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XXIX. The Growth of Ice Crystals from the Vapour

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Imperial College, London*

[Received November 18, 1954]

Summary

The growth and evaporation of single ice crystals have been studied in relation to the temperature and supersaturation of the environment. The sequence of crystal forms which occurs in clouds as the temperature falls from 0°C to −40°C has been reproduced in these experiments. Measurements of the growth rates of individual faces of crystals growing under fixed environmental conditions reveal that $dR^2/dt = \text{const.}$ where $R$ is a linear crystal dimension, but the growth rates of basal and prism faces are different to a degree depending mainly on the temperature. At constant temperature, $dR^2/dt$ varied as the square of the supersaturation $\sigma$ over the range of $\sigma$ studied. A critical supersaturation for growth was found which varied in an apparently random manner from face to face, there being no systematic difference between the values for the prism and basal faces. The crystal habit, defined by limiting values of the axis/diameter ratio, was found to be controlled very largely by the temperature, the supersaturation having a much smaller, non-systematic effect.

§ 1. Introduction

The growth of ice crystals, besides being an interesting and difficult study in crystal physics, is of particular importance to the cloud physicist concerned with the development of the ice phase in natural clouds. Snow crystals occur in a wide variety of shapes and forms and many workers have tried to relate the occurrence of particular crystal forms with the meteorological conditions. That they have not found a clearly marked correlation between the predominant crystal habit and the temperature at the ground is, perhaps, not surprising, since only the conditions prevailing during the growth of the crystals are likely to be of major importance. It is only in recent years that crystals have been collected from different types of clouds having widely different conditions of temperature and water-vapour concentration, and that a correlation between the crystal habit and the environmental conditions has been established. The most comprehensive study of this kind has been made by Weickmann (1947) who took samples at different heights up to cirrus levels where the temperature was below −40°C. His observations are summarized in table 1.

* Communicated by the Authors.
The familiar needle-like crystals, which are not represented in Weickmann’s observations, are observed at the ground when the temperature is only slightly below freezing, and according to Wall (1947) and to Gold and Power (1952), originate from clouds with temperatures between $-3^\circ$C and $-8^\circ$C.

Table 1. Weickmann’s Observations of Predominant Crystal Forms in Different Cloud Types

<table>
<thead>
<tr>
<th>Level of observation</th>
<th>Temp. range</th>
<th>Cloud types</th>
<th>Crystal forms</th>
</tr>
</thead>
<tbody>
<tr>
<td>Lower troposphere</td>
<td>0$^\circ$C to $-15^\circ$C</td>
<td>Nimbostratus, Stratocumulus</td>
<td>Thin hexagonal plates, Star-shaped crystals showing dendritic structure</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Stratus</td>
<td></td>
</tr>
<tr>
<td>Middle troposphere</td>
<td>$-15^\circ$C to $-30^\circ$C</td>
<td>Altostratus, Altocumulus</td>
<td>Thick hexagonal plates, Prismatic columns—single prisms and twins</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Isolated cirrus</td>
<td>Clusters of prismatic columns containing funnel-shaped cavities. Some single hollow prisms</td>
</tr>
<tr>
<td>Upper troposphere</td>
<td>$&lt;-30^\circ$C</td>
<td>Cirrostratus</td>
<td>Individual complete prisms</td>
</tr>
</tbody>
</table>

The existence of a number of basic forms of snow crystals, e.g. hexagonal plates, hexagonal prismatic columns, dendritic forms, and of an almost infinite number of variations on each of these main themes, suggests that their growth and development are complicated matters. As some of these variations may arise from changes in environmental conditions as the crystal falls through the atmosphere, it seems judicious to study the growth of crystals in the laboratory where the temperature and supersaturation of the environment can be controlled.

The growth of crystals in artificially-produced supercooled water clouds has been studied by aufm Kampe, Weickmann and Kelly (1951) and by Mason (1953). The clouds are formed by introducing steam into thermostatically-controlled, room-size, cold chambers and crystal formation is initiated by seeding the cloud with a minute quantity of solid carbon dioxide. In the presence of a water cloud it can be assumed that the crystals are growing in an atmosphere saturated with respect to liquid water, and therefore supersaturated with respect to ice by an amount determined solely by the temperature.

The observed changes of crystal habit with temperature were very similar in both sets of experiments and are summarized in table 2, which shows a marked similarity to table 1 giving the temperature classification of natural snow crystals.
Table 2. Changes of Crystal Habit with Temperature in Artificially-produced Water Clouds (aufm Kampe et al., and Mason)

<table>
<thead>
<tr>
<th>Temperature range</th>
<th>Crystal habit</th>
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<tbody>
<tr>
<td>0°C to −5°C</td>
<td>Single, clear, hexagonal plates with no surface markings. Some trigonal plates</td>
</tr>
<tr>
<td>−4°C to −9°C</td>
<td>Prisms, some showing marked cavities and similarity to needles</td>
</tr>
<tr>
<td>−10°C to −25°C</td>
<td>Hexagonal plates showing ribs, surface markings and tendency to sprout at corners. Sector stars</td>
</tr>
<tr>
<td>−25°C to −40°C</td>
<td>Single prisms, twins, and hollow prisms. Aggregates of prisms and irregular crystals</td>
</tr>
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</table>

The most striking feature of table 2 is the remarkable sequence of habit, plates–prisms–plates–prisms which occurs as the temperature is lowered from 0°C to about −25°C. Such marked changes in habit within this small temperature range, involving changes from preferred growth along the principal c axis for prisms, to preferred growth along the secondary a axes for plates, are apparently peculiar to ice.

To investigate in more detail the factors which control the relative growth rates of different crystal faces, it was decided to make continuous observations on single crystals growing in an atmosphere where the temperature and supersaturation could be controlled and varied independently. The crystals were grown on a metal surface, their growth being recorded by time-lapse photography so that the rates of advance of the different crystal faces could be measured.

§ 2. The Experimental Arrangement

A diagram of the apparatus is shown in fig. 1. The clean, smooth, metal surface on which the crystals are grown is situated in the centre of a cylindrical metal chamber, internal diameter 3 inches and depth 2 inches, the hollow walls of which may be cooled down to a temperature of −55°C by circulating petrol chilled with solid CO₂ through the annular space. The temperature of the chamber walls is controlled by the temperature of a cold bath in which is immersed a long copper spiral forming part of the circuit for the cooling fluid. The base of the chamber is coated with ice, so that the air in the experimental space, which is stirred by a small electric fan, is uniformly saturated with respect to ice at a temperature $T_1$, indicated by a sensitive thermocouple embedded in the surface of the ice layer on the floor of the chamber. The temperature over the floor does not vary by more than 0.2°C, and the thermocouple is located in the coldest spot near the inlet for the cooling fluid.

The experimental surface, a metal plate, is supported on a long copper rod which is insulated from the chamber and dips into a Dewar flask containing liquid air. The plate is thus cooled to a temperature $T_2$ lower than that of the surrounding air, and which can be controlled by
varying the current through a heating coil wound around the copper rod. The temperature $T_2$ of the growing crystals is recorded by a thermocouple soldered immediately below the experimental surface, and the saturation ratio of the air in the immediate vicinity of the crystals is altered by adjusting the temperature difference $T_1 - T_2$, and is measured by the ratio of the equilibrium vapour pressure of ice at the temperatures $T_1$ and $T_2$ respectively.

Fig. 1

Diagram of the apparatus.

The crystals were viewed through a metallurgical microscope fitted with a vertical illuminator and a 4 mm objective, and were photographed at one-minute intervals with a 16 mm camera using a fine-grain film. The growth rates of the crystal faces were determined from measurements made on the negatives with a micrometer eyepiece.

§ 3. Formation of Crystals on the Metal Surface

In order to produce ice crystals on the test surface it was generally found necessary to cool it to the dew point of the surrounding air, so that condensation occurred on the plate. This confirmed the earlier observations of Weickmann (1947) that the great majority of crystals form only when the air is saturated with respect to liquid water, and therefore probably arise by the freezing of the supercooled liquid. Only an occasional crystal formed slightly before the dew point was reached;
in these cases the air was highly supersaturated relative to ice and condensation of very small quantities of water may have occurred in surface cavities of solid nuclei or in fine scratches on the metal surface. To obtain a regular crop of well-formed crystals the metal plate was exposed to a small dose of silver-iodide smoke which provides active nuclei at temperatures below $-5^\circ C$.

Typical examples of crystals forming at different temperatures are shown in fig. 2 (Plate 3). Crystals which grew with their $c$ axes normal to the surface permitted measurement of the rates of advance of three pairs of opposite prism faces. In the case of hexagonal prisms growing with their $c$ axes parallel to the surface, measurements could be made of the growth rates of both the basal faces and the prism faces. Since it was possible to measure only the diameters and length of a crystal, the sum of the growth rates of opposite faces, rather than the growth rate of each face separately, was determined.

It was necessary to work with a sparse deposit of crystals, their spacing being large compared with their linear dimensions, so that the growth of one crystal did not seriously interfere with that of its neighbours.

§ 4. CORRESPONDENCE BETWEEN CRYSTAL FORMS APPEARING ON METAL PLATE AND IN CLOUDS

The first experiments were designed to show whether the various crystal forms appearing in natural and laboratory-produced clouds as described in the Introduction, could be produced in the same temperature ranges on the metal plate.

The plate temperature was held at various temperatures between $-5^\circ C$ and $-40^\circ C$ and crystals were grown at constant temperature, the supersaturation being adjusted to correspond to water-saturation. The crystal habit was found to alternate between prismatic columns and thin hexagonal plates as the temperature was lowered according to the following scheme:

- $-5^\circ C$ to $-9^\circ C$: Prisms,
- $-9^\circ C$ to $-25^\circ C$: Plates,
- below $-25^\circ C$: Prisms.

As a very few crystals appeared at temperatures above $-5^\circ C$, crystals were initially formed at a lower temperature but grown subsequently at temperatures above $-5^\circ C$. Although they may have originated at a temperature normally associated with prismatic columns, when the temperature was raised, they assumed subsequently the plate-like habit which was typical of the temperature range $0^\circ C$ to $-5^\circ C$ in the cloud experiments.

Thus, the variation of crystal habit with temperature followed a very similar pattern to that found in both natural clouds and the model clouds of the laboratory. But, in addition, a new and interesting crystal form quite often appeared on the plate at temperatures between $-4^\circ C$ and
and also below $-22^\circ$C. This took the form of a prismatic column terminated at one end only by a pyramid (see fig. 2). The appearance of such hemimorphic forms, which have been found only very occasionally in natural clouds (usually cirrostratus) but not at all in the small-scale laboratory clouds, is of considerable interest in connection with the possibility of ice possessing a polar lattice (see Owston (1951), Mason and Owston (1952)).

Fig. 3

Plot of the square of the linear dimensions of the crystal against time.

§ 5. GROWTH RATES OF CRYSTAL FACES

The photography at regular time intervals of crystals growing with their c axes either normal or parallel to the metal surface allowed the rates of advance of both the prism and basal faces to be determined. When the square of the linear dimensions $(2c)^2$ or $(2a)^2$ of a crystal were plotted against time, straight lines resulted as shown in fig. 3.

The growth rate of a crystal under steady-state conditions is determined by the balance between the rate at which material diffuses towards the crystal and the rate at which it can be built into the crystal. If the diffusion rate is much greater than the rate of incorporation at the crystal face, the growth is said to be surface limited. If, however, the reverse is the case, the growth rate is determined almost entirely by the rate of diffusion, i.e. we have diffusion-limited growth. Under the latter circumstances, the rate of advance of a particular face should be inversely proportional to some linear dimension of the crystal. The fact that in our experiments the quantities $da/dt$ and $dc/dt$ were inversely proportional to $a$ and $c$ respectively and not to $c$ and $a$ respectively (the ratio $\gamma = c/a$
varied considerably during growth), enables us to describe the growth of
the faces by the equations

$$(2c)^2 = U_c t + \alpha; \quad (2a)^2 = U_a t + \beta$$

where $c$ and $a$ are respectively the semi-principal and semi-secondary
crystal axes, $U_c$, $U_a$, $\alpha$ and $\beta$ are constants. It follows that the crystal
grows towards a limiting habit which may be defined by

$$I = \left| \frac{c}{a} \right|_{\text{lim}} = \sqrt{(U_c/U_a)} = \sqrt{\left[ \frac{\partial}{\partial t} (2c)^2 / \frac{\partial}{\partial t} (2a)^2 \right]}.$$  

The limiting habit, which can be determined experimentally from the
plots of $(2c)^2$ and $(2a)^2$ against time, is the most suitable parameter to
use in discussing the crystal habit because, unlike the actual $c/a$ ratio
of the crystal, it is independent of the period of growth.

![Fig. 4](image)

The growth of three opposite pairs of prism faces of the same crystal, showing
that different faces grow at different rates.

Although the square law of growth held for all crystal faces indicating
that the growth was largely controlled by diffusion, the actual values of
d$a^2/dt$ and $d\sigma^2/dt$ were somewhat different for different crystals growing
under the same environmental conditions, while crystallographically
similar faces of the same crystal sometimes grew at different rates (differences of 50% were not uncommon) as shown in fig. 4. Furthermore, the
growth rate of a particular crystal face sometimes changed abruptly, even
though the temperature, supersaturation and rate of stirring remained
constant. A typical case is illustrated in fig. 5. Very similar behaviour has been observed by Bunn (1949) and by Humphreys-Owen (1949) in the case of crystals growing from solution. These results indicate that the growth of crystal faces is not determined solely by external conditions, but is partly influenced by certain properties of the faces themselves. Thus the variations in habit may be partly attributed to different faces having their own individual characteristics which control the rate at which the available molecules can be built into the crystal structure.

Discontinuities in the growth rate of particular crystal faces.

Detailed examination of all the available data on crystals growing at constant temperature revealed no correlation between the limiting habit of a crystal and its rate of mass increase which is a measure of the contemporary total flux of vapour towards the crystal. This point is of importance in that Weickmann (1950) and also Marshall and Langleben (1954) have suggested that the crystal habit is essentially controlled by the flux of vapour towards the growing surfaces.
§ 6. Variation of Growth and Evaporation Rates with Saturation Ratio

To study how the rate of propagation of a crystal face varied with the supersaturation of the environment, observations were made on individual crystals growing at constant temperature, as the supersaturation, governed by \((T_1 - T_2)\), was changed. The first step was to establish the absolute value of the supersaturation for each experiment. To do this, graphs of \(\sqrt{U_e}\) and \(\sqrt{U_a}\) were plotted against \((T_1 - T_2)\). The resulting straight lines cut the \((T_1 - T_2)\) axis at a distance \(\pm \Delta T\) from the origin which was the same for the plots of \(\sqrt{U_e}\) and \(\sqrt{U_a}\). The intercept \(\Delta T\) was always small in magnitude (<1°C), varied in sign from experiment to experiment and showed no systematic correlation with the plate temperature \(T_2\). These small fluctuations of the straight-line plots about the origin were probably due to slight changes of temperature distribution within the experimental chamber and also small changes in the air temperature within the cold box from experiment to experiment. These results indicated that ideally the supersaturation of \(\sigma\) of the air in the immediate vicinity of the crystals was given by \(\sigma = p(T_1)/p(T_2) - 1\) where \(p(T_1)\) and \(p(T_2)\) are the equilibrium vapour pressures of ice at temperatures \(T_1\) and \(T_2\) respectively; the actual supersaturation prevailing in any one experiment was assumed to be \([p(T_1 + \Delta T) - p(T_2)]/p(T_2)\). In this way it was possible to obtain plots of \(\sqrt{U_e}\) and \(\sqrt{U_a}\) against the supersaturation \(\sigma\), the temperature \(T_2\) remaining constant, for a large number of crystals and for different values of \(T_2\). Typical plots are shown in figs. 6(a), (b), which represent observations made during both growth and evaporation of basal and prism faces. In general, it was found that the quantity \(U\) was proportional to the square of the supersaturation \(\sigma\) (the values of \(\sigma\) employed ranged from 0 to 40%), but with the higher supersaturations there were occasions on which the growth rate was smaller than that which would obtained by extrapolation of the \(U - \sigma^2\) relation.

The behaviour of crystals evaporating under large subsaturations was interesting; under these conditions of rapid evaporation the reciprocity between evaporation and growth suggested by fig. 6 broke down. The crystal developed well rounded corners and eventually lost its external crystalline appearance, the rate of evaporation being much faster than the growth rate under a supersaturation of the same magnitude. It was suggested by Frank (1949) that at moderate subsaturations, dissolution of a crystal face should proceed in a manner closely equivalent to growth, i.e. by unbuilding at molecular steps ending on screw dislocations, but that under large subsaturations, evaporation may arise from attack at the edges and corners to produce curved faces; evaporation then proceeds rapidly and the crystal loses its geometrical shape.

§ 7. Critical Saturation Ratios for Growth and Evaporation

For some of the crystals studied, the saturation ratio \(\alpha = \sigma + 1\) had to exceed or be less than a certain value before growth or evaporation of the
faces could be detected (see fig. 7). The value of the critical saturation ratio was often appreciably different for different faces of the same crystal, but there was no systematic difference between the initial resistance

Fig. 6

\[ \sqrt{|U_a|} \left(10^4 \text{ cm sec}^{-1}\right) \]

Crystal Temperature

-17.6°C

(a)

\[ \sqrt{|U_{cl}|} \left(10^4 \text{ cm sec}^{-1}\right) \]

or \[ |U_{cl}| \left(10^8 \text{ cm}^2 \text{ sec}^{-1}\right) \]

Crystal Temperature

-21.8°C

(b)

Plots of \( \sqrt{|U_a|} \), \( \sqrt{|U_{cl}|} \) and \( |U_{cl}| \) against the supersaturation \( \sigma \).
of prisms and basal faces. This is obvious from fig. 8 in which are plotted the critical values of \( \alpha \) for crystals growing at different temperatures. Again, there was evidence that growth and evaporation occurred more readily on some faces than on others which were crystallographically similar.

Values of \( |U_c| \) plotted against supersaturation showing the critical saturation ratios required for growth and evaporation of crystal faces.

§ 8. VARIATION OF CRYSTAL HABIT WITH TEMPERATURE AND SUPERSATURATION

That the curves of growth rate plotted against supersaturation (figs. 6 (a), (b)) for the basal and prism faces were, in general, of constant slope indicates that the crystal habit defined by \( \Gamma = \sqrt{(U_c/U_a)} \) was generally independent of the supersaturation. This was confirmed by plotting \( \Gamma \) against the supersaturation for each of a large number of
crystals grown at the same temperature; for any given value of the supersaturation, there was a considerable spread in the values of \( \Gamma \) for different crystals but there was no obvious dependence of \( \Gamma \) on the supersaturation. The maximum and minimum values of \( \Gamma \) obtained at various fixed temperatures when the supersaturation was varied are shown in fig. 9. Remembering that values of \( \Gamma < 1 \) indicate a tendency towards plate-like growth and values of \( \Gamma > 1 \), a tendency towards columnar

![Diagram](image)

**Fig. 8**

The critical saturation ratios for growth of basal and prism faces as a function of temperature.

development, fig. 9 suggests that there is a correlation between the temperature and the crystal habit, with a suggestion of the sequence plates—prisms—plates—prisms described in § 3. This indicates that the crystal habit is controlled much more by the temperature than by the supersaturation. The manner in which the crystal habit varied with temperature was brought out more clearly by confining attention to those crystals in each batch which had the largest growth rates (and so were probably least influenced by neighbouring crystals), and to regimes of supersaturation in which both \( \sqrt{U_a} \) and \( \sqrt{U_c} \) were proportional to \( \sigma \), so that \( \Gamma \) was independent of the supersaturation. Values of \( \Gamma \) were obtained from the slopes of the \( \sqrt{U - \sigma} \) plots (e.g. fig. 6), and are shown
as a function of temperature in fig. 10. This indicates a tendency towards plate-like growth between 0 and $-5^\circ$C, for prismatic columns between $-5^\circ$C and $-9^\circ$C, for plates between $-9^\circ$C and $-26^\circ$C and for prisms again below $-26^\circ$C. Thus, in reducing the scatter in the observations occasioned by the individual peculiarities of each crystal, we have shown that the

![Graph](image)

**Fig. 9**

Crystal Temperature $T_2 ^\circ$C

The maximum and minimum values of the limiting habit $\Gamma$ obtained at various fixed temperatures when the supersaturation was varied.

![Graph](image)

**Fig. 10**

Crystal Temperature $T_2 ^\circ$C

The limiting crystal habit $\Gamma = \frac{\partial}{\partial \sigma} \sqrt{U_c} / \frac{\partial}{\partial \sigma} \sqrt{U_a}$ as a function of temperature.

limiting crystal habit is determined largely by the temperature and that although the supersaturation controls very largely the absolute growth rate of a face, it appears to exert only a minor and non-systematic influence on the relative growth rates of the basal and prism faces.
References