MORPHOLOGY OF ICE CRYSTALS GROWING IN FREE FALL AT THE TEMPERATURES BETWEEN −40 AND −140°C

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***Abstract*:** For the sake of studying the morphology and the growth mode of single snow crystals forming in the polar regions, ice crystals have been grown in free fall at the temperatures between −40 and −140°C and their morphology and growth mode have been studied. As a result, in the temperature range from −40 to −70°C, which seem to arise in the polar regions, single ice crystals are predominantly formed at about −40°C, but below about −50°C, polycrystalline ice crystals are predominantly formed; in the case of single ice crystals below 60 μm, hexagonal plate-like ice crystals are predominantly formed. The size ratio c/a of plate-like ice crystals growing predominantly at −50°C hardly depends on crystal size, but that of column-like ice crystals growing at the same temperature increases with increasing crystal size. In addition, the growth mode of column-like ice crystals growing at −50°C is nearly in agreement with that of diamond dust type ice crystals observed in Antarctica by KIKUCHI and HOGAN (J. Meteorol. Soc. Jpn., **43**, 359, 1979). The growth mode of plate-like ice crystals which may be observed in the polar regions should be investigated, too.

1. Introduction

One of the distinctive features of snow crystals forming in the polar regions is that snow crystals are generally nucleated and grow at lower temperatures than those forming in winter in Japan. In addition, the snow crystals in the polar regions have a tendency to be nucleated and grow at low supersaturation because supercooled cloud droplets are hard to be formed in those regions except for the case of the inflow of cyclonic air masses. Moreover, in the polar regions, because of low temperature, inactive materials will be nucleated as ice nuclei. It has been shown that long solid prisms observed in Antarctica (SHIMIZU, 1963) and rectangular snow crystals observed at Barrow, Alaska (HIGUCHI, 1968) grow by screw dislocation mechanism at the supersaturation near ice saturation (GONDA and KOIKE, 1982a). In addition, it has been shown that the habit of diamond dust type ice crystals observed in Antarctica (KIKUCHI and HOGAN, 1979) depends on the supersaturation in air where they are formed. Except for 20-faceted ice crystals observed in Alaska (OHTAKE, 1970), it is inferred that many of single snow crystals forming at low temperature (below −50 to −60°C) in the polar regions are formed without the process of supercooled cloud droplets.

According to the theory of KURODA and LACMANN (1982) and KURODA (1982), the habit of snow crystals growing at the temperatures below −20°C depends on both temperature and supersaturation. When we exclude ice crystals growing at low supersaturation below 1–2%, it has been pointed out that the result obtained from the theory coincides well with the experimental one (GONDA and KOIKE, 1982a, b). The theory indicates that at such low temperature as in the polar regions, plate-like ice crystals are formed at the supersaturation below about water saturation at any temperatures. The purposes of this article are to investigate experimentally the morphology and the growth mode of ice crystals growing at the temperatures between −40 and −140°C and to clarify the morphology and the growth mode of snow crystals forming in the polar regions.

2. Experimental Procedures

Figure 1 shows a cold chamber to form ice crystals in free fall at the temperatures between −40 and −140°C. The chamber is cooled down to a measured temperature by putting a proper quantity of liquid nitrogen into the (F) and (J) portions. The air temperature in a cylinder (G) is kept constant at various degrees by controlling the quantity of liquid nitrogen to put into the (F) and (J) portions, which is carried out by controlling an electric current to flow into a coil of nichrome wire immersed in a reservoir for liquid nitrogen (not shown in the figure). In order to prevent the heat which will flow into the growth chamber, the portion (E) attached to the outside of the cylinder (G) is evacuated by a vacuum pump, and moreover thick thermal insulator

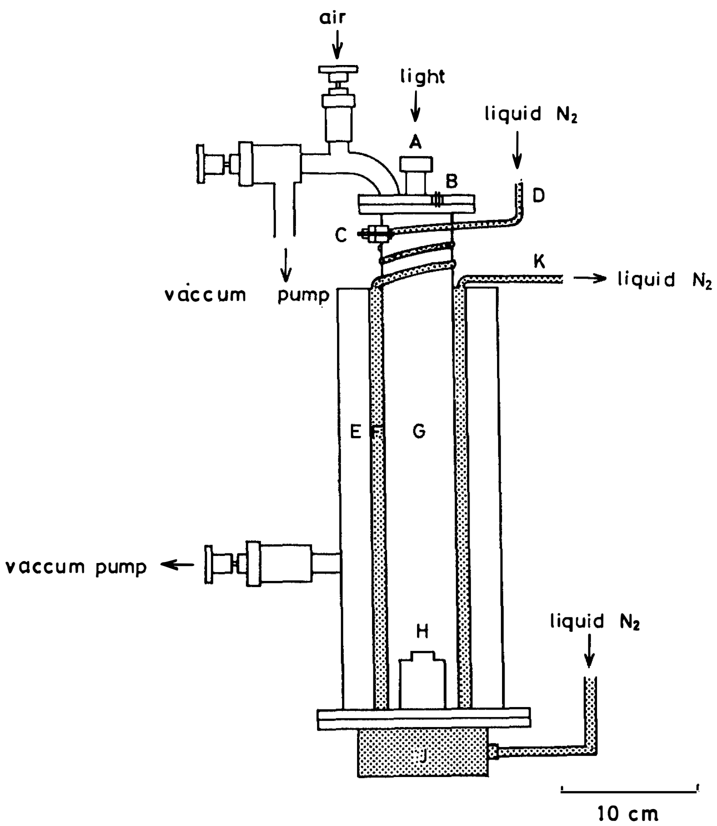

Fig. 1. The cold chamber to form ice crystals at the temperatures between −40 and −140°C.

(not shown) is fastened up to the outside of the vacuum portion (E). By these operations, the temperature gradient along the vertical direction in the chamber is kept at about 0.1°C/cm up to about 40 cm in height from an observation window (H). Then, the temperature in the chamber during the growth of ice crystals is measured at the center of the cylindrical chamber.

Water vapor is supplied by passing an electric current of about 2 amperes for about 7 min to a pipe heater wound with moistened gauze, which is attached at the position of about 15 cm from the top of the chamber. At 1–2 min after the supply of water vapor, silver iodide smoke is generated by passing an electric current of about 6 amperes for about 10 s to a spiral platinum wire which absorbed silver iodide and is attached at the top of the chamber. Prior to the observation of ice crystals, the operations described above are repeated about two times; thereafter ice crystals are nucleated in clean air of 1.0 atm and grown in free fall at various constant temperatures between −40 and −140°C by the same operations as those described above. Though the supersaturation in the chamber is not measured, on the basis of many preparatory experiments and detailed observations of ice crystal, the supersaturation during the growth of ice crystals is estimated to be about 2–3% except for the cases of Fig. 3. Ice crystals that fell on the observation window (H) are photographed by an inverted type microscope through a transmitted light.

3. Experimental Results

3.1. Morphology of ice crystals and ice nucleation

Figure 2 indicates typical examples of plate-like ice crystals with non-hexagonal symmetry grown at various constant temperatures below −40°C. The supersaturation when ice crystals are growing is unknown, but except for ice crystals shown in Fig. 2 it is inferred that the supersaturation is low so as not to produce the corner growth at each corner of ice crystals. As shown in the figure, one of the distinctive features of ice crystals forming at low temperatures is that ice crystals with non-

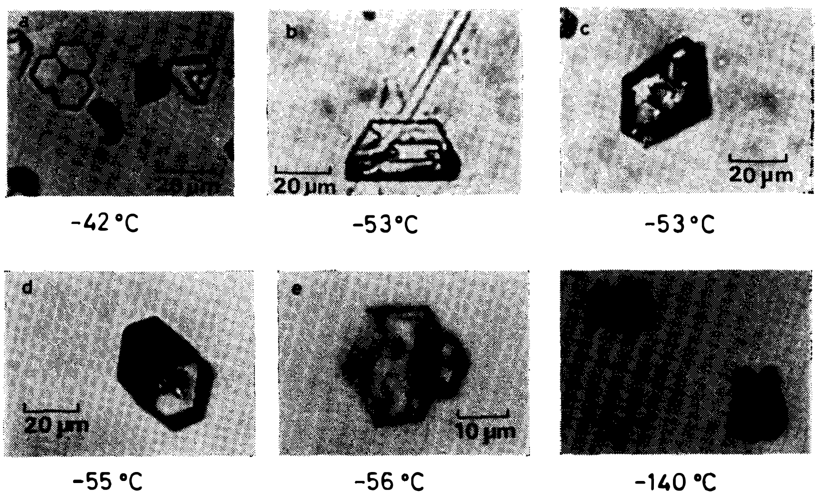

Fig. 2. Typical examples of plate-like ice crystals with non-hexagonal symmetry grown at various constant temperatures below −40°C.

hexagonal symmetry are formed together with those with hexagonal symmetry. Ice crystals represented by (Fig. 2c) and (Fig. 2d) have the same shapes as those observed in Antarctica by KIKUCHI and HOGAN (1979).

Figure 3 shows typical examples of non-hexagonal ice crystals with broad branches and a rectangular ice crystal, which were formed at various constant temperatures below $-40°C$ and relatively high supersaturation so as to produce the corner growth at each corner of ice crystals. However, the growth frequency of ice crystals shown in this figure is very small. Except for the rectangular crystal with well-developed prismatic faces (Fig. 3e), it is seen that a dissymmetric dendrite (Fig 3a), dissymmetric plates with broad branches and their deformed shape crystals (Figs. 3b–3d, 3f) are formed. In the polar regions, ice crystals with such shapes as shown in this figure shall be difficult to observe because in those regions, relatively high supersaturation is hardly produced. In the case of formation of dissymmetric dendrites (Fig. 3a), especially, it is considered to be important that the high supersaturation near water saturation is produced at any temperature.

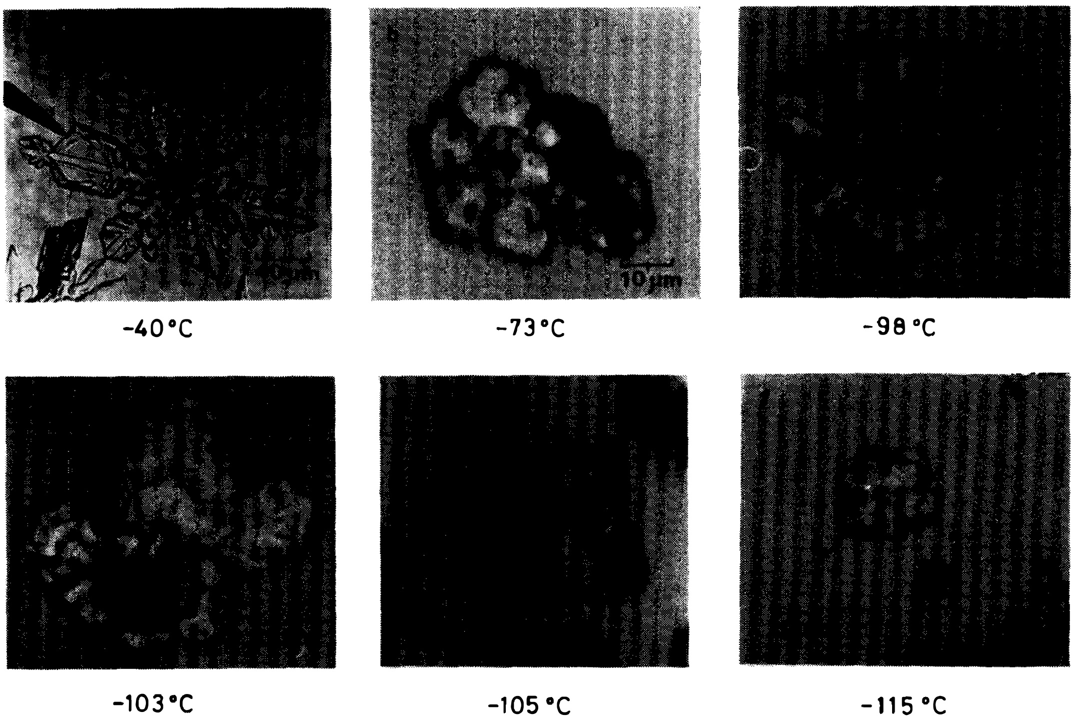

Fig. 3. Typical examples of non-hexagonal ice crystals with broad branches and a rectangular ice crystal grown at various constant temperatures below $-40°C$.

Figure 4 indicates typical examples of columnar ice crystals grown at various constant temperatures below $-40°C$. Though the supersaturation during the growth of ice crystals is unknown, it seems that the ice crystals were formed at considerably low supersaturation as far as we estimate the supersaturation from the external forms of ice crystals. In Fig. 4b, a long solid prism grows at the supersaturation near ice saturation because it grows together with a large number of minute ice crystals. This long solid prism is the similar crystal to that observed in Antarctica by SHIMIZU (1963).

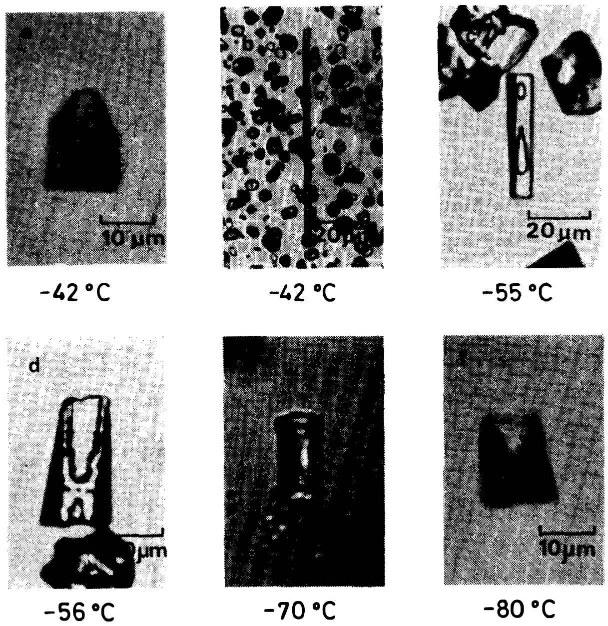

Fig. 4. Typical examples of column-like ice crystals grown at various constant temperatures below −40°C.

A short prism (Fig. 4a) and a relatively long prism (Fig. 4c) are similar crystals to those of diamond dust type ice crystals observed in Antarctica by KIKUCHI and HOGAN (1979). The crystals growing along c-axis with different rates (Figs. 4d, 4f) may be found in polar regions in the future. One of the reasons why the growth frequency of long solid prisms in this experiment is considerably small as compared with that of KOBAYASHI (1965) may be concerned with the fact that screw dislocations are hard

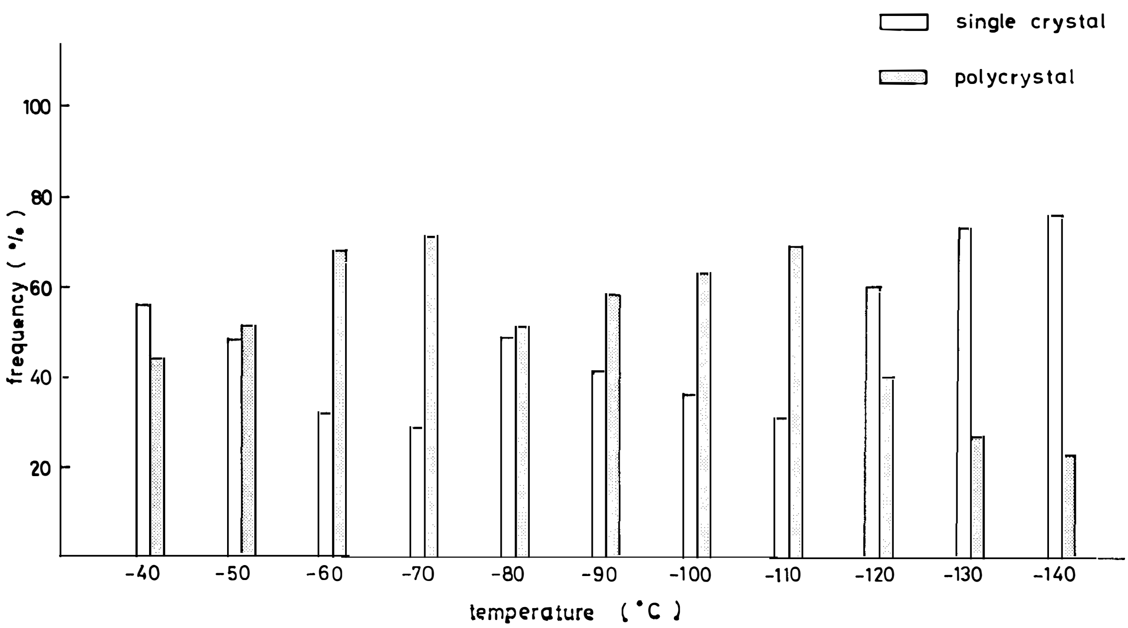

Fig. 5. Temperature variation in the growth frequency of single and polycrystalline ice crystals below about 60 μm in size.

to emerge on the {0001} faces when ice crystals are formed in free fall. Second, it is also considered that the supersaturation during the growth of ice crystals is slightly high and the size of ice crystals observed is small as compared with Kobayashi's experiment (1965).

Figure 5 shows the temperature variation in the growth frequency of single and polycrystalline ice crystals below about 60 μm in size. As shown in a previous paper (Gonda, 1977), at the temperatures above -40°C, single ice crystals grow in high frequency. However, at the temperatures between -50 and -110°C, polycrystalline ice crystals grow in high frequency. This is one of the distinctive features of ice crystals growing at low temperatures. The reason why single ice crystals grow again in high frequency at the temperatures below -120°C is uncertain in the present stage. Figure 6 indicates the temperature variation in the growth frequency of plate-like ice crystals with hexagonal and non-hexagonal symmetry below about 60 μm in size. As shown in the figure, at the temperatures about -60°C, plate-like ice crystals with hexagonal symmetry grow in high frequency. However, at the temperatures between -70 and -140°C, though there are some fluctuations in experimental results, it is seen that the plate-like ice crystals with non-hexagonal symmetry grow in high frequency.

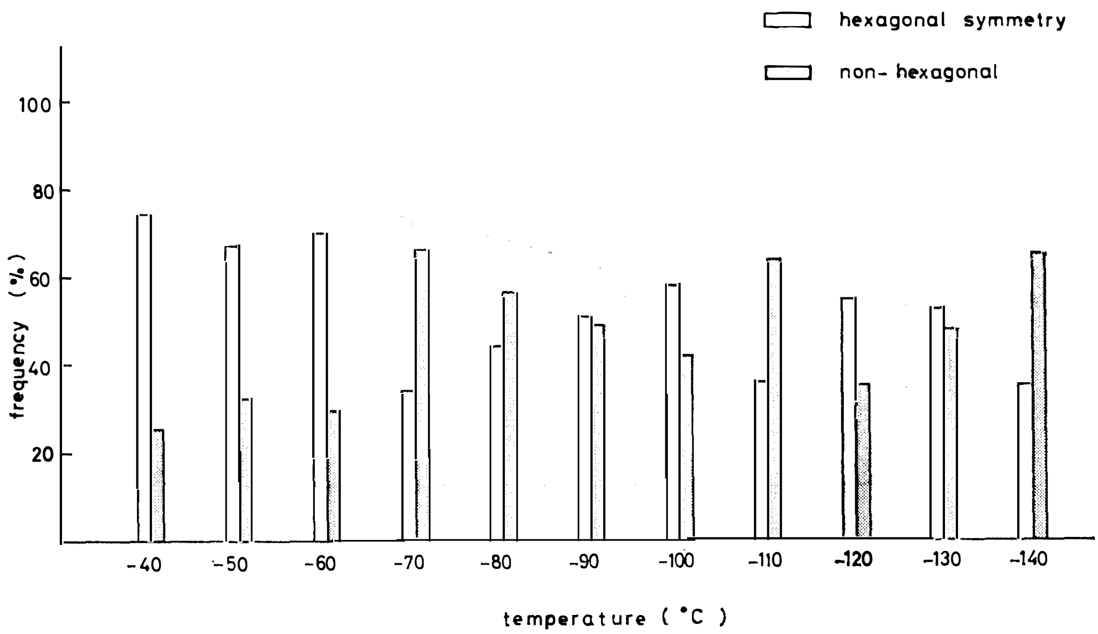

Fig. 6. Temperature variation in the growth frequency of plate-like ice crystals with hexagonal and non-hexagonal symmetry below about 60 μm in size.

3.2. *Growth mode of ice crystals and diffusion field*

Figure 7 shows the temperature variation in the growth frequency of plate-like and column-like ice crystals below about 60 μm in size. At the temperatures above about -110°C, in the case of ice crystals below about 60 μm in size, plate-like ice crystals grow predominantly. However, at the temperatures below -120°C, column-like ice crystals grow predominantly. Figure 8 indicates the temperature dependence on the size ratio c/a of column-like and plate-like ice crystals below about 60 μm in size. Each point shows the mean value of 15 to 20 ice crystals except for the values

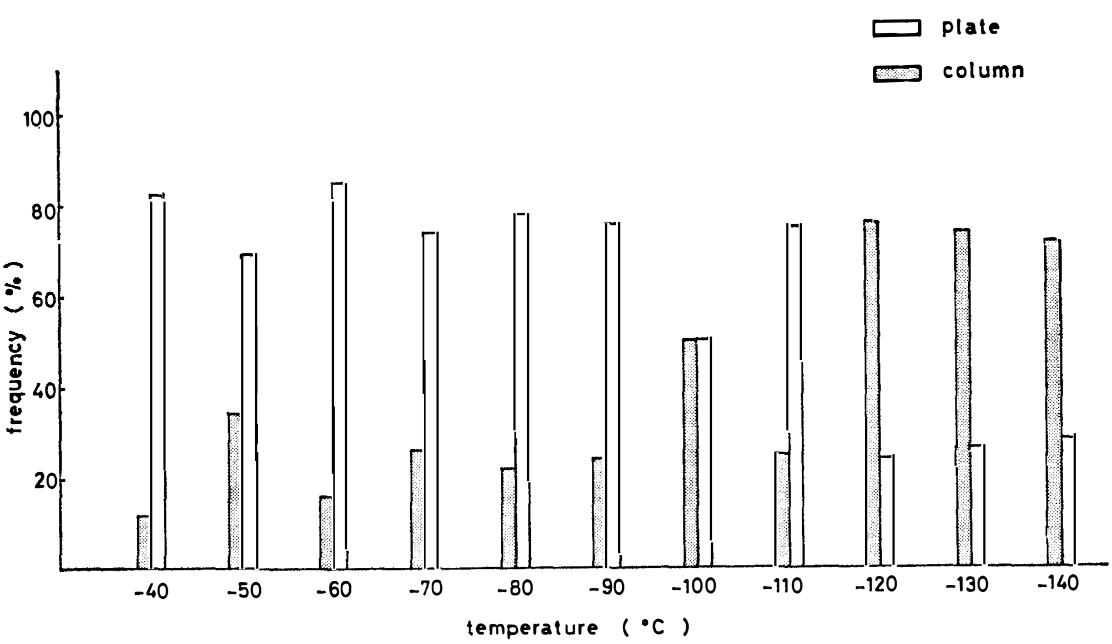

Fig. 7. Temperature variation in the growth frequency of plate-like and column-like ice crystals below about 60 μm in size.

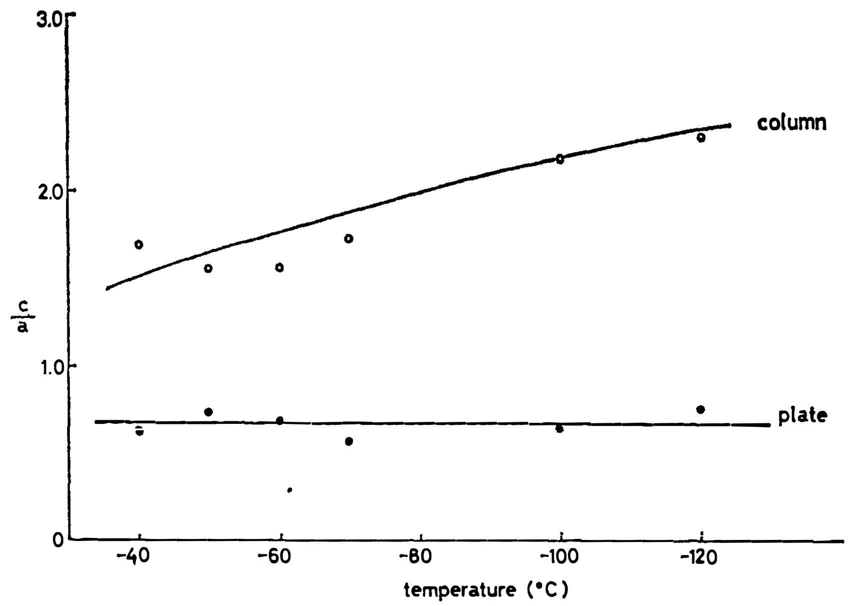

Fig. 8. Temperature dependence on the size ratio c/a of column-like and plate-like ice crystals below about 60 μm in size.

extremely deviated from the mean value. It is understood that the size ratio of column-like ice crystals slightly increases with falling temperature though that of plate-like ice crystals is almost constant with falling temperature.

Figure 9 shows the growth mode of plate-like ice crystals below about 30 μm in size, which grow at the temperature of $-50°C$. As shown in the figure, the length along c-axis of ice crystals seems to be propotional to that along a-axis; that is, in the case of ice crystals below about 30 μm the size ratio c/a of plate-like ice crystals hardly

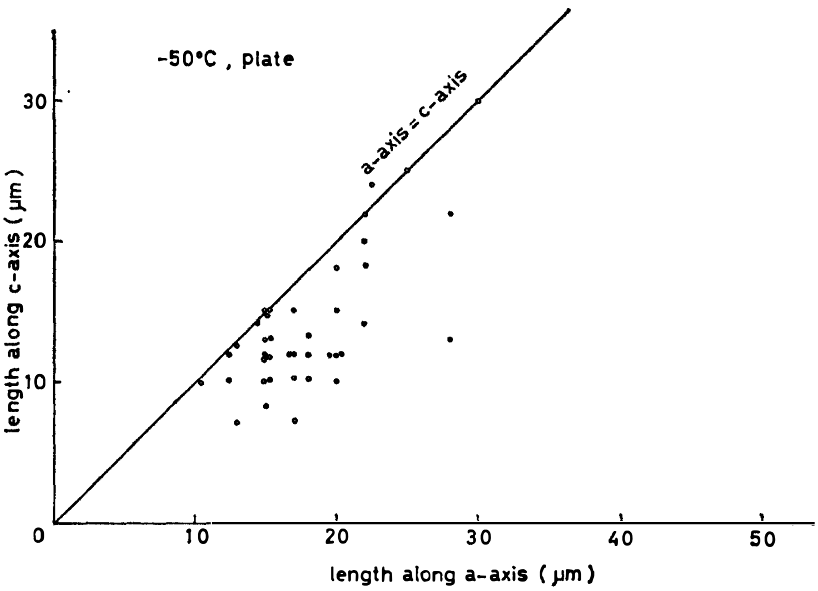

Fig. 9. Growth mode of plate-like ice crystals below about 30 μm in size, wh'ch grow at —50°C.

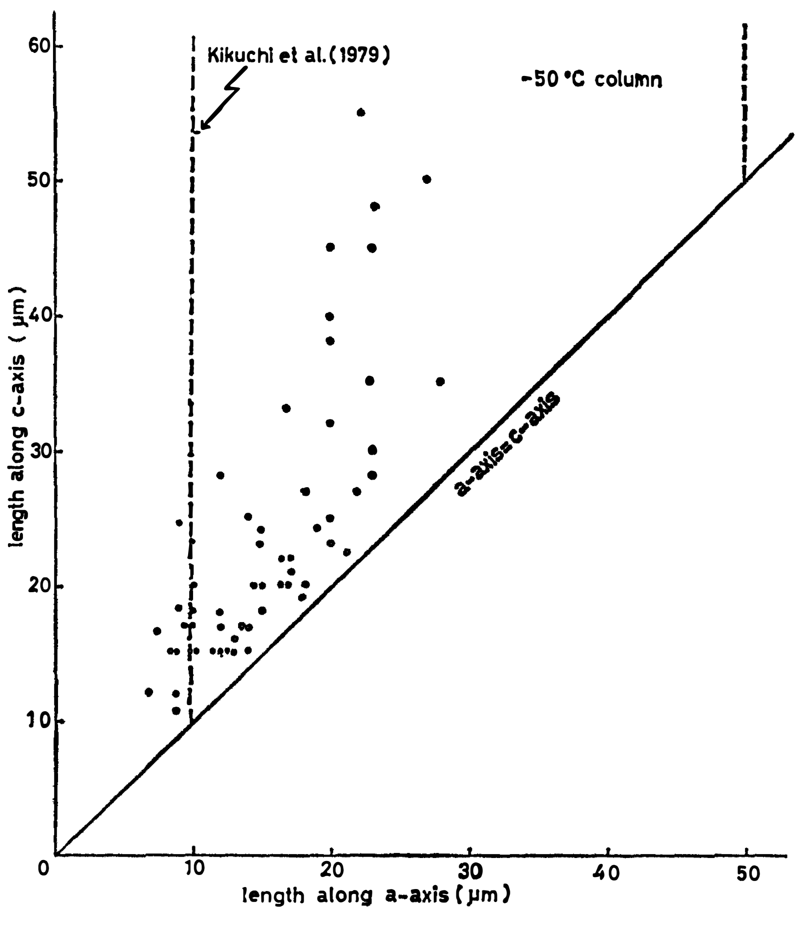

Fig. 10. Growth mode of column-like ice crystals below about 30 μm in size, which grow at —50°C.

depends on crystal size. Figure 10 shows the growth mode of columnar ice crystals below about 30 μm in size, which grow at the temperature of -50°C. As clearly shown in the figure, the length along c-axis of columnar ice crystals increases with increasing length along a-axis; especially, it is understood that the length along c-axis increases rapidly when the length along a-axis grows above about 20 μm in size. That is, the size ratio c/a of columnar ice crystals depends on crystal size. In addition, the growth mode of columnar ice crystals growing at -50°C shifts slightly toward the small crystal size as compared with that of columnar diamond dust type ice crystals observed in Antarctica by KIKUCHI and HOGAN (1979), but the growth mode of columnar ice crystals investigated experimentally coincides roughly with that observed in Antarctica.

4. Discussion

Ice crystals were grown in free fall at various constant temperatures between -40 and -140°C in order to investigate the morphology and the growth mode of snow crystals forming in the polar regions. In the present experiments, the supersaturation during the growth of ice crystals is estimated to be about 2–3% except for the cases of Fig. 3. Accordingly, except for the long solid prism in Fig. 4b, it is thought that ice crystals formed under growth conditions described above grow by the other mechanism than a screw mechanism.

As shown in Fig. 5, at the temperatures above -40°C, single ice crystals are predominantly formed, while at the temperatures below -40°C, polycrystalline ice crystals are predominantly formed; this is not because the difference in supersaturation in the chamber occurs with falling temperature but because aerosol and inactive particles in air have a tendency to act as ice nuclei with falling temperature. Furthermore, as shown in Fig. 6, at the temperatures above about -60°C, plate-like ice crystals with hexagonal symmetry grow predominantly, while at the temperatures below about -70°C, plate-like ice crystals with non-hexagonal symmetry grow predominantly. Generally speaking, it is considered that ice crystals are formed either through the process of supercooled cloud droplets or through the process of direct sublimation of water vapor onto ice nuclei. One of the reasons why plate-like ice crystals with non-hexagonal symmetry grow predominantly below -70°C may be concerned with direct sublimation of water vapor onto ice nuclei. That is, it is concerned with the nucleation process that polycrystalline ice crystals and ice crystals with non-hexagonal symmetry are predominantly formed with falling temperature.

As shown in Fig. 7, plate-like ice crystals grow predominantly at the temperatures above about -110°C and the supersaturation of about 2–3%. This experimental result coincides well with the theoretical diagram showing the relation between the habit of ice crystals and the growth conditions by KURODA and LACMANN (1982) and KURODA (1982). In Fig. 8, the size ratio of column-like ice crystals slightly increases with falling temperature though that of plate-like ice crystals is almost constant with falling temperature. This fact means that surface kinetics of the $\{0001\}$ and $\{10\bar{1}0\}$ faces does not change with falling temperature.

In the next place, as inferred from Figs. 9 and 10, the size ratio c/a of plate-like ice crystals growing at -50°C hardly depends on crystal size, while that of columnar ice crystals increases rapidly with increasing crystal size when ice crystals grow beyond about 20 μm and columnar ice crystals are transformed into long prismatic columns. As pointed out by KURODA and LACMANN (1982) and KURODA (1982), this result may be concerned with the opinion that the flux of water vapor onto the $\{10\bar{1}0\}$ faces is two-dimensional, but that onto the $\{0001\}$ faces is three-dimensional. As a result, it is considered that columnar ice crystals grow remarkably in the $\langle 0001 \rangle$ directions because of the shape effect of diffusion field of water vapor. In addition, the growth mode of columnar ice crystals growing at -50°C shifts slightly toward the small crystal size as compared with that of columnar diamond dust type ice crystals observed in Antarctica by KIKUCHI and HOGAN (1979). This is because the sizes of ice crystals formed experimentally are smaller than those of KIKUCHI and HOGAN (1979).

5. Conclusions

In order to investigate the morphology and the growth mode of single snow crystals forming in the polar regions, ice crystals were grown in free fall at various constant temperatures between -40 and -140°C using the cold chamber cooled by liquid nitrogen. The results obtained experimentally are as follows:

(1) At the temperature of about -40°C, single ice crystals are predominantly formed, while below about -50°C polycrystalline ice crystals are predominantly formed.

(2) At the temperatures above about -60°C, plate-like ice crystals with hexagonal symmetry are predominantly formed, while below -70°C plate-like ice crystals with non-hexagonal symmetry are predominantly formed.

(3) At the temperatures above about -110°C, the habit of single ice crystals below about 60 μm is plate-like. The experimental fact coincides with the theoretical diagram showing the relation between the habit of ice crystals and the growth conditions by KURODA and LACMANN (1982) and KURODA (1982). Below about -120°C, the habit of single ice crystals is column-like. Moreover, the size ratio c/a of plate-like ice crystals is almost constant with falling temperature, while that of column-like ice crystals slightly increases with falling temperature.

(4) The size ratio c/a of plate-like ice crystals below about 30 μm in size, which grow at -50°C hardly depends on crystal size. However, that of column-like ice crystals growing at the same temperature increases rapidly with increasing crystal size and column-like ice crystals are transformed into long prismatic columns.

(5) The growth mode of columnar ice crystals growing at -50°C is slightly shifts toward the small crystal size as compared with that of columnar diamond dust type ice crystals observed in Antarctica by KIKUCHI and HOGAN (1979); but the growth mode of columnar ice crystals experimentally formed coincides roughly with that observed in Antarctica. The growth mode of plate-like ice crystals which may be observed in Antarctica should be investigated, too.

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References

GONDA, T. (1977): The growth of small ice crystals in gases of high and low pressures at −30 and −44°C. J. Meteorol. Soc. Jpn., **55**, 142–146.

GONDA, T. and KOIKE, T. (1982a): Morphology of single snow crystals growing in air at low temperatures. Mem. Natl Inst. Polar Res., Spec. Issue, **24**, 148–156.

GONDA, T. and KOIKE, T. (1982b): Growth rates and growth forms of ice crystals grown from the vapor phase. J. Cryst. Growth, **56**, 259–264.

HIGUCHI, K. (1968): Kyokuchi ni furu yuki (Snow crystals forming in polar regions). Shizen (Nature), **23**(2), 38–46.

KIKUCHI, K. and HOGAN, W. (1979): Properties of diamond dust type ice crystals observed in summer season at Amundsen-Scott Pole Station, Antarctica. J. Meteorol. Soc. Jpn., **57**, 180–190.

KOBAYASHI, T. (1965): Vapor growth of ice crystal between −40 and −90°C. J. Meteorol. Soc. Jpn., **43**, 359–367.

KURODA, T. (1982): Growth kinetics of ice single crystal from vapor phase and variation of its growth form. J. Meteorol. Soc. Jpn., **60**, 520–534.

KURODA, T. and LACMANN, R. (1982): Growth kinetics of ice from the vapor phase and its growth forms. J. Cryst. Growth, **56**, 189–205.

OHTAKE, T. (1970): Unusual crystals in ice fog. J. Atmos. Sci., **27**, 509–511.

SHIMIZU, H. (1963): "Long prism" crystals observed in precipitation in Antarctica. J. Meteorol. Soc. Jpn., **41**, 305–307.

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