Growth of snow crystals from frozen water droplets

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Abstract

Growth of frozen water droplets, falling freely in a supercooled cloud, was investigated by using a large cloud chamber 6.5m in height. The size of frozen water droplets was between 20-100 μm in diameter. Frozen water droplets grew to various shapes. Two modes were confirmed at the initial growth stage. In one mode, twenty circular crystal faces appeared during the growth. They were 2 basal, 6 prism and 12 pyramidal faces. {1120} crystal faces, whose existence has been suggested by theory, were confirmed in this experiment. In the other mode, steps or irregular patterns appeared in place of circular pyramidal and prism faces. After further growth frozen water droplets grew to short column-like ice crystals in both modes, and finally grew to snow crystals with the crystal habit as shown by Nakaya's diagram.

Keywords: Snow crystals, Crystal morphology, Growth from vapor, Frozen water droplets, Free fall

1. INTRODUCTION

Nakaya(1954) has already found that many natural dendritic snow crystals had a small hexagonal plate at the center. He also observed the growth process of such a crystal in the study of artificial snow crystals. At first a short column was formed and one of the basal faces developed to a dendritic crystal. The other basal face grew only to a small hexagonal plate because of competition for the available moisture. Snow crystals with such double hexagonal plates are now called double plate crystals. Weickmann, Katz and Steele(1970) concluded from laboratory experiments, and Auer(1970) from field observations that double plate crystals developed from single-crystalline frozen water droplets. Their conclusion was supported by other laboratory experiments. Pitter and Pruppacher(1973) and Parungo and Weickmann (1973) showed that water droplets of cloud droplets size froze into single-crystalline particles with very high probability when they were nucleated by contact with clay particles or AgI particles at temperatures above -20℃.

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The growth process of single-crystalline frozen water droplets were investigated by Gliki and Eliseev(1962) and Gliki, Eliseev and Marcheko(1962). They observed that circular crystal faces(basal, prism and pyramidal faces) appeared on a spherical surface at the initial stage of the growth of frozen water drops. In their experiments, investigated water drops were much larger than cloud droplets and water vapor supply was not uniform because water drops were suspended on a string. In order to study the growth of snow crystals from frozen water droplets more nearly under atmospheric conditions, the experiments were made for frozen water droplets in free fall. The preliminary results were reported in Yamashita and Takahashi(1972). They observed twenty circular crystal faces(2 basal, 6 prism and 12 pyramidal faces) at the initial growth stage. Gonda and Yamazaki(1978) made in-situ observation of the growth of frozen water droplets and showed the diagram of the growth mode. Since their experiment was made by using a diffusion chamber, the growth of frozen water droplets was lack of symmetry. The present study was carried out to observe the growth of frozen water droplets in the state close to natural clouds by falling water droplets in a supercooled cloud produced in a large cloud chamber of about 6.5m in height.

2. EXPERIMENTAL APPARATUS AND PROCEDURES

Experiments were made in a large cloud chamber as shown in Fig.1. It consisted of a stainless steel pipe of about 6.5m in height and 28 cm in diameter. It was cooled down to -33℃ by circulating cold air around it and its temperature was kept uniform within ±1℃ from the level 50cm below the top to the bottom. Supercooled cloud was produced by a semipermeable cellulose bag containing about 400ml of hot water of about 70℃. The bag was introduced from the top, hung down to the bottom and removed from the top. Supercooled cloud remained for about 10 minutes in the chamber after these operations. Water droplets containing silver iodide in suspension were dropped from the top by using a spray. Silver iodide suspensions were made by mixing potassium iodide and silver nitrate solutions. Solutions of the concentration 10^{-2} g/l were used. Water droplets were nucleated by silver iodide in suspension at about -5℃. To nucleate water droplets at lower temperatures silver iodide solutions of concentration 10^{-5} g/l was also used. Most of water droplets produced from silver iodide solution of this concentration were nucleated below -20℃ (Takahashi and Yamashita,1970). The size of water droplets ranged between 20-100 μm in diameter. After freezing completely, they grew to ice crystals and were collected in silicon oil at the bottom of the chamber. They were covered with a coverglass to prevent evaporation and observed under a polarizing microscope.

3.RESULT

Frozen water droplets grew to various shapes when they were collected at the bottom of the chamber. Two growth modes were observed. In one mode twenty circular crystal faces appeared on the surface of frozen water droplets. In the other mode, steps or irregular patterns appeared in place of circular crystal faces.

3.1 Formation of circular crystal faces

It is possible to follow the growth process by observing ice particles classified into this mode. Fig. 2 shows examples of these ice particles of various shapes and Fig.3 shows the diagram of this growth process.

First stage　Twenty circular crystal faces appeared(Fig.2b). They were 2 basal, 6 prism and 12 pyramidal faces. The pyramidal faces corresponded to {1011} faces. Another crystal faces were observed between to prism faces in a few cases. They corresponded to {1120} faces. The size of {1120} faces was about 1/4 of the size of prism faces.

Second stage　A prism face connected with two pyramidal faces after further growth (Fig.2c). The size of pyramidal faces, which was not so much small compared with that of prism faces at former stage, was apparently smaller than that of prism faces. A small hollow was observed at the center of pyramidal faces. A step was usually formed between a basal face and pyramidal faces.

Third stage　Adjacent prism faces connected. {1120} faces disappeared and small cavities were observed between two prism faces as a trace of {1120} faces. Pyramidal faces also disappeared and hollows remained as a trace of pyramidal facesFig.2d).

Forth stage　Frozen water droplets grew to short column-like crystals(Fig.2e). Afterward they grew to snow crystals of well-known habit corresponding to the temperature and supersaturation. In the temperature range where needles or columns grew, the development of prism faces was favored(Fig.2f and2g). In the temperature range where plates or denndritic crystals grew, the development of two basal faces was favored and double plate crystals grew(Fig.2h and 2i). Two basal faces did not always grow with hexagonal symmetry. Each basal faces developed into opposite direction as shown in Fig.4 . Sometimes plates developed from a step formed between a basal and pyramidal faces(Fig.4a and 4b) and three or more plates developed from a frozen water droplet(Fig.4c). These plates or dendritic crystals without hexagonal symmetry might be an embryo of malformed snow crystals observed in the field.

3.2 Formation of steps

In place of circular prism and pyramidal faces, several steps were formed(Fig.5b and 5c). They were sometimes observed only as irregular patterns(Fig.5a). These steps and irregular patterns disappeared with further growth and frozen water droplets grew to column-like crystals. This mode of the growth was frequently observed when silver iodide solution of the concentration 10^{-5} g/l was sprayed. In this case, most of water droplets were nucleated below -20℃. The surface of frozen water droplets nucleated at low temperatures seemed not to be smooth compared with those nucleated at temperatures close to 0℃.

Both circular prism and pyramidal faces and steps appeared on the same frozen water droplet in several cases. Fig.6 shows a typical example. Circular pyramidal faces appeared on the upper half and steps appeared on the lower half. Such non-symmetrical growth between upper and lower halves of frozen water droplets seem to be caused from the different moisture supply to each part of a frozen water droplet.

4. DISCUSSIONS

4.1 {1120} crystal faces

The existence of {1011} and {1120} crystal faces are suggested from the γ-plot of ice crystals by Higuchi(1961). He indicated these crystal faces are metastable and disappear during the growth process. {1011} crystal faces were observed in the previous works, but {1120} crystal faces was observed for the first time. Two conditions are favorite for the appearance of {1120} crystal faces in this experiment. One is that the size of frozen water droplets was large. Since the size of {1120} crystal faces was small(about 1/8 of the diameter of frozen water droplets), it would be missed in case of the growth of small frozen water droplets. The other is that air temperatures when {1120} crystal faces appeared during the growth were low. Kobayashi(1965) observed the pyramidal faces frequently at temperatures below -40℃. It seems that low temperatures are favorite for the appearance of metastable crystal faces.

4.2 Formation of hollows at the center of pyramidal faces

After pyramidal faces enlarged to observable size, they changed into flat faces with a hollow at the center. As prism faces enlarged and connected with pyramidal faces, the growth of pyramidal faces stopped. Finally pyramidal faces disappeared and cavities were left frequently as a trace of a pyramidal faces. In the experiment by Gonda and Yamazaki, these hollows or cavities were not observed. In their experiment, the growth frozen water droplets was slower than that in the present experiment because moisture supply was small in the diffusion cloud chamber compared with the growth in a supercooled cloud in free fall. Ohtake(1970) also observed ice particles with flat pyramidal faces. This ice particle probably grew from frozen water droplets in ice fog under low supersaturation. Thus the moisture supply to frozen water droplets causes the difference of the growth mode.

4.3 Formation of malformed snow crystals

Among natural snow crystals, those without regular hexagonal symmetry were frequently formed. Nakaya(1954) named those crystals by malformed snow crystals. As described in section 3.3, two basal planes developed into opposite direction. One plate growth prevents the growth of another plate, and finally this ice crystal might grow to a malformed crystal of the three- or four-branched type. If the initial frozen water droplets cracked at the center as shown by Takahashi(1975), they split into two halves and produces three- or four- branched snow crystals.

5. CONCLUSION

Frozen water droplets grew in free fall in a large cloud chamber about 6.5m in height. It is possible to follow the growth process by observing ice particles of various shape collected at the bottom of the chamber. There are two growth modes. In one mode, twenty circular crystal faces appeared. They are 2 basal, 6 prism and 12 pyramidal faces. {1120} crystal faces were observed in a few cases when the size of frozen water droplets was large and the air temperature was about -30℃. Pyramidal faces and {1120} crystal faces disappeared during the growth. A small hollow was observed at the center of these metastable faces and they remained as cavities frequently. In the other mode, steps or irregular patterns appeared in place of circular prism and pyramidal faces. This growth mode was likely to occur when frozen water droplets were nucleated at low temperatures(below about -20℃). In both growth modes, frozen water droplets grow to short column-like crystals and grew to snow crystals with the crystal habit corresponding with air temperatures. In plate growth region, asymmetrical growth of dendritic branches from frozen water droplets suggests the formation of three- or four-branched snow crystals.

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Legend

Fig.1 Large cloud chamber

Fig.2 Ice particles grown from frozen water droplets, accompanied with the development of circular crystal faces. Ice particles grew at temperatures, a:-27.0, b:-31.1, c:-25.5, d:-28.8, e:-28.4, f:-33.2, g:-6.7, h:-18.9, i:-12.0℃. scales=50 μ m.

Fig.3 Diagram of the growth process of the frozen water droplets

Fig.4 Asymmetrical growth of plate or denndritic branches from frozen water droplets. Ice particles grew at temperatures, a:-12.0, b:-15.1, c:-18.9℃. scales=50 μ m.

Fig.5 Ice particles grown from frozen water droplets, accompanied with the development of steps or irregular patterns at the initial stage. Ice particles grew at temperatures, a:-31.0, b:-33.2, c:-22.0℃. scales=50 μ m.

Fig.6 Ice particles with both steps and circular pyramidal faces, grown at temperature -22.0℃. scales=50 μ m.

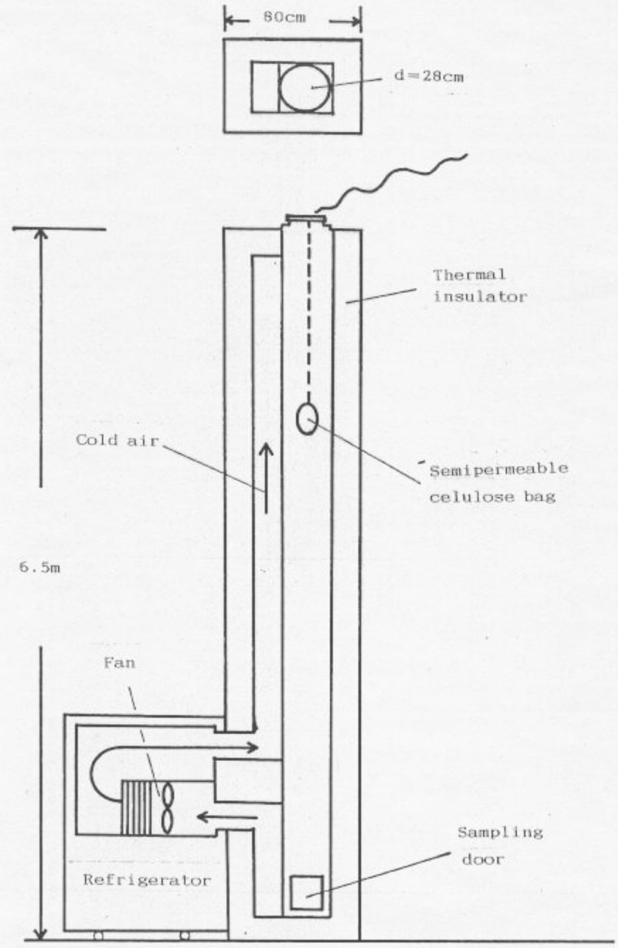
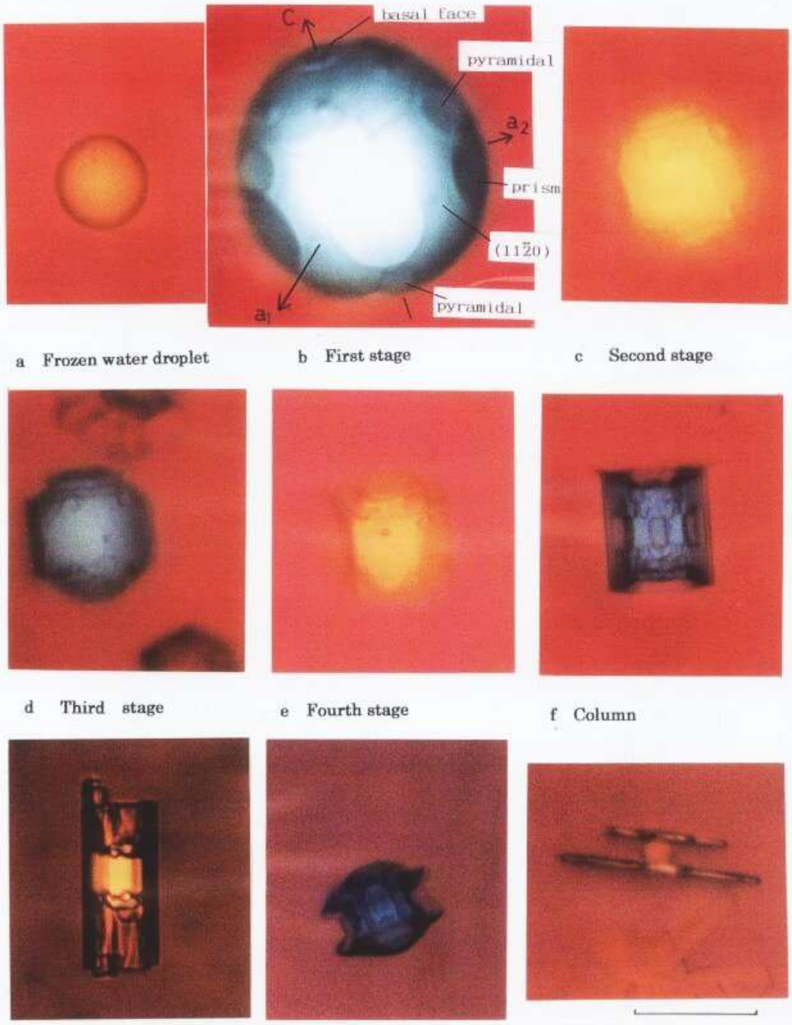


Fig. 1 Apparatus: Large cloud chamber



a Frozen water droplet b First stage c Second stage

d Third stage e Fourth stage f Column

g Sheath h Plate i Dendrite

Fig 2

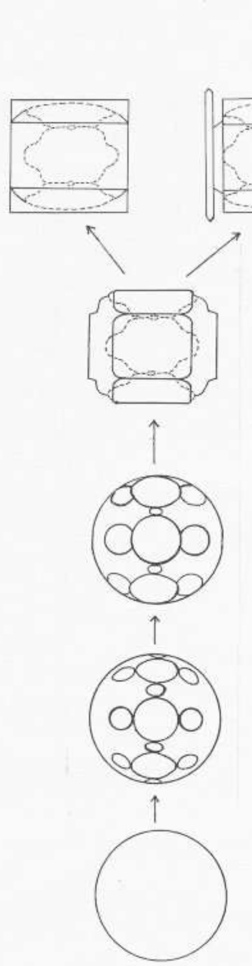


Diagram of the growth of frozen water droplets

Fig 3

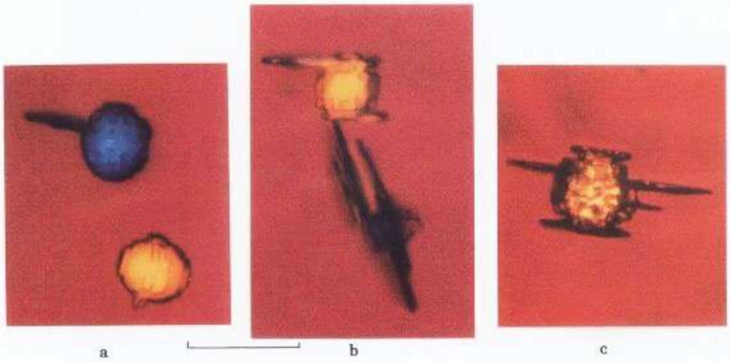


Fig.4

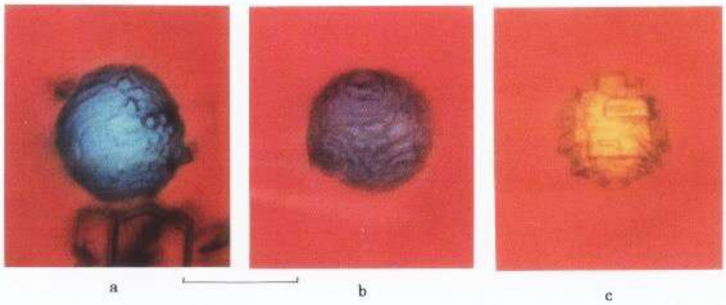


Fig.5

Fig 6