Experimental Researches on the Snow Crystal Habit and Growth by Means of a Diffusion Cloud Chamber

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(Manuscript received 31 May 1957)

Abstract

With the object of examining whether or not Marshall and Langleben's theory on the snow-crystal habit is in agreement with experimental fact, we carried out a series of experiments on artificial snow crystals by means of a diffusion cloud chamber where the cloud droplets had been previously removed. The ambient vapour density was measured by a "drop hygrometer" method.

It was found (1) that the snow-crystal habit is primarily determined not by the excess of the ambient vapour density but by the ambient temperature; (2) that snow crystals produced in aerosol-free air show tendencies to grow either in sheath form or in dendritic form with sector-like structure; (3) that the trace of foreign vapours e.g. silicone or acetone remarkably affects the snow crystal habit.

1. Introduction

Marshall and Langleben⁽¹⁾ interpreted the experiments of Nakaya and his coworkers⁽²⁾ as evidence in favour of the view that the snow crystal habit is determined principally by the "effective" excess of ambient vapour density over that in equilibrium with the growing ice crystal.

Nakaya defined the "supersaturation"—the ordinate of the so-called Nakaya diagram—as the percentage ratio of total water content (vapour plus tiny water droplets) to the vapour content in equilibrium with ice⁽³⁾, which he derived from the measurement by the use of a convective-mixing type of snow-making apparatus.

On the arbitrary assumption, however, that Nakaya's supersaturation was the one relative to *water* equilibrium, Marshall estimated the true vapour excess over water equilibrium from Nakaya's experimental data, and obtained constant loci of effective vapour excess over the growing ice, which well fitted in with Nakaya diagram. Ac-

cording to his argument, changes in crystal type occur first to edge growth and secondly to corner growth, when this vapour-density excess is sufficient to overcome the inhibitions which must exist inasmuch as the edges and corners have higher saturation vapour-densities than the flat faces of the crystal.

In spite of the apparent coincidence with Nakaya's experimental data, Marshall's result must be subjected to critical examination, since his theory is based, as mentioned above, on the assumption that Nakaya's supersaturation should be relative to *water* equilibrium, while, according to Nakaya, it was not actually the case.

Marshall and Gunn⁽⁴⁾ tried to measure the ambient vapour density by means of a "drop-hygrometer" and observed some crystal growth in the cold chamber. It seems, however, that no definite evidence has been obtained from their experiment to justify Marshall's theory.

The present investigation of artificial snow crystals by the use of a diffusion cloud

chamber is intended to examine the relation between the crystal forms and the ambient temperature and the vapour density excess over the crystal and to clarify the influence of tiny cloud droplets and several kinds of vapour traces on the snow-crystal habit and growth. In this paper, the habit of snow-crystals which are formed in the diffusion field free of cloud droplets is mainly described.

2. Apparatus and experimental procedure

The snow-making experiment was carried out in a diffusion cloud chamber (Photo. 1). The main part of the chamber is constructed of double walls of transparent Acrylite*

(inside measure: 18 cm × 18 cm × 22 cm) provided with three pairs of glass windows for the photomicrographic observation. The bottom made of thick copper walls on which the Acrylite walls are to be placed is immersed in a cold brine-bath. The temperature of the brine bath (T_b) is maintained at any desired temperature down to -40°C within the accuracy of $\pm 0.5^\circ\text{C}$, by the aid of a 1/4 H.P. refrigerating machine charged with Freon 22. The top of the chamber is covered with a number of sheets of wetted gauze, the temperature of which (T_w) can be regulated by electric heating up to 40°C . By adjusting T_b and T_w , we are able to set up any temperature profile in the chamber.

Fig. 1

- A; Acrylite double wall
- B; brine bath
- C; copper wall
- D; black chiffon-velvet
- E; water reservoir
- F; cooling pipe
- G; wetted gauze
- H; Heater
- I; thermal insulator
- J; thermocouple
- K; camera with extra-extension tube
- L; light beam
- M; manipulator
- N; guide tube
- O; glass window
- P; fine glass-supporter
- Q; rabbit hair
- R; fine thermometer
- S; snow crystal

In the preceding paper⁽⁵⁾, we pointed out that the snow crystals produced in the water-droplet-free air always grew in the form of sheath or needle at any temperature level down to -40°C . Thereafter, we ascertained that the tendency to grow in the sheath or needle-like form is due to the influence of vapour trace of the Dri-film

which was used for the hydrophobic coating on the Acrylite walls. For the sake of the present experiment, the Acrylite walls were reconstructed and special precautions were taken in avoiding the contamination of the chamber by foreign vapour. Trichloroethylene used for the chilled bath in the preceding experiment was replaced by CaCl_2 brine.

* Acrylic-acid resin.

Within the chamber, stratification of air was found to be very stable. When the chamber, once operated, was left intact half a day long, practically all the supercooled cloud-droplets were rained out and a considerably higher supersaturation in only vapour phase was realized.

In order to get isolated snow crystals, as Nakaya did, we used a fine rabbit hair which was previously fixed to one end of a fine glass supporter and thoroughly dried in a vacuum desiccator, and then put into the chamber through the glass guide fixed to the upper lid of the chamber. It was set up near the same level as the junction of Acrylite wall with the copper box where the temperature gradient was the steepest, consequently the supersaturation was the highest. The camera with an extra-long extension tube was set on a horizontal microscope stage so as to be on a level with the hair. The other end of the glass supporter was fixed to a mechanical device with a vernier scale, by which the rabbit hair could be moved up and down.

Since the crystals which sporadically appear along the whole length of the hair (2~3 cm) grow within the air layer where the temperature gradient is the greatest (ca. $3^{\circ}\text{C}/\text{cm}$), the measurement of accurate location of each crystal was necessary for the precise determination of T_a , the temperature of the air where the crystal is formed. The temperature profile near the crystal-growing region was determined by the use of four thermocouples at intervals of ca. 2 cm and three fine alcohol-thermometers as supplement.

In a few minutes or so after the rabbit hair was set in the chamber, several germs of snow crystals began to appear. The successive stages of crystal growth were observed through microscope and photographs were taken at proper intervals.

When the crystal had grown up to an appropriate size, the crystal was moved upward to the vicinity of the wetted gauze

where the air temperature was above 0°C . After the snow crystal melted to form a spherical water drop, it was put back to the place where it had been, and the growing or evaporating process of the drop was observed at proper time intervals. From this change of drop diameter, the ambient vapour density was calculated by Marshall and Gunn's formulae⁽⁶⁾.

In our experiment, the snow crystals were formed in 2~3 cm range centering around the camera focusing level, so that it was necessary to determine the vapour-density as well as the air-temperature profile by the measurements at two different levels at least. The vapour density at the camera level was actually measured, while that of the other level was estimated on the following assumption, on account of technical difficulty. Since the inner walls of the copper box are always covered with frozen droplets sprayed by an atomizer, the air within the box is maintained approximately at ice saturation. On the lowest thermocouple set within the wall frost-crystals are scarcely observed, while on the other three thermocouples there appears frost. Therefore, it may well be supposed that the air at the lowest thermocouple level is just at ice saturation. The gradient of vapour density is then assumed to be linear between these two levels, and the supersaturation s at each level is determined from the temperature profile previously given.

The abrupt change in condition in the course of crystal formation can easily be brought about by changing the position of crystals vertically. This is one of the reasons why the diffusion cloud chamber is to be preferred for the investigation of the changes in growth type of snow crystals. In our chamber, however, the temperature- and vapour-density-profile cannot be changed *independently*.

3. Crystal forms as a function of T_a and s

The values of s and T_a being thus determined for each crystal formed under various conditions, we are able to represent the form of crystals as a function of T_a and s .

Several examples of snow crystals produced on the rabbit hair are shown in Photos. 2~7. The stepwise changes in growth type caused by changing the position of the crystal up and down in our chamber are illustrated in Photo. 8 and Fig. 2. Photo. 9 and Fig. 3 also show the process of gradual development from a sector to a dendrite with increasing ambient temperature and vapour-density excess.

The result obtained in our present experiment is plotted in T_a - s diagram, as shown in Fig. 4, where s represents the supersaturation with respect to *ice*. The dotted lines in Fig. 4 indicate the boundary which was experimentally determined by Nakaya. The general aspect of snow crystal habit is in conformity with Nakaya diagram; es-

pecially good agreement is observed in regard to the temperature variation of the crystal type. But Nakaya's lower boundary with respect to the supersaturation axis for each crystal-type region is found to be considerably lower than that of our data. We have been informed that the degree of supersaturation obtained by Nakaya was somewhat underestimated owing to the errors inherent to his method of measurement. If allowances are made of this fact, the result represented in Fig. 4 shows that the ordinate s in Nakaya's T_a - s diagram means, without doubt, the supersaturation with respect to *ice*.

In Fig. 4 we see that the air-temperature range for each crystal-type region is clear-cut while the values s for each region are widely scattered. It may, therefore, be considered that the principal factor controlling the snow crystal habit should be the air temperature. As can be seen also from Fig. 4, crystal germs, with the exception of those marked with +, grow on the rabbit hair only when the condition s is attained, which is above (or slightly below) saturation with respect to water, over the whole range of temperature down to -35°C . The marks + show those snow crystals which were brought into the spot from other working region above water saturation where the germs had started to grow. Thus, so far as the development of crystals on a rabbit hair is concerned, it might be considered that the crystal germs were produced by the freezing of liquid water which condensed on the rabbit hair through capillary condensation.

4. Influences of tiny water droplets

In spite of good agreement between Nakaya's and our experimental data there surely exist some significant differences in growth type of crystals, which probably depends on whether the tiny cloud droplets are present (as in the case of Nakaya's convection-type apparatus) or not (as in the case

Fig. 4. T_a - s diagram, showing the conditions for the formation of various shapes of snow crystals which develop in the aerosol-free air.

of our diffusion chamber). Conspicuous feature of crystal formation in our chamber consists in the tendency to the growth of sheath-type crystals and dendrites with sector-like structure.

In our diffusion cloud chamber the sheath-type growth (Photo. 10) is predominantly observed over wide temperature range excepting the plate-growth region ($-12 \sim -18^\circ\text{C}$). It has been ascertained, as described in our previous paper, that the tendency to growth in c -axis is accelerated under the influence of vapour trace of silicone and the like, but it seems undoubted that the tendency is still present even in the absence of foreign vapour, only if the cloud droplets are excluded.

Fig. 5. Distinction between dendritic and sector crystals

- a) true dendritic crystal
- b) dendritic crystal with sector-like structure.

Nakaya distinguished the crystals, which had been arbitrarily called dendrite, into two kinds; namely "dendritic" and "sector" form according to whether the branch tip has round figures (dendritic) or straight-line structure (sector). All the dendritic crystals produced in our apparatus belong to the sector form according to Nakaya's classification. As Nakaya⁽⁷⁾ has emphasized, the tiny water droplets or aerosols in contact with the crystal surface are likely to play an important role for the "dendritic" growth, and his recent observations of snow crystals in the expedition to the Mauna Loa are also in support of our experimental results⁽⁸⁾.

5. Snow crystal habit in the diffusion field of water vapour

The condition in our diffusion chamber under which the artificial snow crystals are produced is quite similar to that assumed in Marshall and Langleben's theoretical treatment. So it is interesting to compare our experimental result with their theory. Marshall and Langleben's loci of constant $\rho_v - \rho_i$ ⁽⁹⁾ bound the regions favourable for the formation of dendrites, plates, columns and needles in Nakaya diagram.

As pointed out in the foregoing section, however, Marshall's evaluation is based on the faulty assumption with regard to Nakaya's supersaturation. In our experiment employing the diffusion cloud chamber, ambient vapour-density ρ is determined by drop-hygrometer method, so that $\rho_v - \rho_i$ is easily obtained by the graphical method⁽¹⁰⁾, since $\rho = \rho_v$ in our case where no cloud droplets exist or $\rho_c = 0$. It will be readily understood from our experimental data shown in Fig. 6, where the ordinate represents $\rho_v - \rho_i$ in place of s in Fig. 4, that the snow crystal habit cannot be interpreted by Marshall's diffusion theory.

Fig. 6. $T_a - (\rho_v - \rho_i)$ diagram, transformed from Fig. 4. Dotted lines indicate Marshall's boundary of crystal-type domains.

The result leads to the following conclusion as to the snow crystal habit in diffusion field of water vapour: The principal factor controlling the snow-crystal habit is the

temperature of the air where the crystal grows, while the water-vapour excess secondarily controls the growth rate, hence the type of growth in each temperature range. The manner in which the crystal habit varies with temperature is represented clearly in Table 1.

Table 1

Temperature range	(a ₁) above -4°C	(c ₁) -4~- -10°C	(a ₂) -10~- -20°C	(c ₂) below -20°C
Direction of preferred growth	a	c	a	c
Crystal form	plate	scroll sheath needle	thick- plate sector dendrite	sheath side- extension

The hexagonal plates produced at temperature above -4°C were omitted from Fig. 4, because the ambient vapour density could not be estimated. It was frequently observed that long needles grow horizontally inwards or slightly upwards from the Acrylite wall. Usually the end of the needle became thick in the form of assemblages of hollow column. There was only one case, however, where the thin plates were observed to develop at the ends of the needles which extended slightly upwards from the wall (Photo. 11). The temperature of the air layer where the plate crystals were formed was surely higher than that at the root of the needles, being -5°C.

Table 1 indicates that a remarkable sequence of transformation, plates—prisms—plates—prisms, involving changes from preferred growth along the principal *c*-axis as prisms to preferred growth along the secondary *a*-axis as plates, or vice versa, occurs as the temperature is lowered from 0°C to about -35°C. These changes of crystal habit with temperature are very similar to those observed in the experiments conducted by aufm Kampe et al.⁽¹¹⁾ and Shaw and Mason⁽¹²⁾. Their results as well as ours are summarized in Fig. 7.

Fig. 7. Representation of snow-crystal habit manifested in aerosol-free air, as compared with Nakaya, aufm Kampe et al. and Shaw and Mason's experimental data.

The condition that the snow crystals grow only through diffusion and sublimation of water vapour in aerosol-free air belongs, it is true, to a simplified and idealized case, but it must be considered fundamental. It will afford an important basis for the future investigation, in which the influence exercised by cloud droplets, foreign vapour traces and also barometric pressure must be taken into account.

6. Influence of the foreign vapour traces

As already mentioned, the sheath-type growth observed in the last year's experiment was due to the influence of the Dri-film vapour. This was established by following observation:

As we at first suspected that the needle and sheath-like crystals which appeared in the aerosol-free air might have been formed under the influence of trichloro-ethylene vapour which was used for cooling bath, the effect of the vapour was thoroughly examined under various conditions of T_a and s . But no remarkable influences were observed, except for frequent occurrence of thick plates within the plate-growth region.

In the meanwhile the effect of silicone vapour was suggested by Dr. U. Nakaya. When it was ascertained that the dendritic or sector form of the snow crystal had been

produced on a rabbit hair, a fine glass rod coated with a drop of Dri-film was put into the chamber. Then, after a while, sheath-type needles were observed to grow from the basal plane passing through the intermediate state of scroll (Photo. 12). Thereafter, when a rabbit hair was newly set in, only sheath- or blade-type crystals were formed in the whole range of the temperature down to -35°C . The effect was very remarkable and astonishing.

Many kinds of silicone-oil and grease and also several kinds of organic solvent were put to test in order to clarify the influence of

these vapours upon the snow-crystal habit*. Among them silicone oil of low viscosity and acetone proved to be most favourable for the sheath-type growth (Photo. 13) and methyl-phenyl silicone oil yielded a curious form of the crystal (Photo. 14).

The investigation on the influence of vapour trace is now in progress under the guidance of Prof. U. Nakaya. Detailed report will be published in the near future.

* Thereafter the author was informed that Schaefer⁽¹³⁾ and Vonnegut⁽¹⁴⁾ have reported on the influence of organic vapours (acetone, nitric acid, acetic acid, higher alcohol and silicone, etc.) upon the form of ice crystals produced by cloud seeding in the laboratory.

References

- 1) **Marshall, J. S. and M. P. Langleben**, 1954: A theory of snow-crystal habit and growth, *J. Meteor.*, **11**, 104-120. —2) **Nakaya, U.**, 1951: The formation of ice crystal. *Compendium meteor.*, Boston, *Amer. Meteor. Soc.*, 207-220. —3) **Nakaya, U.**, 1954: *Snow Crystals, natural and artificial*. Harvard Univ. Press. 249. —4) **Marshall, J. S. and K. L. S. Gunn**, 1955: *A first experiment on snow crystal growth*. A paper presented at the conference on the physics of cloud and precipitation particles, Woods Hole, Mass. Sept. 1955. —5) **Kobayashi, T.**, 1956: Experimental researches on the snow crystal habit and growth by means of a diffusion cloud chamber (Preliminary report). *Low Temp. Sci. Ser. A*, **15**, 1-11 (in Japanese). —6) Reference (4). —7) **Nakaya, U.**, 1955: Snow crystal and aerosols. *Journ. Fac. Sci., Hokkaido Univ. Ser. II*, **4**, 341-354. 8) — **Nakaya, U., J. Sugaya and M. Shoda**, 1957: Report of the Manua Loa expedition in the winter of 1956-57. *Journ. Fac. Sci. Hokkaido Univ. Ser. II*, **5**, 1-35. —9) Reference (1), Fig. 7, 109. —10) Reference (1), Fig. 3, 107. —11) **aufm Kampe, H. J., H.K. Weickmann and J. J. Kelly**, 1951: The influence of temperature on the shape of ice crystals growing at water saturation. *J. Meteor.*, **8**, 168-174. —12) **Shaw, D and B. J. Mason**, 1955: The growth of ice crystals from the vapour. *Phil. Mag.*, **46**, 249-262. —13) **Schaefer, V. J.**, 1949: The formation of ice crystals in the laboratory and the atmosphere, *Chem. Rev.*, **44**, 291-320. —14) **Vonnegut, B.**, 1948: Influence of butyl alcohol on shape of snow crystals formed in the laboratory. *Science*, **107**, 621-622.

No. 1.

The diffusion cloud chamber

No. 2.

Needles;
 $T_a = -5.1^\circ\text{C}$
 $s = 114\%$ over ice
 $t^* = 2\text{ hr } 30\text{ min.}$

$\times 7.7$

No. 3.

Sheath-type;
 $T_a = -6.9^\circ\text{C}$
 $s = 124\%$ over ice
 $t = 102\text{ min.}$

$\times 14$

No. 4.

Column;
 $T_a = -27.2^\circ\text{C}$
 $s = 129\%$ over ice
 $t = 16\text{ hr } 10\text{ min.}$

$\times 20$

No. 5.

Thick plate;
 $T_a = -15.4^\circ\text{C}$
 $s = 113\%$ over ice
 $t = 15\text{ hr } 20\text{ min.}$

$\times 12$

No. 6.

Sector (Tsuzumi-type); $\times 22$
 $T_a = -14.2^\circ\text{C}$
 $s = 115\%$ over ice
 $t = 56\text{ min.}$

No. 7.

Sheath-type and columns with
extended side plane; $\times 11$
 $T_a = -29.0^\circ\text{C}$
 $s = 131\%$ over ice
 $t = 16\text{ hr } 50\text{ min.}$

$\times 11$

* The time taken for the development of the crystal to the stage shown in the photographs, measured from the moment the hair was exposed to the supersaturated air.

No. 8. Successive stages in the course shown in Fig. 2

a) Needles; growing thick at the ends

b) develop into sector

c) grow into scroll

d) needles start. 1 mm

Fig. 2.

No. 9. Successive stages in the course shown in Fig. 3.

a)—b) Sector-form

c) edge growth starts d) develops into dendritic form

e) Dendritic crystal radially developed. 1 mm

Fig. 3.

No. 10.

Multi-layer structure of sheath-type crystal $\times 20$

No. 12.

Influence of Dri-film vapour; Sheath-type needles is beginning to grow from the basal plane of the dendritic crystal. $\times 8.6$

No. 11.

Hexagonal plates formed at the ends of needles. $\times 4.4$
 $T_a = \text{above } -5^\circ\text{C}$

No. 13.

Sheath-type crystals produced under the influence of silicone oil (Shin-etsu Silicone, KF. 96, 5 c.s.) $\times 9.9$
 $T_a = -13.5^\circ\text{C}$

No. 14.

A curious form of an artificial snow-crystals produced under the influence of methyl-phenyl silicone oil, (Toshiba Silicone, 0.65 c.s.) $\times 4.8$
 $T_a = -13^\circ\text{C}$