ON THE MEASUREMENT OF THE THERMOELECTRIC EFFECT ON ICE

(A comment on an experiment described by Dr J. Latham, Quart. J. R. Met. Soc., 96, pp. 266–274)

By C. D. Strow and P. H. Symns*

1. INTRODUCTION

A thermal-gradient diffusion chamber of advanced design has been constructed by Symns (1973) as part of a programme to provide a comprehensive laboratory facility for studies in atmospheric physics. In order to evaluate the chamber and gain some experience in its use, it was decided to repeat some of the experiments of Hallett and Mason (1958) concerning ice-crystal growth and then to perform the experiment of Latham (1964), who measured the thermoelectric effect of an ice needle suspended on a quartz fibre in an electric field. The latter experiment proved impossible to perform and a closer investigation of the work of Latham revealed some discrepancies and problems as described below.

Ice crystals grown in the diffusion chamber or described in the literature are designated by the nomenclature of Magone and Lee (1966) to minimize ambiguities which tend to arise through mere verbal description of their form.

2. OBSERVATIONS ON GROWING ICE CRYSTALS

After Hallett and Mason, ice crystals were permitted to grow from the vapour on a fine quartz fibre suspended from the top of the diffusion chamber. Variations in crystal habit with temperature were noted which agreed well with the observations of other workers. In addition, the occurrence of short columns [C1] at a temperature just below $-3^\circ$C was noted, under conditions of slow growth. Between $-3$ and $-6^\circ$C grew long needle-like crystals, both bundles of elementary needles [N1b] and the prismatic sheaths [N1c]. Compact hollow-ended prisms (i.e. sheaths) occupied the remainder of the fibre down to a temperature of about $-8^\circ$C.

The bundles of needles [N1b] are a degenerate form of prism and are the cause of the fibrous appearance of many of the longer growths observed, giving rise to very non-uniform diameters. On the other hand, the growth of elementary sheaths [N1c] was observed to be much more orderly. The sheaths exhibited a clean profile and, after the initial stages of growth, were noted to have a constant diameter of about 0.3 mm regardless of their length.

In their descriptions, Hallett and Mason do not clearly distinguish between needles and elementary sheaths. Both types of crystal are referred to under the general term of 'needles', though, in fact, the longer needles are actually clusters of crystals, not monocristalline-like sheaths or solid columns.

3. THE EXPERIMENT TO DETERMINE THE THERMOELECTRIC EFFECT ON ICE

Latham (1964) describes an experiment which he performed in a thermal-gradient diffusion chamber to measure the thermoelectric effect in ice. An ice needle was suspended on a quartz fibre and irradiated to produce a temperature gradient between its ends, and an electric field was applied to produce a torsion force on the crystal which then rotated about a vertical axis. The torsion force is the sum of the forces acting on induced polarization charges and charges separated by the temperature gradient in the ice specimen.

Latham describes his ice specimens as being 'needles', which, according to the preceding section, could be interpreted as meaning either sheaths or bundles of elementary needles. Latham quotes the typical dimensions of a 'needle' as being 3 mm diameter and 40 mm in length. From his description, in which the needles are admitted to be irregular in shape, it is apparent that the specimens he used were clusters of multiple crystals and not monocrystalline as would be the case with sheaths.

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Latham and Stow (1967) have shown that, in irregularly shaped ice crystals, the temperature gradients due to non-uniformities will lead to highly asymmetric charge distributions. This means that the earlier assumption by Latham that his specimens are uniform cylinders may be a quite invalid approximation.

Sym's noted further, during studies of ice crystal growth, that crystals, once grown, had a preferred direction within the chamber. Rotation of the fibre from which the crystals were suspended showed that this effect was due to induced polarization charges caused by static charges on the perspex walls of the diffusion chamber. Further experiments showed that the electric field inside the chamber could easily reach values of $10^4$ V m$^{-1}$.

The chamber used by Latham also had walls constructed of perspex and it is not unreasonable that internal field strengths of the order $10^4$ V m$^{-1}$ should have occurred in his chamber also. Latham states that his whole apparatus was surrounded by an earthed metal gauze to eliminate stray electric fields, but there is no mention of internal shielding or static charges on the wall and it may be that this point went unnoticed. Latham found it necessary to weight his quartz fibres in order that they would hang vertically, though this could have been a measure taken to remove minor kinks in the fibre and not because the fibre was attracted to the walls. Since the impressed electric fields used by Latham to measure the thermoelectric effect were of the order of $2$ V m$^{-1}$, it would seem essential to provide internal electrostatic shielding of the chamber contents.

Sym's provided a brass-mesh shield inside his chamber to eliminate stray static fields and found no observable attraction of a 4 $\mu$m-diameter quartz fibre to the chamber walls, and no preferred orientation for growing ice crystals. The effect of the brass-mesh shield on the temperature gradient within the chamber was to raise temperatures at those points in the chamber originally below $-5^\circ$C, but the effect is appreciable only in regions where the temperature gradient of the chamber is already non-linear.

From previous discussions, it follows that ice specimens of the form used by Latham are rather unsatisfactory for use in the investigations performed. On the other hand, the only other type of useful specimen which may readily be grown in a diffusion chamber is the elementary sheath. Some simple calculations may be performed to compare with values quoted by Latham and also to examine the possibility of using sheaths as a substitute for the highly irregular needle structures.

The surface charge density on the plane ends of a uniform cylindrical monocrystalline ice specimen along which a uniform temperature gradient $\frac{dT}{dx}$ is maintained has been calculated by Latham and Mason to be

$$\sigma = 1.65 \left( \frac{dT}{dx} \right) \times 10^{-10} \text{ C m}^{-2}$$

and assumes an average specimen temperature of 270 K.

If it may be further assumed that $\frac{dT}{dx} = 4$ deg C cm$^{-1}$, then

$$\sigma = 6.6 \times 10^{-10} \text{ C m}^{-2}.$$ 

If the impressed electric field is $1.8$ V m$^{-1}$ (the value used by Latham) then the following may be calculated:

$q_T =$ charge on specimen ends due to thermoelectric effects  
$q_p =$ charge on specimen ends caused by external polarization  
$M_T =$ turning moment in the field due to $q_T$  
$M_p =$ turning moment in the field due to $q_p$.

A realistic minimum turning moment to give a detectable (but not accurately measurable)

| TABLE 1. SEPARATED CHARGE AND TURNING-MOMENT VALUES CALCULATED FOR CYLINDRICAL ICE SPECIMENS OF DIAMETER 3 mm AND 0.3 mm COMPARED WITH THE QUOTED VALUES OF LATHAM (1964) |
|-----------------|-----------------|-----------------|-----------------|
| Quantity       | quoted value    | calculated values | Units           |
|                | (Latham 1964)   | $d = 3$ mm       | $d = 0.3$ mm    |                 |
| $q_T$          | 66              | 47               | 0.47            | $\times 10^{-16}$ C |
| $q_p$          | 13              | 10               | 0.47            | $\times 10^{-16}$ C |
| $M_T$          | 5               | 3.4              | 0.034           | $\times 10^{-16}$ N m |
| $M_p$          | 1               | 0.72             | 0.034           | $\times 10^{-16}$ N m |
rotation of the specimen is $M_{\text{min}} - 10^{-16}$ N m. Table 1 shows charge and turning moment values as quoted by Latham and as calculated for a specimen of diameter 3 mm (the size of an irregular needle) and of diameter 0.3 mm (uniform sheath).

There is some discrepancy between the values quoted by Latham and the calculation performed, which is reduced if it is assumed that his specimen had a diameter in excess of 3 mm, though some inconsistency in his calculations still remains.

It can be seen that the turning moment of an irregular needle is detectable, but with very low resolution, whereas for sheaths the turning moments are both undetectable. In the latter case, no advantage ensues from increasing the magnitude of the impressed electric field, since $q_r$ will increase but not $q_T$, and the corresponding unwanted moment, $M_{P_r}$, will dominate.

Bearing in mind that the conditions under which the quantities in Table 1 were calculated represent an optimum, the experiment is not a satisfactory one. This conclusion is supported by Fig. 1, which summarizes the results of Latham by comparing measured charges due to the thermoelectric effect with theoretical values.

The use of finer quartz fibres to increase the rotation for a given turning moment does not seem practical. To give a significantly better result, a fibre of diameter less than 1 $\mu$m would be desirable; however, such a fibre could not support a suitable ice specimen and would in any case be very difficult to handle. A higher value of $dT/dx$ could be impressed, but could not be increased sufficiently to make the experiment satisfactory. Difficulties also arise if parts of the specimen become warmer than about $-7^\circ$C owing to a marked increase in electrical conductivity above this temperature.

A possible solution to this problem may arise if a large single crystal of ice with plane ends were fabricated (as this is what the theory of Latham and Mason requires). Such a specimen could be made by seeding supercooled water in a quartz tube of the required diameter. The induction method of Brownscombe and Mason (1966) required the growth of such crystals, although theirs were held by a brass clamp rather than a fibre. Because of the large dimensions that could be obtained, problems of handling the specimen and of achieving adequate resolution of $M_T$ would be obviated, if Latham's method were used. Furthermore, the investigation of single ice crystals whose charge carrier densities have been altered by doping with trace quantities of impurities (such as HF) could be pursued.

REFERENCES

CORRESPONDENCE AND NOTES


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REPLY

By J. Latham

In assessing the possible usefulness of the experiments performed by Latham (1964), Stow and Syms should perhaps bear in mind the objectives of this study, as defined in the paper. These were to determine whether the theory of the thermoelectric effect in ice, which had recently been developed by Mason, could be confirmed for pure ice specimens under conditions in which electrode contacts could not produce spurious results. The justification for undertaking such an experiment was that impurities and electrode potentials had been found to provide such wide variations in the sign and magnitude of the thermoelectric power of ice that Mason’s theory could not, at that time, unequivocally be accepted as proven. The experiments described by Latham provided quantitative confirmation of Mason’s theory for pure, electrode-free ice to within about 50%. As pointed out in that paper greater accuracy could not be achieved principally because of the non-uniformity of the ice specimens, the fact that the charge separated under the influence of the thermoelectric effect was close to the limit of measurement, and errors in the determination of the applied temperature gradient. The same motivation prompted Browncombe and Mason (1966) to perform a different experiment, again with no direct electrode contacts, which also confirmed Mason’s theory, in this case to within about 20%.

We now turn to more specific points raised by Stow and Syms: they state that Latham’s thermoelectric experiment proved impossible to perform and that closer examination of his work revealed some discrepancies and problems. This conjunction of statements could be somewhat misleading because the failure of Stow and Syms to obtain a detectable thermoelectric effect is presumably simply attributable to the fact that their ice specimens were too thin to permit the separation of measurable charge; unfortunately they do not provide details of the dimensions of the ice specimens used in their electrical experiments or of the procedures employed in this work. If, however, they had used specimens of comparable thickness to those employed by Latham (diameter 3 mm) a measurable effect should have been produced, as indicated by their own calculations. Also, the numerical discrepancies which Stow and Syms report to exist between their calculated values of charge transfer and those of Latham are an artifact resulting from a misinterpretation of the results given by Latham. Stow and Syms have taken the ‘typical value’ of separated charge, employed by Latham in a preliminary calculation to illustrate the general feasibility of his experiment, as an average value of charge transfer. They should have taken the individual values listed...
in Latham's Table 1, in which the theoretical charge transfer for each specimen used was calculated from its measured dimensions. In this case no discrepancy exists between the theoretical values.

The possibility, raised by Stow and Symns, that the results obtained by Latham were spurious because there may have been high charges on the perspex walls of the diffusion chamber can also be rejected. As mentioned in his paper a number of precautions were taken by Latham to ensure that the observed deflections were caused primarily by the thermoelectric effect. Reversal of the applied field was found to cause reversal of the sense of rotation of the ice cylinder if the direction of the temperature gradient remained constant. Conversely, if the direction of both the temperature gradient and applied field were changed at the same time no change in the sense of rotation of the ice specimen occurred. These observations are entirely consistent with separation of charge under a thermal gradient, but completely inexplicable in terms of the effects of extraneous charges. In addition, no 'preferred direction of growth' was observed in Latham's experiments, suggesting that strong electric fields associated with charge on the windows were not present in this work.

In conclusion, while we agree with Stow and Symns that Latham's experiment is not a satisfactory method of making accurate measurements of the thermoelectric power of ice — and in fact it was never purported to be so — it did, in our opinion, fulfil its objective of providing confirmation of Mason's theory for pure ice specimens in conditions where electrode contacts could not produce spurious results. The 'discrepancies and problems' noted by Stow and Symns have been shown to be illusory. Finally, I cannot agree with Stow and Symns that it would be worthwhile to undertake a further, similar, experiment using prefabricated ice specimens of large size. Aside from the fact that problems of surface contamination could arise, there are fundamental difficulties with such an experimental arrangement which severely limit the quantitative accuracy which can be achieved. In my opinion more precise measurements of the thermoelectric power can be obtained much more effectively either by using passive electrodes (on which problem Gross has made important contributions in recent years) or by a simpler, indirect technique, such as that utilised by Brownscombe and Mason.

REFERENCES

Brownscombe, J. L. and Mason, B. J. 1966 'Measurement of the thermoelectric power of ice by an induction method,' Phil. Mag., 14, pp. 1037–1047.


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COMMENT ON THE PAPER BY P. R. BRAZIER-SMITH, S. G. JENNINGS AND J. LATHAM 'THE INFLUENCE OF EVAPORATION AND DROP-INTERACTIONS IN A RAINSHAFT'

By H. P. ROESLI* and M. A. PEDDER

Brazier-Smith et al. (1973) present model calculations of the influence of evaporation and drop-interactions on the drop-size distribution in a rainshaft. Whereas we are not in a position to comment specifically on the drop-interaction part of their paper, we wish to point out some anomalies in their evaporation calculations which might seriously affect their conclusions about the combined effect of interactions and evaporation:—

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