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On the Positive Electrification of Snow Crystals in the Process of Their Melting (IV)

—Charge of droplets produced from bursting of air bubbles in ice specimen—

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Abstract

Following the previous papers (Magono and Kikuchi, 1963, 1965; Kikuchi, 1965), studies on positive electrification associated with the melting of the ice specimen were carried out by experimental method. During the experiment, it was observed that a number of droplets burst from the melting surface of ice specimen. And it was observed that these droplets carried negative charge, and the magnitude of the negative charge was in the order of 10^{-7} to 10^{-5} esu. On the basis of this result, the relationship between air bubble concentration and positive charge in melting ice (Kikuchi, 1965) was clarified.

1. Introduction

In the previous paper (Kikuchi, 1965), it was found that the magnitude of positive electric charge of melting ice specimen was approximately proportional to the air bubble concentration in the ice specimen. Furthermore, it was found in the experiment that air bubble diameter smaller than $150\,\mu$ contributed to the charge generation and almost all of bubbles larger than 1 mm exerted no effect. In order to clarify the mechanism of the positive electrification of melting ice, microscopic observation of the melting ice was made.

2. Experimental apparatus and method

The experimental apparatus is shown schematically in Figure 1.

The main part of this apparatus consists of a metal cylinder 5 cm in diameter. The cylinder has two vertical electrode plates. Horizontal potential difference of ± 400 volt was applied between the electrode plates as shown in the Figure. When droplets from a bursting bubble were charged, they were deflected by the horizontal field, depending on the sign of their charge. In addition to the sign, the magnitude of the charge on the droplets was measured by means of Wells and Gerke's method (1919). For this purpose, a horizontal field of A.C. 2000 volt (2 cm)$^{-1}$ was superposed on the D.C. field. Consequently, the sign and magnitude on the droplets were obtained readily from a photograph.

Ice specimens with a size of $2\times1.5\times1\,cm^3$ of high air bubble concentration which were prepared from distilled and tap water were placed in a container, suspended at the center between the two electrode plates. For the convenience of photographing, the gap space between the container and the ice specimen was filled with distilled water to keep the upper surface of the ice specimen at a constant level. The upper surface of the ice specimen was kept at the level which was little higher than the upper edge of the container at all time.

Prior to melting, dust particles in the main part were removed by a suction fan at the bottom, because otherwise they would have been mixed with droplets in the field of view of the photograph. Furthermore, in order to avoid the evaporation of droplets, water immersed filter papers were placed above the main part (Price. 1960, Day. 1964). Evaporation of the droplets was prevented and melting conditions were held to simulate natural conditions of melting snow crystals. The ice

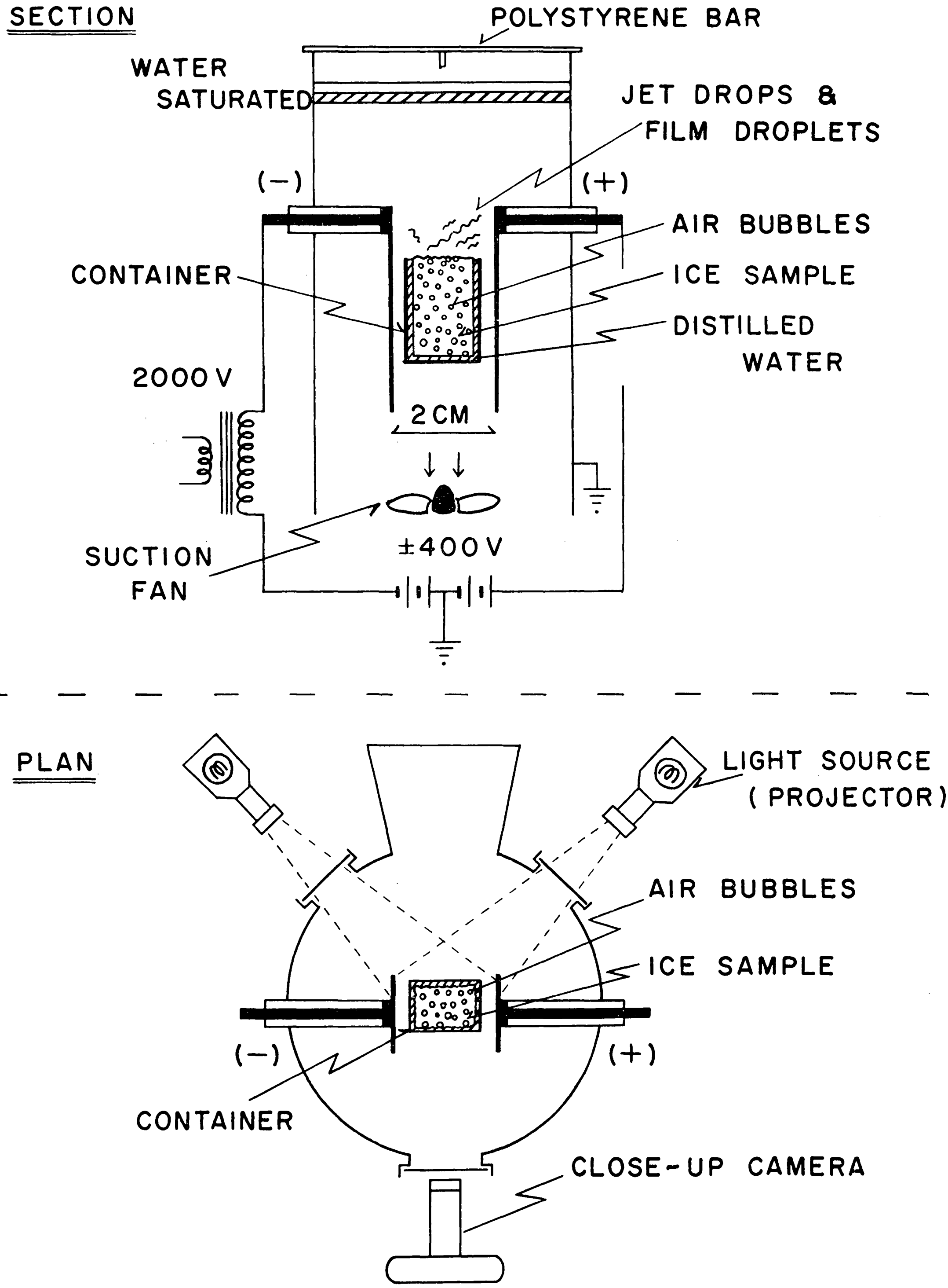


Fig. 1. Experimental apparatus

specimen was melted by an infra-red lamp at first. In the case of the infra-red lamp, smaller droplets were evaporated rapidly by a strong beam and only relatively large droplets were observed. Later, melting was done naturally at room temperature (+18°C).

Concerning the sign of charge a remarkable difference was not recognized in both melting method. Accordingly, almost all of series in this experiment were made at room temperature without using an infra-red lamp. These droplets were photographed by a close-up camera shown at the bottom of Fig. 1. Dark field illumination was made by a lighting system of two slide projectors. This illumination method was convenient since the projectors had both heat absorbing filters and light condensors.

3. Measurable range and accuracy of measurement

The resolving power of droplet size by close-up camera used in this experiment was 15 μ in diameter, and the lower limit in amplitude of wave length of the waved trajectory was 50 μ. Accordingly, the resolving power in e/m, charge per unit mass in Wells and Gerke's method (1919), of the apparatus was about 50 esu·gram^{-1} (Magono and Kikuchi, 1961) where the distance between electrodes was 2 cm and the potential difference was 2000 volt. For instance, the lower limit of measurable range was 2×10^{-7} esu in the case of diameter 20 μ, and 3×10^{-5} esu in the case of diameter 100 μ.

However Wells and Gerke's formula (1919) is applicable for the falling particles with terminal velocity. In the present experiment, droplets did not obtain their terminal velocity in the field of view, because bursting droplets at first flew up and then fell down. Furthermore, droplets were not always ejected on a focal plane. In other words, their trajectories were often oblique to the focal plane. Therefore, two causes of underestimation of an amplitude and a wave length were considered. Such underestimation was presumed to be included in calculations of droplet mass by Stokes' Law. In order to avoid such errors, only waved trajectories of uniform wave length and uniformly sharp focusing were used for the analysis. However, if underestimation of one half was made in the measurement of an amplitude and a wave length, underestimation of one fourth in charge may result. Therefore, in the experiment, margins of error as described were included.

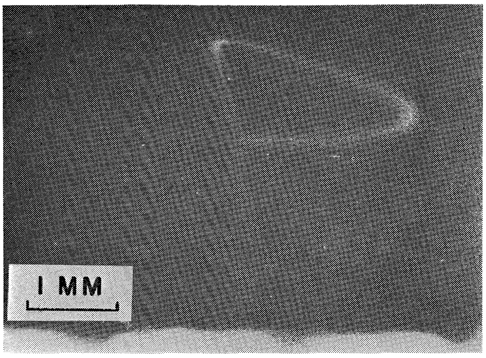
4. Results

Several examples of trajectories of droplets are shown in Photo. 1. In the photographs, white meniscus seen at the bottom of these photographs are the upper surfaces of the ice specimen. The trajectories were taken during ascent. Photo. 1 (a) shows a trajectory of a droplet produced from a bursting bubble in no electric field. Almost all droplets took irregular trajectories as seen in this example. Photo. 1 (b) to (f) show examples of trajectories of charged droplets under the electric field. Photo. 1 (b) shows a trajectory of a positive droplet and the other four photographs show several negative droplets. Because the melting proceeded from the upper surface of ice specimen, the surface was covered by a thin water layer. Many of the small air bubbles which emerged from the ice specimen burst at the interface between air and thin water layer, and a number of droplets were produced there.

The experiment was carried out for both

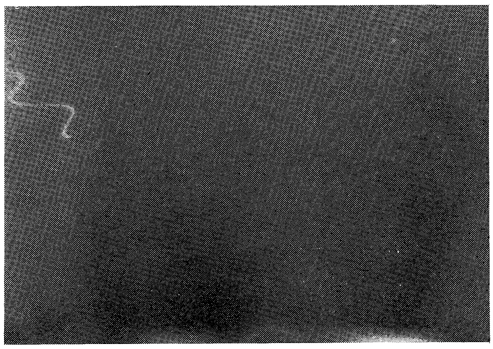
Table 1. Charge of droplets.

ice specimens	electrification	numbers	signs	mean diameters (μ)	numbers	mean charges (esu)	average charges (esu)
distilled water	charged	138	+ −	38 38	36 102	2×10^{-6} 8×10^{-6}	-5×10^{-6}
	not charged	30					
tap water	charged	237	+ −	50 47	37 200	1×10^{-5} 3×10^{-5}	-2×10^{-5}
	not charged	42					



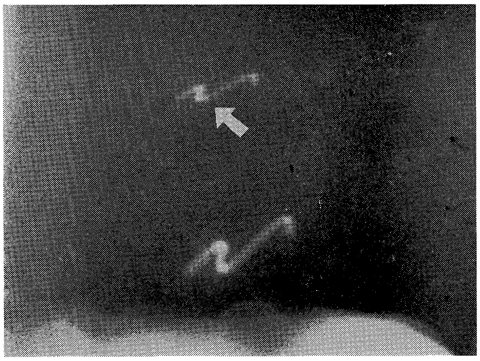
(a) no electric field

(−) (+)



(b) $R=23\,\mu$, $Q=+1\times10^{-5}$ esu

(−) (+)

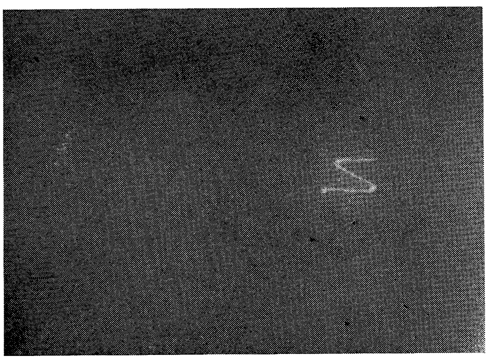


(c) $R=21\,\mu$, $Q=-3\times10^{-6}$ esu

(−) (+)

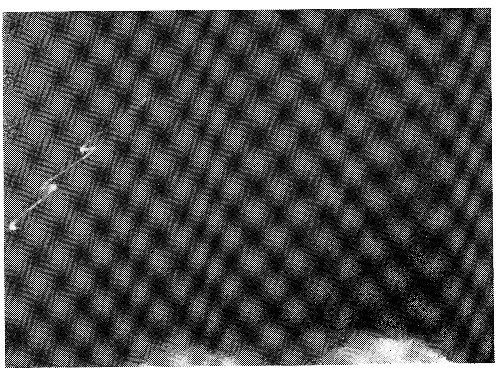
(d) $R=23\,\mu$, $Q=-4\times10^{-6}$ esu

(−) (+)



(e) $R=16\,\mu$, $Q=-5\times10^{-6}$ esu

(−) (+)



(f) $R=22\,\mu$, $Q=-3\times10^{-6}$ esu

Photo. 1. Trajectories of small particles produced by bursting air bubbles.

ice specimen made from distilled and from tap water. The results are shown in Table 1.

Regardless of whether the specimens were made from distilled or tap water, it may be seen that the majority of droplets were charged negatively. The diameters of charged droplets ranged from 20 to 60 μ and their charge was from 10^{-7} to 10^{-5} esu. The average charge for the all measured droplets including positive and not charged droplets was -5×10^{-6} esu for ice specimens produced from distilled water and -2×10^{-5} esu for ice specimens produced from tap water. As described above, these values were underestimated to some extent. It is noted that the charge and size of droplets were of the same order as natural warm cloud droplets measured by Magono and Kikuchi (1961) at the top of Mt. Teine. These were 58 μ in mean diameter and 1×10^{-5} esu in mean charge.

5. Considerations

It was ascertained that droplets carried negative charge statistically when they were produced from bursting air bubbles in melting ice. According to some researchers (Newitt *et al.* 1954, Kientzler *et al.* 1954, Mason. 1954, Blanchard and Woodcock. 1957, Toba. 1959, 1961), there are two processes of water droplets production from the water surface, namely "film droplets" and "jet droplets". After Newitt *et al.* (1954), Mason (1954) and Toba (1959), it is known that the production of film droplets are related to the cap area of a bursting air bubble, and the minimum equivalent diameter of the effective bursting air bubble is 2 mm. It was considered that air bubbles in ice in the present experiment were much smaller than 2 mm, in other words, most of them were smaller than 150 μ, because the ice specimens were prepared in a similar way to those in the previous experiment (Kikuchi. 1965). In such small air bubbles no film droplet process occurs. Therefore, the droplets in the present experiment are considered to be produced by the jet droplets process.

According to Blanchard and Woodcock (1957) and Toba (1961), the number of jet droplets produced from the bursting of one air bubble is approximately four, not depending on the size of air bubbles in the range of the size smaller than 1.5 mm in their diameter. Therefore, in the present work, it will be expected that the number of the droplets from one air bubble is approximately four, because the maximum diameter of air bubbles concerned with the charge generation was 150 μ with the mean volume diameter of 52 μ. Accordingly, it is considered that the charge of $4\times(-5\times10^{-6})$ esu is generated from the bursting of one air bubble in the ice specimen that was produced from distilled water. This value is 20 times larger than 1×10^{-6} esu which was obtained in the previous experiment by ice melting (Kikuchi, 1965). Regarding to this difference, it is thought as below. The diameter of the jet droplets observed were all larger than the lower limit (15 μ), with mean diameter of 38 μ. However it is supposed that the majority of the jet droplets actually produced were much smaller than 38 μ, because according to Toba (1961), the diameter of the jet droplets was nearly one tenth of the diameter of the bursting air bubbles, and the mean volume diameter of the air bubbles of the present experiment was 52 μ. And also, it is usual that the smaller the size, the smaller the charge. Therefore, it is considered that the difference was originated from the fact that the small charges of droplets smaller than 15 μ were removed from the calculation of the mean of the present experiment. Accordingly the mean value 1×10^{-6} esu from one bursting air bubble in the previous paper (Kikuchi, 1965) will be representable one for the charge which was obtained by melting of snow crystals.

The next problem left is to explain why the droplets acquired negative charges selectively when they were ejected. At present, it seems that there are at least two different mechanisms. One is the mechanism concerning the temperature gradient (Yosida. 1944, Reynolds *et al.* 1957, Brook. 1958, Takahashi. 1962). Since the melting phenomenon always proceeds from the surface to the interior of ice both in the cases of natural snow crystals and of ice specimen experiment, it is assumed that a temperature gradient sets up near melting ice surface, that is to say, the surface of the melting ice is always warmer than the interior of the ice. According to Latham and Mason (1961, a), negative ions concentrate on the surface of the ice. Consequently, some negative charge concentrated on the surface of the ice by this mechanism are carried away from the ice by droplets which are ejected from the surface of ice. Thus the melted ice acquires some positive charge.

However, since almost all of the experiments of charge generation on ice by the temperature gradient mechanism were carried out at low temperature where the ice was considered to be not wet, there are left some questions whether this mechanism is applicable to the case of melting ice, because it is considered that some kind of thin water layer exists on the melting ice surface. Latham and Mason (1961, b) ascertained experimentally that negative charge was separated on artificial soft hail by the temperature gradient mechanism when colder supercooled water droplets were impacted to and rebounded from artificial soft hail. It is considered that the charge generation of their experiment was the same as the author's experiment, because some liquid layer must have existed on the surface of the soft hail. Furthermore, Mason and Maybank (1960) explained the charge generation in the case of freezing of water drops by the temperature gradient. Recently, Latham (1964) ascertained experimentally that charge transfer in ionic solutions was explained by temperature gradient theory.

Another mechanism is related with the ejection of droplets by bursting of air bubbles from water surface. According to Magono and Takahashi (1959), water droplets seemed to carry negative charges when they were ejected from a water surface colder than 80°C independent of the temperature gradient.

Regardless of the mechanism, we know experimentally that droplets carry off negative charges from melting ice.

After the observation at the top of Mt. Teine (Magono and Kikuchi, 1965), snow crystals acquired the charge of $+2\times10^{-4}$ esu per crystal on the average in their melting process, and most of them became positive raindrops, depending on the original charge of the snow crystals. Furthermore, it was observed that usual snow crystals produced approximately 50 air bubbles per crystal in their melting process, and complicated snow crystals, snowflakes and graupel produced air bubbles much more than 50 in the process. So, it is computed that the usual snow crystals acquires the charge with amount of $50\times(1\times10^{-6})$ esu, namely 0.5×10^{-4} esu on the average when they melt into positive raindrops. This value is a little smaller but nearly the same as the observed value ($2\times$ $\times10^{-4}$ esu). And it is considered that the snowflakes and graupel will acquire positive charge more than 0.5×10^{-4} esu.

Thus the author considers that the most of the positive charge obtained by melting of snow crystals in the experiments at the top of Mt. Teine are resulted from the bursting of air bubbles in the melting process.

After Chalmers and Pasquill (1938) and Orikasa (1962), the mean charge of raindrops in continuous rainfall was $+5.3\times10^{-4}$ and $+5\times10^{-4}$ esu respectively. These values are of the same order of the mean value of charge which was produced by jet droplets in the melting process of snow crystals ($+2\times10^{-4}$ esu at Mt. Teine, and $+0.5\times10^{-4}$ esu in the laboratory experiment).

Therefore, the author considers the positive electrification of melted snow crystals plays an important role on the positive charge on raindrops in continuous rainfall.

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References

Blanchard, D.C. and A.H. Woodcock, 1957: Bubble formation and modification in the sea and its meteorological significance. *Tellus*, **9**, 145-158.

Brook, M. 1958: Laboratory studies of charge separation during ice-ice contact. *Recent Advances in Atmospheric Electricity*. 383–389.

Chalmers, J.A. and F. Pasquill, 1938: The electric charges on single raindrops and snow-flakes. *Proc. Phys. Soc.*, **50**, 1–16.

Day, J.A. 1964: Production of droplets and salt nuclei by bursting of air-bubble films. *Quart. J. Roy. meteor. Soc.*, **90**, 72–78.

Kientzler, C.F., A.B. Arons, D.C. Blanchard and A.H. Woodcock, 1954: Photographic investigation of the production of droplets by bubbles bursting at a water surface. *Tellus*, **6**, 1–7.

Kikuchi, K. 1965: On the positive electrification of snow crystals in the process of their melting (III),—The relationship between air bubble concentration and charge generated in ice specimen during their melting—. *J. meteor. Soc., Japan*, Ser. II, **43**, 343–350.

Latham, J., 1964: Charge transfer associated with temperature gradients in ionic solution. *Nature*, **202**, 686–687.

———, and B.J. Mason, 1961 a: Electric charge transfer associated with temperature gradients in ice. *Proc. Roy. Soc.*, A. **260**, 523–536.

———, and ———, 1961 b: Generation of electric charge associated with the formation of soft hail in thunderclouds. *Proc. Roy. Soc.*, A. **260**, 537–549.

Magono, C. and K. Kikuchi, 1961: On the electric charge of relatively large natural cloud particles. *J. meteor. Soc. Japan*, Ser. II, **39**, 258–268.

———, and ———, 1963: On the positive electrification of snow crystals in the process of their melting. *J. meteor. Soc. Japan*, Ser. II, **41**, 270–277.

———, and ———, 1965: On the positive electrification of snow crystals in the process of their melting (II). *J. meteor. Soc. Japan*, Ser. II, **43**, 331–342.

———, and T. Takahashi, 1959: The electric charge on condensate and water droplets. *J. Meteor.*, **16**, 167–172.

Mason, B.J. 1954: Bursting of air bubbles at the surface of sea water. *Nature*, **174**, 470–471.

———, and J. Maybank, 1960: The fragmentation and electrification of freezing water drops. *Quart. J. Roy. meteor. Soc.*, **86**, 176–185.

Newitt, D.M., N. Dombrowski and F.H. Knelman, 1954: Liquid entrainment I. The mechanism of drop formation from gas or vapour bubbles. *Trans. Inst. Chem. Engrs.*, **32**, 244.

Orikasa, K. 1962: On the disturbance of the surface electric field caused by rain and snow fall. (in Japanese) *Geophys. Bull. Hokkaido Univ.*, **9**, 123–160.

Price, P.B. 1960: On the growth of cadmium crystals from the vapour. *Phil. Mag.*, **5**, 473–484.

Reynolds, S.E., M. Brook and M.F. Gourley, 1957: Thunderstorm charge separation. *J. Meteor.*, **14**, 426–436.

Takahashi, T. 1962: Electric charge generation by breaking of ice piece. *J. meteor. Soc. Japan*, Ser. II, **40**, 277–286.

Toba, Y. 1959: Drop production by bursting of air bubbles on the sea surface (II), Theoretical study on the shape of floating bubbles. *J. Ocean. Soc. Japan*, **15**, 121–130.

———, 1961: Drop production by bursting of air bubbles on the sea surface (III), Study by use of a wind flume. *Memoirs of the College of Science, Univ. Kyoto*, Ser. A, **XXIX**, 313–344.

Wells, P.V. and R.H. Gerke, 1919: An oscillation method for measuring the size of ultra-microscopic particles. *J. Amer. Chem. Soc.*, **41**, 312–329.

Yosida, Z. 1944: The mechanism of charge generations in friction and destruction of ice and in thunderstorms. (in Japanese) *Teion Kagaku*, The Institute of Low Temperature Science, Hokkaido Univ., Ser. **I**, 149–187.

降雪の融解による正荷電について (IV)

—氷に含まれる気泡の破裂によって生ずる微水滴の電荷について—

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これまでの研究 (Magono and Kikuchi, 1963, 1965; Kikuchi, 1965) に引続き，氷の試料の融解の際に発生する正電荷についての実験が行なわれた。

その結果，融解中の氷の表面から，氷に含まれていた気泡の破裂によって生じた多くの微水滴が飛び散るのが観測された。それらの微水滴の多くは，$-10^{-7}\sim-10^{-5}$ esu の電荷を有するものであることがわかった。これらの微水滴が "jet droplets" 機構によってできるものと仮定すれば，気泡量と電荷量との関係 (Kikuchi, 1965) がよく説明される。