A New Effect of Evaporation Accelerating the Ice Phase in Supercooled Water

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1. Introduction

There are some reports in the literature of the increase of nucleating ability when supercooled droplets are in the state of evaporation (Vonnegut and Weickmann, 1960). Study, by the present authors, of the freezing characteristics of rainwater drops has shown that the

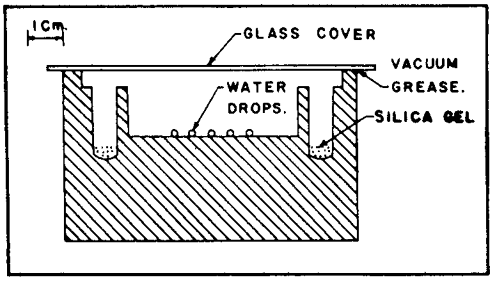

FIG. 1. Experimental dish used for study of the freezing characteristics of water drops.

drops freeze more readily under simultaneous supercooling and evaporation than under supercooling alone, even when the temperature of the drops is essentially the same. This observed improvement in the nucleation of the ice phase (drop freezing probability), which may be called the "dynamic effect of evaporation," is likely to be of value, when fully confirmed, for obtaining a better understanding of the physics of cold clouds, *viz.*, for the reconciliation, at least partially, of the unusually large concentrations of ice crystals as compared to those of ice nuclei, noticed in cumulus clouds (Murgatroyd and Garrod, 1960; Koenig, 1963; Braham, 1964; Mossop, 1970). We therefore make a brief report here of our finding.

2. Experiment

Millimeter size drops, produced by a hypodermic needle, are taken in equal numbers of 30 each in two identical metal (aluminium alloy) dishes, each provided with a special groove all around the region in which the drops are placed (Fig. 1). The groove provided in each dish is deep bottomed, sufficient to isolate its surface from the rest. This precaution was taken to minimize entry, by direct means, of any particulates possibly ejecting from the groove into the dish region in which the drops are placed. Both dishes are similarly coated on their inside bottoms with a water repellent (Dow Corning silicone lubricant) before placing the drops. A small quantity of previously evacuated indicator silica gel is placed in the groove of one of the dishes so that, when the dish is covered from the top, the drops in that dish become subject to the process of evaporation due to the moisture adsorbing property of the silica gel. The amount of silica gel placed in the groove, about 2 gm, is small compared to the weight of the dish (~590 gm) and this does not add materially to the thermal load of the dish. Both dishes are then sealed from the top with glass covers and then subjected immediately to identical rates of cooling by transferring them simultaneously to a cold stage kept cooled to a predetermined temperature. The number of drops freezing in each dish at successive intervals of time are noted with the naked eye until all 30 drops in each dish have frozen.

The temperature difference between the dishes as well as the actual temperatures of the dishes individually, with the full contingent of the drops within, are measured with thermocouples made from SWG-36 copper-constantan wires, using a Hewlett-Packard recording micro-voltmeter. In addition, the temperature difference between two single drops, each in a separate dish, as well as the actual temperatures of the drops are measured, in the same way, by thermocouples inserted in an identical manner inside the drops.

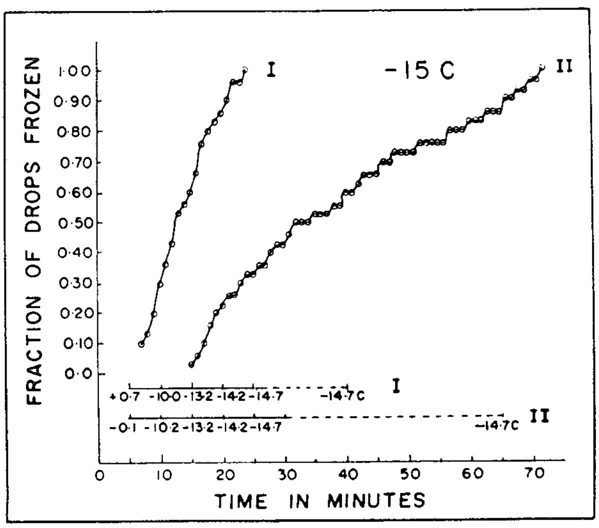

FIG. 2. Fraction of drops frozen when the cold stage was initially at −15°C. Curve I relates to freezing events noticed in the dish which contained silica gel in the groove and curve II to the other dish (which did not contain silica gel). Temperatures are given at 5-min intervals for both the dishes separately.

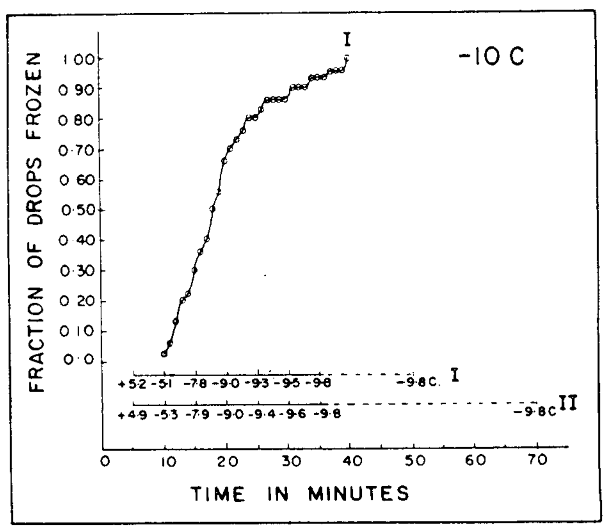

FIG. 3. Fraction of drops frozen when the cold stage was initially at −10C. Curve I relates to freezing events noticed in the dish which contained silica gel in the groove. No freezing event was observed even after 100 min in the other dish. Temperatures are given at 5-min intervals for both the dishes.

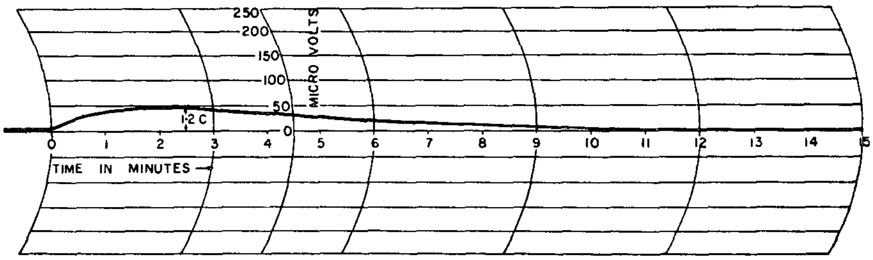

Fig. 4. Variation in the temperature difference between the two experimental dishes. The potential difference remains in the positive direction so long as the dish which contained silica gel in the groove is at a warmer temperature. Only the recording obtained for the first 15 min is given. Values of potential differences recorded after 10 min, until the end of the experiment, are nearly zero.

3. Measurements

The fraction of drops frozen at successive intervals of time, corresponding to initial cold-stage temperatures of −15 and −10C, as noticed in a typical run of the experiment, is given, respectively, in Figs. 2 and 3. As shown in Fig. 2 freezing began in the dish containing silica gel in the groove 8 min earlier (corresponding to a temperature of −5.4C) than it began in the other dish where freezing did not take place until the drop temperatures fell to −13.2C; there was then a net gain of 7.8C in the freezing temperature. The last drop in the dish, which contained silica gel in the groove, froze 48 min earlier than the last drop in the other dish. In the case where the cold stage was at −10C (Fig. 3), freezing began in the dish containing silica gel in the groove at the end of 9 min (time is counted from the moment the dishes are transferred to the cold stage), corresponding to a temperature of −5.1C. Freezing was faster than in the case when the temperature of the cold stage was initially at −15C and with no silica gel in the groove. Moreover, none of the drops in the other dish had frozen even after 100 min.

The fact that, of the two dishes used for observing freezing, the one containing silica gel in the groove has always shown freezing events occurring, more rapidly raises the question as to whether the drops in that dish were not reaching systematically lower temperatures during the cooling process. Although no such feature has been noticed, precaution has been taken to ensure that no such possibility exists.

For this purpose, the positions of the two dishes on the cold stage, insofar as their proximity to the inside wall of the cold chamber are concerned, were so chosen that the dish with silica gel in the groove initially develops a small temperature bias in the positive direction. As a result, while undergoing cooling, the dish with silica gel will be held, for sometime at least, at a temperature slightly higher than that of the other (Fig. 4). Although this may mean introducing a change in the effective rate of cooling of that dish with respect to the other, and, therefore, of the drops placed in that dish with respect to those in the other dish, the change introduced will actually be so small (about 0.1C at a 6C min^{-1} cooling rate in the first 5 min) that is of no consequence to the drop freezing probability in that dish. A typical recording of the temperature difference between single drops each placed in the dishes, for an initial cold-stage temperature of −10C, is given in Fig. 5. The recording shown refers to the case with silica gel in the groove of one of the dishes. The difference in the temperature between the drops, though gradually narrowing after attaining a maximum value of 1.2C by the third minute, is clearly seen to remain, as anticipated, in a positive direction. This is to say that

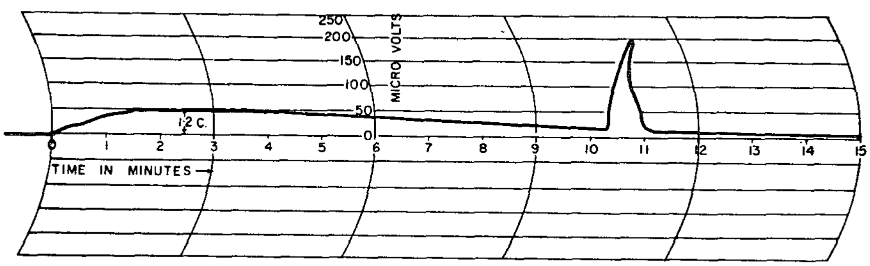

Fig. 5. Variation in the temperature difference between single drops each placed in the two experimental dishes. The potential difference remains in the positive direction as long as the dish which contained silica gel in the groove is at a warmer temperature. Only the recording obtained for the first 15 min is given. Values of potential differences recorded after 14 min, until the end of the experiment, are nearly zero. The sudden increase in the difference noticed between 10 and 11 min is due to the latent heat released when the drop in the dish with silica gel in the groove froze. The drop in the other dish did not freeze even after 100 min.

the drop which was placed in the dish with the silica gel, i.e., the dish in which freezing occurred, remained at a higher temperature until sometime after it froze. Further, the drop which was placed in the other dish never froze although it was held, from the beginning, at a lower temperature; this drop also subsequently reached a temperature (−9.8C) which was much lower than that (−5.1C) at which the drop in the dish with silica gel, had frozen. It is confirmed, therefore, that the observed improvement in the ice nucleation cannot be attributed to the fact that the drops in the dish containing silica gel in the groove may have reached lower temperatures than the drops in the other dish.

4. Conclusion

The results obtained from the drop freezing events at constant temperatures in the present study which showed that all the drops considered eventually froze at that temperature under certain conditions (i.e., in both dishes at −15C and in one dish at −10C) appear to be in disagreement with the results reported from experiments of a similar type (Vonnegut, 1948; Vali and Stansbury, 1966; Salt, 1966), where, at relatively warm temperatures, not all drops of a given sample froze even if held for some length of time. While the reasons for this apparent disagreement are not clear, one possibility which can be considered is that the experimental conditions in the present study were not exactly identical with those in the other studies reported.

It has not been possible to offer any interpretation for the improvement of the ice-phase nucleation noticed in the present experiments. One possibility to be considered is whether the water substance is undergoing changes in its physical properties during the period when the supercooling drops are kept subject to evaporation.

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