

## Crystallographic Orientation Distributions in Accreted Ice

W. C. MACKLIN AND P. J. RYE

*Dept. of Physics, University of Western Australia, Nedlands*

1 October 1973

### ABSTRACT

The  $c$ -axis orientation distributions of ice formed by the accretion of supercooled droplets have been determined for a wide range of air and deposit temperatures. There are peaks in the distributions which may be related to the air temperature. Consequently, quantitative information on the growth environment of a hailstone can be obtained from its crystallographic structure.

### 1. Introduction

By measuring the number of crystals as a function of the angle between the  $c$ -axes of the crystals and the radial growth direction, Levi and Aufdermaur (1970) determined the  $c$ -axis orientation distributions in accreted ice formed on cylinders rotating in an icing tunnel. They found that there were peaks in the distributions which could be broadly related to the temperature of the accreted droplets. It is known that the direction of growth of dendrites in bulk supercooled water is inclined at an angle to the basal plane, the angle increasing with supercooling (Macklin and Ryan, 1965; Pruppacher, 1967). To explain their results Levi and Aufdermaur suggested that those droplets which freeze with the initial dendritic growth parallel to the accreting surface give rise to preferred orientations in the bulk ice. This produces peaks in the crystallographic orientation distributions corresponding to the growth angles of the dendrites. However, Levi and Aufdermaur found that, at an air temperature of  $-22^{\circ}\text{C}$ , the peak in the distribution occurred at an angle of  $45^{\circ}$  which is considerably higher than the angles so far measured in the freezing of bulk water.

A theoretical study of the factors affecting the development of crystallographic orientation distributions

in accreted ice has been made by Rye and Macklin (1973). They defined two functions: 1) the analogue of the measured distributions,  $f(\phi)$ , and 2)  $h(\phi) = f(\phi) / (2\pi \sin\phi)$ . The latter function avoids the geometrical effect which results in  $f(\phi)$  being zero parallel to the growth direction. When they analyzed the experimental data of Levi and Aufdermaur and of Knight and Knight (1968), who determined the crystallographic orientation distributions in natural hailstones, Rye and Macklin found two peaks in the  $h(\phi)$  distributions. One of these occurred at an angle of  $0^{\circ}$  and the other at an angle between  $27^{\circ}$  and  $40^{\circ}$ . The zero-degree peaks were considered to be due to the rapid warming of the droplet-ice interface which occurs when the droplet is first accreted. Assuming that the warming raises the interface temperature to about  $0^{\circ}\text{C}$ , then those crystals whose  $c$ -axes are normal to the ice surface produce dendrite sheets which are parallel to the surface. Such crystals are always in a preferred orientation. The higher angle peaks were again assumed to correspond to the angle of growth of the initial dendrites in bulk water of the same supercooling as that of the droplets. Rye and Macklin further suggested that the ratio of the sizes of the two peaks may also vary with the conditions under which the deposits were grown.

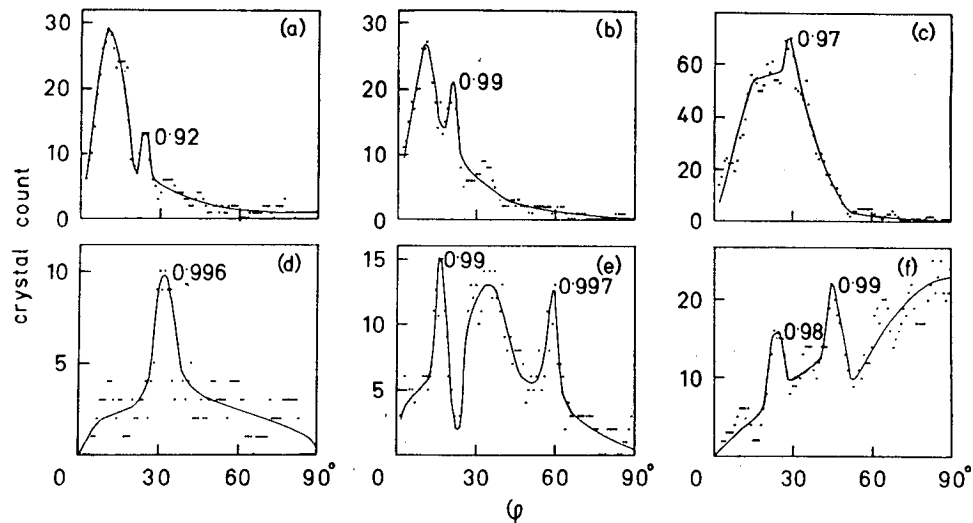


FIG. 1. *c*-axis crystallographic orientation distributions for various air and deposit temperatures. The crystal count is plotted as a function of the angle between the *c*-axis and the radial growth direction. The respective air and deposit temperatures are (a)  $-10, -7^{\circ}\text{C}$ , (b)  $-15, -9^{\circ}\text{C}$ , (c)  $-20, -6^{\circ}\text{C}$ , (d)  $-25, -3^{\circ}\text{C}$ , (e)  $-20, -13^{\circ}\text{C}$ , (f)  $-30, -5^{\circ}\text{C}$ .

The experiments described here were carried out to determine the quantitative dependence of the position of the growth-angle peaks, and of the ratio of the sizes of the two peaks on air temperature and other variables possibly involved in determining the crystallographic orientation distributions in accreted ice.

## 2. Experimental

The icing tunnel used to produce the samples of accreted ice was that described by Bailey and Macklin (1968). This is a vertical wind tunnel housed in a large cold room. Water droplets were injected into the tunnel from an array of sprays situated below the tunnel entrance. The liquid water concentration was varied either by using a different number of sprays or by varying the rate of flow of water through the individual sprays. Liquid water concentrations up to  $10\text{ gm m}^{-3}$  were obtained in this way. The median volume diameter of the droplet distribution was  $30\text{ }\mu\text{m}$  and the maximum diameter  $65\text{ }\mu\text{m}$ . For most experiments an airspeed of  $32\text{ m sec}^{-1}$  was used. Experiments were conducted also at an airspeed of  $15\text{ m sec}^{-1}$ .

The deposits were formed on a brass cylinder, 2.5 cm in diameter, which could be rotated at either 30 or 180 rpm. A thermistor was attached to the inner surface of the cylinder and its output fed to a chart recorder so that the deposit temperature was continuously monitored. The temperature at which a particular deposit was formed was determined by the air (i.e., droplet) temperature and the liquid water concentration. Each deposit was grown to a thickness of about 10 mm and the deposit temperature was held approximately constant by varying the liquid water concentration. The air temperature was also continuously

recorded and for each experimental run both the air and deposit temperatures were held constant to within about  $0.5^{\circ}\text{C}$ .

Sections of each deposit, 2–3 mm in thickness, were made in the usual manner (see, e.g., Levi and Aufdermaur, 1970) and the crystallographic orientations of the crystals in the sections determined using the etch-pit method. Two procedures were used to form the etch pits, the Formvar technique described by Higuchi (1958) and the vacuum etching technique, using a perforated foil, described by Knight (1966). While the latter technique is more controllable, it does not give sufficiently sharp etch pits in densely opaque ice samples because of the air bubbles present. Accordingly, the technique used was varied depending on the opacity of the sample. The etch pits formed were photographed through a microscope and the appropriate angular measurements made from projected images of the film. To facilitate the analysis of the data, the conversion of the angular measurements to crystallographic orientations and the plotting of the distributions was carried out by electronic computer.

In order to obtain maximum possible resolution from the data it was found necessary to use large numbers of crystals per sample, typically 150–200, and to devise a more complex representation of the *c*-axis orientation distributions than the simple histogram form used previously (e.g., Levi and Aufdermaur, 1970; Knight and Knight, 1968). The distributions were obtained by counting the crystals in intervals,  $4^{\circ}$  in width, centered on each whole degree from  $2^{\circ}$  to  $90^{\circ}$ .

## 3. Results and discussion

Typical distributions for various air and deposit temperatures are shown in Fig. 1. At the warmer air

temperatures ( $-10$  to  $-15^{\circ}\text{C}$ ) and at deposit temperatures below about  $-2^{\circ}\text{C}$  there are two peaks in the distributions, one at about  $10^{\circ}$  and the other at  $22^{\circ}$  (Figs. 1a and 1b). The former becomes the zero-degree peak in the  $h(\phi)$  distribution while the latter is considered to be the peak corresponding to the growth direction of the initial dendrites. At air temperatures above  $-10^{\circ}\text{C}$  the growth angle peak was too small, and at too small an angle, to be distinguished from the zero-degree peak. At deposit temperatures above  $-2^{\circ}\text{C}$  the distributions were more spread and a hump near  $90^{\circ}$  developed, indicative of the onset of wet growth (see Levi and Aufdermaur; Fig. 1d). At air temperatures between  $-20$  and  $-25^{\circ}\text{C}$  the growth-angle peak becomes more dominant and shifts toward  $32^{\circ}$ , while the  $0^{\circ}$  peak becomes rounded off (Figs. 1c and 1d). The significance of the growth-angle peaks was tested using binomial statistics and the appropriate probabilities are shown in the figures. In fitting the curves to the points, only peaks having a significance level higher than 0.90 have been indicated. At moderately low air temperatures ( $\sim -20$  to  $-25^{\circ}\text{C}$ ) and low deposit temperatures ( $\lesssim -8$  and  $-3^{\circ}\text{C}$ , respectively), there are two sharp peaks in the distribution, apart from a broader central maximum, which appear to be significant (Fig. 1e). At low air temperatures ( $\sim -30^{\circ}\text{C}$ ) the distribution tends to become sinusoidal although other significant peaks are still discernible (Fig. 1f). The appearance of these other peaks, which occurred in all the distributions determined at these temperatures, is as yet unexplained. The overall sinusoidal component

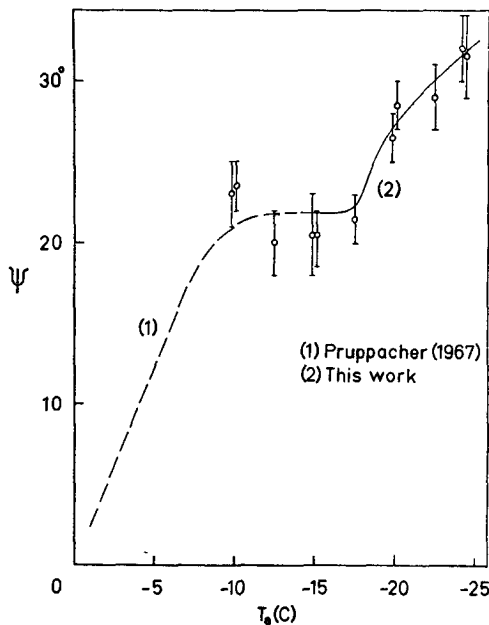


FIG. 2. The position of the growth angle peak ( $\psi$ ) as a function of air temperature ( $T_a$ ). Pruppacher's (1967) values for the angle between the growth direction and the basal plane of ice dendrites growing in bulk supercooled water are also shown.

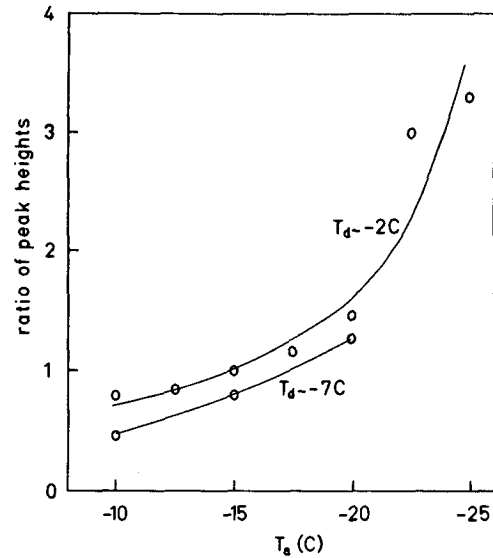


FIG. 3. The ratio of the heights of the growth-angle peak to the zero-degree peak as a function of air temperature. The two lines shown are for deposit temperatures of  $\sim -2$  and  $-7^{\circ}\text{C}$ .

is undoubtedly due to pronounced crystallographic reorientation (see Rye and Macklin, 1973). At all temperatures the position of the growth-angle peak was found to be independent of the deposit temperature, over the range of deposit temperatures in which the peak occurred. The position of the growth-angle peak was also unaffected by variation of airspeed and cylinder rotation rate.

The relation between the position of the growth-angle peak and air (i.e., droplet) temperature is shown in Fig. 2 together with Pruppacher's (1967) curve for the angle of growth to the basal plane of ice dendrites growing in bulk supercooled water. In the region of overlap, where in fact the growth angle is approximately independent of temperature, there is good agreement between the two sets of data. However, the present results indicate that the growth angle begins to increase again at supercoolings greater than about  $17^{\circ}\text{C}$ , reaching a value of  $32^{\circ}$  at a supercooling of  $25^{\circ}\text{C}$ . It is interesting to note that there is a discontinuity in the growth velocities of ice dendrites growing in supercooled water and that this occurs in the range of supercooling from about  $10$  to  $13^{\circ}\text{C}$  (Pruppacher, 1967). This has been interpreted as being due to a change in the molecular mechanism involved in the growth process (Macklin and Ryan, 1969). There is no evidence of a growth-angle peak occurring at an angle as high as  $45^{\circ}$ , implicit in Fig. 1c of Levi and Aufdermaur (1970). However, at about these temperatures ( $-20$  to  $-25^{\circ}\text{C}$ ) a broadening of the  $c$ -axis distribution due to crystallographic reorientation has been observed. This results in a broad central maximum (Fig. 1e) and it is suggested that this, and not the growth-angle peak, is what Levi and Aufdermaur observed.

TABLE 1. Characteristics of *c*-axis orientation distributions for deposit temperatures  $T_d$  less than  $\sim -2\text{C}$ .

Air temperature $T_a$	Characteristics
$\geq -10\text{C}$	No growth-angle peak.
$\sim -10$ to $-15\text{C}$	Growth-angle peak at $22^\circ$ and ratio of growth-angle to zero-degree peak heights less than unity.
$\sim -15$ to $-17\text{C}$	Growth-angle peak at $22^\circ$ and ratio of peak heights increasing (Fig. 3).
$\sim -17$ to $-25\text{C}$	Position of growth-angle peak varying (Fig. 2) and ratio of peak heights increasing rapidly (Fig. 3). Crystallographic reorientation affecting distribution at low deposit temperatures ( $-9$ to $-3\text{C}$ depending on ambient temperature).
$\leq -25\text{C}$	Distribution has overall sinusoidal component due to crystallographic reorientation.

The ratios of the heights of the growth-angle peak to the zero-degree peak have been determined and are shown as a function of air temperature in Fig. 3. There is a slight dependence on the deposit temperature which is considered to be due to varying surface roughness. The ratio of the peak heights begins to increase significantly at air temperatures below  $-15\text{C}$ .

The sizes of the individual crystals (lengths and widths) were also measured. The information obtained was essentially similar to that presented by Levi and Aufdermaur (1970, Fig. 8).

#### 4. Conclusions

The present investigation has shown that there are peaks in the *c*-axis orientation distributions of accreted ice which may be related to the air temperature. There are, in general, two peaks in the distributions: a "zero-degree" peak and a peak at an angle which corresponds to the direction of growth of ice dendrites formed at the droplet supercooling. The position of the peaks is independent of the temperature of the ice deposit over a wide range, the airspeed, and the rate of rotation of the accreting object. Since the growth angle deduced from ice deposits formed from  $30\text{-}\mu\text{m}$

droplets is the same as that of ice dendrites growing in bulk supercooled water, the position of the growth-angle peak is inferred to be also independent of droplet size.

Levi and Aufdermaur (1970) have shown that the crystallographic orientation distributions in accreted ice reach equilibrium in a fraction of a millimeter of growth thickness. Consequently, it should be possible to infer from the crystallographic analysis of hailstone layers 1–2 mm in thickness, the air temperatures, or range of air temperatures, at which the layers were formed. The characteristics of the *c*-axis orientation distributions for deposit temperatures less than  $\sim -2\text{C}$  are summarized in Table 1.

*Acknowledgments.* This work was supported by the Australian Research Grants Committee. One of us (P. J. R.) is indebted to the Commonwealth Scientific and Industrial Research Organization for the provision of a Postgraduate Studentship. We are indebted to Mr. J. N. Carras for assistance in making the ice deposits and to Dr. C. A. Knight for supplying the perforated foil used in etching the sections.

#### REFERENCES

- Bailey, I. H., and W. C. Macklin, 1968: The surface configuration and internal structure of artificial hailstones. *Quart. J. Roy. Meteor. Soc.*, **94**, 1–11.
- Higuchi, K., 1958: The etching of ice crystals. *Acta Metall.*, **6**, 636–642.
- Knight, C. A., 1966: Formation of crystallographic etch pits on ice and its application to the study of hailstones. *J. Appl. Meteor.*, **5**, 710–714.
- , and N. C. Knight, 1968: Spongy hailstone growth criteria I. Orientation fabrics. *J. Atmos. Sci.*, **25**, 445–452.
- Levi, L., and A. N. Aufdermaur, 1970: Crystallographic orientation and crystal size in cylindrical accretions of ice. *J. Atmos. Sci.*, **27**, 443–452.
- Macklin, W. C., and B. F. Ryan, 1965: The structure of ice grown in bulk supercooled water. *J. Atmos. Sci.*, **22**, 452–459.
- , and —, 1969: Interpretation of the growth rates of ice dendrites in supercooled water. *J. Chem. Phys.*, **50**, 551–2.
- Pruppacher, H. R., 1967: Growth modes of ice crystals in supercooled water and aqueous solutions. *J. Glaciology*, **6**, 651–662.
- Rye, P. J., and W. C. Macklin, 1973: Interpretation of crystallographic orientations in accreted ice. *J. Atmos. Sci.*, **30**, 1421–1426.