Freezing of Freely Suspended, Supercooled Water Drops by Contact Nucleation

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ABSTRACT

The freezing of freely suspended, supercooled water drops by contact nucleation has been studied. Water drops were balanced in an updraft of a large vertical wind tunnel and allowed to supercool to the ambient temperature. Ice crystals introduced into the updraft were, as expected, the most effective nucleants in freezing the drops at ambient temperatures colder than and up to 0C. The results of this experiment using silver iodide and clay as contact nucleants closely agree with earlier work performed with a constant rate of cooling apparatus. The effective temperature for 100% nucleation efficiency in the case of AgI particles was −4 to −5C and for silicate particles −7 to −10C.

1. Introduction

Earlier results of the experiments on droplet freezing by contact nucleation were reported by Gokhale and Goold (1968). A constant rate of cooling apparatus was used for that study. Silver iodide particles sprinkled on supercooled, millimeter-size water drops were effective in freezing the drops at −5C. Particles of naturally occurring silicates were found to be effective in the range −7 to −10C, indicating that dry particles are much more effective in freezing drops by contact than are particles embedded in the drops.

In the experiments described above, the drops were supported on a metal plate with paraffin coating which was cooled to the desired temperature. The freezing of freely suspended supercooled drops in a large vertical wind tunnel involves a different experimental technique. Many drops were freely suspended and allowed to supercool to the wet-bulb temperature. Particles of different materials were then introduced into the updraft to determine their effectiveness in freezing the drops by contact nucleation.

2. Design and air flow of the large vertical wind tunnel

A large vertical wind tunnel was constructed for the study of the interactions of hydrometeors suspended in an updraft. Its vertical cylinder is 6 ft in diameter and 10 ft in length. To streamline the air flow, cross-hatched plywood was placed in the base of the vertical section of the tunnel. Three metal screens were fixed at a distance of 1, 3 and 5 ft, respectively, above the plywood hatching. At the top of the tunnel, a 3-inch thick hexagonal cell honeycomb was placed to further reduce the turbulence within the air flow. The cells of this honeycomb have an equivalent diameter of $\frac{3}{8}$ inch. More details of the design and construction of this tunnel have been discussed by Spengler and Gokhale (1970).

The updraft velocities were measured using a Hastings-Raydist hot wire anemometer. Updraft profiles are such that drops can be suspended in the tunnel for several minutes. The size of the drops suspended depends on the updraft velocity which is controlled by the fan speed. It can be maintained constant at any value between a few meters per second and 13 m sec^{-1}.

3. Temperature measurement

This large vertical tunnel is located in an unheated airplane hangar, and the large fan draws in air from outside the hangar. Thus, the air coming through the tunnel is at the temperature of the outside air. Temperature measurements are made by three thermocouples located at the mouth of the tunnel outside the building, halfway up the vertical section of the tunnel, and at the top of the tunnel. When in operation, the temperature at the outlet or opening of the tunnel is generally within 0.5C of the temperature at the inlet to the tunnel. One important correction necessary in determining the temperature of supercooled drops is the ventilation or cooling effect on the drops in subsaturated air. In unsaturated air, drops will cool to the wet-bulb temperature. However, during these experiments on contact freezing, the wick of the wet-bulb thermometer would freeze; therefore, the ice-bulb temperature was recorded. The temperature of the water reservoir was also monitored.

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4. Experimental procedure

The purpose of the experiment was to determine the warmest temperature at which contact nucleation would cause freezing in freely suspended drops. Tests were conducted over an ice-bulb temperature range from 0 to −14C.

In these experiments, drops were obtained by using a small vibro-staltic pump which pulses a stream of water. This pump generated about 120 drops of uniform size per second, and the accumulated concentration was controlled by the running time of the pump. The drops used in the experiments had an average equivalent diameter of 4–6 mm. They were injected into the updraft from a water reservoir where the water temperature was maintained slightly above 0C. Twenty-five seconds after injection into the updraft, the cooling drops were assumed to have reached their thermal relaxation temperature. The thermal relaxation times for our particular experiment were calculated by modifying the equations given by Kinzer and Gunn (1951). The thermal relaxation time of evaporating drops was found to vary with the size of the drop, and the difference between the initial drop temperature and the environmental wet-bulb temperature. For these experiments, 25 sec was found to be the maximum thermal relaxation time. Therefore, all tested drops were at least $1/e$ of the difference between their initial temperature (T_i) and the wet-bulb temperature (T_w). Fig. 1 is a plot of $T_{\mathrm{Drop}}=T_w+(T_i-T_w)e^{-t/\tau}$, where t is time in seconds and τ is the calculated thermal relaxation time for the different drop sizes. The curves show the rate of cooling for large water drops in the unsaturated updraft of the wind tunnel. In most cases the drops tested had cooled for over 60 sec, thus insuring that they were within 1C of the environmental wet-bulb temperature. The water reservoir temperature, and the dry and the ice-bulb temperature of the updraft were continuously monitored during the experiment.

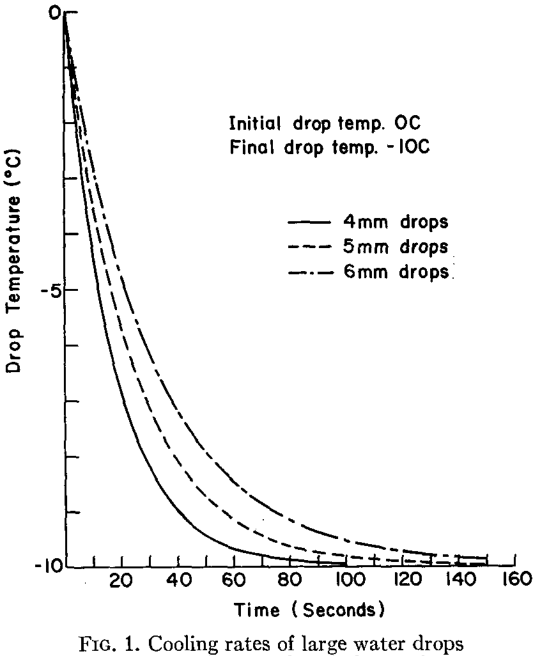

FIG. 1. Cooling rates of large water drops in the updraft of the wind tunnel.

TABLE 1. Average diameter (μ) of particles used in contact nucleation studies.

Material	Mean diameter
AgI	2.8
Clay	3.2
Red soil	2.7
Sand (crushed rock)	5.4
Sand (industrial)	3.2

The materials tested in this experiment were ground with a mortar and pestle, placed in a 470-ml plastic bottle and stored in the same environment as the vertical wind tunnel. The materials tested were within 1C of the ambient air temperature. (It should be noted that a few tests with warmer particles indicated that this precaution was not necessary.) The plastic bottles were fitted with a rubber stopper and a hollow glass tube ~35 cm long.

Grinding of materials usually produces a log normal distribution of particle sizes. While no efforts were made to measure actual size distributions, a mean measured diameter of the particles was determined for each material. Using a method similar to the experimental procedure, the plastic bottles were shaken and then squeezed, forcing a jet to impact on a clean glass slide. This method and the optics of the microscope used are biased against small particles. This is evident in the complete exclusion of particles $<1\,\mu$ diameter from our measured particles. Counting larger particles ($>20\,\mu$) with a sonic counter indicates that there are approximately 10 such particles in a 40 cm^3 volume of aersol sample. What has been measured optically then is representative of the size of particles actually participating in the contact nucleation. Table 1 gives these measurements.

From previous work (Gokhale and Goold, 1968) it was concluded that particles of micron size seem to be more effective than sub-micron particles in freezing drops by surface nucleation. See Fig. 4 from that paper.

While the drops were cooling in the updraft, the bottle containing the ground material was vigorously shaken. Anytime after 40 sec the end of the glass tube was positioned at a distance of about 1 m below a single drop. Then, the plastic bottle was squeezed in such a manner that the particles released from the bottle would contact the drop. A 40-cm^3 volume was displaced during each squeeze of the bottle and with a little practice one could be assured that the plume of particles hit the drop. If freezing occurred within five seconds after contact with particles, it was attributed to contact nucleation. It was observed that drops were freezing immediately upon contact by

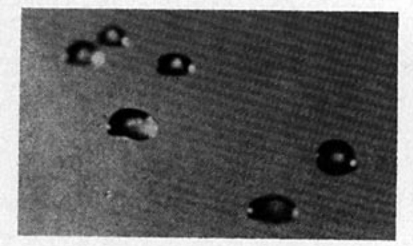

Fig. 2. Large liquid drops and frozen pellets suspended in the wind tunnel. The drop on the right is liquid (~5 mm equivalent diameter) while lowest and largest drop in focus on the left is frozen.

particles. Only six drops, which did not freeze immediately but did freeze within 5 sec, were considered to have been frozen by contact nucleation. This number represents less than 1.5% of all the drops tested in this experiment. At temperatures colder than −5C a drop contacting effective material would instantly turn opaque in appearance. When freezing was produced with silver iodide at temperatures near −3C, the growth of the ice crystal structure could be visibly seen propagating over the surface of the drop. In the intermediate range of temperatures, freezing could be detected by a change in reflectivity of the drop, by complete cessation of drop oscillations, or by physically capturing the suspected drop and examining it for ice.

High-speed movies taken at 500–2000 frames sec^{-1} were used to study the contact events. One print from this film is included here (Fig. 2).

5. Results

Fig. 3 summarizes the effectiveness of the materials tested in contact freezing of over 400 drops. The solid triangle indicates the warmest ice-bulb temperature at which freezing would be produced by contact nucleation of a particular substance. The snowflake symbol indicates the warmest ice-bulb temperature at which the particular material used had a 100% chance of causing freezing. The intermediate range of temperatures is the transition region from 0 to 100% nucleating efficiency of the materials. It should be recognized that there is a possibility that the particles may have a 100% nucleation effectiveness at slightly warmer temperatures. Since the temperature of the cooling drop approaches the wet-bulb temperature asymptotically, it will be slightly warmer, when it actually freezes, than the ice-bulb temperature used as the reference temperature. The ice-bulb and wet-bulb temperatures were calculated over the range of humidities and ambient air temperatures encountered in this experiment. The maximum discrepancy between the ice-bulb temperature and the drop temperature, which was assumed to be the wet-bulb temperature, was 0.3C.

With this experimental procedure it is impossible to determine the exact number of particles contacting a drop. A rough estimate of the number of particles reaching any particular drop is between 1 and 500. Fig. 1 in the paper by Gokhale and Goold (1968) indicates that as the number of particles (in this case they were clay particles) contacting the surface of the drop increases, the probability of its freezing at warmer temperature increases. The effect, however, only occurs for temperature increases of the order of 2C. It has also been observed in microcinematographic studies of contact nucleation (Gokhale and Lewinter, 1971) that nucleation may commence from more than one particle simultaneously. The implications of these facts are discussed in the results.

Ice crystals which were either drawn in by the fan or deliberately introduced into the updraft always caused freezing at ice-bulb temperatures $\leqslant -2$C. At drop temperatures equal to 0C none of the drops froze, as expected. Freezing of freely suspended supercooled drops with ice crystals dramatically demonstrated the effect of evaporational cooling. Drops were frozen when the dry air temperature was as warm as +2C.

Silver iodide particles were the next most effective nucleating material. Freezing could be initiated with few drops at a −2.5C ice-bulb temperature, but did not obtain a full 100% effectiveness until the drops cooled to at least −4C.

Unexpectedly, crushed red soil (collected in Arizona) also caused freezing at temperatures as warm as −2.5C. However, this material did not reach full effectiveness until −6.5C.

A brief one-sentence description of a 1939 cloud seeding experiment performed by Findeisen appears in a paper by Schulz and Kissingen (1948). Virga is reported to have been produced from a cloud whose top was not colder than −9C when it was seeded with crushed sea sand. This inspired the authors to test

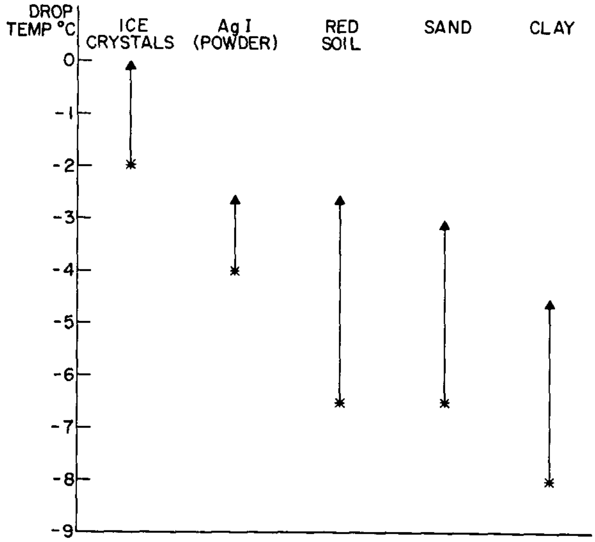

Fig. 3. The effectiveness of various materials in nucleating freely suspended, supercooled water drops. See text for explanation of symbols.

ground sand. The two types used in this experiment were commercial-industrial sand and sand made from crushed rock (Pennsylvania Glass-Sand Co.). Both had about the same ability in freezing drops. The sand became effective as a contact nucleant at −3C and reached full effectiveness at a −6.5C ice-bulb temperature.

A clay sample (collected in Albuquerque, N. M.) was the least effective material tested in contact nucleation. It became effective between −4.5 and −5C and did not reach full effectiveness until the drops contacted had cooled to −8C.

Several other materials were tested, but not systematically, over a range of temperatures. However, the results are interesting enough to include in this report. Glass spheres approximately 19 μ in diameter were not effective at −7 or −8C but were 100% effective at −13.5C. Soluble materials like table sugar and common salt were 100% effective at −11C and −13.5C, respectively.

It was also apparent, at the warmer freezing temperatures, that when the materials were first effective, the largest number of particles had to be used to initiate nucleation. When the material became 100% effective, the supercooled drop could be as much as 2 m above the source of particles and would still freeze. This meant very few particles were necessary to nucleate the drop.

6. Conclusions

The temperatures at which AgI and clay reach their 100% nucleating effectiveness in this experiment coincide within 1C of the temperatures reported in earlier work (Gokhale and Goold, 1968), where drops were cooled at a constant rate on a metal plate and sprinkled with particles of the contact material. AgI particles were found to be 100% effective at −4C in freezing freely suspended supercooled drops, and at −5C in freezing supercooled drops supported on a metal plate. Clay was found to be about 60% effective at −7C in the wind tunnel experiments, and about 40% effective at −7C in the metal plate experiments.

Another interesting conclusion was that red soil particles and ground sand reached full effectiveness at −6.5C in nucleating freezing in suspended supercooled drops.

Thus, the present as well as the earlier experimental results on the freezing of supercooled water drops by contact nucleation demonstrate that drops freeze at much warmer temperatures by the contact nucleation mechanism than by other ice nucleation mechanisms.

Best results will be obtained in freezing water drops in clouds by contact nucleation if silver iodide particles are introduced in the region where the degree of supercooling is at least −5C.

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