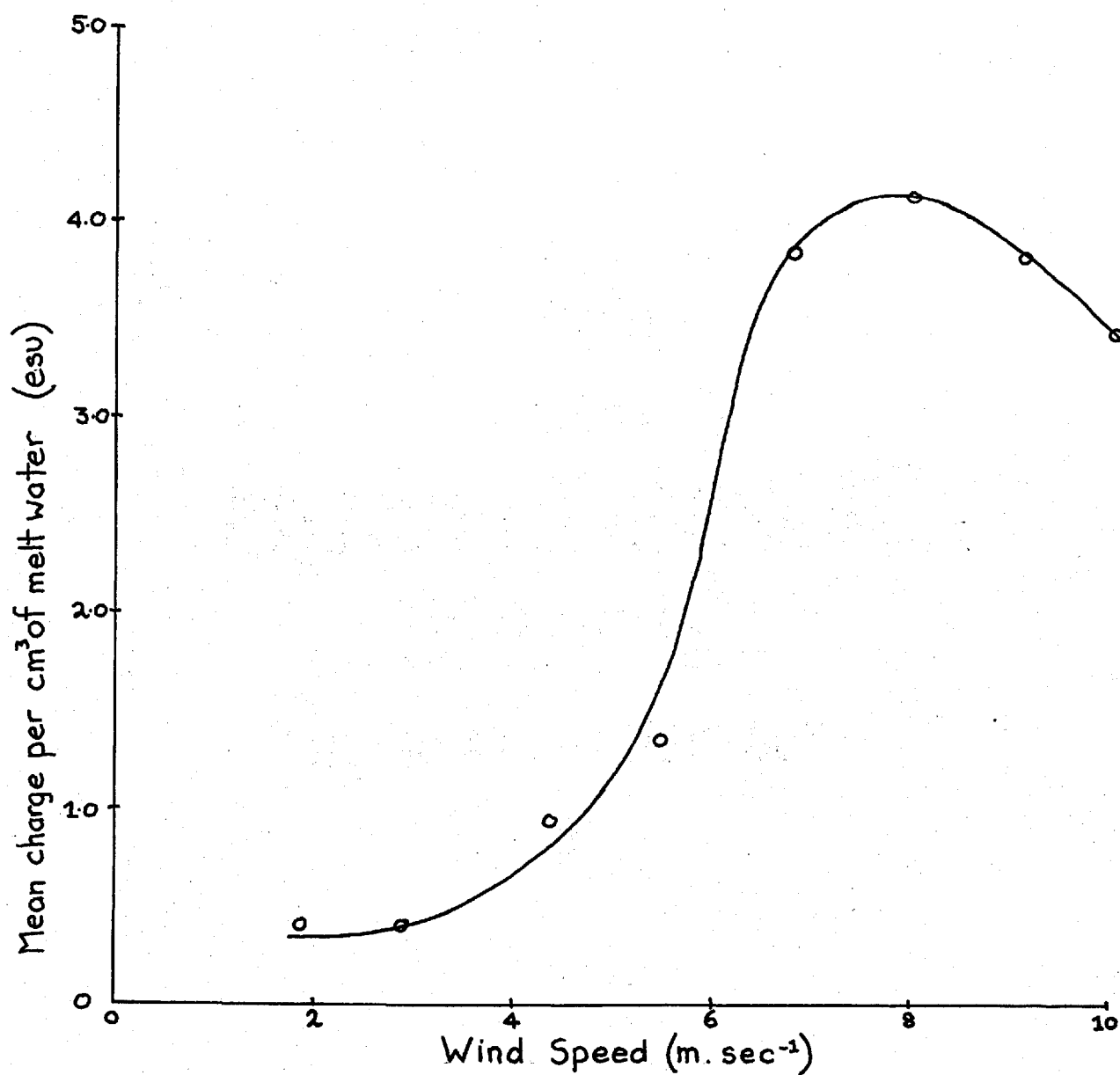


Fig. 6.8 The mean charge per cm<sup>3</sup> for melting ice specimens made from 'pure' water, as a function of wind speed. Melting stream +14°C. Freezing temperature -5°C. Nitrogen environment.

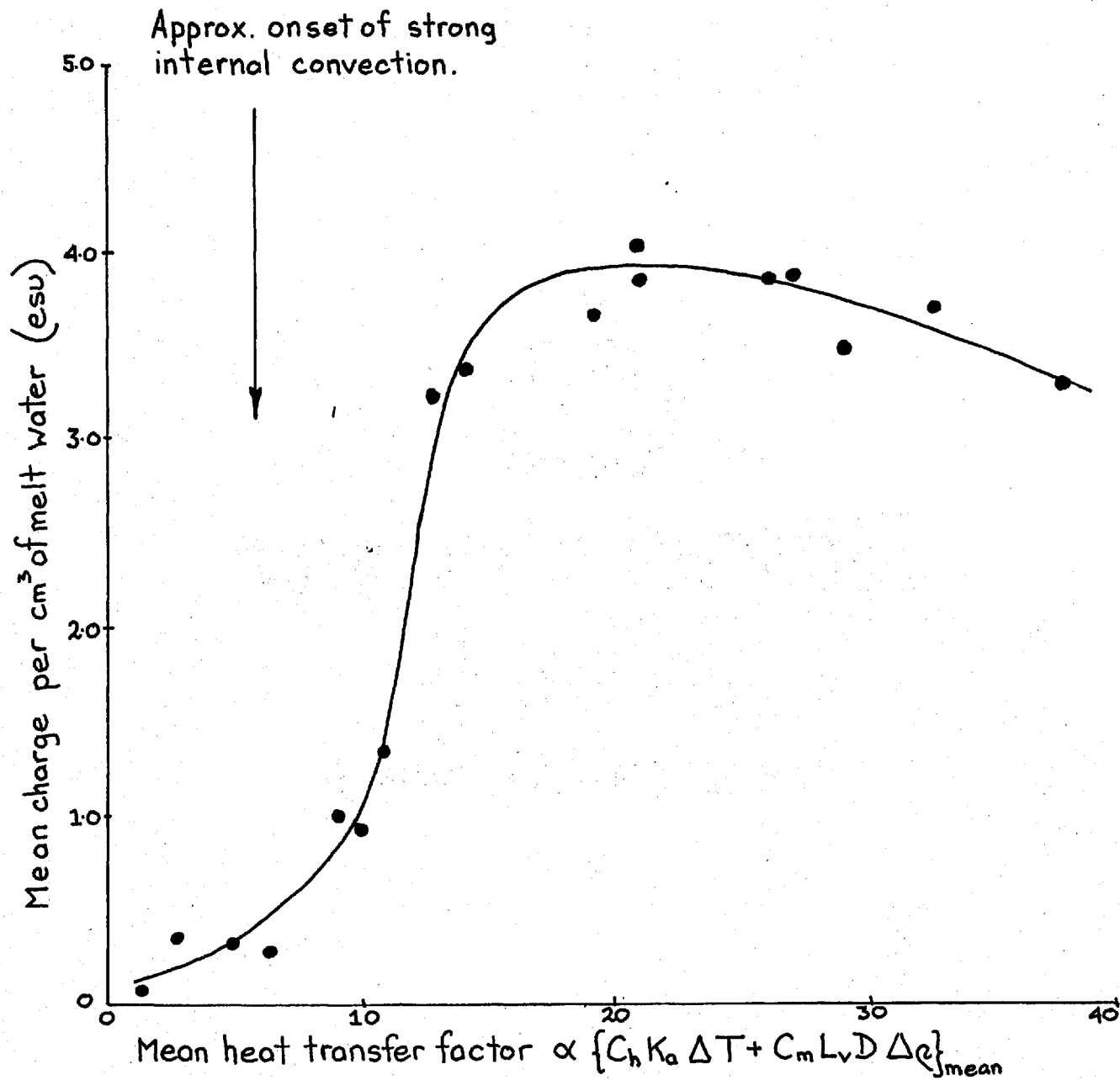


Fig. 6.9 Graph showing charge separation as a function of the mean heat transfer factor for melting. i.e. Rate of melting.  
Freezing temperature  $-5^{\circ}\text{C}$  at  $8 \text{ m. sec}^{-1}$

where  $T_w$  is the wet-bulb temperature of the air stream. It can be seen that there is a definite correlation of the charge per  $\text{cm}^3$  of melt water with the rate of melting of the specimens. The reason for the slow decrease after passing through the maximum may be that the time between release of the gas bubbles from the ice, and the subsequent bursting at the surface, is comparable with the relaxation time for formation of the electric double layer around the bubbles. This time between release and bursting was observed to be less than 1 second for temperatures greater than about  $+12^\circ\text{C}$ . Iribarne (1966a) estimated the time for formation of the electric double layer to be of order a few tenths of a second in the case of pure water.

The effect of freezing temperature on the charge production is indicated in Fig.(6.10). The upper limit of temperature ( $+2^\circ\text{C}$ ) was fixed by the consideration that the ice crystals used for nucleation failed to reach the specimen, either melting or evaporating in the warm gas stream. The lower limit was set by the temperature of the cold room used as the heat exchanger ( $-15^\circ\text{C}$ ). A single wind speed of about  $8 \text{ m sec}^{-1}$  was used for freezing the specimens, corresponding to the approximate natural fall speed for specimens of the mean size used. The curve shows a slight maximum in the region of  $-3^\circ\text{C}$ . If the freezing process is interpreted

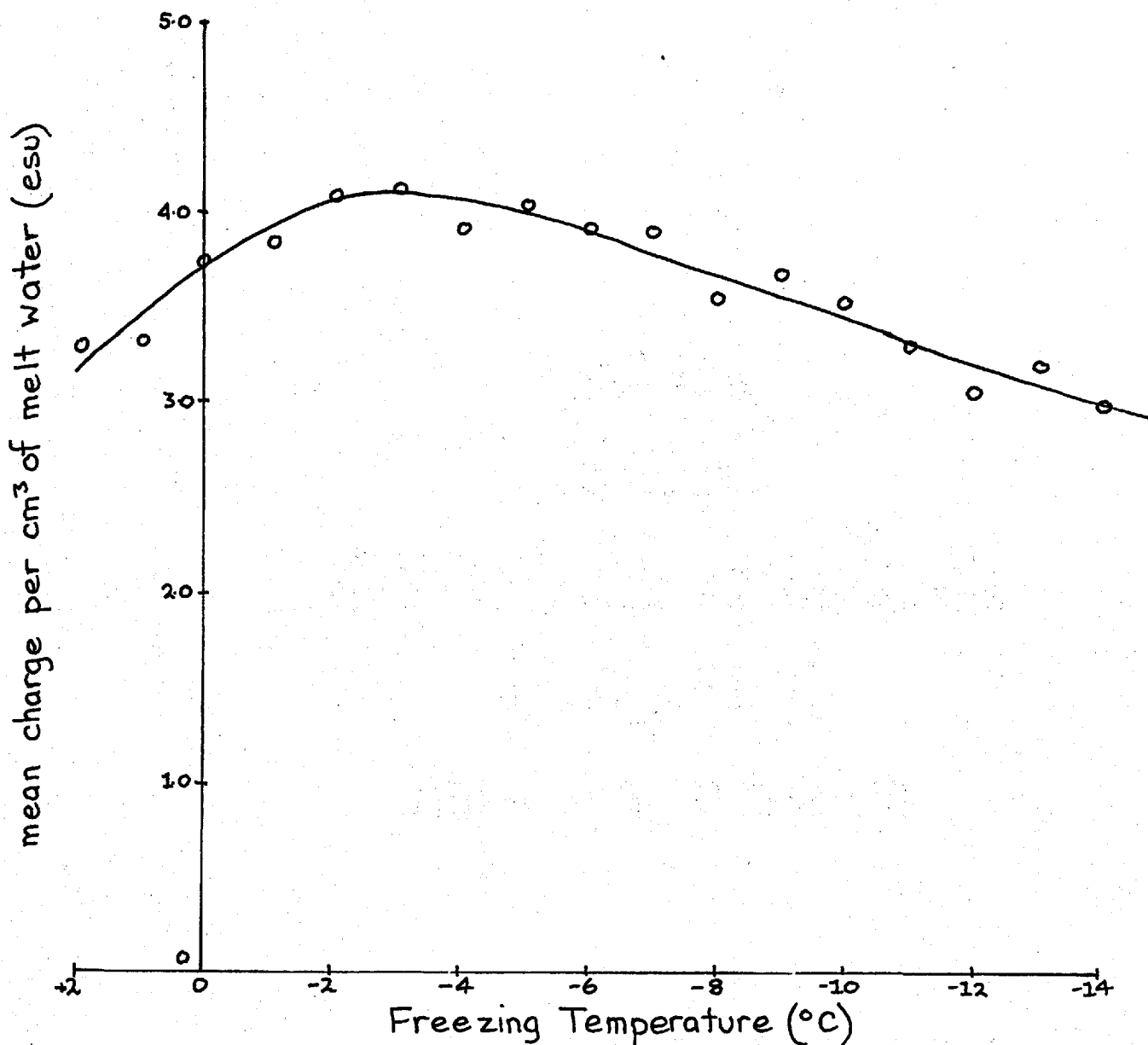


Fig. 6.10 The mean charge per cm³ for melting ice specimens made from 'pure' water, as a function of freezing temperature.

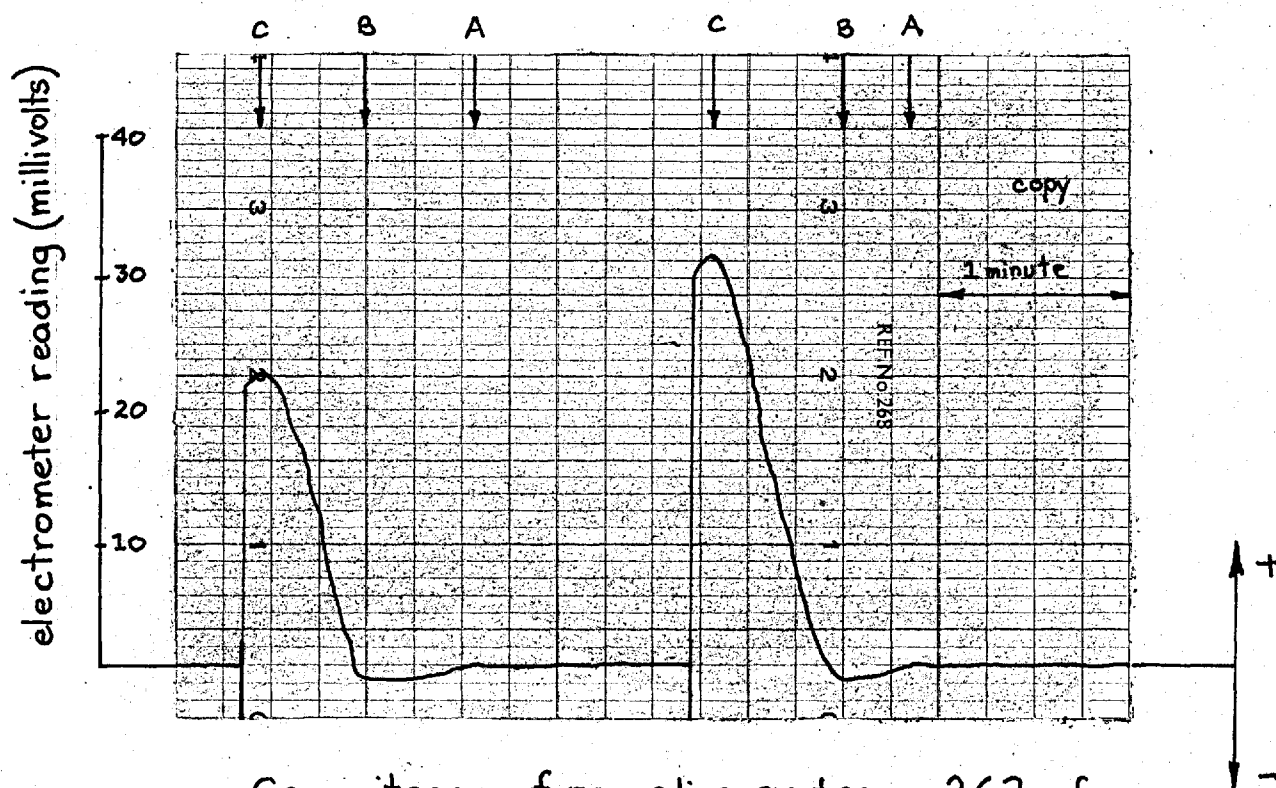
Melting stream +14°C. Wind speed 8 m. sec<sup>-1</sup>

in terms of heat factors, then the experimental range ( $+2^{\circ}\text{C}$  to  $-14^{\circ}\text{C}$ ) at very low humidity corresponds approximately to ( $-5^{\circ}\text{C}$  to  $-30^{\circ}\text{C}$ ) at 100% humidity. This latter range covers the region in which freezing might typically take place in a cloud. However, consideration of the possible application of the results to the generation of atmospheric charge is deferred until after discussion of the effects of impurities and nature of the melting gas are considered in the next chapter.

#### 6.4 Other effects

(1) It was again found that, as in the case of the large wind tunnel, the slightest touch of the support with a finger reduced the charge generation by as much as an order of magnitude.

(2) When the melting was carried out using a cylinder of compressed air, the same final results were recorded as in the case of melting in nitrogen. This is as expected, as any  $\text{CO}_2$  present in the cylinders would be completely liquified at the pressures used. (100 lb/in<sup>2</sup> to 2000 lb/in<sup>2</sup>). However, it was observed in this case that the charging records did not show the sudden start some seconds after melting began. See Fig.(6.11).. Instead a small negative charge was recorded as soon as the melting began (of order 0.1 esu per cm<sup>3</sup>), the positive charging beginning at approximately the same time as before.. Discussion of this effect is delayed until the Appendix.



Capacitance of recording system = 262 p.f.

Spheres frozen at  $-5^{\circ}\text{C}$

Melted at  $+12^{\circ}\text{C}$ . Wind speed  $8 \text{ m. sec}^{-1}$

A Melting begins

B Convection begins in water

C Melting ends

Fig. 6.11 Typical records for spheres melting in air from cylinder.

(3) In order to test the effect of prolonged exposure of the specimens to the air stream, and the possible accumulation of surface contaminants, a single specimen was subjected to a series of meltings and re-freezings. It was found that a small reduction of charge took place over a period of about 20 minutes, (5 meltings), amounting to approximately 10% of the initial charge.

(4) In order to test the possible influence of the support used for the specimens on the magnitude of the charge separation, several loops of different wire and having different diameters were used. No significant change in the mean charge under any given conditions could be detected for loops made from copper, contra,, stainless steel and manganin, and having diameters between 0.5 mm and 2.5 mm.. No correlation could be found between the size of the loop and the degree of convective activity in the water portion of the melting specimen.

## CHAPTER SEVEN

### Impurity effects and change of environment

#### 7.1 Change of environment

##### 7.1.1 Melting environment

The hot inlet line was disconnected from the cylinder fitting and attached to a vacuum cleaner pump open to the laboratory. Specimens were prepared in an identical manner to those of Chapter Six, and melted in a mixed environment of cold nitrogen and hot laboratory air. The ratios of mixing were not determined. Tests conducted with a few selected values of freezing temperature, gas temperature for melting and wind speed showed that the charging levels were slightly increased ( $\sim 10\%$ ) over the former case, and that the maximum associated with the temperature of the melting environment was shifted towards lower values. This latter effect was interpreted as due to the increased humidity of the melting environment, leading to an increase in the rate of melting at any given gas temperature. The increase in the charging levels was both unexpected and surprising, as it was thought that the  $\text{CO}_2$  content of the air would have a reducing effect. The level of  $\text{CO}_2$  in the laboratory air was probably much greater than the normal ground level concentration in the mean atmosphere, due to the use of solid  $\text{CO}_2$  in other parts of the building, and

the housing of the experiment being in a basement of a laboratory in Central London.

Blocks of solid  $\text{CO}_2$  were then introduced into the dust bag space of the vacuum cleaner and the experiments repeated at isolated points. There was some evidence that the charging levels were further increased, although the increase was only on the border line of significance (about 3%). However, in view of the experience of previous workers on the poisonous effects of  $\text{CO}_2$  on the charge production, the fact that the charge was not decreased is highly important.

The slight negative excursion at the beginning of the charging records was again in evidence.

#### 7.1.2 Freezing and melting environment

The cold gas inlet was now connected to a second vacuum cleaner mounted inside the cold room. Before use, all other apparatus was removed from the cold room and the walls, floor and ceiling cleaned. The experiments were then repeated for approximately the same ranges of freezing temperature and wind speed as in the case of 'pure' water in nitrogen. The range of air temperature used for melting was much lower due to the increased humidity of the melting environment. The maximum levels of charge generated were, in general, increased by a factor of 1.5 to 2.0 over the case of preparation and melting in nitrogen. The increase varied considerably with the ambient humidity

of the laboratory air. Typical results for the 'pure' water are shown in Figs.(7.1) (7.3) (7.4): these figures are discussed in detail later in comparison with those for water containing known impurity concentrations.

The small negative excursion on the charging records was again noticed when melting had just begun.

## 7.2 Introduction of ionic impurities

Because of the variation of the charge with ambient humidity mentioned in section (7.1.2) it was realised that in order to compare the results for different samples of water, it was desirable to carry out the experiments in a period of time in which the humidity was approximately constant. This was not entirely possible due to the number of specimens required for each complete set of results. The system adopted was to choose a few water samples, and to restrict the number of individual results for each point on the graphs to five. Using all the water samples, results were taken at a single wind speed and temperature as quickly as possible, the support being cleaned between use of each water sample. In this way a complete set of curves for all samples could be obtained in a continuous period of about 10 hours. The experiments were restricted to days when the weather situation over the South East of England was reasonably stable; even so, considerable variation of humidity sometimes took place during the course of the experiments, especially during

the early morning. In these cases it was necessary to repeat the earlier results. The results presented later in this chapter are those for which the variation of humidity over the experimental period was less than about 10% at any given air temperature, except where stated.

Various samples of water were prepared by adding ionic salts to 'pure' water in known concentrations. These were stored for use in cleaned glass bottles with glass stoppers. To prepare an ice specimen for melting, a clean glass pipette was used to suck a small sample of water from the storage bottles. The lid was then removed from the wind tunnel and a drop of diameter about 2 to 3 mm was positioned on the wire loop support. The lid was then replaced and the air stream controls adjusted to give a specific freezing temperature and wind speed. After supercooling for a few seconds the water drops were nucleated in the manner previously described and allowed to freeze. The freezing behaviour was essentially similar to that described for freezing in a nitrogen stream. The observation and recording of the charge generation took place as before.

The impurity concentrations of the bulk water samples were checked using a dip-type conductivity cell, as well as by calculation from the weight of dissolved salts. The solutions containing  $\text{CO}_2$  were prepared by bubbling  $\text{CO}_2$  through the water: this was done continuously

throughout the experimental period because of the small time constant for solution of  $\text{CO}_2$  in water. The exact  $\text{CO}_2$  concentration was not recorded, as by the time the specimens were nucleated in the wind tunnel the concentration was unlikely to be anything like the concentration in the bulk samples. It was inferred from the freezing behaviour however, that the specimens contained a concentration higher than the equilibrium concentration of the air stream. The ice specimens prepared from  $\text{CO}_2$ -bubbled water contained much larger bubbles for a given freezing temperature and were more prone to produce large bulges and spikes. The spikes, in particular, sometimes showed peculiar distortions in direction during growth and resembled writhing serpents in appearance..

The results of the charge generation and its dependence on wind speed, air temperature, freezing temperature and impurity concentration are shown in Figs.(7.1) to (7.7), which are discussed below.

### 7.3 Graphical results

NOTE In interpreting the results it is essential to avoid comparing the magnitudes of the charges generated between different figures, as misleading conclusions will be drawn due to differences in ambient humidity in the laboratory on different days. However, the general shape of each curve was consistent over a wide range of humidity and may be compared..

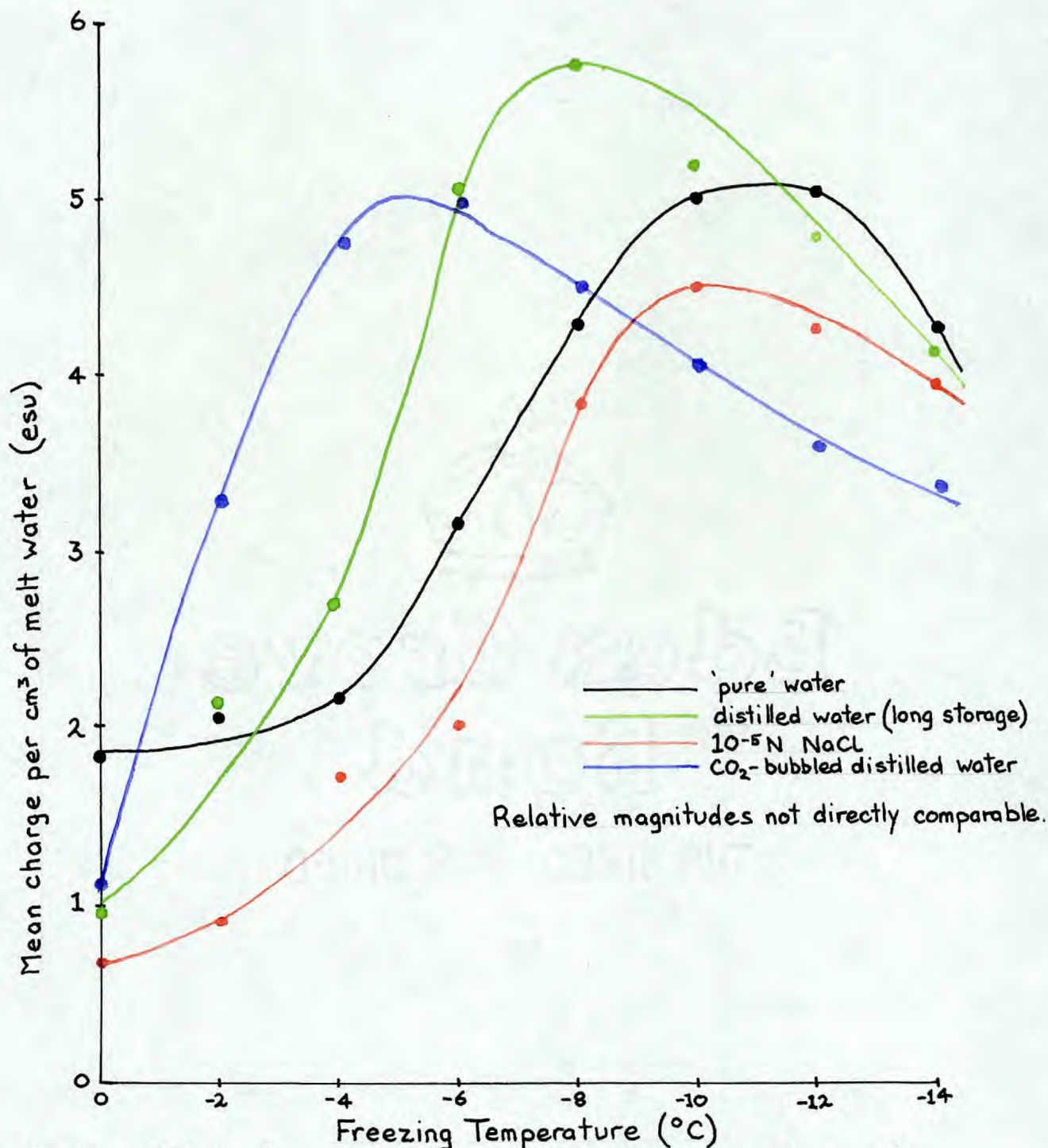


Fig. 7.1 Typical curves showing charge separation for different water samples as a function of freezing temperature, for melting in laboratory air. Melted at  $+8^{\circ}\text{C}$ ; wind speed  $\sim 8 \text{ m. sec}^{-1}$

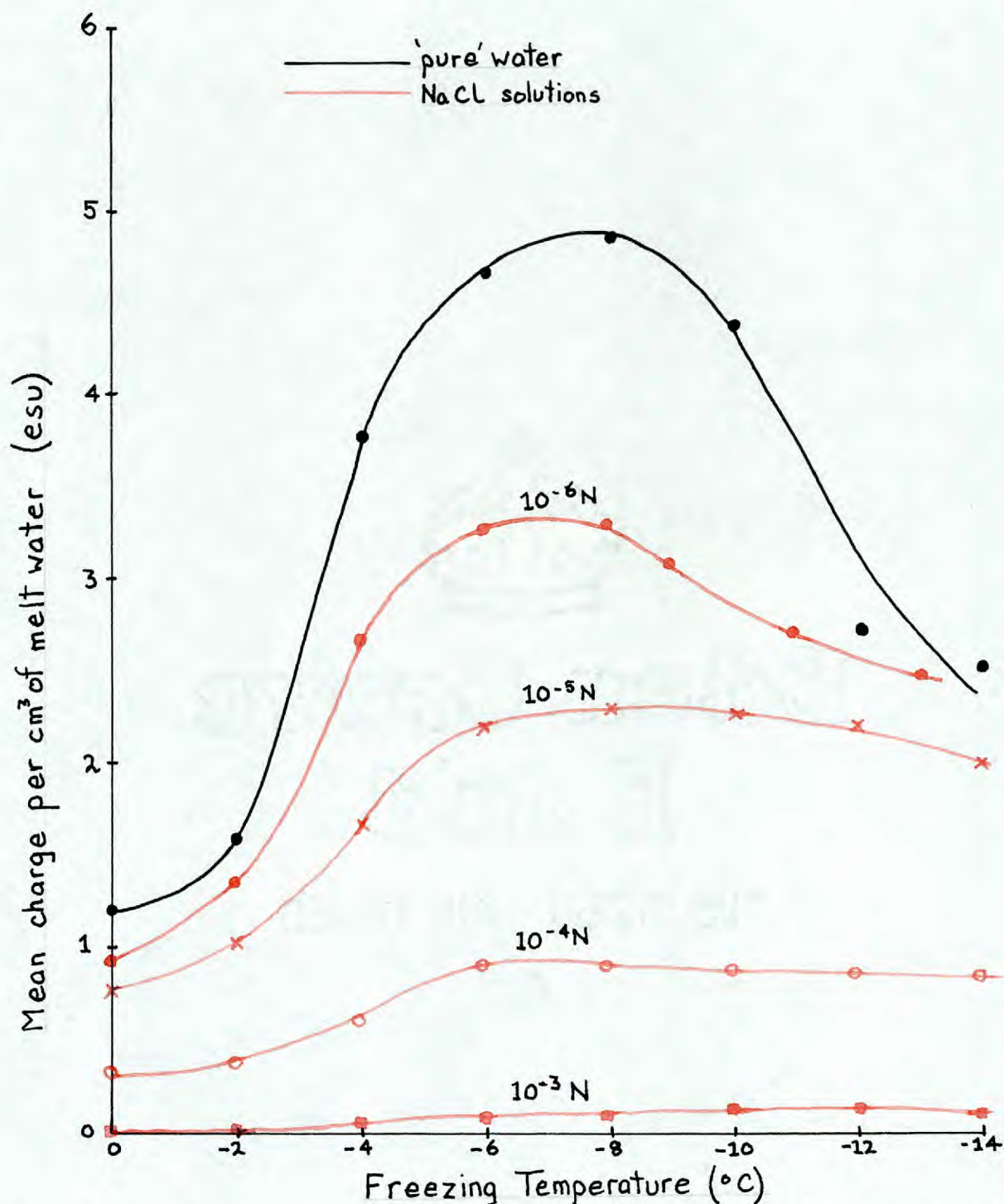


Fig. 7.2 Typical family of curves for different NaCl concentrations showing charge separation as a function of freezing temperature, for melting in laboratory air.  
Melting stream  $+8^{\circ}\text{C}$  : wind speed  $\sim 8 \text{ m. sec.}^{-1}$

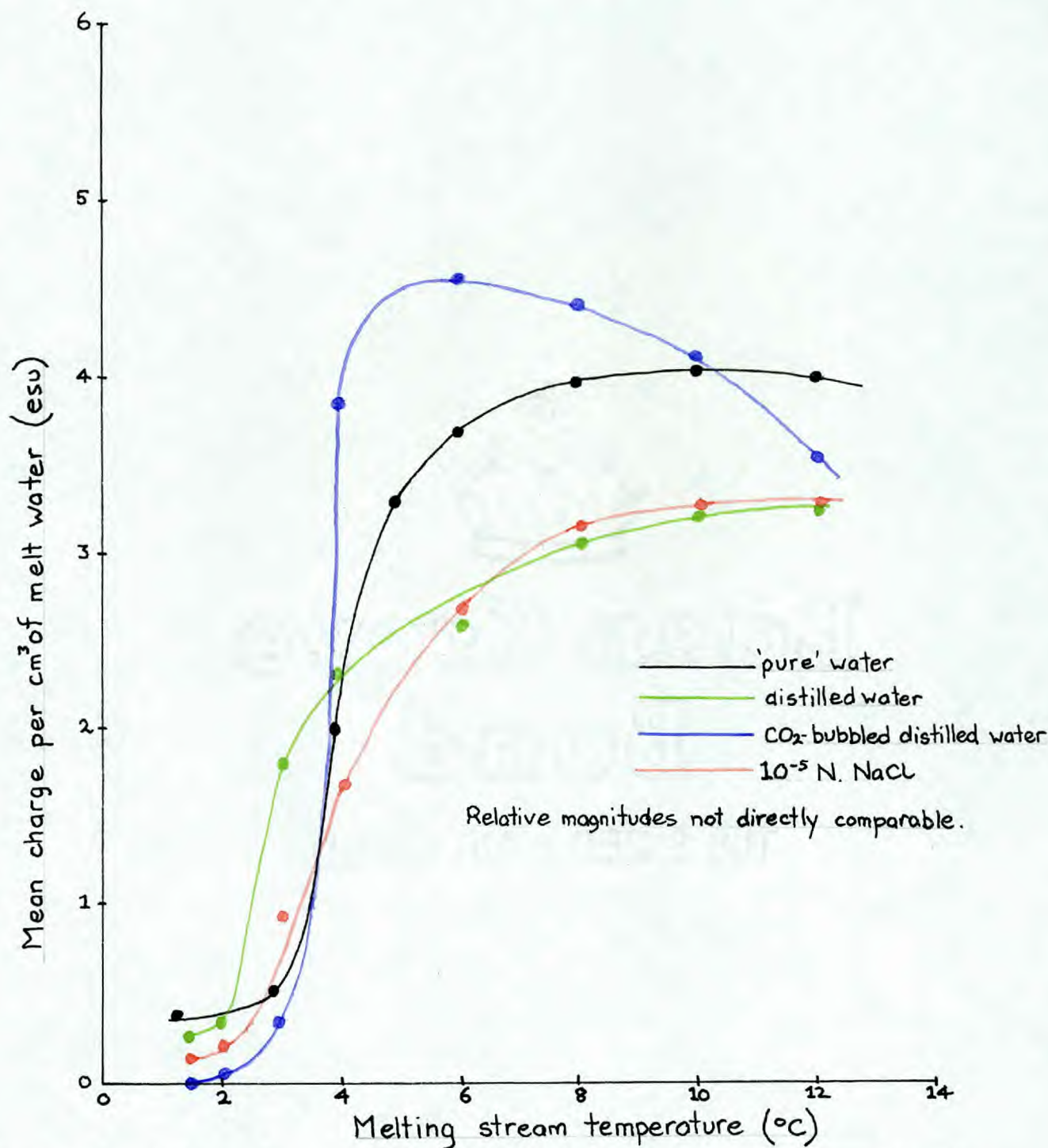


Fig. 7.3 Typical curves showing charge separation as a function of air temperature used for melting. Wind speed  $\sim 8 \text{ m. sec}^{-1}$ . All specimens frozen at  $-6^\circ \text{C}$ .

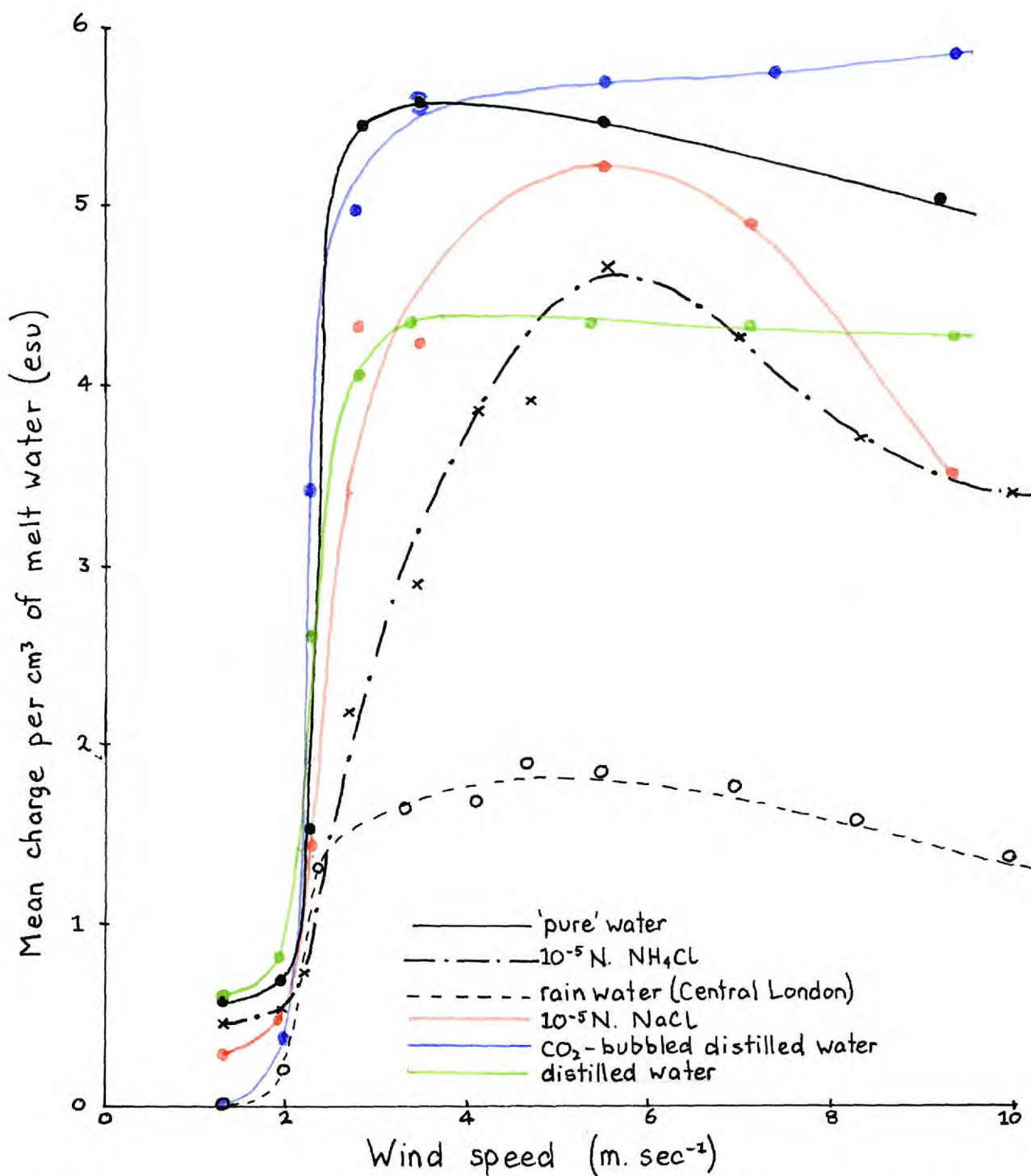


Fig. 7. 4 Typical curves showing charge separation as a function of wind speed for melting in air  
Freezing temperature  $-6^{\circ}\text{C}$ : melted at  $+6^{\circ}\text{C}$ .

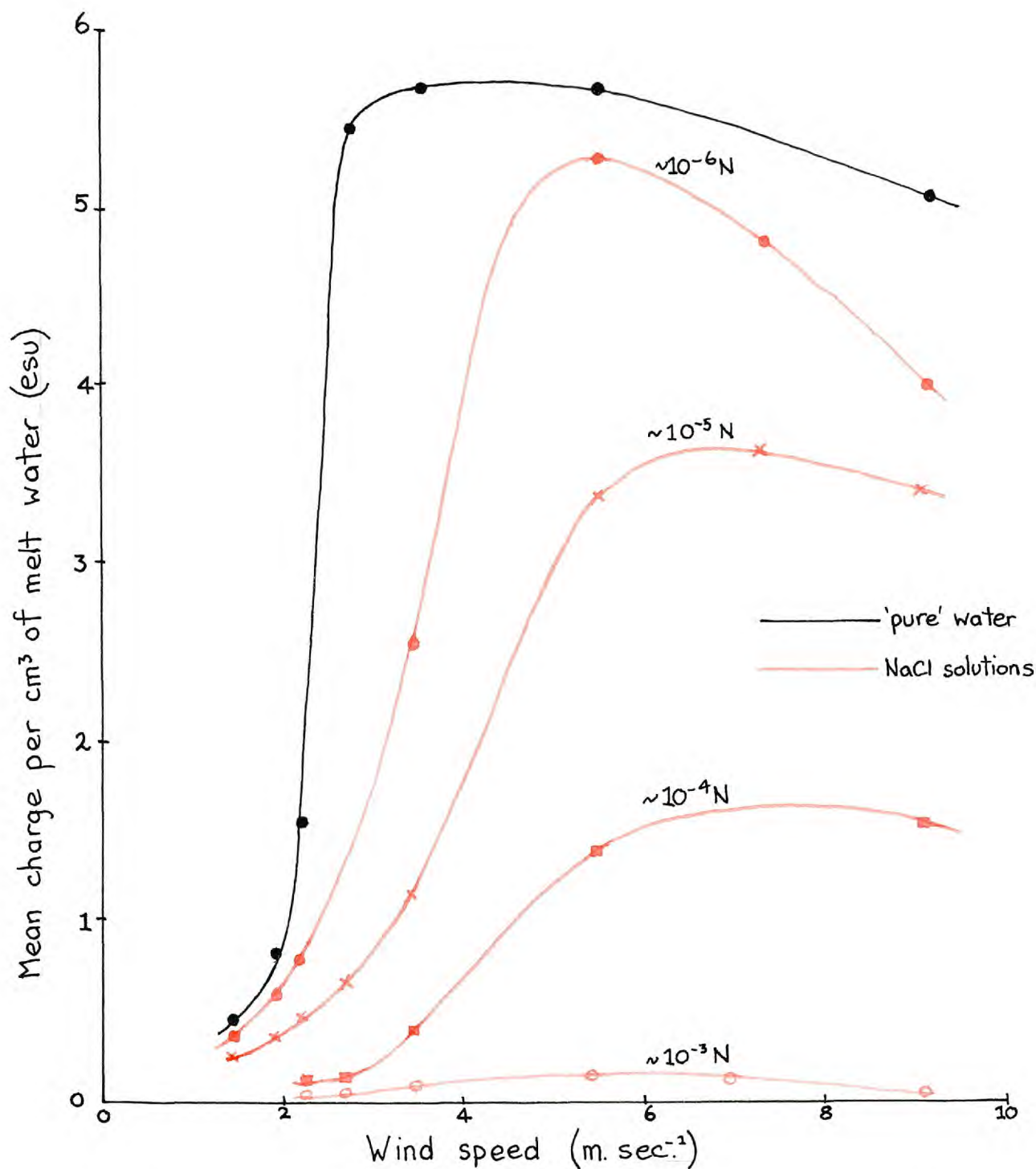


Fig. 7.5 Typical family of curves showing charge separation as a function of wind speed used for melting in laboratory air, for different concentrations of NaCl.  
Freezing temperature  $-6^{\circ}\text{C}$ : melted at  $+6^{\circ}\text{C}$ .

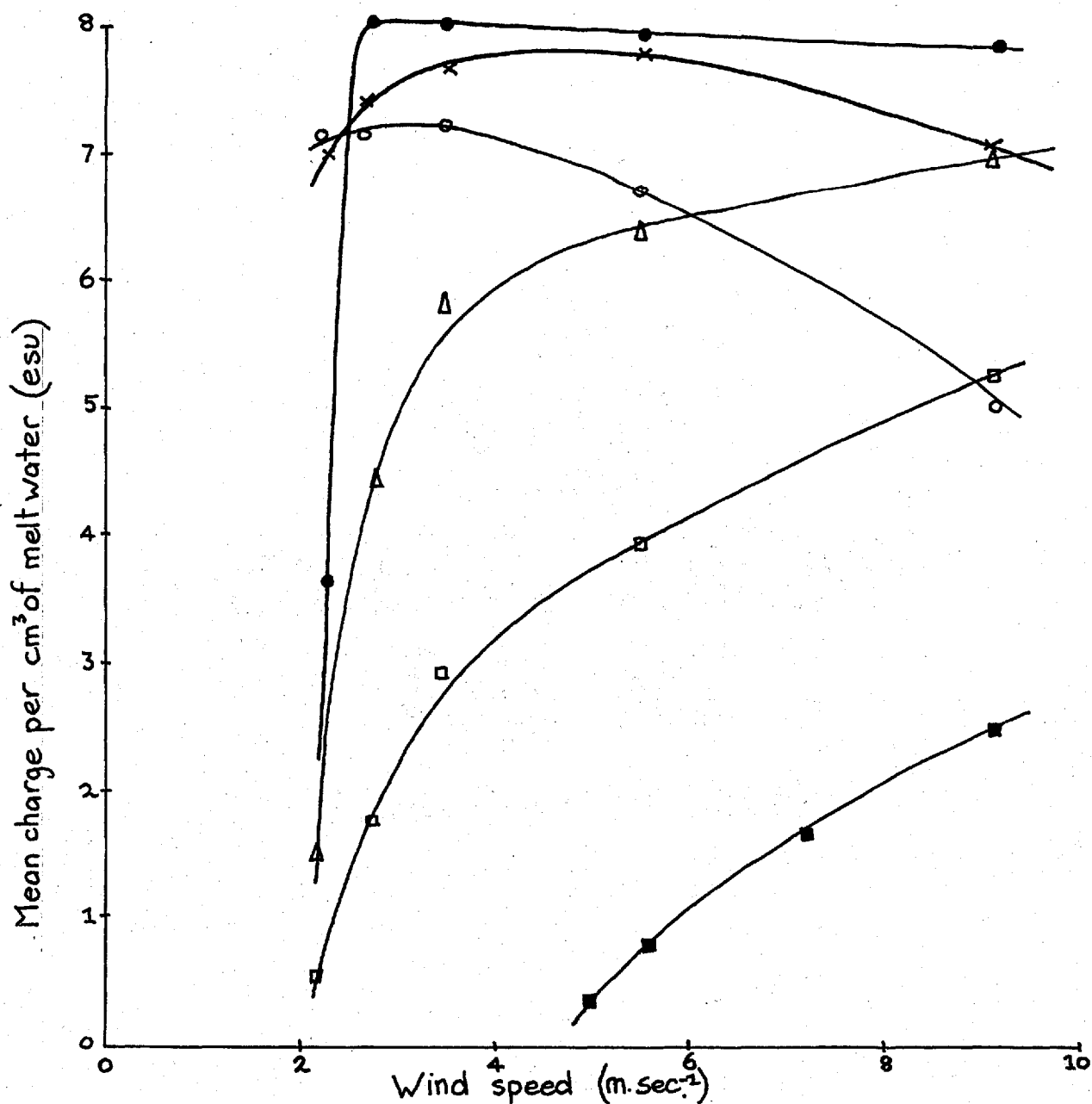


Fig. 7.6 Family of curves showing charge separation as a function of wind speed and air temperature, for 'pure' water specimens.  
All specimens frozen at  $-10^\circ\text{C}$  and  $8 \text{ m} \cdot \text{sec}^{-1}$

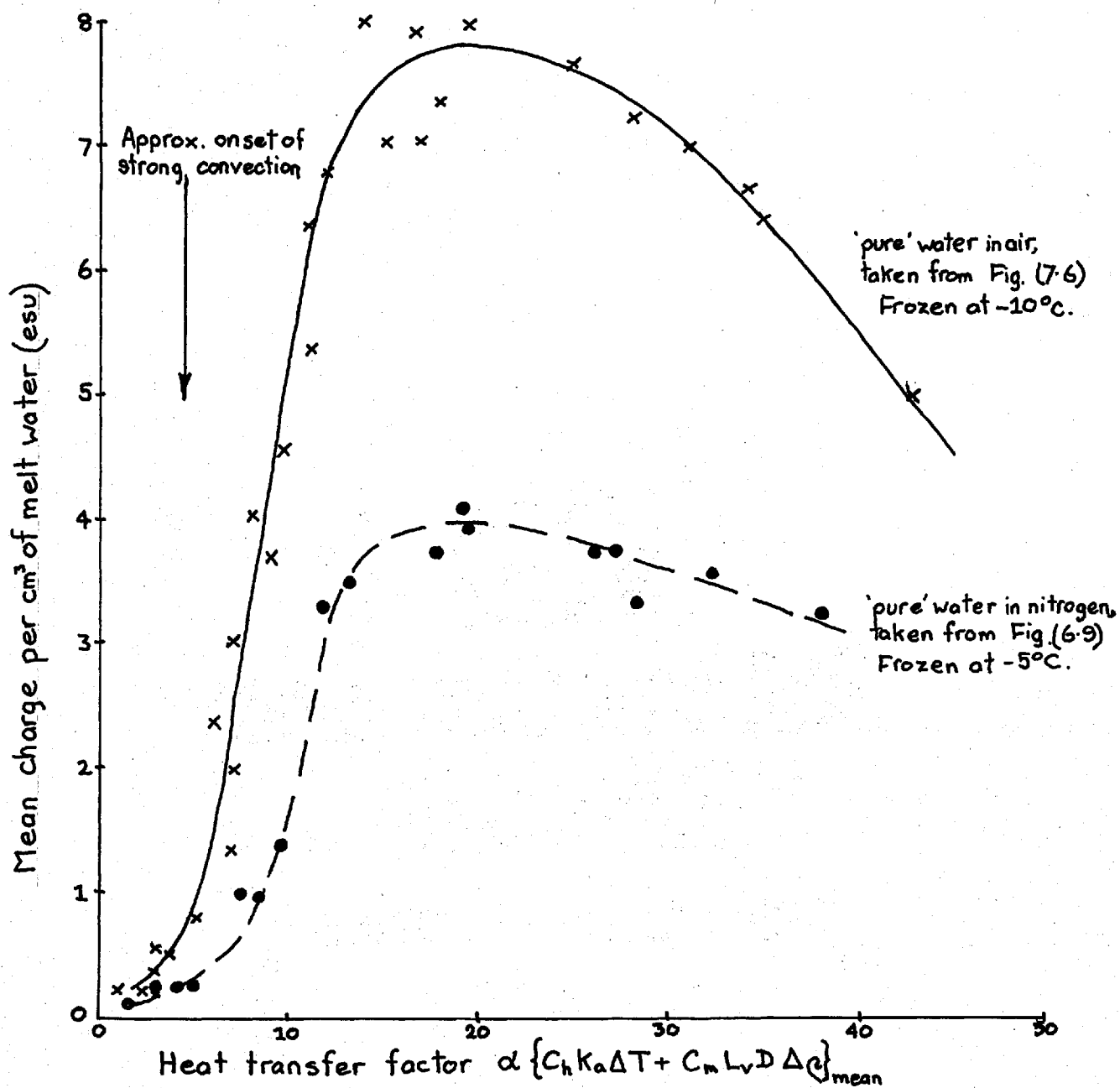


Fig. 7.7 Graph showing charge separation as a function of the heat transfer factor for melting, i.e. rate of melting. Rates of freezing were similar in the two cases. Difference in magnitude attributed to  $\text{CO}_2$ .

In all figures, the charge referred to is the equivalent charge per  $\text{cm}^3$  of melt water calculated from the final value recorded by the electrometer. The small negative excursion at the start of melting has been entirely neglected for the purpose of construction of the graphs as this is not considered to be due to the bursting of bubbles. This effect will be discussed in detail later in the Appendix after Chapter Eight.

### 7.3.1 Freezing temperature

Figs.(7.1) and (7.2) show the typical effect of freezing temperature on the charge generation for various water samples melted at a single temperature and wind speed. The curves are all essentially similar in shape having maxima in the region  $-8^{\circ}\text{C}$  to  $-11^{\circ}\text{C}$ , except for the case of the  $\text{CO}_2$ -bubbled distilled water which has a maximum in the region  $-4^{\circ}\text{C}$  to  $-5^{\circ}\text{C}$ . In all cases the specimens were frozen at a wind speed of approximately  $8 \text{ m sec}^{-1}$ , this value being chosen as the approximate natural fall speed of the specimens. The maxima were found to lie in approximately the same position when other combinations of air temperature and wind speed were used.

The difference in the position of the maximum in the case of 'pure' water in nitrogen, Fig.(6.10), and the curves of Fig.(7.1) excluding the  $\text{CO}_2$ -bubbled case, is interpreted as being due to the great difference in

humidity in the two cases. When the results are compared for equivalent rates of freezing, rather than gas temperature, the maxima show a near correspondence in position.

The variation of the charge per  $\text{cm}^3$  is considered to be an effect of the difference in the mean bubble sizes and concentrations, due to the different rates of freezing and the equilibrium air concentrations, both of which vary with the temperature. In the case of the  $\text{CO}_2$ -bubbled distilled water, the quantity of dissolved gas was greater than the equilibrium concentration at any given temperature.

The actual distribution of bubble sizes in the ice specimens was not measured. It was found that initial attempts to do this extended the length of time for each experiment so that no more than two or three points could be plotted on any one day with any degree of accuracy. It was found that the distributions only showed small variation for differences in freezing temperatures of a few degrees, and it was necessary to examine sections from a large number of separate specimens in order to distinguish between two different distributions. For the complete range of freezing temperatures, the bubbles in the ice had diameters typically between 10 and 500 microns. The total number of bubbles per mm size drop was of order  $10^1$  to  $10^3$ , and the median diameter was typically

between 100 and 200 microns..

Since the gases released during the freezing of the specimens will be under considerable pressure due to the formation of the ice shell, the actual bubble sizes in the final ice specimens will not necessarily be the same as the bubble sizes in the melt water on subsequent melting. Also, there will probably be considerable coalescence of bubbles in the melt water after release and before bursting. The combined effect will be to alter the distribution in an unpredictable way. For these reasons a detailed knowledge of the bubble spectrum in the ice prior to melting would not lead to a quantitative estimation of the mean charge per bubble bursting, for direct comparison with the work of Iribarne (1966a). In the discussion and comparison which follows in this work, the sizes referred to are the mean range of sizes found in the ice specimens prior to melting, typically 10 to 500 microns diameter, except in the special case of the specimens containing a high concentration of  $\text{CO}_2$ .

### 7.3.2 Air temperature used for melting

Fig.. (7.3) shows a typical set of curves demonstrating the variation of charge with air temperature used for melting, for specimens frozen under identical conditions and melted at a constant wind speed. The relative magnitudes of this set of curves should not be directly compared as the results are taken from different experimental

occasions. The form of the curves is not independent of the wind speed used, and they will be discussed in relation to the variation with wind speed in the next section.

### 7.3.3 Wind speed

Typical sets of results for specimens frozen under identical conditions and melted at a single air temperature but with different wind speeds are shown in Figs.(7.4) and (7.5). In each case the charge rises very rapidly to a maximum or plateau value, though the positions of the maxima are not always in the same place. The general shape of the curves is similar in appearance to the melting temperature characteristics, and suggests that the behaviour is a function of the rate of melting rather than the wind speed and temperature independently, as in the case for melting in nitrogen..

For this reason the combined effect of wind speed and temperature was investigated and the results presented in Fig.(7.6). The complete programme using a single sample of 'pure' water took approximately 14 hours to complete,, and several attempts were made before the condition of the change of ambient humidity during the experiment being less than about 10% at any given temperature was met. The results have been replotted in Fig.(7.7) in terms of the heat transfer factor for melting. This factor was calculated on the assumption that the mean contribution

to the melting rate by evaporation or condensation was negligible. This is not an unreasonable assumption, as the humidity at 20°C was approximately 70%, and the calculations of Chapter Two indicate that the contribution to the melting rate even for humidities of 100% is only of order 20%. The mean temperature difference between the sphere and the air stream was assumed to be equal to half the wet-bulb temperature of the air stream. For comparison, the curve of Fig.(6.9) for the case of 'pure' water prepared and melted in nitrogen has also been drawn on the same scale.

#### 7.4 Discussion of results

##### 7.4.1 General shape of curves

The analysis of the variation of charge with the freezing conditions, and the associated bubble structure, has been dealt with in section(7.3.1).

The curves of Fig.(7.7) indicate that the charge separation is a function of the rate of melting, rather than the separate parameters air temperature, humidity and wind speed. They also show that there is a critical melting rate, associated with the development of strong internal convection in the melting specimens, at which the total separated charge increases by an order of magnitude. As the melting rate is increased above this critical value, the separated charge passes through a maximum and begins to decrease. Possible explanations of

some of the facets of the typical behaviour are discussed below.

i) The presence of some effect not explained by the theory of the electric double layer as expounded in Chapter Four. This point will be considered in detail in section (8.2) when the effect of impurity concentration on the separated charge has been considered.

ii) Since the rapid increase in the charge with melting rate appears to be associated with the development of strong internal convection in the melting specimens, it may be that the whole dynamical process of bursting is altered. Although the actual bursting could not be seen, the bubbles appeared to be distorted on the surface prior to bursting, probably due to the shearing forces in the liquid and the drag in the boundary layer round the sphere. This may well affect the mechanism of break up of the central jet and the rupture of the bubble cap. An investigation of these possibilities was beyond the scope of this work.

iii) The strong convective activity may well have had the effect of continuously renewing the water surface at which the bubbles burst, thus reducing the effect of surface impurities. The results of experiments with prolonged exposure to the gas stream (section 6.4) indicated that surface contaminants were present even with the cleanest conditions available in the laboratory.

iv) The strong convective activity will lead to a shorter time between release of bubbles from the ice and their subsequent bursting at the surface. This may significantly alter the build up of the electric double layer around the bubbles. The estimated time constant for this build up is of order 0.1 second in the case of 'pure' water. (Iribarne 1966a). This time is comparable with the time between release and bursting of bubbles for the higher range of melting rates.

v) The motion of the bubbles through the moving liquid also affects the electric double layer formation. It was seen in section (4.1) that there is a compact layer of charge rigidly attached to the bubble surface, and a diffuse layer that is not resistant to shear motions in the liquid. It is possible that this diffuse layer is removed by the motion of the bubbles in the liquid.

vi) The shorter lifetime of bubbles at the higher rates of melting may prevent the accumulation of impurities at the bubble surface, from the interior of the liquid.

Of all these possibilities, or any combination of them, it is felt that the one having the major significance in explaining the sudden increase in charge with melting rate is the renewal of the water surface by the convective activity. The experiments with the large wind tunnel in the cold room failed to show this increase, (except in

a few isolated cases), and it was suspected that this was due to the presence of surface films. It was also seen that by touching the support lightly with a finger, the charging levels were reduced by approximately an order of magnitude to a value equivalent to that of the initial tail of the curves of Fig.(7.7).

Experiments were also conducted in which the same specimen was melted and refrozen in the same air stream a number of times under identical conditions. The effects on the generation of charge are shown in Fig.(7.8). It is seen that there is a gradual decrease in the charge with the number of meltings and/or the length of exposure to the air stream. The positive charge generation is entirely eliminated over a period of approximately 40 minutes, and only the initial small negative excursion at the onset of melting remains. This is interpreted as due to the gradual absorption of surface impurities from the laboratory air stream. (The surroundings included two compressor units, a workshop and proximity to a major road. The laboratory was situated in Central London).. Similar experiments conducted with nitrogen and compressed air drawn from cylinders showed only a very slow decrease in the generated charge with length of exposure to the gas stream. (See section 6.4)..

The possibilities presented above, apart from that of the effect of surface impurities, might serve to explain

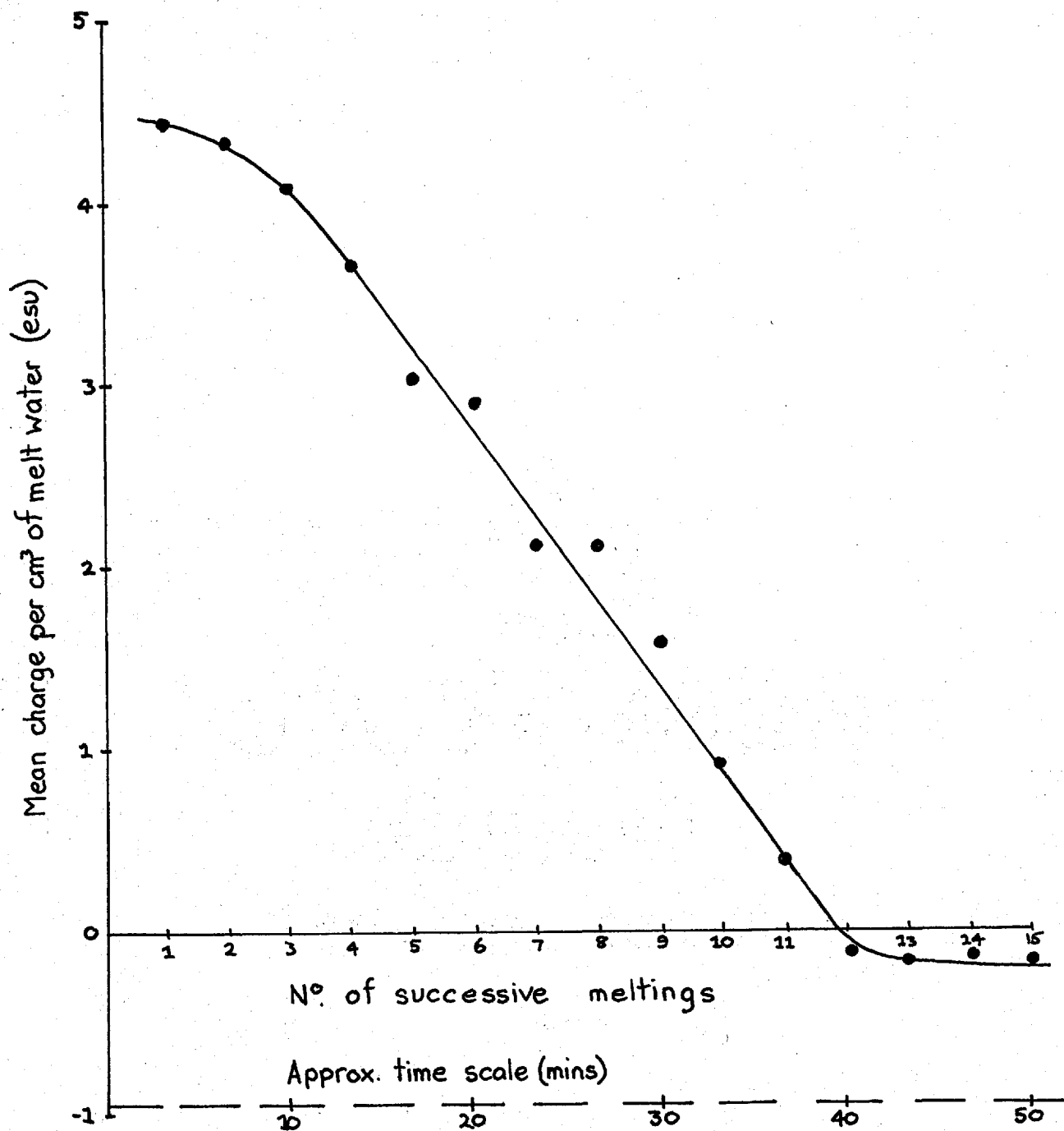


Fig. 7.8 Graph showing effect of prolonged exposure to laboratory air stream on the separation of charge.

either an increase or decrease in the separated charge with rate of melting. Since neither the nature of formation of the electric double layer, nor the mechanism of bubble bursting are fully understood even in the case of gentle passage of the bubbles through the water before bursting at the surface, it is not possible to eliminate or finally decide between any of the several possibilities..

#### 7.4.2 Effects of ionic impurities

##### Ionic salts:-

experiments were conducted using solutions of NaCl,  $\text{NH}_4\text{Cl}$  and  $(\text{NH}_4)_2\text{SO}_4$ . The results were essentially similar for the three different solutions, and the detailed discussion and figures are restricted principally to solutions of NaCl..

In general, it was found that the shapes of the various characteristics of charge separation were not significantly altered by changing the concentration of the dissolved salt. The curves of Figs. (7.1) (7.3) (7.4) and (7.5) show a systematic decrease in magnitude of the separated charge with increase in concentration of the solutions, the charge being undetectable for concentrations above about  $10^{-2}$  moles/litre. (Less than about  $10^{-4}$  esu per  $\text{cm}^3$  of melt water). This type of behaviour is consistent with the behaviour found by Iribarne (1966a) in his experiments, but the levels of charge do not correspond and in no case in this work was a change of sign of the

charging effect found.

Fig.(7.5) shows that for an NaCl concentration of about  $10^{-3}$  moles/litre, the charge separated for melting rates above the critical value is of order 0.1 esu per  $\text{cm}^3$  and is always such as to leave the melt water with a positive charge. This means that the mean charge per bubble bursting event in these experiments is such that the debris carries away a negative charge. Iribarne found that a reversal of sign occurred in the charge separation per bubble for ionic concentrations greater than  $10^{-4}$  moles/litre over the complete range of bubble sizes 20 to 200 microns radius, such that the debris carried away a net positive charge. See Fig.(4.4). Even for concentrations as low as  $10^{-5}$  moles/litre, a reversal of sign took place over part of the bubble size range that would suggest the mean charge separated in the melting of the ice specimens in these experiments should have been such as to leave the melt water with negative charge, or a much smaller positive charge than actually recorded.

These combined effects suggest that the results for gentle bubble bursting at a water surface cannot be applied to the case of bursting when strong convection and shear are present in the water.

The recording system was not sensitive enough to determine whether Iribarne's results are consistent with the charging levels found with melting rates less than the

value for which strong internal convection appears in the melting specimens. However, the general trend of the results at these slow melting rates indicates that there is a qualitative agreement.

Since the ice specimens used in these experiments with air as the melting environment will have contained  $\text{CO}_2$  concentrations in equilibrium with the air stream, the difference in behaviour might be attributed to this. However, the theory of the electric double layer in its simple form suggests that the different ion concentrations will have an additive effect, and this would further increase the discrepancy between the two cases. A direct comparison of these results with the work of Iribarne is not quantitatively possible, but calculations using the approximate values of the mean bubble sizes and densities in the ice specimens suggest that the inconsistency is as much as two orders of magnitude in the mean charge per bubble, these results being the greater.

#### Carbon dioxide:-

Ice specimens made from water containing a high concentration of  $\text{CO}_2$  gave greater charge than those for which the  $\text{CO}_2$  content was in equilibrium with the air in the laboratory. Also, in comparing the results for specimens melted in air from the laboratory and those for specimens prepared and melted in an atmosphere of nitrogen, the charges are increased by a factor of 1.5 to 2.0 for

the same rates of melting.

These several observations can only lead to the conclusion that the presence of  $\text{CO}_2$  in the water from which the specimens are made, or in the environment in which freezing takes place, leads to an increase in separated charge. This is a surprising conclusion in view of the suspicion of previous workers (Dinger 1964, Matthews and Mason 1964) that  $\text{CO}_2$  had an extremely poisonous effect on the charge separation. However, it was noted for the case of the  $\text{CO}_2$ -bubbled water, in the initial tail of the characteristics at slow melting rates, the curve was the lowest, indicating a reducing effect. See Figs. (7.3) and (7.4). This may well be the reason for the suspicion of these workers, as their experiments have been conducted using rigid containers for the melting specimens coupled with slow wind speeds. Under these conditions the strong convective activity associated with the melting in this work will not have developed.

#### Rain water:-

Fig. (7.4) shows the effect of wind speed on the charge separation for a sample of rain water collected from a shower over Central London. The ground temperature at the time was  $+18^\circ\text{C}$ . The water was collected using a wide-necked glass funnel and glass bottle, supported about 3 metres above the ground and remote from surrounding trees and buildings to avoid collection of splash water.

The shower took place at approximately 2.30 p.m.. No analysis of the salt content of the water was made, but vigorous shaking of the sample gave a bubble persistency at the surface before bursting greater than 1 second, indicating the presence of surface impurities..

The shape of the characteristic using this sample is similar to all the other characteristics for the region where strong internal convection is present. As in the case of the CO<sub>2</sub>-bubbled water, no significant charge was recorded for melting rates below the critical value, and this is interpreted as due to the surface contamination.

## CHAPTER EIGHT

### Conclusions and application of results

#### 8.1 Preliminary results

The results of the experiments performed in the horizontal wind tunnel described in Chapter Five were found to be unsatisfactory for a number of reasons previously discussed. The main factor inhibiting the generation of charge was felt to be the constraint on the dynamics of the melting process due to the use of a rigid container for the melting specimens. When the large wind tunnel inside the cold room was used, it was suspected that the results were affected by the presence of surface contamination from the surroundings. Since no correlation of the mean charge per  $\text{cm}^3$  of melt water could be found with the rate of melting of the specimens used in this case, it is concluded that the convective activity was not sufficiently strong in the water portion to have the effect of continuously renewing the surface at which the bubbles burst; except in the few isolated cases when large charges were recorded. It was found that small increases in the  $\text{CO}_2$  content of the melting environment did not significantly alter the mean levels of charge recorded.

When the restrictions of dynamics and surface contamination were greatly reduced by the use of the small vertical wind tunnel outside the cold room, much higher

levels of charge were recorded. The effects of impurities on the magnitude of the charge separation in this case raise doubts as to the applicability of the simple double layer theory to describing the events.

## 8.2 Electric double layer theory

In order to discuss the results of the latter experiments in terms of electric double layer theory, it is convenient to divide the melting behaviour into two regions. The first in which the melting takes place slowly and strong convection does not occur in the water portion of the melting specimen: the second, the regime in which this strong convection does occur.

### 8.2.1 Slow melting without strong convection

The general behaviour of the experimental results for the slow melting case could be approximately interpreted in terms of the simple electric double layer theory expounded in Chapter Four. Increases in the concentration of ionic impurities led to a systematic decrease in the separated charge, and the magnitudes of the charges found in this region were comparable with the values predicted by the experiments of Iribarne (1966a) when the reducing effect of  $\text{CO}_2$  is taken into account. The presence of strong concentrations of  $\text{CO}_2$  in the water samples reduced the charge separation in this region.

Due to the relatively short time constant of the recording system, comparable with the time of melting of

the specimens, charges greater than about  $5 \times 10^{-3}$  esu per  $\text{cm}^3$  of melt water were not detectable. For ice specimens of a few mm diameter, and containing about 500 bubbles, the smallest mean charge per bubble detectable would have been of order  $10^{-6}$  esu per bubble. Positive charges on the debris from bubble bursting, of this order of magnitude, were detected by Iribarne for ionic salt concentrations greater than about  $10^{-5}$  moles/litre and  $\text{CO}_2$  concentrations greater than about  $10^{-5}$  moles/litre. This would predict a negative charge being left on the melt water in these experiments with melting ice specimens, for concentrations of ionic salts and  $\text{CO}_2$  of the above values. In no case in these experiments was a net negative charge on the melt water recorded, though in all cases a small negative charge occurred at the onset of melting, and this will be discussed in the Appendix.

Hence we may conclude that although the results show a qualitative agreement with the predictions of the electric double layer theory in this gentle melting case, they do not entirely agree with the results expected from the work of Iribarne. However, the disagreement occurs at the limit of accuracy of this experiment.

### 8.2.2 Rapid melting with strong convection

For this regime of melting, in which strong internal convective motions were present in the melt water, there seems to be very little correlation of the results with

the simple electric double layer theory. The only feature immediately reconcilable with this theory is the observation that increasing concentration of ionic salts led to a systematic decrease in the separated charge.

Against this must be weighed the fact that the presence of  $\text{CO}_2$ , in either the environment or in the water, led to an increase in the charge where the theory predicts a decrease, and that even for concentrations of ionic impurities as high as  $10^{-3}$  moles/litre, no reversal of sign of the mean charge per bubble had occurred.

Also, when compared with the results of Iribarne, the magnitude of the mean charge per bubble for melting in the presence of the laboratory air, is approximately two orders of magnitude greater than expected from the typical concentrations of  $\text{CO}_2$  in the specimens.

### 8.2.3 Conclusions

For both the regimes of melting, and particularly for the more rapid case, the results cannot be completely reconciled with the theory of the electric double layer in its simple form, nor with the experiments of Iribarne (1966a) for the bursting of nitrogen bubbles at a water surface. However, this is not to say that the effect is not one of double layer separation, for the experimental conditions have introduced dynamical conditions that are not considered in either the theory, or in the experiments of Iribarne. Even so, it will be difficult to reconcile

the increase in charge in the presence of  $\text{CO}_2$ , as in all treatments of the double layer effect the suggestion is that its presence would lead to the same type of behaviour as with other ionic compounds; i.e. a decrease in the layer thickness and a decrease in the surface charge density and volume charge density in the diffuse region. It thus seems that there is a possibility of there being some other effect present, that is associated with the presence of  $\text{CO}_2$ , particularly when there is strong convection in the water.

### 8.3 Application of results to atmospheric melting of snow

Although experiments with snow were restricted to the melting of a few samples in rigid containers in the horizontal wind tunnel, the observations made in the first half of this thesis on the behaviour during melting while loosely supported in the vertical wind tunnel, together with the experiments on solid precipitation elements in the small vertical wind tunnel, allow certain predictions to be made.

In the first part of this thesis, the observations on the melting of snow flakes in the large vertical wind tunnel showed that even for melting rates greater than that typical of the atmosphere, convection did not occur in the melt water except during the last few seconds. The crystal framework retained its form until these last few seconds, and the structure then collapsed to form

drops of water. Because of this behaviour, even if the flakes were to remain partly unmelted by the time they have fallen to a temperature level in the atmosphere where the rate of melting was sufficient to develop strong convection in the melt water, the melting is unlikely to separate charges of the same order of magnitude as observed in the case of solid specimens. Also, because of the framework type of structure, drops ejected by bubble bursting in the centre of the melting flakes are likely to be recaptured by the members in the outer region.

Experiments by Magono and Kikuchi (1965) on the charges residing on the melt water for the case of natural snow flakes in free fall, found that small charges of both signs were present but that an excess of positive charge was recorded over a long period of time. In these experiments, the flakes were entirely melted over a fall path of some several centimetres by application of a radiative field. The resulting water drops were subjected to a strong horizontal electric field, and the charge on the water drops inferred from the displacement of the drops in this field. In view of the results of the experiments conducted in this work, in which the charge separation is highly dependent on the rate of melting of the specimens, it is probable that the charges recorded by Magono and Kikuchi are not typical of the atmospheric melting case.

However, an analysis of all the literature on the

electrical charge generation of melting snow suggests that the melt water acquires predominantly positive charge under all circumstances of melting, and it is thus probable that the bursting of bubbles during the melting process accounts, at least in part, for this effect. It is also reasonable to assume that the negative space charge found over melting snow is due to the same effect.

#### 8.4 Melting of hail and thunderstorm charge generation.

In considering the generation of charge during the melting of solid hydrometeors, and its application to the thunderstorm, we have to consider the following factors.

- a) Rate of freezing and bubble structure.
- b) Rate of melting.
- c) Effects of ionic impurities.
- d) Effects of surface impurities.

These are dealt with below under their separate headings.

##### 8.4.1 Freezing and bubble structure

The results of the experiments using different freezing temperatures (rates of freezing), suggest that there is an optimum temperature region (bubble structure) for the maximum charge separation to occur on subsequent melting. In the real atmosphere, the situation is extremely

complex due to the combination of freezing and coalescence of the various elements during their passage through different regions of the cloud. Examination of large hailstones often indicates a pronounced layer structure of bubbles representing several different growth regimes with different freezing characteristics. On the other hand, low density graupel gives a bubble structure having two distinct regions in its spectrum: one from the bubbles in the individual frozen drops that comprise the pellet and the other from the air spaces between these individual drops.

Because of the complexity of the atmospheric situation it is not possible to predict possible magnitudes of the charge separation on an analysis of the temperature conditions alone. However, the bubble sizes in the specimens used in these experiments, of order 10 to 500 microns diameter, correspond approximately to the range of bubble sizes found in hail and graupel. It is thus reasonable to assume that the levels of charge suggested by these experiments will be appropriate to that generated by the melting of hail and graupel in the atmosphere, on consideration of bubble structure alone.

#### 8.4.2 Rate of melting

Given that the typical bubble structure is the same, and neglecting for the moment the effects of the various

impurities, then we can examine the likely charge separation from the melting of hail and graupel in terms of the melting rate.

An important aspect of the melting is that the charge generation did not begin until convection began in the melting specimens, and the maximum separation occurred when the convective activity was strongly established. In the real atmosphere, in which the temperature and humidity profiles vary with height, we may represent this effect by a hypothetical 'critical' isotherm at which the main charge separation begins. Particles melting entirely before reaching this critical temperature may be expected to develop charges equivalent to the small values in the initial tail of the characteristic of Fig.(7.7). In the case of a saturated atmosphere, the isotherm will lie in the region of  $+2^{\circ}\text{C}$ , and the results of Chapter Two then suggest that there will be little charge separation for particles smaller than about 1 mm diameter, as they will have melted completely by the time this isotherm is reached.

For ice specimens greater than about 1 cm diameter, the melt water will be shed in the air stream. This will mean that the ice is covered by only a thin layer of water during the initial stages of melting, and strong convection will not be able to develop. When the size is reduced by this water shedding to a stable size for the

remainder of melting, the temperature level that the particles will have reached will be such that the rate of melting is extremely rapid, i.e. it is well past the position of maximum charge separation shown as a function of melting rate in Fig.(7.7). Hence it appears that the maximum charge separation will occur for particles in the intermediate range, of a few mm diameter. Particles of this size will melt completely over a distance of fall of about 2 Km from the 0°C level, and we may thus conclude that from considerations of rate of melting alone there is likely to be a region of maximum charge density between about +2°C and +12°C, associated with the melting of particles of a few mm diameter.

#### 8.4.3 Effects of ionic impurities

The presence of ionic salts did not affect the general shape of the characteristics of Figs.(7.1) to (7.7), and it is thus reasonable to assume that the general arguments of the two previous sections will not be modified in this case, except in determining the relative magnitude of the separated charge.

If we consider the mean volume concentration of typical salts found in rain water at ground level, then the range is typically  $10^{-4}$  to  $10^{-6}$  moles/litre.(Junge,1963). For the upper limit of  $10^{-4}$  moles/litre the reduction in charge from the case of specimens made from 'pure' water would be a factor of 2 to 3 for any given melting conditions.

For the lower figure there would be practically no reduction. In the melting region of the atmosphere, before scavenging takes place from the lower layers, the concentrations of these salts are likely to be nearer the lower values. Any difference in the concentration of  $\text{CO}_2$  between these experiments and real atmospheric particles is unlikely to alter the value of the maximum charge separation by a significant amount, since, except in the case of complete exclusion of  $\text{CO}_2$  the results were not altered by more than 15% even in the case of the  $\text{CO}_2$ -bubbled distilled water.

From these arguments we may conclude that although the application of the results to the atmosphere will depend on the presence of the various ionic impurities, the levels of charge are not likely to be decreased by more than 50% in the worst possible case from the values suggested by these experiments.

#### 8.4.4 Surface impurities

It was found that the experiments were highly sensitive to the presence of surface contamination, which in all cases reduced the separated charge. However, for the sample of natural rain water collected from a shower over Central London, charges equivalent to about one third of the values recorded for the 'pure' water were obtained when using it to make specimens for wind tunnel melting. With the ground temperature of  $+18^\circ\text{C}$ , the rain

will have fallen approximately 3 Km from the melting level, through an atmosphere likely to contain a much higher concentration of potential surface active material than in an average location. Also, in the laboratory experiments the air was drawn from this same London atmosphere, and although it was filtered, must have contained surface contaminants as demonstrated by the results shown in Fig.(7.8). It is thus probable that the presence of the surface contamination will not reduce the charges on real atmospheric particles from the values found in the laboratory.

#### 8.4.5 Application to the thunderstorm

The combined results of sections (8.4.1) to (8.4.4) suggest that during the melting of solid ice hydrometeors in the atmosphere there will be a region of maximum charge separation. This will be associated with ice particles of a few mm diameter, and will lie in the region  $+2^{\circ}\text{C}$  to  $+12^{\circ}\text{C}$  approximately. i.e. a layer about 2 Km deep.. The separation of charge is such as to leave the melt water positive. The magnitude of the charge separation is dependent on the presence of impurities in the ice or on the surface, but is likely to lie within the limits 4 esu per  $\text{cm}^3$  to 8 esu per  $\text{cm}^3$  of melt water.

In a thunderstorm, evidence that intense electrical activity is associated with the appearance of small hail and graupel at the base of the cloud, and that the centre

of this activity coincides with the region of heaviest precipitation, is given by Kuettner (1950), supported by Workman and Reynolds (1950). A typical value for the liquid water content of this region is about  $4 \text{ g m}^{-3}$ , (see Chapter One), and if this is assumed to be due to the melting of small hail and graupel, then the experimental values of 4 to 8 esu per  $\text{cm}^3$  of melt water would give charge densities of between  $5 \text{ C Km}^{-3}$  and  $10 \text{ C Km}^{-3}$ . The figure calculated as necessary to explain the presence of the subsidiary pocket of positive charge in a thunderstorm, in Chapter One, was  $8 \text{ C Km}^{-3}$ ..

The experiments suggest that the location of the region of positive charge will be between about  $+2^\circ\text{C}$  and  $+12^\circ\text{C}$ , and this is consistent with the observed location of the positive charge pocket in real thunderstorms.. The depth of the region is suggested as about 1.5 to 2 Km for normal lapse rates. In order to give a typical volume of about  $0.5 \text{ Km}^3$  for the region of positive charge, a typical horizontal dimension would have to be about 0.5 Km. This is not an unreasonable value for the horizontal extent of the region of most intense precipitation.

It thus appears that the mechanism of bubble bursting is capable of giving a reasonable explanation of the main features of the pocket of positive charge located at the bases of thunderstorms, starting from neutral conditions, i.e. magnitude and sign of the charge density,

location and physical dimensions of the region.

APPENDIX"Some Unexplained Observations"Initial negative charging of specimens

In all the cases of melting where air was used as the melting environment, whether drawn from the laboratory by a vacuum cleaner pump or from compressed air cylinders, the charging records had a slight negative excursion at the onset of melting. When nitrogen was used as the melting environment gas, the effect was not present. An examination of all the charging records showed that there appeared to be no significant variation in the magnitude of the charge per  $\text{cm}^3$  with concentration of ionic impurity in the specimens. The charge was typically of order  $10^{-1}$  esu per  $\text{cm}^3$  of melt water, and showed considerable variation about the mean value for identical samples treated in identical fashion. Since no explanation of this could be given the effect was investigated further.

Ice specimens were made on the support in the small vertical wind tunnel in the usual fashion, and allowed to partially melt in the air stream and then slowly refrozen. In this way, a fairly thick shell of nearly bubble free ice was formed on the specimens. The specimens were then re-melted and the charging records examined. In each case it was found that the negative excursion was still present, and began as soon as the melting began. It was concluded

from this that the charge was not due to the bursting of bubbles.

An examination of the records used to construct the curve of Fig.(7.8) showed that the magnitude of the negative charge did not alter significantly with number of meltings of the same specimen, or length of exposure to the air stream. This indicates that it is independent of the absorption of surface impurities, at least in small quantities. The effect could be eliminated by adding a drop of liquid soap solution to a litre of 'pure' water and using this to make the specimens. This also eliminated the positive charging effect.

No significant variation in the magnitude of the effect could be found with either wind speed or melting temperature in the range 1.5 to 10 m sec<sup>-1</sup> and +2°C to +10°C.

The results of all the investigations have not revealed the reason for the behaviour, and although the effect is too small to be of importance in generating charge in the atmosphere, it is important simply because it defies explanation at this stage.

#### Charging without melting

On some occasions when wind speeds above about 9 m sec<sup>-1</sup> had been used, it was noted that a charging effect was present before the ice specimens began to melt. A short investigation of this effect was made.

Ice spheres in the mm diameter range were made from

three samples of water in the manner previously described in Chapter Seven. 'Pure' water distilled water and tap water were used. The ice spheres were allowed to sit in a gas stream of temperature below  $0^{\circ}\text{C}$ , while any generation of charge was recorded.

No charge was recorded for any of the samples when the cold gas stream came from either a cylinder of compressed air or nitrogen, under all conditions of wind speed and temperature.

No charge was recorded for any conditions when the spheres were made from 'pure' water.

The spheres acquired positive charge when made from distilled water, and subjected to air from the laboratory, at all temperatures below  $0^{\circ}\text{C}$  and for wind speeds greater than about  $10 \text{ m sec}^{-1}$ . The rate of charge generation was typically of order  $1 \text{ esu per cm}^3$  of ice per minute at a wind speed of  $15 \text{ m sec}^{-1}$ , and increased slightly with increasing wind speed. The generation rate was steady and showed no significant change after a period of about 15 minutes, by which time the ice sphere had evaporated to approximately half its original volume.

Ice spheres made from tap water showed similar behaviour to that of the distilled water case, except that the sign of the mean charge acquired by the sphere was negative, and the generation rate was not steady but had stationary periods and occasional positive excursions

lasting a few seconds.

These few experiments suggest that the effect is impurity dependent, as no charge could be detected under any conditions with spheres made from water containing no impurities. Also, since the effect did not occur in air or nitrogen obtained from commercial cylinders of these gases, it was thought that it might be due to either the presence of  $\text{CO}_2$  or particulate matter in the air stream. The artificial introduction of increased concentrations of  $\text{CO}_2$  did not appear to modify the behaviour. The removal of the cotton wool filter from the inlet tube of the wind tunnel led to a small increase in the rate of charge generation.

A similar charging effect has been reported by Brownscombe (1966), during the passage of air from a cold room over the surface of large ice specimens supported in a vertical wind tunnel.

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# REFERENCES

- Adamson, A.W. (1960) "Phys. Chem. of Surfaces."  
Interscience, New York.
- Battan, L.J. (1953) J. Met. 10, 311
- Blanchard, D.C. (1963) Prog. in Oceanography. Vol. 1  
Pergamon Press.
- Brownscombe, J.L. (1966) Private communication.
- Chapman, D.L. (1913) Phil. Mag. 25, 475
- Debye, P. and Huckel, E.  
(1923) Physik. Z. 24, 185
- Dinger, E.J. (1964) Q.J.R.Met.Soc. 90, 208
- Dinger, E.J. (1965) J.Atmos. Science. 22, 162
- Dinger, E.J. and Gunn, R.  
(1946) Terr.Mag.Atmos.Elec. 51, 477
- Feteris, P.J. and Mason, B.J.  
(1956) Q.J.R.Met.Soc. 82, 446
- Findeisen, W. (1939) Met. Z. 55, 121
- Gouy, G. (1917) J. Phys. 9, No.4, 457
- Grenet, G. (1947) Annales de Geophys. 3, 306
- Gunn, R. (1951) J. Met. 8, 71
- Gunn, R. (1960) J. Geophys. Res. 54, 57
- Gunn, R. and Kinzer, G.D.  
(1949) J. Met. 6, 243
- Harper, W.R. (1957) Advances in Phys. 6, 365

- Hilpert, R. (1932) VDI Forsch Hft. 355
- Iribarne, J.V. (1966a) Trans.Farad.Soc. (in press)
- Iribarne, J.V. (1966b) Publication prepared for Q.J.R.Met.Soc.
- Jayaweera, K.O.L.F. (1965) Ph.D. Thesis, Univ. of Lond.
- Junge, C. (1963) Air Chem. and Radioactivity. Acad. Press.
- Kerker, M., Langleben, M.P.  
and Gunn, K.L.S. (1951) J. Met. 8, 424
- Kientzler, C.F., Arons, A.B.,  
Blanchard, D.C. and  
Woodcock, A.H. (1954) Tellus 6, 1
- Kinshott, E.J. (1950) Met. Mag. 79, 205
- Knelman, F., Dombrowski, N.  
and Newitt, D.M. (1954) Nature 173, 261
- Kuettner, J. (1950) J. Met. 7, 322
- Latham, J. (1965) Q.J.R.Met.Soc. 91, 369
- Macklin, W.C. (1963) ibid. 89, 360
- Magono, C. and Kikuchi, K.  
(1963) J.Met.Soc.Jap. 41, 270
- Magono, C. and Kikuchi, K.  
(1965) ibid. 43, 331
- Marchand, E. (1903) Bull.Soc.Ramond Toulouse. 8, 12
- Mason, B.J. (1954) Nature 174, 470
- Mason, B.J. (1956) Q.J.R.Met.Soc. 82, 209

- Mason, B.J. (1957) "The Physics of Clouds"  
Oxford Univ. Press.
- Mason, B.J. and Howorth, B.P.  
(1952) Q.J.R.Met.Soc. 78, 226
- Matthews, J.B. (1963) Ph.D. Thesis, Univ. of Lond.
- Matthews, J.B. and Mason, B.J.  
(1963) Q.J.R.Met.Soc. 89, 376
- Matthews, J.B. and Mason, B.J.  
(1964) ibid. 90, 208
- McCready, P.B. and Proudfit, A.  
(1965) ibid. 91, 54
- Monteith, J.L. (1953) Proc.Phys.Soc. 67, 217
- Pasquill, F. (1949) Q.J.R.Met.Soc. 75, 239
- Powell, R.W. (1940) Trans.Inst.Chem.Engnrs.  
Lond. 18, 36
- Ranz, W.E. and Marshall, W.R.  
(1952) Chem.Eng.Prog. 48, 141,173
- Simpson, G.C. (1909) Phil.Trans.A. 209, 379
- Simpson, G.C. (1949) Geophys. Mem.(Met. Off. Lond.)  
4, No.84
- Smith, L.G. (1955) Q.J.R.Met.Soc. 81, 23
- Stern, O. (1924) Z. Elektrochem. 30, 508
- Thorpe, A.D. and Mason, B.J.  
(1966) Brit. J. App. Phys. 17, 541
- Vonnegut, B. (1953) Bull.Amer.Met.Soc.  
34, 8, 378

Woodcock, A.H., Kientzler, C.F.

Arons, A.B. and Blanchard, D.C.

(1953) Nature 172, 1144

Workman, E.J. and Reynolds, S.E.

(1950) Proc. Conf. on Thund. Elect.  
Chicago, 29