Experimental Studies on the Growth of Small Ice Crystals

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**ABSTRACT**

The fall velocities and growth rates of freely falling ice crystals were measured at various intervals for about 1 min after ice crystal nucleation of a supercooled fog. Measurements were also made at various degrees of supercooling.

The results showed that: 1) the velocity varied linearly with time t for 60 sec after the first 12-sec period, with two falling velocity plateaus being found at −5 and −8, and −10 and −18°C; 2) the mass varied as \( \beta \) except at −18 to −19°C, a small mass growth rate peak being found at −6.5°C; 3) the larger peak shifted toward colder regions as the crystal grew; 4) tsuzumi crystals formed at around −18°C; and 5) at the temperatures where two maxima of the mass growth occurred, the apparent densities were at minima.

Current theory failed to describe the mass growth rate of ice crystals. The existence of an unknown factor was detected and introduced into the rate equation of ice crystal growth.

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1. Introduction

In the process of ice phase precipitation development, growing ice crystals play important roles (Bergeron, 1935). The growth of ice crystals has been studied by various researchers and some of the specific problems appear to be fairly well understood (Mason, 1965; Byers, 1965). However, such problems have often been handled individually under simplified conditions rather than by directly studying the complex behavior of ice crystals in an actual cloud environment. As a result, the interpretation of this information presents technical difficulties when one tries to understand the whole process of ice phase precipitation. The results naturally contain a large degree of uncertainty.

On the other hand, while some success has been and is being realized by increasing efforts put into direct field or airborne study of the processes in natural clouds, the great complexity of the cloud system requires a considerable improvement in observational methods, i.e., instrumentation, before more useful information can be obtained.

The purpose of this study was to fill gaps in our previous fundamental knowledge by making experimental studies of ice crystal growth under conditions which closely simulate those of the atmosphere. The new and more reliable data are expected to assist in the development of numerical models of ice phase precipitation which, in turn, will help to fill the gaps of knowledge in laboratory and field studies.

2. Experimental procedure

The work was concerned with the measurement of three major factors affecting the growth of ice crystals in supercooled fog, i.e., fall velocity, shape and size, and mass. In each case measurements were made at various supercooled fog temperatures as a function of time, for a period of about 1 min after nucleation.

The experimental apparatus used for this study is shown in Figs. 1 and 2. It is a double-walled chamber made of Plexiglas with inside dimensions of 9.6×9.6×61.5 cm³ and outside dimensions of 15.5×15.5×61.5 cm³.

![Diagram of experimental apparatus](image-url)
The chamber is cooled with 70% aqueous glycerine solution chilled to a predetermined temperature and circulated by a cooling unit. A wooden frame supports a black cloth about 6 mm from the inside wall of the chamber to prevent frost formation. Near the bottom, the frame supports two black 8-mesh screens 2 cm apart. A slide holder was placed under the screens.

A cooled copper chamber with inside dimensions of $10.5 \times 10.5 \times 50.5$ cm$^3$ and outside dimensions of $14 \times 14 \times 50.5$ cm$^3$ was placed on top of the Plexiglas cloud chamber to serve as a fog source and precooler. Electrically warmed water in a cellulose acetate semipermeable membrane supplied moisture at the top of the copper chamber, the fog being formed on condensation nuclei existing in room air. Two thermocouples were used to measure fog temperatures which were recorded on a strip chart recorder. The temperature throughout both chambers is uniform to within ±0.2°C except within the top 20 cm of the copper chamber.

It was interesting to note a temperature drop of several tenths of a degree when fog first reached the lower cloud chamber. This was assumed to be due to cooling caused by evaporation of the fog droplets.

Since minute amounts of organic or inorganic vapor can change the crystal growth habit, extreme care was taken to avoid such contamination. All volatile chemicals were removed from the laboratory and no plastic replica methods were used.

Seeding of the supercooled fog with ice crystals was accomplished at the opening near the bottom of the upper chamber by means of a brass rod 3 mm in diameter indirectly chilled by dry ice. A rapid circular swing of the rod in the fog produced minute ice crystals in a well dispersed manner which immediately started growing at water saturation.

The number of ice crystals was adjusted to about 1–10 cm$^{-3}$ to avoid moisture exhaustion.

### a. Fall velocity measurements

For the fall velocity measurements, the cloud chamber was illuminated through a slit with a light beam from a mercury lamp filtered with a solution of cupric sulphate and chopped at a known speed. A Polaroid Land camera was placed in a position perpendicular to the illumination system. At predetermined time intervals after seeding, with the chopper operating, photographs were taken at a slow shutter speed (1 sec) so that the falling ice crystals were recorded as chopped streaks of light (see Fig. 3). The distance on the photograph was calibrated by a photograph of a ruler taken from the same distance.

The measurements were made over time intervals of 15–20, 25–30, 35–40, 45–50, 55–60 and 65–70 sec after seeding at about 2°C temperature intervals from approximately −3 to −20°C.

Fogging of the outside Plexiglas wall within the optical system was avoided by attaching another Plexiglas plate with a few millimeter gap of trapped air. The two metal screens served to suppress turbulence.
in the chamber, the air between them being motionless when tested by smoke during the fall velocity measurements. While ice crystals always grew on the screens, the number of fragments coming from them were low and their sizes were such that they could be easily detected on the photographic plates.

b. Growth rate measurement

For the study of the growth rate of ice crystals, the apparatus was placed in a chest-type freezer so that the lower half of the Plexiglas chamber was in the freezer air at a temperature of about −15°C. The chamber was illuminated by the same light source with the help of a pair of mirrors.

A glass slide carrying precooled silicone oil—a in a circular cavity 18 mm in diameter and 0.8 mm deep was inserted in a slide holder and set in the bottom part of the cloud chamber. When temperature equilibrium had been established between the slide and the chamber, the fog was seeded, and after removal of a sheltering plate, the ice crystals were allowed to fall on the exposed silicone oil for a known period of time. The slide holder was then taken out of the cloud chamber and a precooled, silicone oil coated cover glass was placed on the crystal-carrying silicone oil. By this operation, although growing ice crystals are received with supercooled fog droplets, the crystal growth is so slowed that no observable growth takes place within a short observation period. The observation was made on a freezer-cooled microscope stage.

Direct readings and analysis of the photomicrographs were used to measure the ice crystal dimensions which, in the case of single ice crystals, were the values along the a and c axes. Since the ice crystals usually rotated slowly in the liquid, they could be observed in many directions. In order to obtain accurate values of ice crystal mass, the crystals were melted by a puff of hot air over the cover glass and the diameter of the produced droplets measured directly or photomicrographically (Fig. 4). The hot air was produced by passage of air through a copper tube 2 mm in inside diameter attached to a heater. Since the density of the silicone oil is higher than that of ice, the ice crystals in the oil slowly rise toward the cover glass. If one waits too long, the ice crystals reach the cover glass and the melting results in spherical caps of attached water on the glass instead of freely suspended spheres. However, such hemispherical water droplets can easily be distinguished by the appearance of the rims. The rim of a free spherical drop looks darker in the bright background due to the total reflection of the transmitted light.

Measurements were made for time intervals of 15–20, 25–30, 35–40, and 45–50 sec after seeding at temperature intervals of about 1.5–2°C.

3. Results and discussion

a. Ice crystal habit

The ice crystals grown in the cloud chamber showed marked habit changes, photomicrographs of the crystals being shown in Fig. 5. Although typical single ice crystals are shown in Figs. 5a–I, polycrystalline particles (Figs. 5m–n, and the left part of Fig. 4) were rather common and in particularly high concentration.
at low temperatures. Only at temperatures near the melting point did the single crystals become dominant.

The observed changes of ice crystal habit were the same as those obtained by aum Kame et al. (1951) with CO₂, AgI, CdI₂ or CsI seeding in a room-size cold chamber and those by Mason (1953) with dry ice seeding in a 2-ft³ cold box, except in a temperature range between −19 and −20°C. Since our method allowed observation of the formed ice crystals from almost all directions, capped columns or tsuzumis were found instead of stellar or dendritic plates. An ice crystal tends to fall and eventually lands in a position of maximum aerodynamical resistance. Aum Kame et al. applied direct photomicrography to the ice crystals collected on glass plates and Mason used Formvar solution for replication. Such methods allow ice crystal observation from only one direction, and it is possible that they overlooked the tsuzumis crystals.

To apply these results to the real atmosphere, the effect of atmospheric pressure has to be considered (Kobayashi, 1958). A good summary of the ice crystal habits and their classification in the real atmosphere is available (Magone and Lee, 1966).

Fig. 6 gives the size distribution of fog droplets in the cloud chamber. The concentration of fog droplets was between several hundred and 1000 cm⁻³.

### b. Fall velocity

The results of the fall velocity measurements are shown in Fig. 7. Within about 12 sec after seeding, the fall velocities of all the ice crystals were more or less the same regardless of the temperature of the supercooled fog. This appears to be the period just before marked growth habit differences start to appear.

After this initial 12-sec period, the velocities increased linearly with time, at least for the next 60 sec, such that

\[ u \approx 0.9 + C_1(t - 12), \]

where \( u \) is the fall velocity, \( C_1 \) the slope, and \( t \) is the time.

The terminal velocity of a particle falling through air is given as

\[ u = C_2 \frac{\rho - \rho_A}{\eta} \left( \frac{C_D \text{Re}}{Re} \right), \]

where \( C_2 \) is a constant, \( \rho \) and \( \rho_A \) the densities of the particle and the air, \( \eta \) the dynamic viscosity, \( g \) the gravitational acceleration, \( A \) the cross-sectional area which the particle presents to the flow, \( C_D \) the drag coefficient, and \( \text{Re} \) the Reynolds number; \( (C_D \text{Re})_0 \) denotes the quantity for very small particles. The drag
The coefficient is expressed as

$$C_D = \frac{mg}{\frac{1}{2} \rho_A u^2 A},$$

where $m$ is the mass of the particle, and

$$\text{Re} = \frac{ud \rho_A}{\eta},$$

where $d$ is the diameter of the sphere which just encloses the ice particle.

At ground level,

$$u \propto \frac{(C_D \text{Re})_0}{(C_D \text{Re})},$$

Using the expressions

$$m \propto t^1,$$
$$A \propto m^1,$$

which we shall develop later, (5) becomes

$$u \propto \frac{(C_D \text{Re})_0}{(C_D \text{Re})},$$

Since $u$ is experimentally linear between 12 and 70 sec after seeding, $(C_D \text{Re})_0 / (C_D \text{Re})$ must be constant within this interval, and we have

$$C_D \propto \frac{1}{\text{Re}},$$

which is typical for Stokes particles.

In order to apply these data to the real atmosphere, the above relations have to be considered together with the mass growth rate. For Stokes particles, from (3), (4) and (8), we have $u \propto \eta^{-1}$ and $\eta \propto T^{-1}$, $T$ being the temperature; whereas, according to Cornford (1965), the corresponding relation for precipitation particles, which are no longer in Stokes range, is $u \propto \rho_A^{-1}$.

Fig. 8 shows the fall velocity of ice crystals at various temperatures 45–50 sec after seeding. There are two plateaus, one between −5 and −8°C and the other between −10 and −18°C. The former appears due to the single or polycrystalline hollow prism and the latter due to the dendritic structure. As we shall see both of them correspond to the observed growth rate peaks.

c. Growth rate

The observed mass growth rates of ice crystals are plotted on a log-log scale in Fig. 9. The growth rates fell reasonably well on straight lines such that

$$\log_{10} m / \log_{10} \omega = 1.5, \text{ or } m \propto \omega^1,$$

except for the interval from −18.0 to −19.5°C. This is the relation (6) referred to earlier.

Around an ice crystal growing from the vapor, a steady-state diffusion of water vapor and a steady-state thermal conduction are established in some short period of time, i.e.,

$$\nabla^2 \rho_w = 0,$$
$$\nabla^2 T = 0,$$

where $\rho_w$ is the density of the water vapor.
Fig. 7. Fall velocity of ice crystals at various temperatures vs time after seeding.

Considering the steady-state flow of heat and vapor, the rate of ice crystal growth by vapor diffusion is currently expressed as

\[
\frac{dm}{dt} = 4\pi C f_1 f_2 (S - 1) \left\{ \frac{L^2 M}{KRT^2} + \frac{RT}{DM_p} \right\},
\]

where \( C \) is the capacity factor for the crystal, \( f_1 \) the ventilation factor of the crystal in the air flow, \( f_2 \) the correction factor of the vapor field to that of super-cooled fog (Marshall and Langleben, 1954), \( S \) the saturation ratio, \( L \) the latent heat of sublimation, \( M \) the molecular weight of water, \( K \) the thermal conductivity.

Fig. 8. Fall velocity of ice crystals vs temperature (45-50 sec after seeding).

\(^2\) Although Hallett (1965) included the accommodation coefficient into a growth rate equation as a proportional factor, it does not appear to control the rate in such a simple manner (cf. Rooth, 1957).
of air, $R$ the gas constant, $D$ the diffusion coefficient, and $p_s$ the saturation vapor pressure over a plane surface of ice at the ambient temperature $T$. The ventilation factor may be taken as

$$f_1 = 1 + 0.22 R e^{1/2}.$$  \hspace{1cm} (12)

Within the range of present study, this factor increases the growth rate at the most by only 13%; it was therefore neglected. For the correction factor, we have

$$f_2 = 1 + kr,$$  \hspace{1cm} (13)

where

$$k = (4\pi \sum_{i} r_i)^{1/4},$$  \hspace{1cm} (14)

and $r$ and $r_i$ are, respectively, the radii of the ice crystal and the fog droplet. The increase of the growth rate by this factor is at the most 5% here and was also neglected.

Since $C \propto d \propto m^1$, and assuming other factors are all independent of $m$ and $t$, Eq. (11) integrates into the expression (6).

The rather deviating behavior of the growth plot between $-18$ and $-19^\circ C$ appears mainly due to the enhancement of the $C$ term in Eq. (11) in the mid-course of the growth change from column to tsuzumi crystals or from combined prisms into radiating assemblages of tsuzumi type dendrites.

Fig. 10 shows ice crystal mass after known time periods as a function of temperature. The vertical lines for the data 45-50 sec after seeding show the range of $\pm \sigma$, the standard deviation, from the mathematical means of the observed crystal masses.

At first glance, one notices a small peak at around $-6.5^\circ C$, which appears to be the same as noted by Hallett (1965) and Todd (1964), although their observations were, respectively, at $-4$ and $-5^\circ C$ with the peak being sharper. In the mass plot of single crystals shown in Fig. 11, the same peak may be seen. The observed small peak is in the temperature range where hollow prisms appear (cf. Fig. 5) and the temperature matches well with that of the apparent density minimum in Fig. 12, the density values being esti-
Fig. 11. Mass of single ice crystals vs temperature (45–50 sec after seeding). The broken line shows the mass of a spherical ice crystal obtained by integration of Eq. (11) for 47.5 sec after seeding at an environmental pressure of 1000 mb and $f_s/f_0 = 1$.

Estimated by dividing the mass of melted ice crystal in Fig. 11 by the apparent volume of ice crystal obtained from Fig. 13. It is clear that this peak may be attributed to the growth habit of a hollow prism at that temperature. Such a structure is needed to attain a faster mass growth rate with rapid vapor diffusion toward the ice crystal of larger surface area (higher capacity) and rapid dissipation of the released latent heat from the crystal.

Another feature found with the measurement of the mass growth rate is the shift of the highest peak toward colder regions as the crystals grew. At 45–50 sec after seeding, the peak stays at around $-17$ to $-18$C. The possible reason for this, as already partly discussed,
appears to be the nucleation and growth of tsuzumi crystals or their radiating assemblages. When the latter start forming from columns, the capacity is considered to increase abruptly. Since this is the temperature zone where the fastest growth takes place, the latent heat release is rapid. The released heat warms both the local system around the crystal and the whole system. If the temperature of the initial system is slightly below that of the habit change, the tsuzumi crystal would be formed along the direction of warming; however, the estimated warming appears slightly too small for such an explanation.


For a spherical ice crystal, \( C = r \), and under conditions where \( f_1 \) and \( f_2 \) can be neglected, the growth equation (11) integrates into

\[
m = -\frac{8\pi}{3\rho} \left\{ \left( \frac{2}{S-1} \right) \left( \frac{L^2 M}{RT^2} + \frac{RT}{DM_p} \right) \right\}^{\frac{1}{4}}.
\]

Eq. (15) is plotted as a broken line in Fig. 11 for 47.5 sec after seeding at an environmental pressure of 1000 mb with \( f_1 f_2 = 1 \). As can be seen, the calculated mass of the spherical ice crystal is larger above and smaller below –13°C compared with the observed value for single ice crystals.

For miscellaneous shaped ice crystals, if the shape and the apparent density of the ice crystal remain approximately the same\(^4\) for the later part of their growth, Eq. (15) may be written as

\[
m = m_{cr}\left( \frac{C}{C_i} \right)^{\frac{1}{4}} \left( \frac{\rho}{\rho_{AP}} \right)^{\frac{1}{4}},
\]

where \( m_{cr} \) is the mass of compact spherical ice crystal, \( (C/C_i) \) the “shape factor,” is the ratio of the capacity factors between variously shaped and spherical ice crystals of the same mass\(^5\), and \( \rho_{AP} \) the apparent density of the ice crystal.

At −10°C and 47.5 sec after seeding, the calculated mass of the prolate ice crystal, which has the same apparent density as given in Fig. 12 and the same \( a \) and \( c \) ratio as shown in Fig. 13, is about 2.3 times larger than the actual observed value. At −15°C and 47.5 sec after seeding, the calculated mass of oblate ice crystals is about 3.1 times larger than the observed value. These calculations were, of course, made under the assumption that \( f_1 f_2 = 1 \); the ratios could, therefore, have been even larger if \( f_1 \) and \( f_2 \) were considered.

Thus, current theory has failed to describe the observed data. As one can easily notice, the capacity term in the growth equation originally came from the surface area term of the equations of diffusion and heat conduction. Since a sphere has the smallest surface area for the volume, the ice crystals of various shapes should have an advantage in growth rate over a spherically shaped ice crystal. On the contrary, the observed mass growth rates of actual ice crystals are, at warm temperatures, smaller than those of spherical crystals by more than 50%. Neither experimental error nor any known factors can account for such a large discrepancy.

\(^4\) For the temperature range –18 to –19°C or for long runs, this assumption does not hold.

\(^5\) Normalization of the capacity factor will be discussed elsewhere. The capacity values for different shapes may be obtained from the work of McDonald (1963).
In order to fill this gap, a new factor, \( f_s \), needs to be introduced into Eq. (11), i.e.,

\[
\frac{dm}{dt} = 4\pi C f_s f_\alpha f_\beta (S-1) \left\{ \frac{L^2 M}{KRT^2} + \frac{RT}{DM p_s} \right\},
\]

where \( f_s \leq 1 \) and is a function of temperature. Probably, it is a function of the sublimation coefficient on basal and prism planes and controls the vapor pressure over ice under growing conditions.

4. Concluding remarks

As we have seen, the present experimental study and analysis have revealed some new features concerning the growth of small ice crystals which were previously unconfirmed or overlooked. The main points are summarized below:

1) For approximately the first 12 sec, the fall velocities of ice crystals growing at various temperatures under water saturation differed little. For the next 60 sec, however, they behaved entirely as Stokes particles, such that, \( u \propto t \).

2) Two plateaus of fall velocities of ice crystals were observed at temperature intervals between \(-5\) and \(-9^\circ C\), and \(-10\) and \(-18^\circ C\).

3) For the first 50 sec of growth, \( m \propto t^3 \) except from \(-18\) to \(-19.5^\circ C\).

4) A new small growth rate peak was detected at \(-6.5^\circ C\).

5) The largest growth rate peak was observed to shift toward the colder region as the ice crystals grew. About 47.5 sec after seeding, the peak position was at around \(-17.5^\circ C\). It appeared to be attributable to the formation of tsuzumi crystals or their radiating assemblages.

6) The two temperatures where each peak of the growth rate occurred matched the apparent density minima of the formed crystals.

7) The current theory failed to describe the mass growth rate of the crystal. From the gap, the existence of an unknown factor was detected and introduced into the rate equation.

As a new approach, the present experimental technique has proved to be quite satisfactory. However, considering the necessity and importance of experimental data of this kind for the detailed understanding and prediction of ice phase processes in the cloud, further and more accurate studies are urgently needed in a longer growth time zone. Specific studies, both experimental and theoretical, on the newly detected factor are also necessary.

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