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Preliminary Experiments on the artificial Production of Snow Crystals.*

By

Ukitirô NAKAYA, Isonosuke SATÔ and Yatarô SEKIDO.

(Plates I–IV)

1. Introduction.

In the preceding paper, Investigations on Snow No. 6[1], the authors described the results of experiments on the artificial production of frost crystals with reference to the mechanism of the formation of snow crystals. In those experiments the authors attempted to infer the mechanism of producing various types of snow crystals from knowledge gained from the artificial formation of the corresponding frost crystals. In the spring of 1935 the authorities of the Hokkaidô Imperial University began to build a cold chamber laboratory and it was completed at the beginning of 1936. This laboratory is provided with a cold chamber $4\times4\times4$ metres in dimension, the whole chamber being designed to be cooled down to $-50°C$ by a refrigerating mechanism. The former experiments on the artificial frost crystals were carried on in this cold chamber and further the authors succeeded in producing a snow crystal which has six dendritic branches extending outwards almost symmetrically from a nuclear centre. The nucleus of this artificial snow crystal was suspended by being attached to a thin rabbit hair or cotton filament. Various types of snow crystals were produced by varying the temperature of the air where the crystal is made and that of the ascending water vapour. The results are described in this paper, although they are still of a preliminary nature.

2. The Apparatus.

Two sets of apparatus were used for this experiment. Apparatus No. 1 is almost the same as that used in the experiments described in report No. 6 and is shown in Fig. 1. B is a cooling vessel made of copper sheets and

* Investigations on Snow, No. 10.

1) U. NAKAYA and I. SATÔ, this Journal, 1 (1935) 206.

is filled up with alcohol, which is cooled down far below the freezing temperature, say, to −25°C, by using liquid air. This cooling vessel was used only when the apparatus was used in an ordinary room, the temperature of which was at nearly 0°C. When we use the apparatus in the cold chamber at nearly −30°C, this cooling vessel B is not necessary and it is

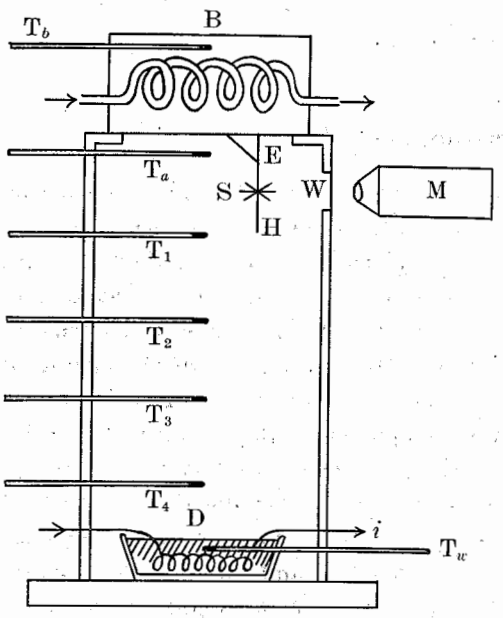


Fig. 1. Apparatus No. 1.

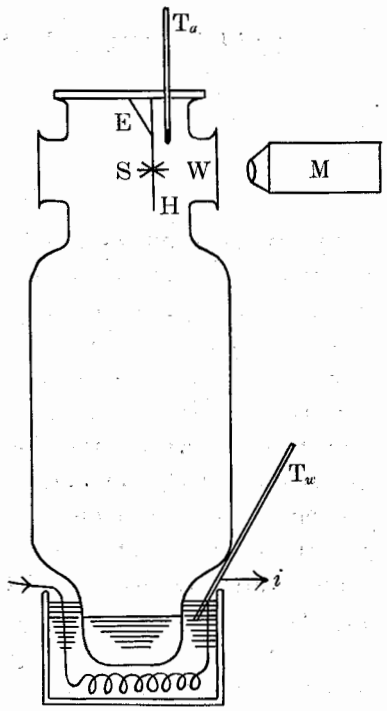


Fig. 2. Apparatus No. 2.

replaced by a simple sheet of copper.

The water vapour supplied from the shallow dish D, which is warmed up by an electric current *i*, condenses on the wedge E and develops in a crystalline form. This crystal is the same as the so called frost crystal. From the standpoint of the crystal habit, this frost crystal is the same as that of the snow, as described fully in report No. 5. In future this crystal will be referred to as "artificial frost".

Further we succeeded in making an ice crystal which has six branches extending outwards in an almost symmetrical manner from a nuclear centre. This sort of crystal was made by being suspended by a fine rabbit hair, which is shown by H in Fig. 1. We shall call this sort of crystal

"artificial snow" in future. These artificial frost and snow crystals are shown schematically in Fig. 3. These crystals were observed by a long focus microscope M through the window W.

The successive stages of the development of a crystal could be thus observed under a microscope without touching the crystal. In the case of taking the microphotographs of the successive stages of a crystal, the artificial snow suspended by a fine filament was not suitable, as the crystal was made to sway about by the ascending air current. Artificial frost, therefore, was chiefly used for this purpose.

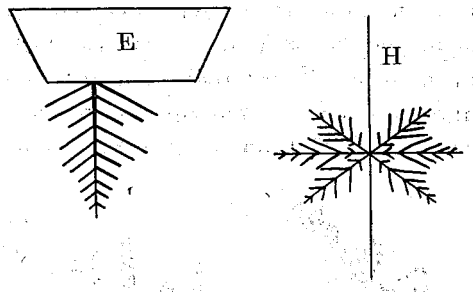


Fig. 3.

a) Artificial frost

b) Artificial snow

Apparatus No. 2 as shown in Fig. 2 was designed for experiments when very pure water or heavy water is used. The whole vessel was made of glass, and the principle is similar to that of No. 1, except for the method of heating the water. T_a and T_w are the thermometers for measuring respectively the temperature of the air where the crystal is made and that of the water in the reservoir. Slender alcohol thermometers were used which were specially designed for this purpose.

3. Supplementary experiments on artificial frost crystals, the room temperature being at nearly 0°C.

Experiments similar to those described in report No. 6 were carried on further with apparatus No. 1. In the former case the frost crystals detached from the cold ceiling had been placed on a cooled glass plate and subjected to a microscopic investigation. In the present case the crystal was observed with a microscope from outside without touching the crystal, so that the successive stages of its development were examined with ease.

In order to determine the condition, the temperature was measured at various places as shown in Fig. 1. One example is shown in Photo. 1. This is the state of the crystal one hour and ten minutes after the start of the experiment. In the former experiment, No. 6, we could obtain a crystal, the branches of which developed in a dendritic manner, but a close examination of a photograph, for example, Photo. 3 of No. 6, shows us that the minute structure of the ray of the crystal is different from that

of ordinary snow. In the case of natural snow two fine continuous canals are usually seen in the middle part of the ray, arranged parallel to one another and extending in the direction of the ray. In the ray of artificial frost, however, these canals were not usually observed in the former experiment and the structure of the ray looks like an assemblage of small flakes in the form of a fish scale, as will be seen in Photo. 3 of report No. 6. The difference in the structure is shown schematically in Figs. 4 A and B. These fine canals observed in the ray of a snow crystal were verified by the present experiment to be obtainable also in the case of artificial frost. One will see in Photo. 1 those canals in question. The conditions under which this crystal was produced are tabulated in Table I. The conditions controlling the formation of these different types of ray structure are not yet clear, and experiments are now in progress to clarify them.

A B

Fig. 4.

Table I.

	t=0, start	t=50 min.	t=1 h 10 min.
T_b	−18.5°C	−18.5°C	−20.9°C
T_a	− 8.5	− 8.6	−11.0
T_1	− 1.0	− 2.3	− 2.7
T_2	− 1.0	− 2.4	− 2.7
T_3	− 1.0	− 2.3	− 2.4
T_4	− 1.2	− 2.1	− 3.0
T_w	+ 8.3	+ 5.6	+ 5.5
s	3.69	3.09	3.81

s=supersaturation ratio, see report No. 6.*

As described in the former report, these frost crystals begin to fall down by themselves, giving the appearance of a snowfall. Formerly it was simply supposed that they become detached from the ceiling by their own weight, and the mechanism of this process was clarified by the present experiment. While observing continuously the stages of development of the frost crystals, it was found that the part near the root of the crystal

* s=The ratio of the saturation vapour pressure of water at T_w to that at T_a.

began to become attenuated. This stage can be clearly seen in Photo. 1. After this stage the attenuated part becomes thinner and thinner, so that after some time it appears that the crystal is suspended by a fine thread of ice, and then this thread is cut so that the crystal falls down by itself. One can see this final state in Photo. 2. In this case, T_w was $+11.5°C$, T_a $-7.5°C$, T_b $-12.3°C$, T_1 $-1.2°C$, T_2 and T_3 and T_4 were all $-1.3°C$. This phenomenon is to be explained as follows. After the crystals that grow in clusters develop to some extent, the water vapour drawn upwards by natural convection cannot get into the space near the roots of the crystals. On the other hand, as there exists a colder metal plate just above the frost crystals, the ice molecules of this part of the crystal sublimate and condense to the minute colder crystals which adhere to the metal plate. The surface tension of the fine edge of the crystal will also accelerate this process. From this result we were lead to conclude that in order to get a well developed crystal of ice, which is very like that of snow, we must remove this cooling box and carry on the experiment by setting the whole apparatus in a large cold chamber.

An interesting fact is observed in this experiment. The structure of the thread of ice above described is not simple and appears like an aggregate of somewhat rounded granules of ice, as will be seen in Photo. 2. Comparing this structure with that of powder snow observable in a snow storm, Photo. 46 of report No. 2, one will see a marked resemblance between the two. In this case the temperature of the air was $-7.5°C$ and it will be understood that this structure like an aggregate of rounded granules of ice, which is often observable in natural snow, can be produced by sublimation even when the temperature is far below freezing point.

4. Further experiments on artificial frost in the cold chamber.

As described in the preceding article, artificial frost can be developed to its full extent when the whole apparatus is set in a large cold chamber. We began to repeat the experiments in the chamber, which can be cooled down to $-50°C$ as described in the Introduction. In this series of experiments the room temperature in the chamber was kept usually between $-30°C$ and $-20°C$, and those who were engaged on them were protected against the cold by specially desigend clothes, boots and caps.

The first aim of this experiment was to observe the successive stages in the development of a crystal. One example of a series of photographs showing the manner in which a dendritic crystal develops is given in Photos.

3*a–d*. In this case apparatus No. 2 was used and the crystal was made to grow on a wooden edge. The water was distilled. T_r was kept at $-20°C$, T_w at $+20.5°C$ and the mean temperature of the air where the crystal was formed, T_a, varied between $-15.3°C$ and $-16.0°C$. Photo. 3*a* shows the crystal about 30 min. after the beginning of the experiment, and this time interval varies to a considerable degree according to the initial condition of the apparatus. The mode of growth of the crystal is shown in Photos. 3*a–d*, taking state 3*a* as the initial state, $t=0$. The rate of growth of the extremity of the crystal is 0.047 mm/min at the state between 3*a* and 3*b*, 0.044 mm/min between *b* and *c*, and 0.045 mm/min between *c* and *d*. In this case the minute structure of the ray of crystal belongs to the type B in Fig. 4, that is, the assemblage of scale-like flakes. The rate of growth of the ray of a crystal in dendritic form varies considerably with the condition of formation, but usually it can be taken as nearly 0.05 mm/min. These rates vary not only with the temperatures of the water and atmosphere, but also with the construction of the apparatus, that is, with the mode of the convection of water vapour. Further investigations on the rate of crystal formation will be described in a future publication. In the case of any crystal it is well known that a dendritic form is quickly formed in a sufficiently supersaturated condition while a crystal in solid form is developed slowly in a less supersaturation. In the case of snow also our crystallographic knowledge suggests that the dendritic form is the fastest to be produced, the plate form requiring more time, and the column or prism even more. The difference in the modes of formation of the dendritic ray and the plate form is clearly shown in Photos. 4 and 5, Pl. II. The plate form represented in Photo. 4 was made with the room temperature at $-25°C$, the temperature of the water reservoir at nearly $0°C$, the apparatus being left overnight. The rate of growth, therefore, is about one tenth that of the dendritic one. The latter type of crystal, shown in Photo. 5 for comparison, was made with T_a at $-16°C$ and T_w at $+10°C$, and the photograph shows the state nearly two hours after the beginning of the experiment. In this case the minute structure of the ray is like that of an ordinary snow crystal, and one can see the continuous canal along the middle line of the ray.

In our country dendritic crystals of snow, developed in a six-petalled flower, are very frequently observed, while plate ones are seldom seen. The plate crystals observable in our climate are very small as described in report No. 8. This is to be attributed to the excessive humidity of our climate. The minute plate is the early stage of the crystal which is made

near the earth's surface. Bentley's observations at Jericho show that relatively large plates are frequently observed there. It can be explained by assuming that in frequent occasions the atmosphere over that country is dry right from its upper layer down to the earth's surface.

Hexagonal plates with dendritic extensions at the corners are reproduced in Bentley's book in large numbers. They are also often observed in our climate. According to the view above described these crystals are made when the upper layer of the atmosphere is relatively dry and the layer near to the earth surface is sufficiently humid. Artificial production of this type of crystal was attempted in the following way. The temperature of the water reservoir was kept at nearly $+5°C$ and the hexagonal plate was obtained; then the temperature of the water was raised to about $+15°C$ by increasing the heating current, so that we could expect dendritic extensions at the corners. In this case it was found that the excessive humidity must be supplied quite suddenly, otherwise the crystal develops in a quite different form from the so called "hexagonal plate with dendritic extensions at the corners" observable in the case of natural snow. Actually a screen was inserted above the water reservoir after the formation of the plate, then the temperature of the water was raised to the required value and the screen was removed, so that the plate crystal was exposed suddenly to the excessively humid air. From this result it will be supposed that the boundary of the dry and humid layers in the atmosphere is fairly discontinuous when this type of snow is falling. One example of artificial frost of this type is described in article 6.

5. Artificial snow crystal made in the cold chamber.

The artificial snow crystals were produced by suspending the germs* of crystal on a fine filament. It was fairly difficult to attach one or more isolated germs to the filament. When we put the filament in the ascending air current supersaturated with water vapour, the latter condenses on the filament as frost crystals. Usually these crystals are distributed all over the filament, giving an appearance as of a hairy caterpillar. In order to avoid this we chose after some trials a fine rabbit hair which was thoroughly dried in a dessicator. Using this filament we could make a few germs attach themselves to the filament and develop artificial snow from these isolated germs. Some other technical precautions are necessary for the reproducibility of this experiment. The theory and practice for getting

* The very early stage of a snow crystal; see the following report No. 11.

an isolated germ of ice crystal on the filament are described in full detail, together with the effect of the nature of the filament, in the next communication.

Photo. 6 shows the mode in which an artificial snow crystal of a dendritic type develops on the filament. In this case the plane of the crystal is nearly parallel to the filament, but in many cases the crystal grows in a plane perpendicular to the filament. The crystal shown in Photo. 6 was obtained in apparatus No. 2 with T_a at $-16.5°C$ and T_w between $+18°C$ and $+20°C$. This photograph shows the state of the crystal 50 min after setting the filament in the apparatus. Photo. 7 is a typical dendritic crystal, as made in apparatus No. 1. The crystal was made in two hours and during that interval T_a was nearly $-16°C$ and T_w varied between $+6°C$ and $+8°C$, the supersaturation ratio being nearly 7.5. The inner structure of the ray is exactly the same as that of the ordinary dendritic snow crystal of a fern-like type. The crystal shown in Photo. 8 was also made in apparatus No. 1. In this case a cotton filament was used and this crystal was obtained at the tip of the filament. T_a was nearly $-16°C$ and T_w varied between $+7.6°C$ and $+9.0°C$. This is quite similar to the plane crystal of snow which is described as the broad branch type in report No. 8. A plane crystal of snow with three branches is sometimes observed in natural snow, and it was made clear that this type is produced when a crystal develops from two nuclei arranged one close on top of the other and the crystal is separated afterwards, as described in full detail in report No. 7. A similar crystal is also obtained in the case of artificial snow. One example is shown in Photo. 9, which is made in apparatus No. 1 with T_a at nearly $-17°C$ and T_w at $+6.5°C$. The supersaturation ratio is nearly 7 in this case.

Decreasing the rate of the supersaturation of water vapour, the dendritic crystal above described were transformed into a plate type. Under the same condition as the plate crystal a cup crystal was sometimes obtained, which type is so often observed in the crystals of depth hoar; that is, a sublimation product growing beneath the surface of the fallen snow. The microphotographs of the depth hoar can be seen in Seligman's book[1]. One example of this cup crystal is reproduced in Photos. 10 *a* and *b*, Pl. III, *a* being the front and *b* the side view. This crystal takes the form of shallow hexagonal dish. In this case T_a was nearly $-15°C$ and T_w $0°C$, the surface of the water in the reservoir having been covered

1) G. SELIGMAN, Snow Structure and Ski Fields, London, 1936, pp. 62–65.

with a thin sheet of ice. The supersaturation ratio was nearly 3.3. This cup was found on the wedge in apparatus No. 1, the bottom of the cup adhering to the surface of the wedge. This photograph shows the state of the crystal about 7 hours after the beginning of the experiment. In the same experiment it was noticed that the cotton filaments hanging down form the wedge were coated with minute crystals of ice. The microphotograph of the same, Photo. 11, shows that those minute crystals are an assemblage of columns and pyramids. In this case the given amount of water vapour that was supplied to the neighbourhood of the filament, was distributed among so many crystals that the rate of growth was particularly slow. After seven hours they attained an average length of only 0.3 mm. This type of snow observable in nature is also very small in size as described in report No. 8.

The thick plate type of snow crystal is frequently observed in our climate. As described in report No. 2, it is produced when many water droplets, cloud particles, attach themselves to a plane crystal of snow. A similar type is also obtained in the case of our artificial snow. One example is shown in Photo. 12. In this case the temperature of the water was very high and some of the water vapour brought up by natural convection condensed as fog and attached itself to the crystal. The process is almost the same as the supposed mechanism of formation of this type of natural snow. The crystal shown in Photo. 12 was made in apparatus No. 2 with T_a at $-16.3°C$ and T_w at $+24°C$. It took two hours for the crystal to reach the state shown in the photograph.

Summarizing the results of the experiments above described it may safely be stated that almost every sort of snow crystal can be made in the laboratory. The chief factor governing the formation of various types of snow is the rate of crystal growth and the latter is strongly influenced by the supersaturation ratio. The temperature of the air where the crystal is produced was found in the present experiment to be also of great importance in determining the crystal habit of snow. Besides, the mode of convection of the water vapour also has some noticeable influence. The argument given in report No. 6, to the effect that the form of the crystal is chiefly determined by the supersaturation ratio, was found to be insufficient. These points will be discussed in detail in a future publication.

6. Artificial frost and snow made with heavy water.

Similar experiments to those just described were repeated with heavy water. The heavy water was supplied by the Norsk Hydro-Elektrisk

Kvelstofaktieselskaf of Norway and the content of D_2O was 99.6%. Apparatus No. 2 was used throughout. Heavy water weighing altogether 5 gr was used and it was adequate in bulk for many beautiful crystals to be obtained. No essential difference was observed between the characteristics of crystals made from heavy water and those made from ordinary water. As the melting point of heavy water ice is $+4°C$, the temperature of the water reservoir in the case of heavy water had to be raised a few degrees than is necessary in the case of ordinary water, in order to obtain a similar form of crystal. Using the heavy water, the inner structure of the dendritic branch was liable to take the form of an assemblage of scales, that is, type B in Fig. 4. Photo. 13*a*, made when T_a was $-17°C$ and T_w $+25°C$, is a typical example showing the inner structure like an assemblage of scales. A close examination of a crystal of this type revealed that it was made by the overlapping of one piece of flake on another. The side view of the crystal shown in Photo. 13*a* is reproduced in Photo. 13*b*, which shows nicely the mode of overlapping of component flakes. In the case of type A in Fig. 4, which has fine continuous canals along the middle line of the ray, the whole crystal or at least the main ray grows in one plane.

A crystal of a hexagonal plate with dendritic extensions at the corners was made with heavy water. The process is the same as in the case of ordinary water described in article 4. A hexagonal plate is produced as shown in Photo. 14*a*, Pl. IV, which was made with T_a at $-19°C$ and T_w at $+9.7°C$. Then the temperature of the water in the reservoir is quickly raised to $+22.5°C$. The temperature of the air where the crystal is made rises to $-18°C$, owing to the excessive warm vapour supplied from below. Taking the stage shown in Photo. 14*a* as the initial tsate, $t=0$, dendritic extensions begin to start from the corners of the plate and after 10 min the latter assumes the form shown in Photo. 14*b*. The dendritic rays develop rather quickly under these conditions and after 40 min the crystal takes the appearance shown in Photo. 14*d*, through the stage represented in Photo. 14*c*, which shows the tsate when $t=25$ min. The inner structure of the ray in this case is also type B, that is, an assemblage of scale flakes.

The A type of ray structure is only obtained with difficulty in the case of heavy water, but it can be produced. Photo. 15*a* shows two sorts of artificial snow crystal made with heavy water. The upper crystal belongs to a plane dendritic type and the lower is a spatial assemblage of dendritic branches. In this case the inner structure of the ray belongs to type A and the fine continuous canals can be clearly seen along the middle

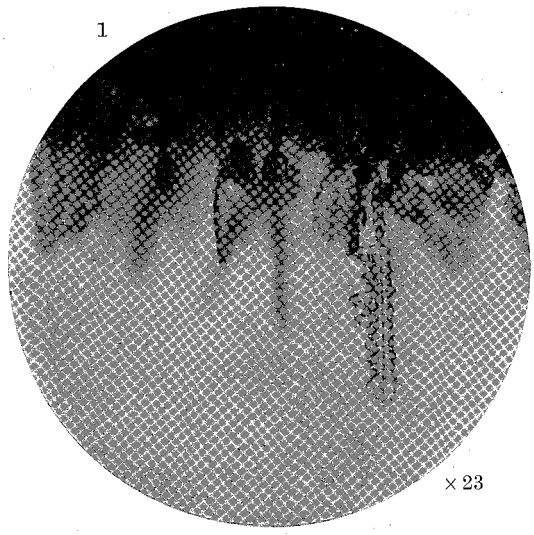
line of the ray. T_a was $-15.8°C$ and T_w $+25.2°C$ and the crystal developed to this state in 25 min. The lower crystal was detached after taking the photograph and the upper one was made to grow under the same conditions. Photo. 15*b* shows the state of the crystal 50 min after the beginning of the experiment.

Summary.

An apparatus was designed, in which water vapour was brought up by natural convection to a cooled space where it condensed into frost crystals. The effect of sublimation was discussed in the case of these artificial frost crystals. A similar experiment was carried out in a cold chamber which can be cooled to $-50°C$. Conditions for giving a plate or dendritic type of crystal were studied and the mode of development of a dendritic crystal was examined by taking microphotographs of successive stages during its formation.

Artificial snow crystals were produced in a similar manner, by suspending the germ of the crystal with a fine rabbit's hair. Almost every sort of snow crystal observable in nature was produced in the laboratory. The relation between the rate of growth and the form of the crystal was examined. Similar artificial snow crystals were made with heavy water. They were almost the same in form as those made with ordinary water.

In conclusion, the authors wish to express their best thanks to Nippon Gakuzyutu-sinkôkai for their financial assistance in this research.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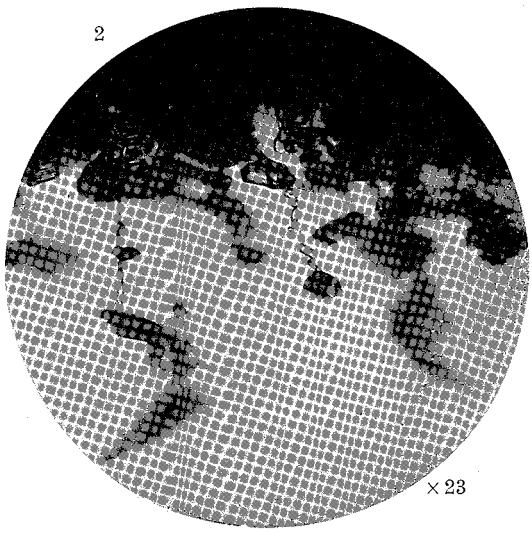
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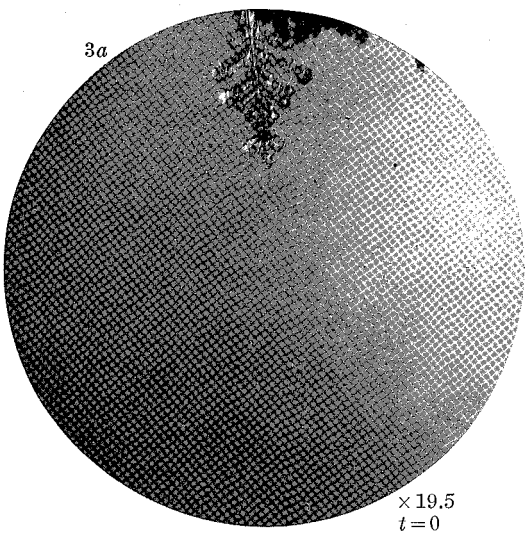
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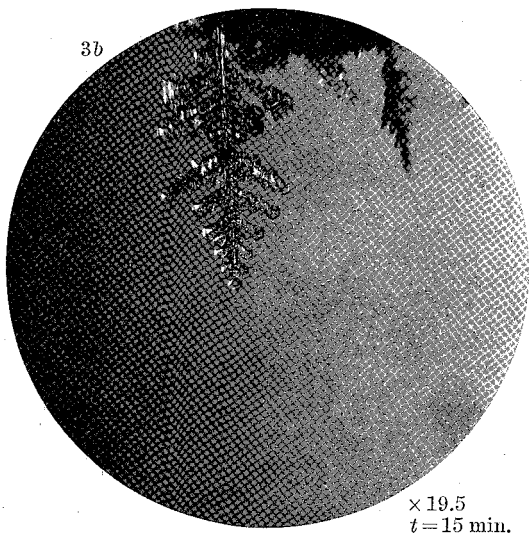
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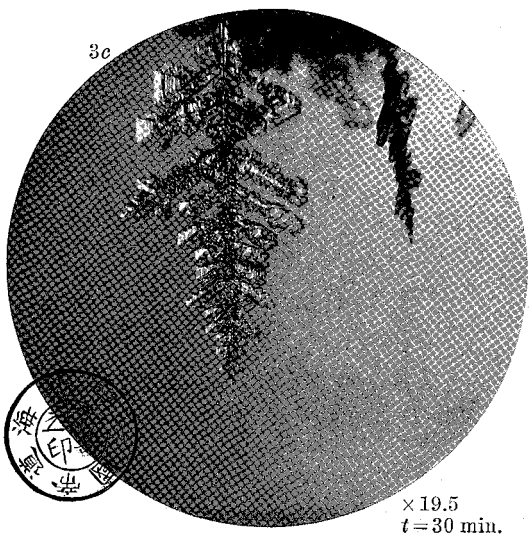
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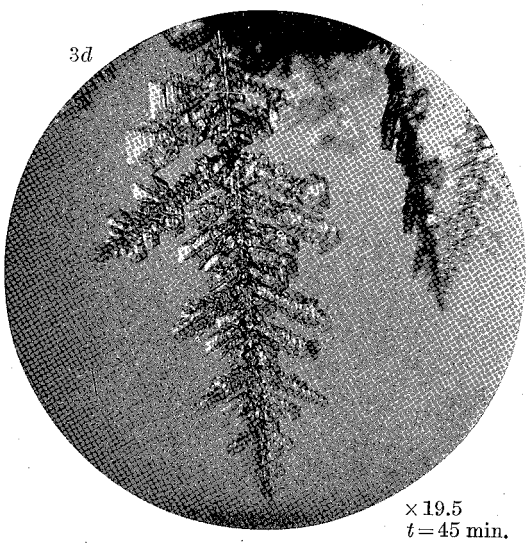
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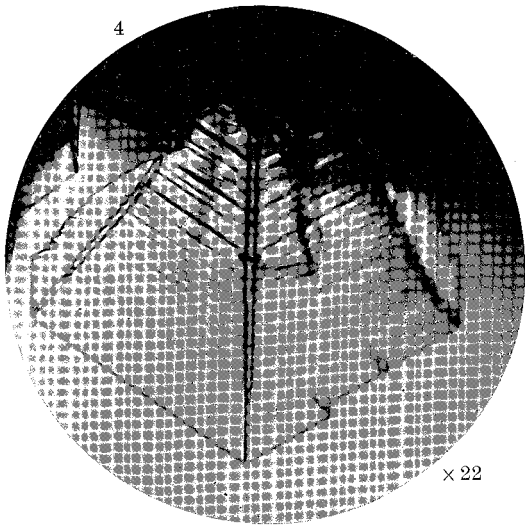
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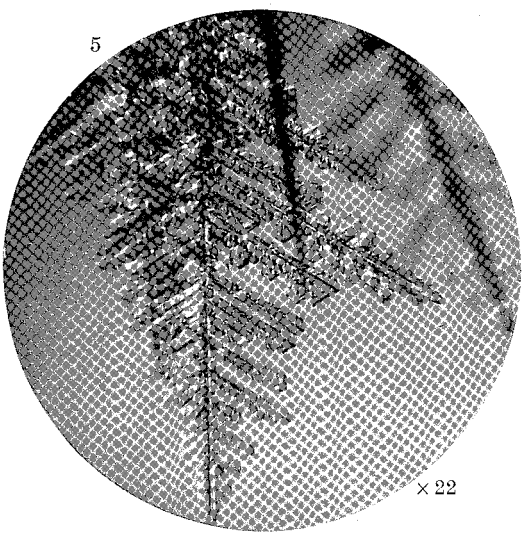
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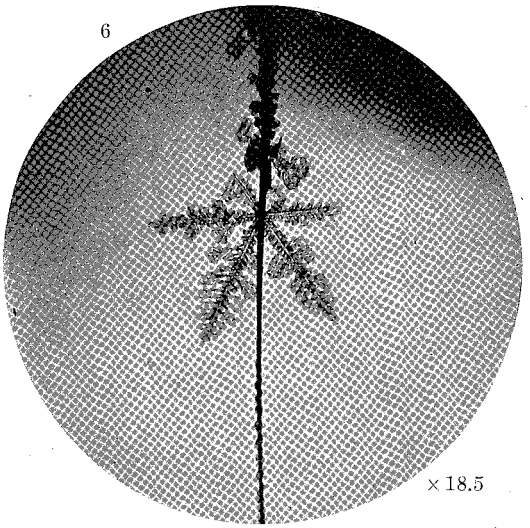
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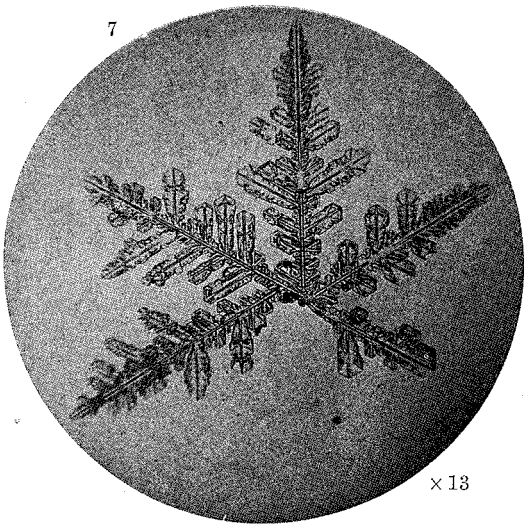
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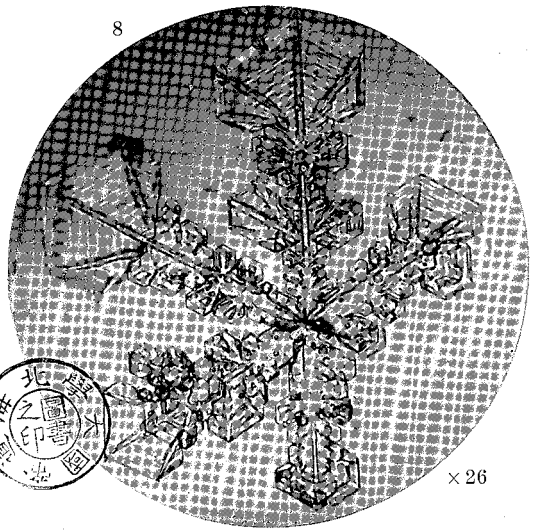
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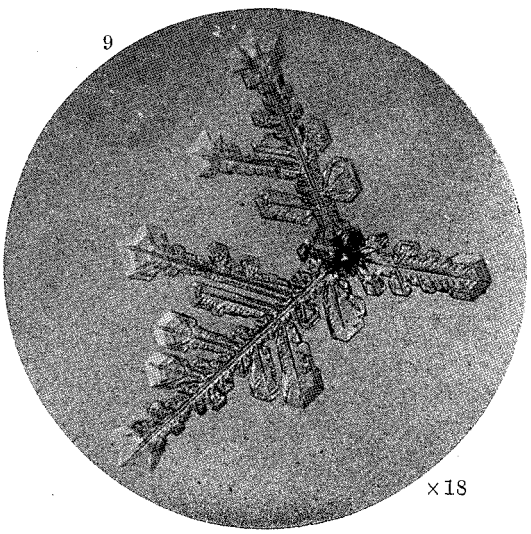
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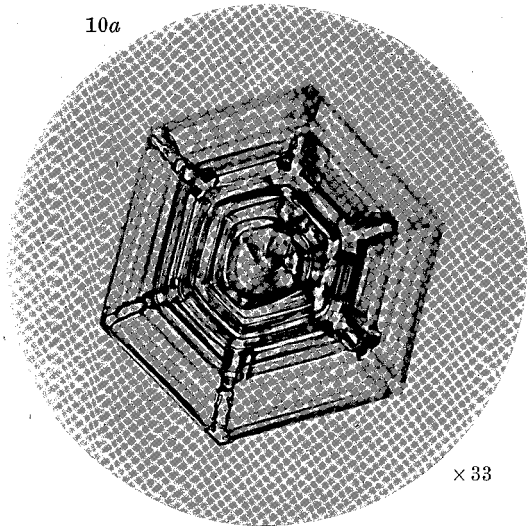
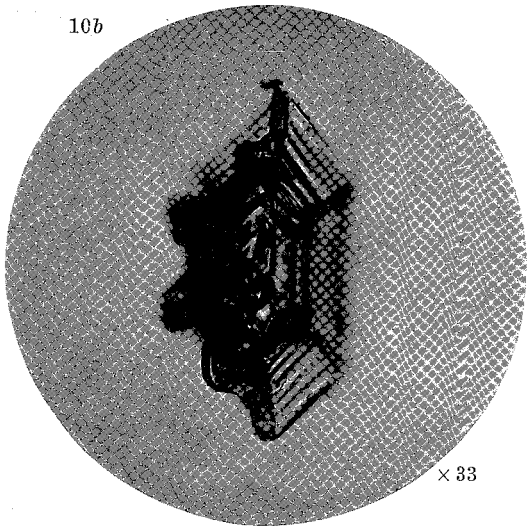
北海道帝國大學圖書之印

10*a*



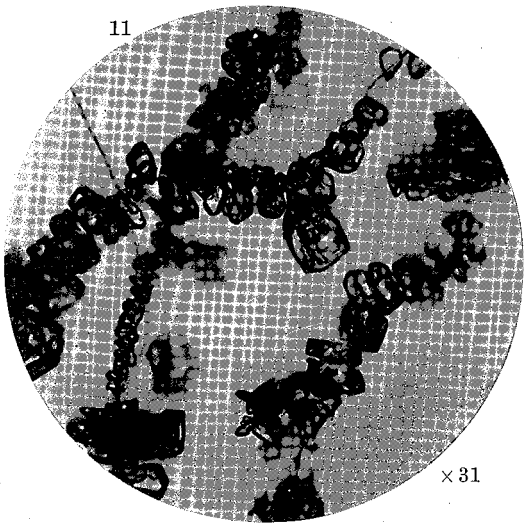
×33

10*b*



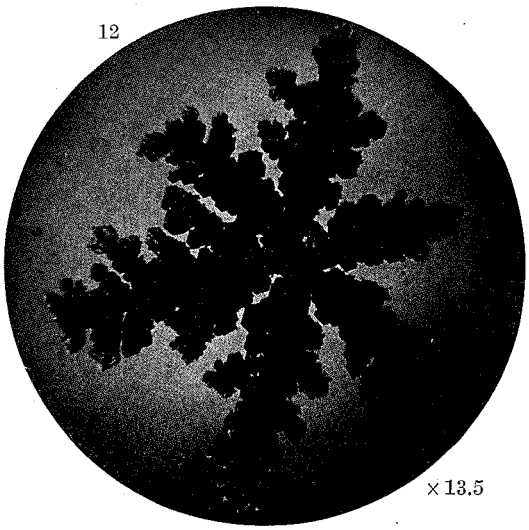
×33

11



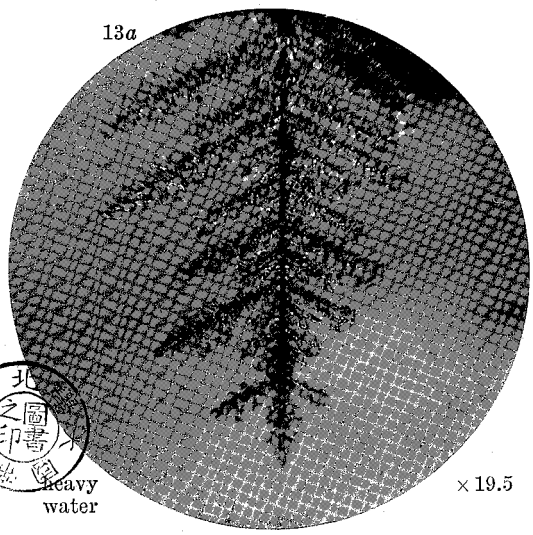
×31

12



×13.5

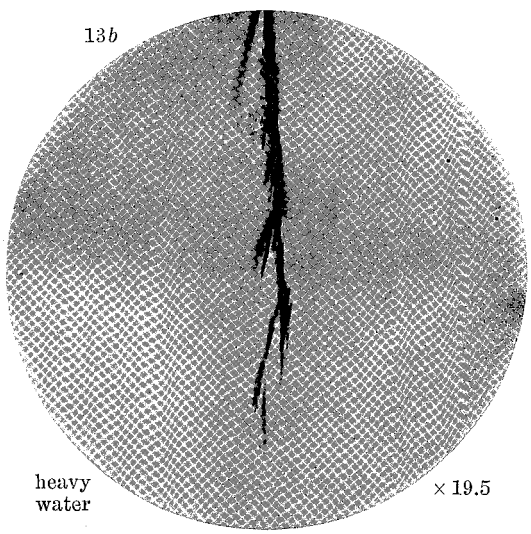
13*a*



heavy water

×19.5

13*b*



heavy water

×19.5

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