[Journal of the Faculty of Science, Hokkaidô University, Japan, Ser. II, Vol. IV, No. 4, 1953]

Cinematographic Study of Ice Crystal Formation in Water

Motoi KUMAI and Kazuhiko ITAGAKI

(Received June 22, 1953)

Experimental work was conducted in the cold chamber laboratory of the Institute of Low Temperature Science, Hokkaido University. Applying shadow photograph method, the authors devised a cinematographic method, which for examining ice crystal formation in the water. By seeding with minute ice crystals on a water surface slightly supercooled, tiny ice-discs in the early stage were produced; according to the surrounding conditions they deformed to various type. The relation between the ice crystal form and the water temperature in supercooling was studied. In this experiment, a number of powdered materials as silver iodide, kaolin, carbon, and clay particles were found to be not active for nucleus of ice crystal directly, but minute ice crystals and splinters were active.

§ 1. Introduction

The formation of ice-discs has long been a problem among engineers and physicists, and the direct observation of such formation by various methods has been recommended by many workers. But no paper has yet appeared, at least on the observation by cinematograph of ice-disc formation in detail. The work of BARNES[1] and ALTBERG[2][3] contains one of the most detailed summaries of available data in this field. SCHAEFER[4] also reported concerning the formation of frazil and anchor ice under natural conditions, and he described his studies on their properties by seeding supercooled water with small fragments of solid CO_2. IMAI[5] observed by microscope the formation of ice-discs in the cold water of a small vessel. ARAKAWA and HIGUCHI[6] devised a shadow photograph method to study water freezing, and successive photographs were taken of freezing initiated by artifical seeding. In the present paper, the results are given of observation by a cinematograph, of ice formation in the cold water of a small tray. The photographs were taken in the cold chamber laboratory.

§ 2. Cinematography of ice formation in the water

The present authors developed a method for taking cinematographs of the process of ice crystal formation in the water. A camera of the Le Palvo type was used for this purpose. Arrangement of the apparatus is shown in Fig. 1. Removing the lens from the camera, we turned it to face upwards, and put a shallow glass tray on the hole, under which the film was to be run through. The hole rim was originally to be used to hold a filter glass, so the tray was made as the same size as the filter glass. It was $3 \times 4\ cm^2$ in dimension and 1 cm in thickness. A small electric torch bulb of 6 V and 0.8 A was used as a light source. By using a slightly higher voltage (about 6.8 V), its brightness could be increased. It was set at 80 cm above the water surface. All the apparatus was mounted in the cold chamber laboratory, where the air temperature was kept about $-7°C$.

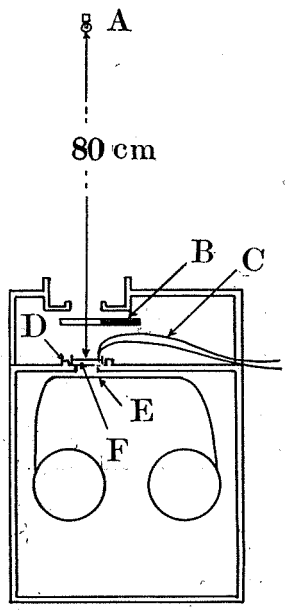


Fig. 1.
Arrangemgement of apparatus
A torch bulb
B shutter
C thermo-junction
D hole rim
E film
F water

The experimental procedures were as follows. At first, 3 c.c. water was poured into the tray, corresponding to a depth of about 0.3 cm. When the water temperature measured by a thermo-couple indicated some slight supercooling of the water, film driving was started in the camera. Next, the water surface was seeded with minute ice crystals[7] gerenerated around a thermometer stem previously cooled by liquid air to the temperature of $-50°C$. As soon as the seeding was carried out, many ice crystals began to grow in the water. The process of their growth was recorded on the cinefilm. As the speed of film driving is reduced to one or two frames per second, one can see the fast motion picture of the process.

In such a way, a study was made especially of the earlier

stage of crystal formation in the water. In spite of the rapidity of the occurrence of phenomena, the mode of deformation of ice crystal had been exactly clarified by this method. The photograph in ARAKAWA's report[6] is a negative one, but that in the present paper is a positive.

§ 3. Method of seeding

By preparing dried particles in the cold chamber which is kept between −6°C and −10°C, seeding was done with certain particles on the slightly supercooled water surface. They were silver iodide, kaolin, carbon, and clay particles. Then, no ice crystal appeared. They were not active as nuclei of ice crystal in this experiment. According to SMITH-JOHANNSEN,[8] the freezing temperature was below −6.8°C for a bulk water into which was introduced a number of particles of powdered material such as silver iodide, graphite or zinc oxide, etc. Present authors observed that crystallization started from the edge of tray, but not from the particle on the water surface. By agitating with an ice piece on the slightly supercooled water surface, ice germs[9] were grown in its trace, and they were grown to ice-discs. In the case of more supercooled water, it was strongly agitated to make ice-discs. Thus, ice-discs were grown at −1.2°C always. In this experiment, as described above in §2, the authors seeded on the water surface sligetly supercooled with mixed minute ice crystals and supercooled fog generated around a thermometer stem previously cooled by liquid air to the temperature at −50°C as shown in Fig. 2.

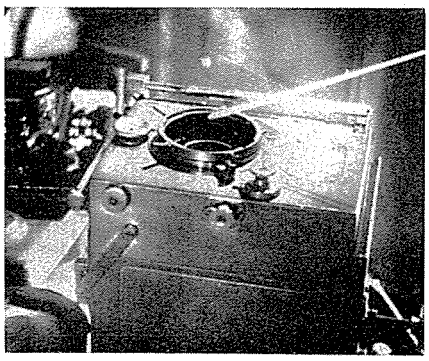


Fig. 2. Apparatus and seeding on the water by cooled thermometer stem.

Minute ice crystals were active as nuclei for the freezing of circular ice-discs. By this method, cinematograph of ice formation in the water were taken.

§ 4. The formation of ice crystals in water

In this series of experiments the room temperature in the chamber was kept usually between −6°C and −10°C. The first aim of this experiment was to observe the successive stages in the development of crystals. The results are described as follows.

1) Ice-disc crystal

One example of a series of photographs showing the manner in which thin circular ice-discs develop is given in Photos 1–9. In this case the apparatus as shown in Fig. 1 was used and ice-discs grew near the surface of water. The water was distilled. The room temperature was kept at −7°C, the water volume in the tray was always 3 c.c. The rate of cooling of the water was 0.034°C/sec at 0°C. Photo 1 shows the crystal about 50 seconds after the seeding with thermometer stem cooled to −40°C by liquid air. The temperature of water at the instant of seeding was at −0.14°C, and then it rose to −0.05°C with latent heat liberated by growing crystal as shown in Fig. 3. The mode of growth of ice-discs is shown in

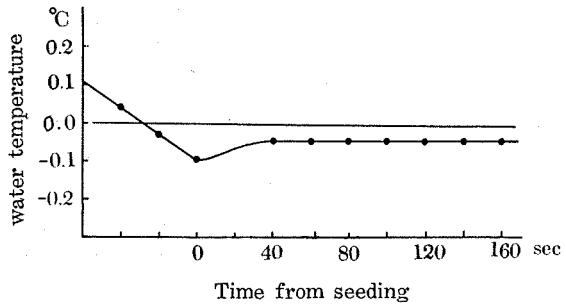


Fig. 3. The relation between the water temperature and the time from seeding, about the crystal marked A in Photos 1–9.

Photos 1–9. At first, circular ice-disc as shown in Photo 1 marked A is formed by seeding, and this developed to ice-disc with notches as shown in Photo 6 marked A. Then the ice-disc with notches developed sometimes to dendritic crystals like those seen in Photo 31. Taking condition in Photo 1 as the initial state, the rate of growth of ice-discs marked A is 1.7×10^{-3} cm/sec, that of deformed

ice-disc marked B is 3.9×10^{-3} cm/sec and of that marked C is 3.6×10^{-3} cm/sec in the stages between Photo 1 and 3. In this case, the circular form of ice-disc A is kept longer than that of B and C, crystals B and C are deformed soon to irregular form because they exsist near the surface. The rate of growth of ice-discs varies considerably with the condition of formation, but usually it can be taken as nearly $1 \sim 3 \times 10^{-3}$ cm/sec for circular ice-disc, $3 \sim 4 \times 10^{-3}$ cm/sec for deformed ice-disc in this apparatus. These rates are varied not only by the rate of cooling of the water and the number of ice-discs in a unit volume, but also by whether the crystals float on or slightly under the cold water surface and by their movement, that is, with the mode of the conduction and convection of latent heat by crystal formation. Fig. 4 shows the growth of ice-disc marked A in Photos 1–9. This ice-disc developed to a circular ice-disc 2.1 mm in dimension with notches at 140 seconds from the seeding time. The rate of growth of circular ice-disc and circular ice-disc with notches was the same 1.7×10^{-3} cm/sec, but that of rounded pattern in the circular ice-disc with notches was decreased to 6.7×10^{-4} cm/sec as seen in Fig. 4. For the ice-disc with notches or stellar crystal, the rate of growth of rounded pattern of upper layer is smaller than that of under layer.

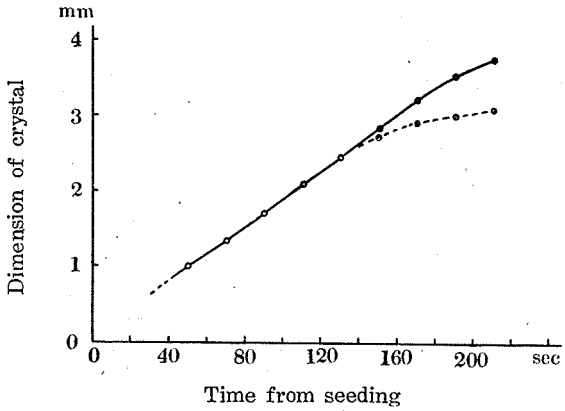


Fig. 4. Growth from the cirular ice-disc to the circular ice-disc with notches about the crystal marked A as shown in Photos 1–8.

(○ circular ice-disc, ● circular ice-disc with notches, ⊙ rounded pattern in the crystal. The same marks are used for the other Figs. 6, 8, 9.)

Fig. 5 shows the relation between the water temperature and the time from seeding. In this case, the crystal developed from circular ice-disc to circular ice-disc with dendritic branches (stellar crystal). The water temperature in Fig. 5 is lower than in Fig. 3. In the case of Fig. 5, the relation between the crystal form

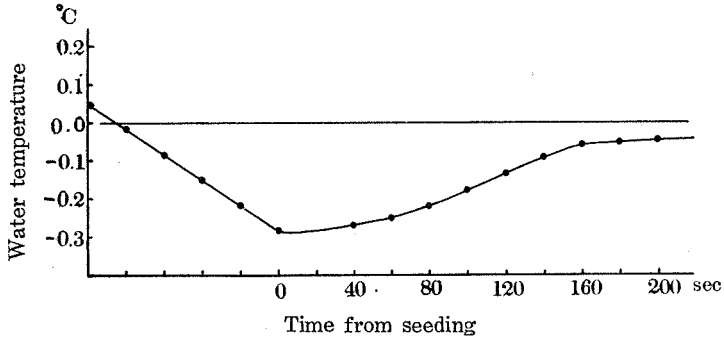


Fig. 5. The relation between the water temperature and the time from seeding, about the crystal in Fig. 6.

and the rate of growth is shown in Fig. 6. Photographs of crystal used in Fig. 5–6 are omitted in this paper. The room temperature was kept at −8.5°C, the cooling rate of water was 0.036°C/sec at 0°C. Only one circular ice-disc grew by seeding; at about 80 seconds from the seeding time needle-like crystal grew on the bottom of the vessel. The rate of growth of this circular ice-disc was 2.6×10^{-3} cm/sec, and then that of circular ice-disc with notches and dendritic crystals was the same 5.7×10^{-3} cm/sec. The rate of growth of the rounded pattern in the stellar crystal was the same as that of circular ice-disc. The number of crystals in a unit volume in the case of Fig. 6 was smaller than that in Photo 1, and also the rate of growth of the

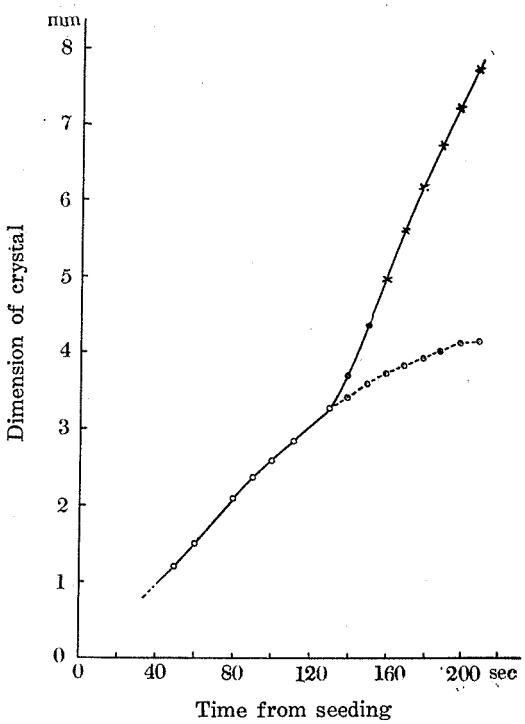


Fig. 6. Growth from the circular ice-disc to the stellar crystal in water. (× stellar crystal)

circular ice-disc in the former was larger than in that of the latter. The rate of growth of the stellar crystal in the water was of the same order as that in a 1.7 per cent solution of sodium chloride as seen in Photos 29–34. In Photo 5, the ice-disc A was showing a start of dendritic growth and only the ice-disc marked A was slightly under the cold water surface, but many other ice-discs were floating upon the surface. The complex structure in ice-discs marked B, C, etc., as seen in Photo 5 are showing openings in the water cover. These tears deformed rapidly.

The number of notches around the disc was 48, but this was not constant for each crystal. The notches grow around the under layer of the ice-disc. Photos 5–9, and 10–14 show the mode of growth of the notches and the internal pattern of ice-discs. When an ice-disc grows the notches, the width of the edge of the ice-disc increases as seen in Photo 5 crystal A and Photo 10 crystal D. Sketches of the crystal marked A shown in Photos 6–8 and the crystal marked D shown in Photo 14 are given in Fig. 7. The rate of growth of the rounded patterns of upper layer in the ice-disc marked D is smaller than that of the notches of under layer as seen in Photos 10–14 or Fig. 7 b. Fig. 7 a consists of double layers and Fig. 7 b consists of several layers. The ice-disc D is slightly under the cold water surface. But the ice-disc marked E has a complex structure, which is floating on the water surface. In these photographs, long ice needles always grew out from the edge; they were growing on the bottom of the tray.

Fig. 7. a) Sketch of the crystal marked A in Photos 6–8.
b) Sketch of the crystal marked D in Photo 14.

SCHAEFER[4] and LAMBOR[10] have both reported that frazil ice particles often showed a visible "mote" in their center. The ice-discs in Photos 15–16 showed "a black point" in their center. It is doubtful whether "black point" is a "mote". As stated above, such particles as silver iodide, kaolin, carbon or clay were not active for nuclei in our experiment.

Photo 17 shows the crystals about 15 seconds after the over seeding with thermometer stem cooled to −50°C, and Photo 18 shows the same 40 seconds after. In this case the room tempera-

ture was kept at −7°C. The rate of cooling of the water was 0.034°C/sec at 0°C, the temperature of water at the seeding was −0.57°C. As shown in Photo 17, there are some needle-like crystals and many semicircular ice-discs in addition to the circular ones. It seems that some needle-like crystals deform to semicircular ice-disc crystals as shown in Photos 17 and 18. Some needle-like crystals have a shadow beside themselves as shown in white in Photo 18. This is a reflection of light by edge of the crystal. The condition of the ice formation is different from Photos 1–16 at the water temperature of seeding and the number of seeding nuclei. As described above, circular ice-discs were formed in the slightly supercooled water, but needle-like crystals were formed in the more supercooled water as described later. In this case, it seems that at first needle-like crystals, then semicircular ice crystals and at last, circular ice-discs grow during successive seedings.

2) **Needle-like crystal**

In 1949, IMAI[5] proposed an explanation regarding the growth of an ice-disc crystal into a needle-like form. His explamation is based on the inclination of a disc crystal in relation to the water surface. In the present experiment, also our knowledge of crystals seems to suggest that the formation from a circular ice-disc to a needle-like form is based on the inclination to the water surface. The appearance of this deformation is shown by the ice-disc crystal marked F in Photos 19–22. The crystal F was deformed from the circular ice-disc in photo 19 to the semicircular ice-disc in photo 20, and to needle-like crystal in photo 22 and then to the wing-like form as shown in Photos 23. The side with a ice-disc of the wing-like crystal marked F in Photo 23 is inclined into the water, and the other side shows upward tendency. The inclination was shown from the cinematograph.

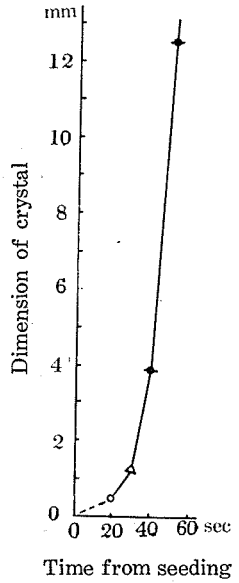


Fig. 8.
Growth from the circular ice-disc to the needle-like crystal, about the crystal marked F in Photos 19–22.
(△ semi-circular ice-disc. ●— needle-like crystal.)

The two sides of the crystal are unsymmetrical.

The conditions of crystal formation in the present experiment are as follows. Photo 19 shows the crystals at 20 seconds after the seeding with thermometer stem cooled to −50°C. In this case the room temperature was kept at −7°C, the water temperature at the seeding was −0.38°C. This was changed to −0.19°C after 120 seconds. The deformation from needle-like crystal to the wing-like crystal is shown in Photos 24–28. Photo 24 shows the crystal 25 seconds after the seeding. In this case, the rate of cooling of the water was 0.02°C/sec at nearly 0°C. The water temperature was −0.76°C at the seeding, and it was changed from −0.45°C to −0.10°C between 55 seconds and 115 seconds after the seeding.

3) **Stellar crystal**

Photos 29–34 show an example of dendritic crystal formation. This was made in a 1.7 per cent solution of sodium chloride dissolved in distilled water, by seeding with minute ice crystals generated around the thermometer stem cooled to −50°C. In this case, the room temperature was kept at −9°C and the rate of cooling of the water was 0.025°C/sec. The water temperature at the seeding was −1.33°C. The mode of growth from circular ice-disc to dendritic crystal is shown in these photographs. In this case, ice-discs did not grow as large as in distilled water. They have notches around themselves in the early stage, which are showing the start of dendritic growth. The rate of growth of the circular ice-disc marked G shown in Photo 29 was 1.0×10^{-3} cm/sec, and then that of the ice-disc with dendritic branches G (stellar crystal) was increased to 9.0×10^{-3} cm/sec as shown in Fig. 9. The rate of growth of the stellar crystal H was 5.4×10^{-3} cm/sec.

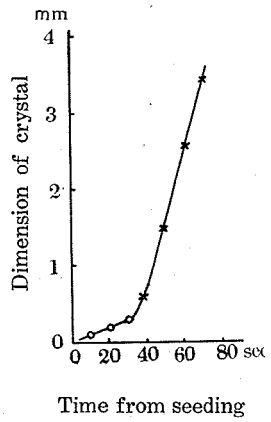


Fig. 9.

Growth from the circular ice-disc to the stellar crystal, about the crystal marked G in Photos 29–34.

4) **Crystal form and water temperature at the seeding time**

It can reasonably be expected that the

form of ice crystal in the water is strongly influenced by the form of the crystal in its early stage. Further the crystal form also in the early stage is influenced by the water temperature at the instant of seeding. The results of investigations on this point are described in this section. The authors know that a certain kind of crystal is observed chiefly within a definite thermal interval of water at the seeding time, but there is more than one type of crystal. One or two other types of crystals are mixed with the prevailing one. For example, as shown in Photo 17, circular ice-disc crystals are mixed with semicircular ice-discs or even with needle-like crystals. In our experiments, the relation between the crystal form and the water temperature at the seeding time was shown in Table 1.

TABLE 1.

Crystal form and water temperature at the seeding time.

Distilled water			1.7% NaCl solu.
Ice-disc chiefly as shown in Photo 1	Mixed with ice-disc, semicircular, needle-like as shown in Photo 17	Needle-like chiefly as shown in Photo 24	Stellar crystal as shown in Photo 29
− 0.05 °C	− 0.33 °C	− 0.57 °C	− 1.33 °C
− 0.10	− 0.38	− 0.76	− 1.33
− 0.10	− 0.57	− 0.81	—
− 0.14	—	− 0.91	—
− 0.14	—	—	—
− 0.19	—	—	—
− 0.20	—	—	—
− 0.23	—	—	—
− 0.24	—	—	—
− 0.29	—	—	—

The water temperature was measured at a corner of the tray as seen in Fig. 1. In this table, circular ice-discs as shown in Photos 1–16 grew chiefly in the water temperature between 0°C and −0.3°C; mixed crystals with circular, semicircular and needle-like crystrls as seen in Photos 17–23 grew in the water temperature between −0.3°C and −0.6°C; and needle-like crystals as seen

in Photos 24–28 grew chiefly in the temperature between $-0.6°C$ and $-0.9°C$.

5) **Crystal form and the rate of growth**

It can be expected that the rate of growth is influenced by the form of ice crystal, the water temperature, the number of ice crystals in a unit volume and the rate of cooling. The results of investigations on this point are described in this section. In this experiment, the cooling rate of water was between 0.036°C/sec and 0.020°C/sec, the volume of water was 3 c.c. The authors knew that some relation exists between the crystal form and the rate of growth. In our experiments, the rate of growth of crystal was $1 \sim 2.6 \times 10^{-3}$ cm/sec for a circular ice-disc under the water surface; $3.5 \sim 4.0 \times 10^{-3}$ cm/sec in mean diameter for a deformed ice-disc near the water surface; sometimes an ice-disc has visible double layer-like growth. In this case, the rate of growth of upper layer (rounded pattern) is smaller than that of the under layer; $5 \sim 9 \times 10^{-3}$ cm/sec in the greatest dimension for the stellar crystal and $4 \sim 5 \times 10^{-2}$ cm/sec in the largest dimension for the needle-like crystal. It is remarkable that the rate of growth is of the same order for each kind of crystal in undistilled water, distilled water and also in 1.7 per cent solution of sodium chloride dissolved in distilled water. The smallest rate of growth is for a circular ice-disc, the largest rate is for a needle-like crystal.

Comparing the present result with ALTBER'S,[3] the later is larger than ours. For example, the rate of growth of circular ice-disc in the water temperature between $-0.1°C$ and $-0.2°C$ is ten times as large as found in the present observation. It seem that this great difference is caused by the difference of the number of ice crystals in a unit volume and by the movement of crystals in the water, that is, by the mode of conduction and convection of latent heat in the process of crystallization.

Summary

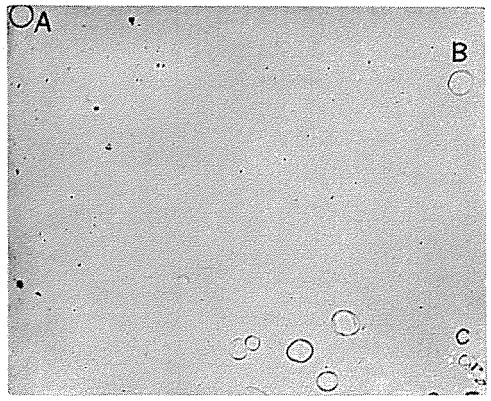
A new method of cinematography was devised by the authors, for taking pictures of the formation of ice crystals in water. By this, the successive stages were observed in the developing of ice

crystals. The conditions for giving a circular ice-disc, stellar crystal and needle-like crystal were studied. In every case, ice-discs were the slowest to be produced, stellar and especially needle-like crystal requiring less time as seen in Photos 19–23 and in Figs. 4–9. Some tiny particles as silver iodide, kaolin, carbon or clay were not directly active for nuclei of ice crystal in this experiment.

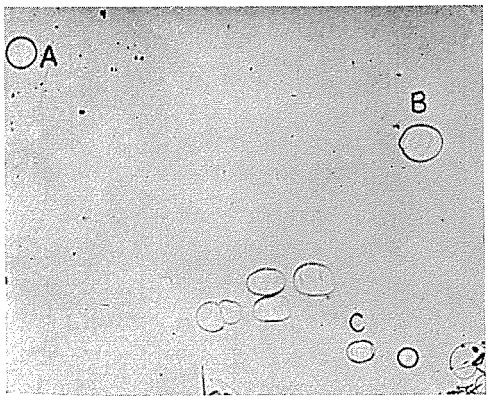
In conclusion, the authors wish to express their best thanks to Prof. U. NAKAYA, Asst. Profs. K. ARAKAWA and A. HIGASHI for their suggestions through this research.

References

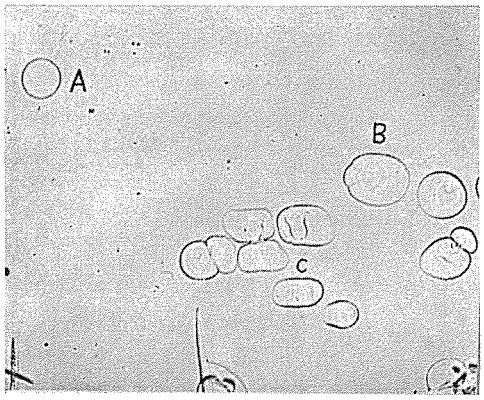
1) H. T. BARNES, *Ice engineering, Montreal,* (1926), 364.
2) W. J. ALTBERG, *Trans. of the Meeting of the Int. Commition of Snow and of Glaciers,* (1936), 373.
3) W. J. ALTBERG, *Acta Physicochimica U. R. S. S.* **9**, (1938), 725. **11**, (1939), 287.
4) V. J. SCHAEFER, *Trans. Amer. Geophys. Union,* **31**, (1950), 885.
5) I. IMAI, *J. of the Japanese Society of Snow and Ice,* **11**, (in Japanese) (1949), 10.
6) K. ARAKAWA, and K. HIGUCHI, *J. of the Fac. of Sci., Hokkaido University,* Series 2, **4**, (1952), 201.
7) V. J. SCHAEFER, *Chem. Rev.* **44**, (1949), 291.
8) R. SMITH-JOHANNSEN, *Final Report Project Cirrus RL* 140, *General Electric Research Laboratory,* (1948), 79.
9) R. GROSS, H. MÖLLER, *ZS. Phys.* **19**, (1923), 375.
10) J. LAMBOR, *Association International d'Hydorogei Scientifique Assemblee de Oslo,* (1948), 367.



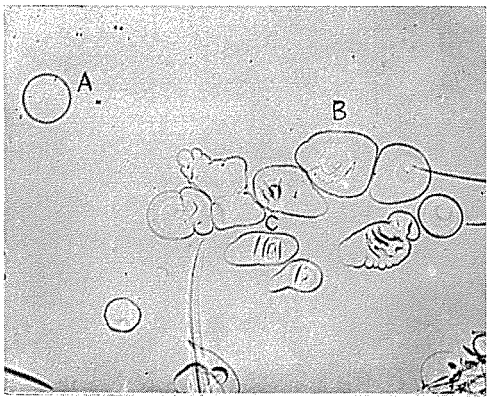
1. At 50 second after seeding × 3
Water temperature −0.05°C



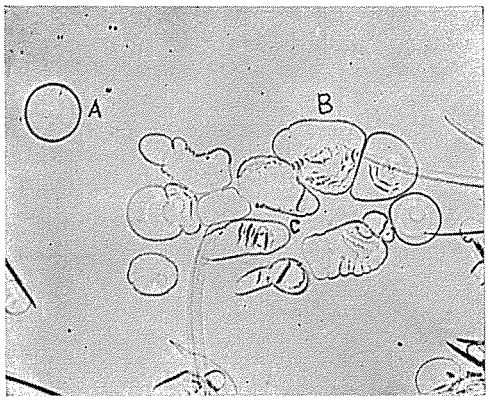
2. At 70 sec. − 0.05°C × 3



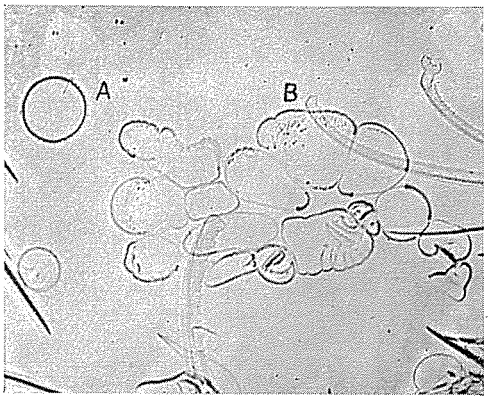
3. At 90 sec. − 0.05°C × 3



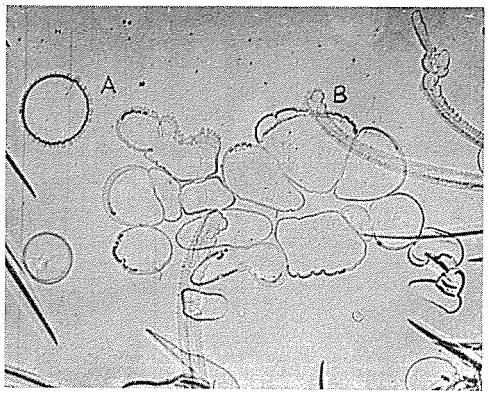
4. At 110 sec. − 0.05°C × 3



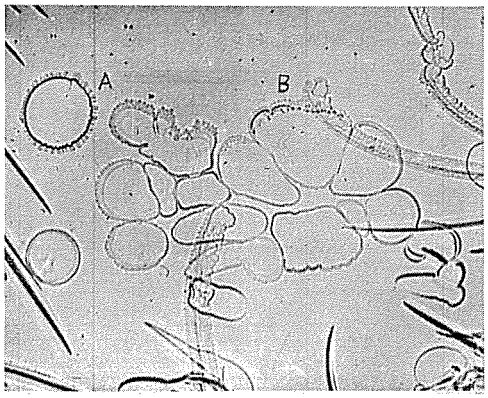
5. At 130 sec. −0.05°C × 3



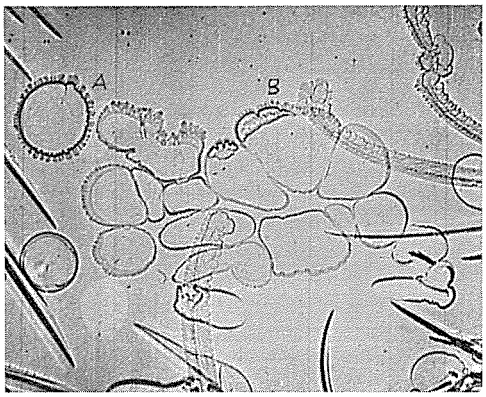
6. At 150 sec. − 0.05°C × 3



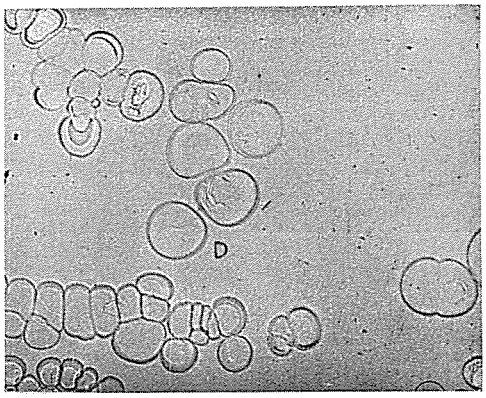
7. At 170 sec. −0.05°C × 3



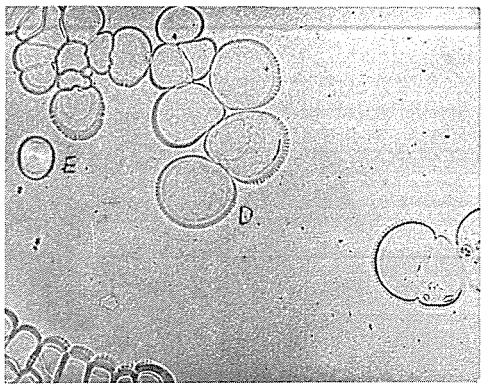
8. At 190 sec. −0.05°C × 3



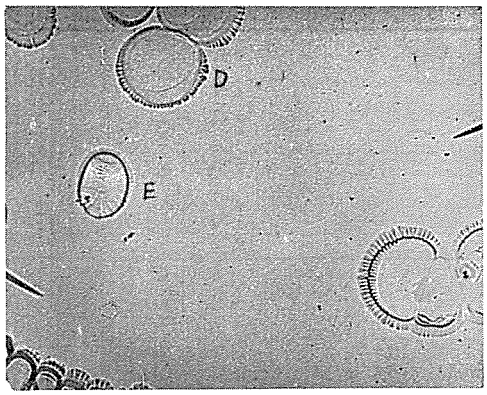
9. At 210 sec. −0.05°C × 3



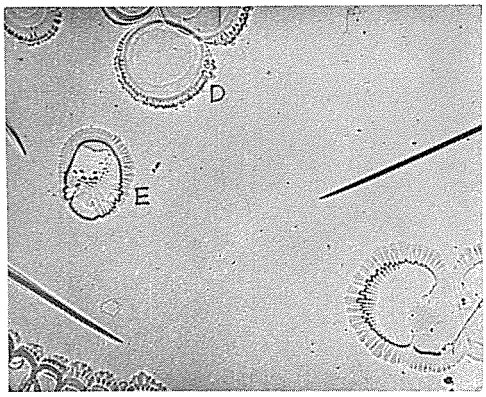
10. At 170 sec. −0.05°C × 3



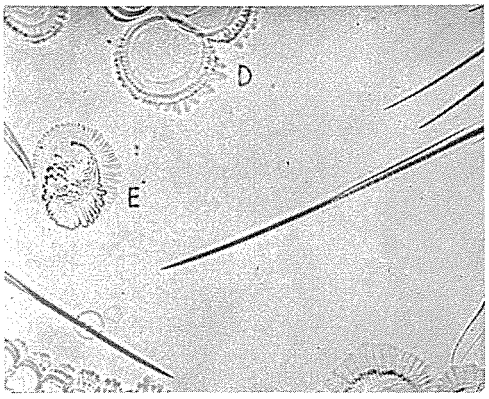
11. At 250 sec. −0.05°C × 3



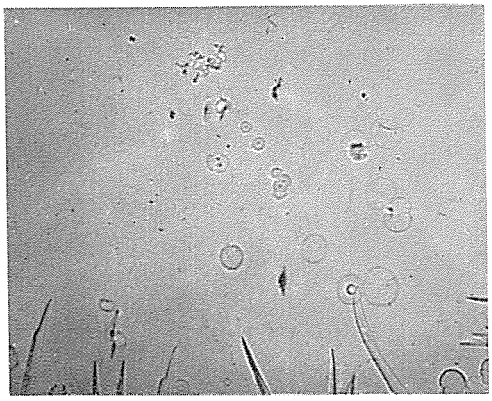
12. At 270 sec. −0.05°C × 3



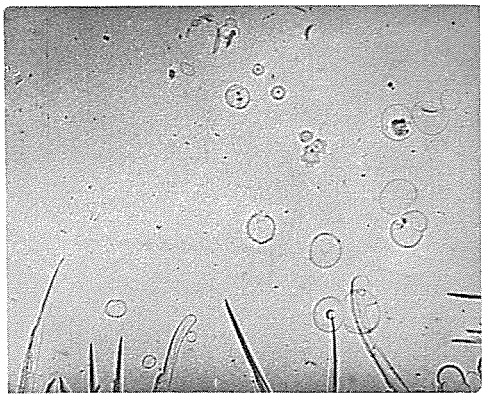
13. At 330 sec. −0.05°C × 3



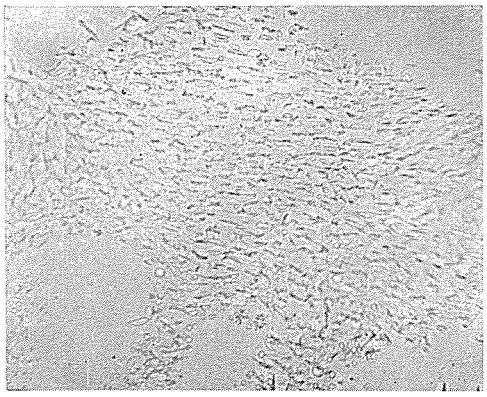
14. At 370 sec. −0.05°C × 3



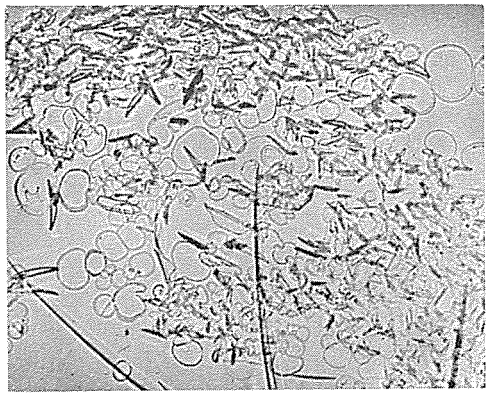
15. At. 50 sec. −0.05°C × 3



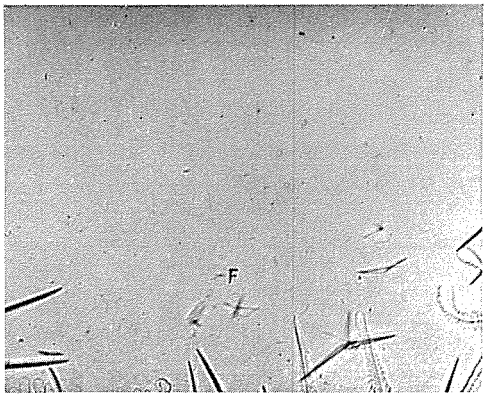
16. At 60 sec. −0.05°C × 3



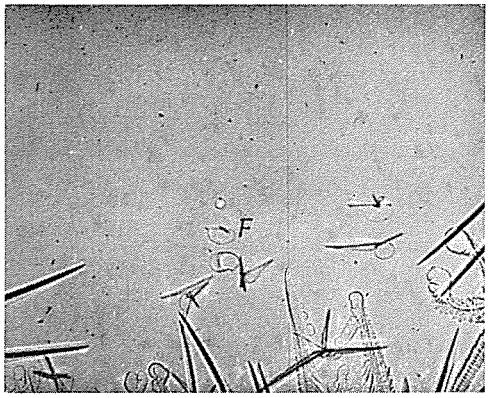
17. At 15 sec. −0.50°C × 3



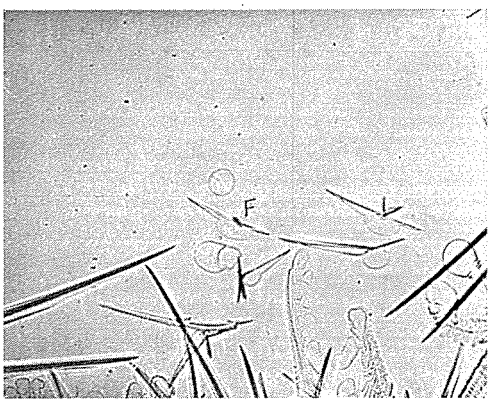
18. At 40 sec. −0.25°C × 3



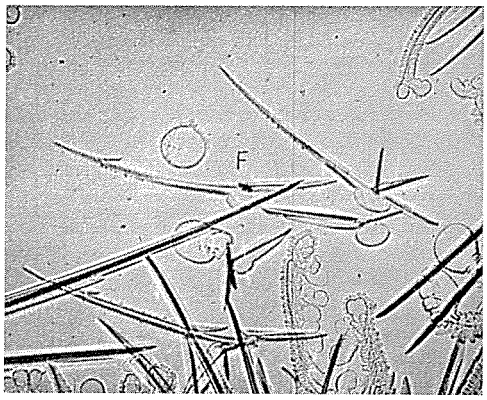
19. At 20 sec. −0.38°C × 3



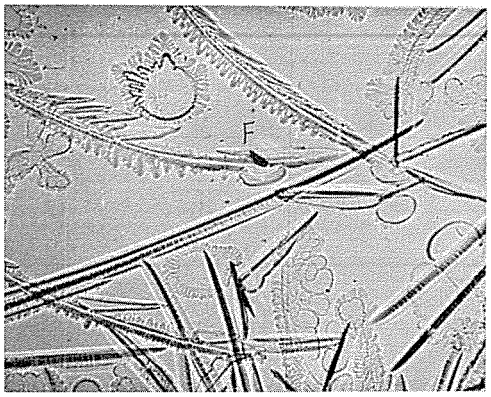
20. At 30 sec. −0.36°C × 3



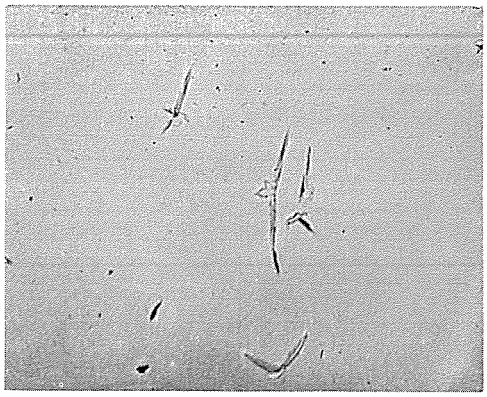
21. At 50 sec. −0.35°C × 3



22. At 80 sec. −0.29°C × 3

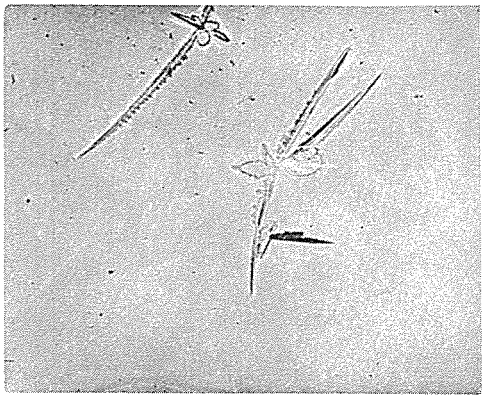


23. At 140 sec. −0.19°C × 3



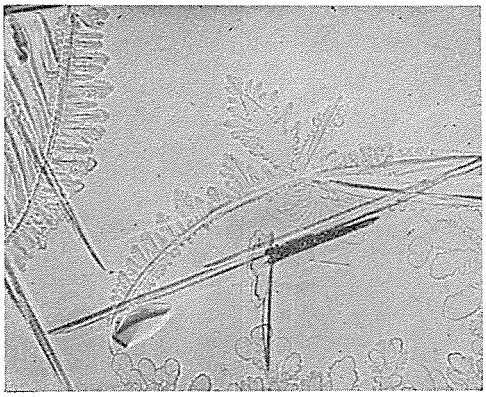
24. At. 25 sec. −0.65°C × 3

25. At 30 sec. −0.60°C × 3

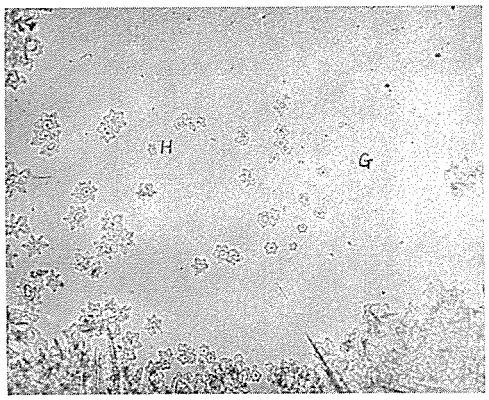


26. At 35 sec. −0.58°C × 3

27. At 55 sec. −0.45°C × 3

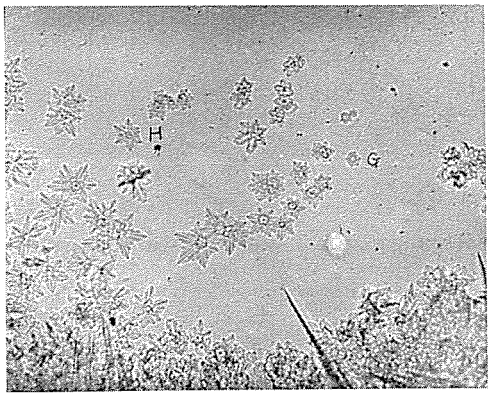


28. At 115 sec. −0.1°C × 3

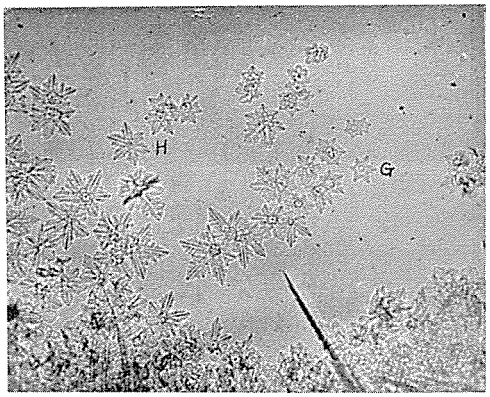


29. At 20 sec. −1.3°C × 3

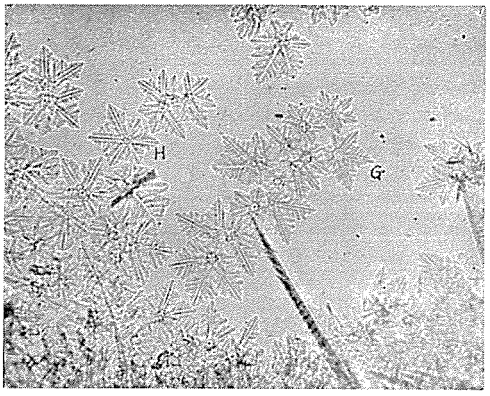
30. At 30 sec. × 3



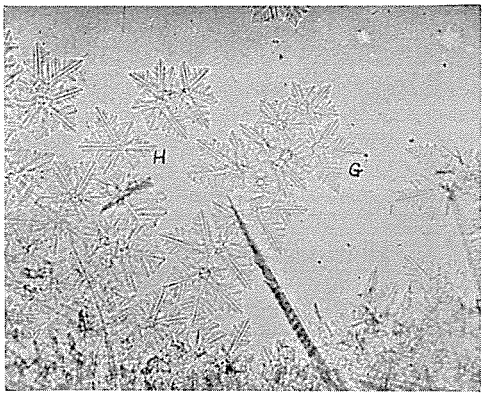
31. At 40 sec. × 3



32. At 50 sec. × 3



33. At 60 sec. × 3



34. At 70 sec. × 3