Growth Rates and Habits of Ice Crystals Grown from the Vapor Phase

D. LAMB AND PETER V. HOBBS
Dept. of Atmospheric Sciences, University of Washington, Seattle
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1. Introduction

Studies of the growth rates and habits of ice crystals grown from the vapor phase are of considerable importance in cloud and precipitation physics as well as being of general interest in the field of crystal growth. Some experimental facts are now well established. The basic habit (plate-like or column-like) of an ice crystal growing from the vapor phase is governed primarily by temperature (Nakaya, 1954; Hallett and Mason, 1958; Kobayashi, 1957, 1961, 1965). As the temperature is lowered from 0 to -90°C the basic habit changes in the sequence: plate, column, plate, column; the approximate temperatures at which the transitions occur are -4, -9 and -22°C. The rate of increase in the mass of an ice crystal, growing at water saturation in air, also varies with temperature, with maxima occurring at about -5 and -15°C and a minimum at about -8°C (Hallett, 1965; Fukuta, 1969).

A satisfactory explanation for the variation of ice crystal habits with temperature has not yet been given. The studies of Bryant et al. (1959), Hallett (1961) and Mason et al. (1963) indicate that ice crystals grow from the vapor by the propagation of steps, a few hundred angstroms in height, across the surfaces of the crystals. In the light of these observations, Mason et al. and Hobbs and Scott (1965) proposed tentative theories for the variations of the habit of ice crystals with temperature. Although both of these theories were based on measurements of the variation with temperature of

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parameters related to the propagation of steps on the basal face, the behavior of steps on the prism face had to be postulated. Moreover, both theories are open to criticism on theoretical grounds.

The temperature dependence of the mass growth rates of ice crystals at water saturation has been explained in the past solely in terms of the variations with temperature of the difference in equilibrium vapor pressures over ice and water together with changes in the electrostatic capacity of a crystal with temperature as the habit varies. Although these two factors probably play important roles in determining the mass growth rate of an ice crystal, account must also be taken of the surface kinetics as reflected in the condensation coefficient at different temperatures.

The purpose of the present note is to draw attention to some recent measurements of the linear growth rates of the basal and prism faces of ice crystals, and to point out their relevance to our understanding of the variations with temperature of the habit of ice crystals and their mass growth rate.

2. Linear growth rates of the basal and prism faces of ice crystals

Full details of the experimental techniques involved in obtaining the measurements have been given by Lamb (1970) and need not be repeated here. Suffice to say that the conditions were such that the growth of the crystals was controlled by molecular events taking place on the surfaces of the crystals and not by the rate of supply of water molecules from the vapor phase or by the rate at which the latent heat of deposition was removed.

The results of the measurements of the linear growth rates of the basal and prism faces of ice as a function of temperature are shown in Fig. 1. The ice crystals were situated in an environment of pure water vapor and the excess vapor pressure was held constant at (10±1) μm of mercury irrespective of temperature. Since the net impingement flux of molecules onto a crystal was the same at all temperatures, the marked variations of the linear growth rates of both the basal and prism faces with temperature are a consequence of changes with temperature in the condensation coefficients of these two faces.

3. Ice crystal habits

When the linear growth rate of the basal face is greater than that of the prism face, column-like crystals grow, and when the reverse holds plate-like crystals grow. The temperatures at which the two curves in Fig. 1 cross over, namely −5.3 and −9.5°C, are in reasonable agreement with the temperatures at which ice crystals grown from the vapor in an environment of air change their basic habit. The alteration of the basic habit of ice crystals with temperature is therefore a consequence of the fact that the linear growth rates of the basal and prism faces of ice fluctuate with temperature in such a way that they cross each other several times as the temperature is lowered.

Comparison of the measurements obtained by Hallett (1961) of the velocity of steps on the basal face of ice as a function of temperature with the fluctuations with temperature of the linear growth rate of the basal face (Fig. 1) reveals a close correspondence. We conclude from this observation that the linear growth rate of the basal face at any temperature is qualitatively proportional to the velocity of steps on the basal face at that temperature. The velocities of steps on the prism face of ice have not been measured, but if, like the basal face, the linear growth rate of the prism face is proportional to the velocity of steps on that face, then the variation with temperature of the linear growth rate of the prism face shown in Fig. 1 also represents the variation with temperature of the velocity of steps on the prism face. In this connection we note that if a crystal face does grow by the incorporation of adsorbed molecules into spiral steps then the linear growth rate of the face is proportional to the velocity of steps on that face.

To complete our understanding of the variations of ice crystal habits with temperature, an explanation is required for the unusual variations with temperature of the linear growth rates and the velocities of steps. Although attempts to explain the latter have been made by Mason et al. (1963), Hobbs and Scott (1965) and Ryan and Macklin (1969), a satisfactory quantitative explanation has not yet been given. It may be readily shown that the classical theory of crystal growth by spiral steps (Burton et al., 1951) fails to predict the observed temperature dependence of the velocity of
steps or the linear growth rates of ice (Mason et al.; Hobbs and Scott).

4. Mass growth rates

The measurements of the linear growth rates of the basal and prism faces of ice which are shown in Fig. 1 can be used to calculate the temperature dependence of the mass growth of an ice crystal situated in an environment of pure water vapor at a constant excess vapor pressure of 10 μm of mercury.

The rate of increase in the mass $M$ of an ice crystal in the form of a solid hexagonal prism is given by

$$\frac{dM}{dt} = \rho [G_B(T)A_B + G_P(T)A_P],$$

(1)

where $\rho$ is the density of ice, $G_B(T)$ and $G_P(T)$ are the linear growth rates of the basal and prism faces, and $A_B$ and $A_P$ the total areas of the basal and prism faces, respectively. If $a$ is the diameter of the inscribed circle around the hexagonal base of the prism and $c$ the length of the prism, then

$$A_B = \frac{3a^2}{2 \cos 30^\circ},$$

(2)

$$A_P = \frac{3ac}{\cos 30^\circ}.$$

(3)

In an environment of pure water vapor the linear growth rates are constant with time at any one temperature so that the values of $a$ and $c$ at time $t$ after nucleation are given by

$$a = 2G_P(T)t,$$

(4)

$$c = 2G_B(T)t.$$  

(5)

Combining (1)–(5) we obtain for the rate of mass accumulation

$$\frac{dM}{dt} = \frac{18\rho}{\cos 30^\circ} G_B(T)G_P(T)t^3.$$

(6)

Substituting the values from the best-fit curves to the experimental measurements of $G_B(T)$ and $G_P(T)$, which are shown in Fig. 1, into Eq. (6), the mass growth rate of a hexagonal ice prism can be determined as a function of temperature. The results for $t = 10^3$ sec are shown in Fig. 2. We refer to this growth rate as the “inherent” mass growth rate since it is determined by the condensation coefficients of the basal and prism faces and not by the supply of water vapor to the crystal. Of particular interest is the fact that the “inherent” mass growth rate attains peak values at temperatures of $-5.5$ and $-12^\circ$C and a minimum value at $-8^\circ$C. These temperatures are approximately the same as those for which the rate of increase in the mass of an ice crystal growing at water saturation in air is observed to attain maximum and minimum values. Clearly, therefore, the variation of the condensation coefficients of the basal and prism faces of ice with temperature must play an important role in determining the temperature dependence of the mass growth rate of an ice crystal in an environment of air at water saturation. In the latter case, the difference between the equilibrium vapor pressures over ice and water and the diffusion of the water vapor through the air will also affect the mass growth rate. Since the difference between the equilibrium vapor pressures over ice and water reaches a maximum value at a temperature of about $-12^\circ$C, this will enhance the peak imposed by the “inherent” mass growth rate at the lower temperature relative to that at the higher temperature.

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