On the Artificial Production of Frost Crystals, with Reference to the Mechanism of Formation of Snow Crystals.*

By

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(Plates I–III)

I. Introduction

In a preceding paper[1] one of the authors remarked that the crystal habits of snow and frost crystals are very similar with each other so that the mechanism of producing a copious variety of snow crystals may be inferred to some extent from the knowledge about the formation of the corresponding frost crystals. In our previous study on the velocity of the fall of individual snow crystals,[2] it was found that they usually fall very slowly; for example, a dendritic plane crystal takes about one hour in falling 1 km. distance, assuming that the crystal keeps throughout the final state observed on the earth surface. At a higher altitude, however, the dimensions of the crystal must be much smaller than those observed on the earth surface, so that the time required for the development of the crystal will be longer than that mentioned above.

In the case of artificial production of snow crystals, the difficulty exists in suspending the crystal in air for such a long while. One way for attacking this problem is to consider the question of crystal development separately from that of the nucleus formation. The former point is treated in this paper. ADAMS[3] succeeded in making columnar crystals of snow in the laboratory by mixing a cold air current with a warm and wet one. He received the nuclei thus formed on a cold glass plate and made them grow to columnar crystals of dimensions observable under a microscope, by letting a stream of wet air pass over them. Unfortunately the conditions under which they were produced are not clearly stated. In the present experiment, the water vapour evaporated from a water surface was

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* Investigations on Snow, No. 6.

1) U. NAKAYA, this Journal, **1** (1935).

2) U. NAKAYA and T. TERADA, Jr., this Journal, **1** (1935).

3) J. M. ADAMS, Phys. Rev., **35** (1930) 113. Proc. Roy. Soc. A, **128** (1930) 588.

brought up by natural convection and deposited as frost crystals on the bottom surface of a metal box cooled with liquid air. By varying the temperature of the box and that of the water, various forms of frost crystals were produced which correspond to the various types of snow crystals.

II. Experimental Procedure

As the first series of experiments, an attempt was made to produce frost crystals on a cooled glass plate by letting a warm and moist air current pass over it. First a window frost was formed on the plate and when the supply of water vapour was continued, minute frost crystals grew from the base frost projecting upwards into the air. These crystals could not be developed to a size suitable for the study of their crystal habits. Then this apparatus was left to be used chiefly for the investigation of the window frost, the result of which will be reported later in a separate paper. The apparatus was then modified to bring the water vapour to the cooled space by natural convection, which diagram is shown in Fig. 1. In the Figure, A is a closed box made of copper sheet filled up with alcohol. A copper tube for passing the cold air evaporated from liquid air reservoir is wound in a spiral form and immersed in the alcohol. B is a wooden box for containing the copper vessel A, it lacks a bottom plate, the space between A and B being filled up with cotton.

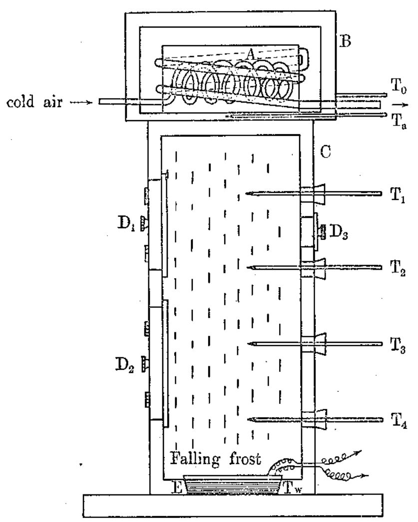

Fig. 1.

This cooling vessel is placed on the top of a tall wooden box C, the front and back sides of which are made of glass, with three doors D_1, D_2, D_3 put on the side walls. This box C lacks a ceiling, so that the bottom plate of the copper vessel A is exposed to the air inside the box C. A dish E is placed in the box C and it is filled with pure water, in which a heating coil is immersed for the purpose of regulating the temperature of the water. For measuring the temperatures of various places seven thermometers are used in total; T_0 for measuring the temperature of the copper vessel, T_a that of the air just outside the bottom of the vessel, T_w immersed in the water for measuring its temperature and T_1-T_4 for those of the air at various points in box C. The room temperature was usually between 0°C and 3°C.

For producing the frost crystals, the vessel A is gradually cooled down to a certain value by passing the cold air, and after T_a reaches a desired value the current of the cold air is regulated to keep it constant. Then the temperature of the water in E is raised to a required value. After waiting a little while, beautiful frost crystals begin to develop on the bottom of the cooled vessel all hanging down in the air. Keeping this condition still longer, fully developed crystals are detached by their own weight and fall down in the box C. These falling frost crystals are usually very numerous and they give an aspect just similar to a natural snowfall. As a matter of fact this phenomenon may be called an artificial snowfall, if the question of nuclei is not taken into consideration. These falling crystals were received on a glass plate which had been previously well cooled. They were then examined under microscope. The crystals which adhered to the bottom plate of the cooling vessel were also detached mechanically with a cold glass plate and brought under a microscopic investigation. These crystals are marked "adhered" in the later description.

III. Crystal Habits and Degree of Supersaturation

The point of this experiment is that the water vapour evaporated from the warmer water surface is brought up to the cold space by natural convection and condensed into frost crystals by sublimation. Two examples of the temperature distribution in the system are shown in Table I, in which it is seen that the intermediate space between the cold spot and the water reservoir is kept at nearly room temperature in both cases when the temperature of the water is higher or lower than the room temperature.

Table I.

	No. 29 Needle Type	No. 46 Plate Type
T_a	−14.0°C	−11.4°C
T_1	1.8	2.4
T_2	2.7	3.0
T_3	3.0	3.0
T_4	3.1	2.7
T_W	11.0	1.1

Even when the temperature of the water is much higher, being 11°C in case of the example cited, no fog is observed in the intermediate space, so that the vapour evaporated from the surface of the warm water must be brought up in a supersaturated state and condensed into a crystal form in the cold space T_a. In this series of experiments attention was paid chiefly to the rate of supersaturation. The ratio of the saturation vapour pressure of water at T_w to that at T_a was chosen as representing the rate of supersaturation and it was called supersaturation ratio s. The difference between the saturation tensions at T_w and T_a may be taken as another indicant, but the result of the present experiments showed that the former was more suitabe for the indication of supersaturation in the discussion of its bearings upon the habits of crystals. It was found that the element of primary importance to influence the formation of various types of crystals was the supersaturation ratio s above described while the temperature of the air where crystals are formed was secondary, though it also influenced the matter not a little. In this preliminary report the discussion is limited to the relation between the supersaturation ratio and the crystal habits, the other elements being disregarded for the present.

When the supersaturation ratio is large the crystal develops into a needle type, but with small supersaturation it tends to gnow in a form of a prism or a pyramid. The plane type, either in dendritic or plate form, is obtained with a supersaturation ratio lying between those for needle and prism. The form of the crystals, the temperature of air where the crystal is formed and the supersaturation ratio are tabulated in Table II for thirty examples.

1) Needle type. Crystals of needle type are formed with the supersaturation ratio between 6 and 8, the mean for five examples being 7.52. Two examples are reproduced in Photos. 1 & 2, Plate I. It is seen in the photographs that the needle is composed of pillars growing in parallel with each other. This characteristic is also observed in case of the natural snow crystal as may be seen in Photos. 27 & 28 of the previous paper reporting physical investigations on snow, Part II.[4] It seems that there are two kinds of needle snow crystals; the one is an elongated prism and the other an assemblage of pillars, the latter being more frequently observed in our climate. In Hokkaido the needle snow crystals fall chiefly when the temperature is comparatively warm and the air is very humid. Mr. HATAKEYAMA, the director of Toyohara Magnetic Observatory in South

4) U. NAKAYA and K. HASIKURA, This Journal, Vol. 1 (1934) 163.

Saghalien, observed the similar weather condition for the occurrance of needle crystals. The result of the artificial production of this type of crystal in laboratory agrees well with these observations for natural snow.

2) Dendritic form. BENTLEY and HUMPHREYS[5)] consider that the columnar crystal of snow is produced by a slow process at a high altitude where the humidity is comparatively small, while the dendritic form is obtained in a humid region near the earth surface. Many others also agree with this view, which is concordant with our present knowledge on crystal habits, but no supposition has been proposed for the order of supersaturation that is prevalent in case of the crystal formation. In the present experiment the dendritic form was obtained with the large supersaturation ratio next to needle crystal, the mean of four examples being 6.57. Two of these crystals are shown in Photos. 3 & 4, Plate I.

3) Intermediate form between dendritic and plate crystals. Decreasing the supersaturation ratio, the dendritic form tends to turn into a plate form, as expected from our knowledge on crystal habits in general. There are of course intermediate states between the two extremities, two examples of which are shown in Photos. 5 & 6, Plate II. The range of the supersaturation ratio is fairly wide, being between 3.5 and 5.5 as shown in Table II. In the photographs one may see clearly the nature of the crystal form belonging to this category.

4) Plate form. The supersaturation ratio for the formation of plate crystals ranges between about 3 and 4. The mean of six examples cited in Table II is 3.45. A complete form of hexagonal plate was very rarely formed and only one example was observed during this series of experiments, the microphotograph of which is reproduced in Photo. 7*a*. By eye observation under microscope it was noticed that a thin strip of frost crystal was attached to nearly the centre of the plate perpendicularly to the plane of crystal. schematical sketch is shown in Photo. 7*b*. Unfortunately this strip melted off before any good photograph was obtained. It is very natural to suppose that the tip of a strip of frost hanging down from the ceiling was a minute hexagonal prism or plate and that this complete hexagonal plate developed perpendicularly to the strip having this tip crystal as a nucleus.

The condition above described is very rarely fulfiled and usually a part of a hexagonal plate is produced, two examples of which are reproduced in Photos. 8 & 9, Plate II. Comparing Photo. 9 with the photo-

5) BENTLEY and HAMPHREYS, Snow Crystals, New York, 1931, p. 8.

Table II.

		No. of Photo.	Ta C	Tw C	s	State of Ocurrence	Mean of s
Needle Type	1	25	− 5.2	21.0	6.31	adhered	7.52
	2	26	−14.3	13.0	8.51	falling	
	3	27	−14.3	12.9	8.45	,,	
	4	28*	−14.1	10.5	7.08	,,	
	5	29*	−14.0	11.0	7.26	adhered	
Dendritic Form	1	40*	−22.3	0.1	7.46	adhered	6.57
	2	42	−21.5	0.9	7.33	,,	
	3	51*	−15.8	4.3	5.42	,,	
	4	52	−17.0	4.3	6.07	falling	
Intermediate Form	1	34	− 8.8	11.5	4.70	falling	4.40
	2	35	− 7.0	11.0	3.89	adhered	
	3	43*	−13.2	1.0	3.38	,,	
	4	47*	−14.5	4.0	4.71	falling	
	5	48	−16.0	4.0	5.41	adhered	
Plate Form	1	44	−12.5	1.0	3.17	adhered	3.45
	2	45*	−12.0	1.0	3.03	,,	
	3	46*	−11.4	1.1	2.89	,,	
	4	54	−12.3	4.1	3.88	,,	
	5	55	−12.0	4.2	3.80	falling	
	6	66*	− 4.5	14.0	3.82	,,	
Pyramidal Form	1	67*	− 3.0	5.8	1.94	falling	2.21
	2	75*	−10.1	0.0	2.37	,,	
	3	79	− 9.5	0.4	2.32	,,	
Prism	1	60*	− 5.5	−0.1	1.58	falling	1.66
	2	63*	− 5.6	0.0	1.60	,,	
	3	64	− 5.3	0.2	1.59	,,	
	4	80*	− 5.5	0.7	1.67	,,	
Prism with extended side plane	1	18*	− 2.3	0	1.21	adhered	1.59
	2	70	− 2.5	5.4	1.81	,,	
	3	71*	− 2.2	5.2	1.74	,,	

* Reproduced in the Plates.

graph of a natural snow crystal, for example Photo. 15 of the previous report Part II, no essential difference is observed between these two crystals either in their forms or in their structures. Accordingly it may not be unreasonable to guess that the most part of this snow crystal is formed under the condition of $s=3.5$.

5) Pyramidal form. A simple pyramidal form of snow crystal is known but it is seldom observed. In laboratory the frost of this type is not difficult to produce. The authors observed three examples during this series of experiments, two of which are reproduced in Photos. 10 & 11*a*, Plate III. Photo. 10 is a good example showing a side view. Photo. 11*a* is a half melted state of the crystal looked down from above. By eye observation this crystal was known to have taken the form shown schematically in Photo. 11*b*, before it began to melt. The supersaturation ratio for the production of this form of crystal was 2.21 as a mean of three examples.

6) Prism. A prismatic or columnar crystal of snow is not rarely observed. This type of frost crystal is obtained with a small supersaturation ratio as expected. The mean of s for four examples observed is 1.66. Several photographs were taken, among which the one reproduced in Photo. 13, Plate III, is a simple prism, that in Photo. 12 is a short prism with pyramid and that in Photo. 14 is a columnar crystal with pyramidal head. The one last mentioned is exactly the same in form as a kind of columnar crystal of snow that is commonly called the bullet type.

7) Prism with extended side plane. This type is produced in laboratory under a similar condition to that for prism. Two examples were observed which are reproduced in Photos. 15 & 16. It is clearly seen in the photographs that a thin plane of crystal is attached to a prism and it looks to be an extension of the side wall of the prism. This type was found always adhering to the cold ceiling of the box. A snow crystal of this type has not yet been known in any country of the world, so far as our literature at hand shows, but the authors had some chances to observe crystals which seemed to be of this type in a few snowfalls at Sapporo and at Mt. Tokati last winter. For example, the snowfall on 16 I 1935 mostly consisted of this peculiar type of snow crystal. Many photographs were taken of these crystals, among which one is reproduced in Photo. 17. Detailed explanation of this type of crystal will be published in a succeeding paper in this Journal.

IV. Provisional Discussion

As described in the foregoing section, it was found that various types of frost crystals corresponding to a copious variety of snow crystal habits are produced in laboratory by varying the supersaturatoin ratio s, that is the ratio of the vapour pressure of water at the temperature of the source to that at the temperature of air in which the crystal is formed. If it is assumed that the air near the surface of the water in the reservoir is saturated at the temperature of water and that it is brought up to the cold spot as it is, and condensed into frost crystals, then the supersaturation ratio defined above will show the actual ratio of the supersaturation when the crystal is formed. This is, however, not the case, as the upward convection current of air supersaturated with water vapour is, so to say, diluted with the surrounding air and the downward convection stream of dry air. Then the supersaturation ratio s used in this paper must be larger in numerical value than the actual ratio of supersaturation for crystal formation, but it may be accepted that those values will show the state of things in their relative values. It is well known that the formation of the various forms of crystals are chiefly governed by the growing velocity of the crystals; for example, VOGEL[6] explained the formation of a dendritic form of crystal when it grows rapidly, by introducing a consideration on the heat of crystallisation. The supersaturation ratio s represents relatively the velocity of crystal growth, at least as a first approximation. The obtained result tabulated in Table II seems to be concordant with our present knowledge on crystal habits hitherto known qualitatively, except the needle type.

As pointed out by WEGENER,[7] the needle crystal of snow has not yet been studied in detail and it cannot be decided whether it is an elongated prism or something else. The authors have been inclined to consider that there are two kinds of needle snow, and the result of the present experiment shows that the one kind appearing to be made up of an assemblage of pillars must be treated as a different sort from a simple elongated prism.

The delicate form and design of a snow crystal must have been formed by developing step by step while it is falling through various strata of the atmosphere, and the authors consider that the structure of the atmospheric layer may be inferred to some extent by investigating in parallel the structure of snow crystals and performing the more detailed experiment of the kind described in this paper.

In conclusion, the authors wish to express their best thanks to Nippon Gakuzyutu-Sinkôkai for the financial aid in this research.

6) VOGEL, ZS. f. Anorg. Chem., **116** (1921) 21.
7) WEGENER, Thermodynamik der Atmosphäre, dritte Auflage, Leipzig, 1928, p. 286.

Explanation of Photographs

T_a = Temperature of the air where crystal is formed.
s = Supersaturation ratio.

Plate I.

1. Needle Type, No. 28, $T_a = -14.1°C$, $s = 7.08$, $\times 32$.
2. „ „ , No. 29, $T_a = -14.0°C$, $s = 7.26$, $\times 32$.
3. Dendritic Form, No. 40, $T_a = -22.3°C$, $s = 7.46$, $\times 49$.
4. „ „ , No. 51, $T_a = -15.8°C$, $s = 5.42$, $\times 55$.

Plate II.

5. Intermediate Form, No. 47, $T_a = -14.5°C$, $s = 4.71$, $\times 52$.
6. „ „ , No. 43, $T_a = -13.2°C$, $s = 3.38$, $\times 48$.

7a. Plate Form, No. 66, $T_a = -4.5°C$, $s = 3.82$, $\times 140$.
7b. Schematical sketch of the crystal 7a.

8. Plate Form, No. 45, $T_a = -12.0°C$, $s = 3.03$, $\times 48$.
9. „ „ , No. 46, $T_a = -11.4°C$, $s = 2.89$, $\times 48$.

Plate III.

10. Pyramidal Form, No. 67, $T_a = -3.0°C$, $s = 1.94$, $\times 140$.

11a. „ „ , No. 75, $T_a = -10.1°C$, $s = 2.37$, $\times 290$.
11b. Schematical sketch of the crystal 11a.

12. Short prism with pyramid, No. 60, $T_a = -5.5°C$, $s = 1.58$, $\times 210$.
13. Simple prism, No. 63, $T_a = -5.6°C$, $s = 1.60$, $\times 210$.
14. Prism with pyramidal head, No. 80, $T_a = -5.5°C$, $s = 1.67$, $\times 210$.
15. Prism with extended side plane, No. 18, $T_a = -2.3°C$, $s = 1.21$, $\times 32$.
16. „ „ „ „ „ , No. 71, $T_a = -2.2°C$, $s = 1.74$, $\times 130$.
17. Natural snow crystal of prism type with extended side planes, Sapporo, 1925 I 16, $\times 35$.

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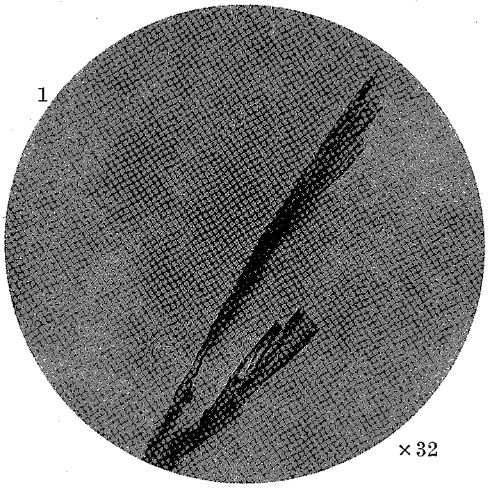

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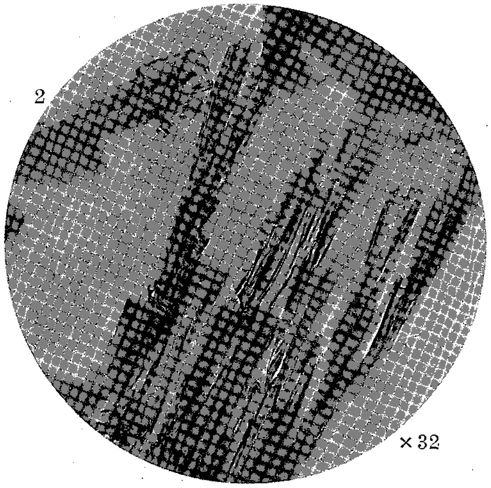

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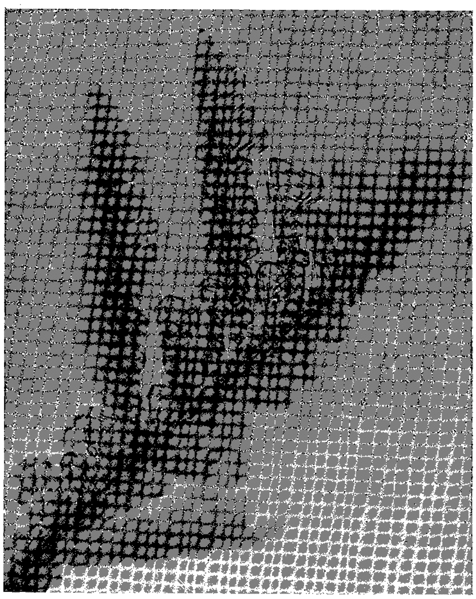

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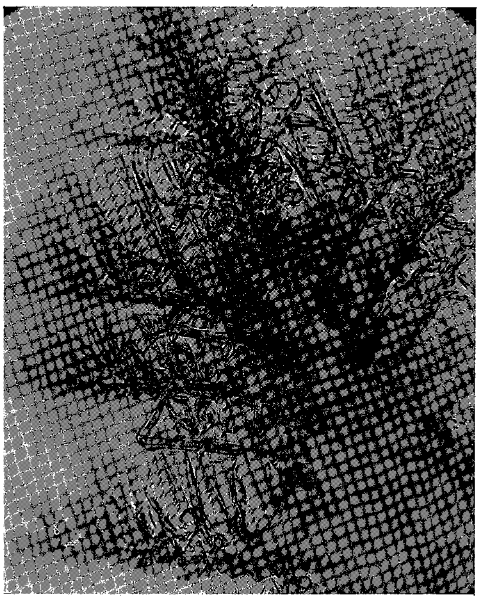

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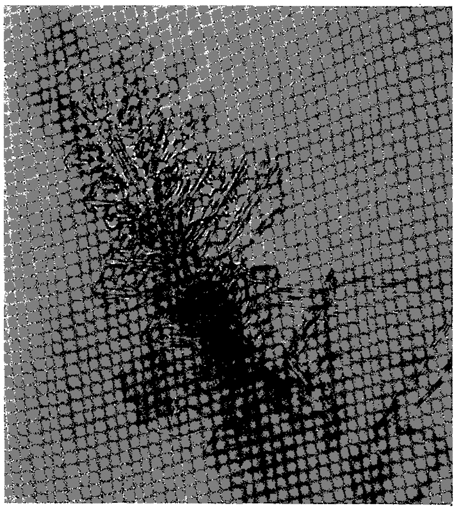

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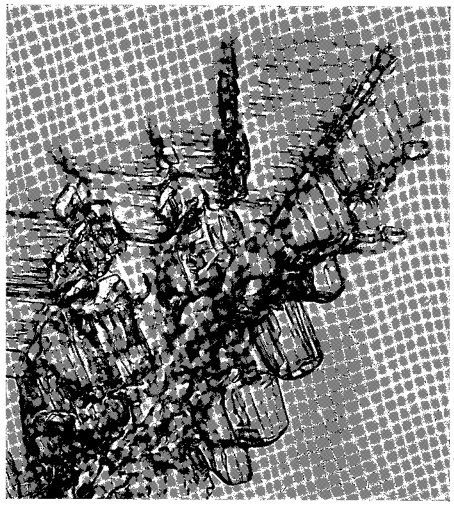

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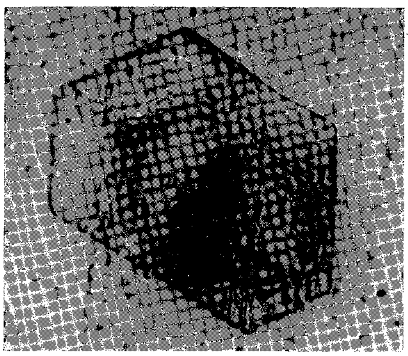
7a



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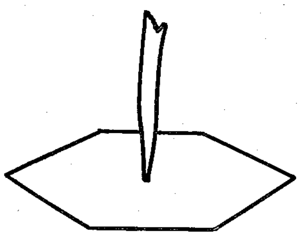

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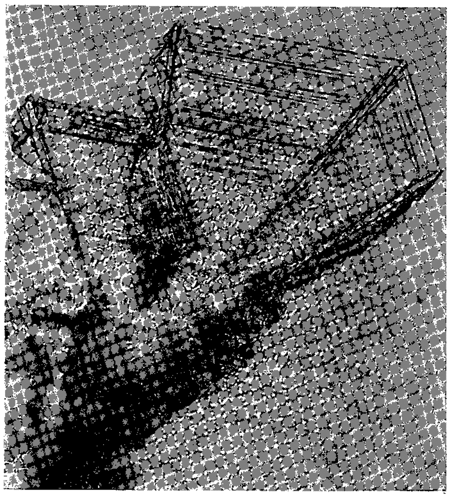

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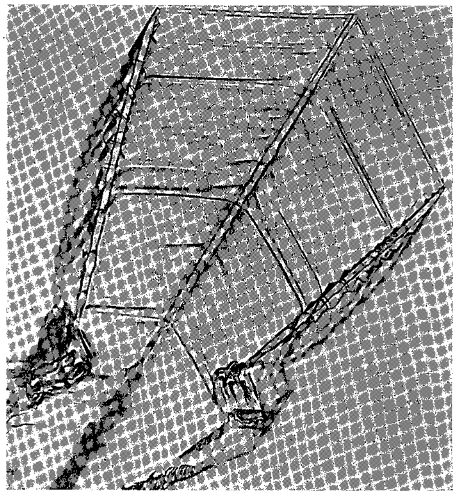

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Jour. Fac. Sc., Hokkaido Imp. Univ., Ser. II, Vol. I, No. 7.

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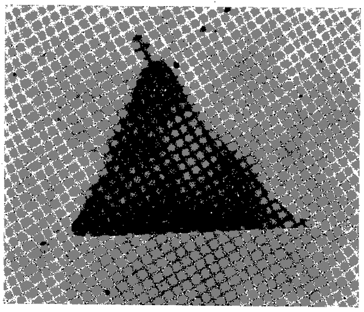

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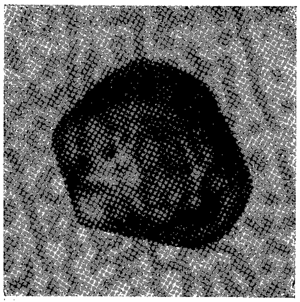

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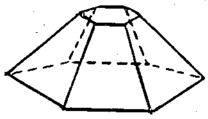
11b



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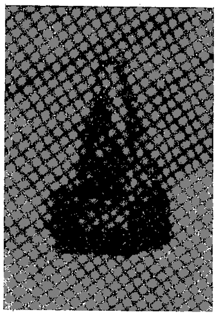

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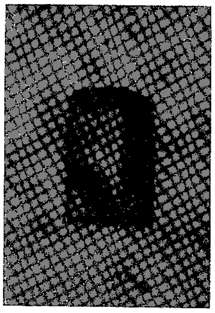

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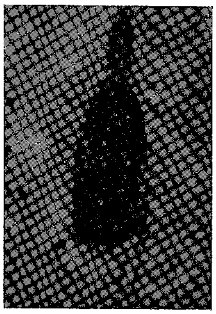

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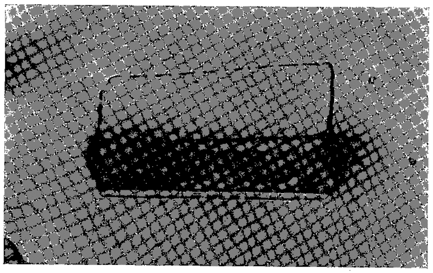

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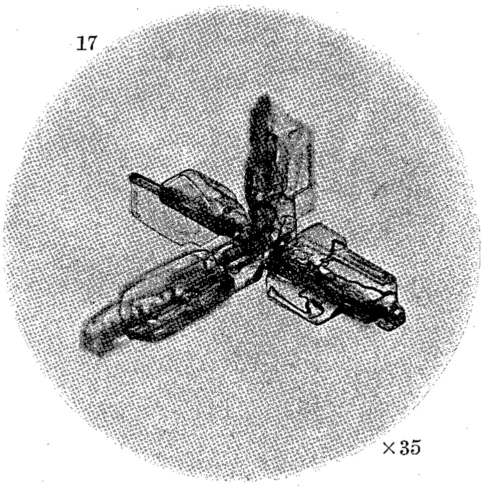

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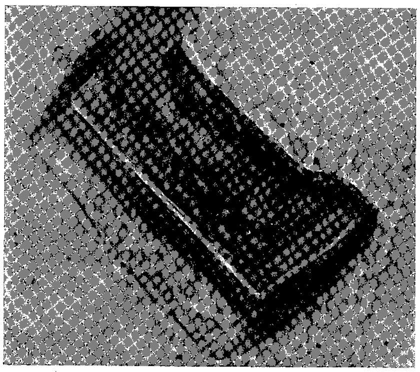

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