A Preliminary Study of Ice Grown by Droplet Accretion Using Water-Insoluble Particles as Tracer

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ABSTRACT

The migration of water-insoluble particles during freezing was used to study the structure of ice grown by droplet accretion. Experiments were performed in a cold tunnel under different growth conditions using nickel powder as a tracer; the resulting deposit was examined using the x-ray microradiograph technique. Analysis of the types of tracer particle aggregates provided a description of the freezing processes. It was concluded that this method could be used as an additional tool in studies of the various ice growth conditions found in nature.

1. Introduction

Hailstone growth is generally classified as dry or wet depending on whether the deposit temperature is below or at 0°C, respectively. However, the microphysical freezing processes which take place during and immediately after accretion of cloud droplets are not yet completely understood.

Concerning dry growth, a theoretical analysis of the microphysical aspects of the accreting process has been performed by Macklin and Payne (1967) who considered initial freezing, spreading of the droplets, subsequent freezing and final cooling. As for spongy growth (a mixture of ice and water), List (1959, 1960) had originally shown that no shedding of liquid water occurs when the limit posed by the heat dissipation rate is exceeded by the accretion rate. Knight (1968) and Knight and Knight (1973) have investigated the mechanism of spongy growth. Detailed analyses of the ice matrices and structures of the spongily grown ice during and after accretion were performed by Roos and Pum (1974). More recently, Macklin (1978) and List (1978) have discussed some of the interpretations of hailstone characteristics attained from laboratory and field hail research.

The main purpose of the present work was to investigate the possibility of determining more details of ice fabric in dry, simple wet and spongy growth by taking advantage of the mechanism whereby insoluble tracer particles migrate within growing hailstones due to rejection by the ice-water interface; the spatial distribution of these particles within ice may contain a record of the freezing process. The migration of water-insoluble particles with the ice-water interface during the bulk freezing of water was originally studied by Hoekstra and Miller (1967).

2. Experimental procedure

A vertical wind tunnel (30 cm \( \times \) 30 cm \( \times \) 2 m high) was connected to a cold chamber and air was circulated through it at a prescribed temperature. The working section of the tunnel where the rotating cylinder of ice is accreted was reduced to 7 cm \( \times \) 7 cm, and the vertical velocity in it was 29 m s\(^{-1}\). The cylinder was rotated at 1 Hz.

Nickel spherule powder\(^\dagger\) was used as the tracer particle; after washing with "particle-free" water (filtered through a 0.22 \( \mu \)m membrane filter) it was suspended in "particle-free" water at a concentration of 5 g l\(^{-1}\) and, while being shaken continuously during the test, was sprayed from a pair of nozzles. The so-called Feret’s statistical diameter of the nickel particles measured at the working section of the tunnel was 12 \( \mu \)m, and there were no individual particles < 3 \( \mu \)m or > 20 \( \mu \)m. About 60% of the

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mass consisted of small agglomerates of 2–4 particles. Examination of drops containing tracer particles, sampled on a gelatin-coated slide at the place inside the tunnel where the accretion was taking place, showed that the nickel particles were all present within droplets. The droplets’ diameters ranged from 5 to 45μm; the mean volume diameter was equal to 12μm. Nickel particles did not freeze supercooled water droplets above a temperature of –18°C and consequently they did not act as freezing nuclei in our experiments.

Another pair of clean spray nozzles was used concurrently to supply enough water droplets to maintain the wet growth conditions. The droplet diameter size range of a clean spray was between 5 and 65μm; the mean volume diameter was equal to 16μm.

The nickel powder was used because of its high x-ray absorption and its very limited size range; thus the particle deposit could be more clearly seen in the x-ray radiograph without any confusion as to individual particles and size and shape of aggregates. A high concentration of tracer particles was used to bring about and enhance the characteristic features that could develop.

The ice cylinder was grown and the final freezing process was completed by continuing rotation at the predetermined air temperature; a cross section of the cylinder was obtained by cutting it with a saw; the slice was then shaved down with a microtome to a uniform thickness of ~2 mm. A picture was taken of the entire deposit, and both a reflected light picture and a contact radiograph (Prodi, 1969) were taken of each slice. The particle deposit and distribution were examined on the radiographs by a microscope. The temperature of the cylinder was continuously recorded during and after growth until the final freezing process was completed. This was done by a thermistor embedded in the rotating cylinder. Although this was not the true average surface temperature, it was assumed to be representative of the average deposit temperature. The correction used by Ashworth and Knight (1978) was not included here because the largest diameter grown in our accretions did not exceed 3 cm and the differences were all less than 1.0°C.

3. Results and discussion

Experiments were divided into three ice growth categories: dry growth, simple wet growth and
spongy growth. Special attention was given to the transition zones between different growth conditions, and these are discussed in a separate paragraph.

a. Dry growth

A simple case of dry growth is shown in Fig. 1. The air temperature $T_a$ was $-15^\circ C$ and the deposit temperature $T_d$ was $-5^\circ C$. Practically no aggregation of particles is noted—even aggregates with only 2 or 3 particles are very few—and the deposits are quite uniform. There is some evidence at times of a layer-like deposit which is due to the slight variations in the rate of droplet spray. Close examination of these layers shows that individual particles are uniformly distributed. In the case of dry growth, therefore, it is evident that the droplets freeze individually upon capture on the surface of ice.

b. Simple wet growth

Fig. 2 shows an example of a simple wet growth where $T_a$ was $-14^\circ C$ and $T_d$ was $0^\circ C$. The thickness of concentric layers is often seen to vary due to slight variations in the rate of droplet spray and subsequent rate of freezing. Lightly and densely populated concentric layers were formed under simple wet growth. The insert in Fig. 2a shows a region of a densely populated layer with tracer particles. Close examination shows that tracer particles in these layers consist mostly of the small aggregates. The percentage of aggregates found in concentric layers ranged from about 50–80% for the dense layers, and about 30–50% for the somewhat less populated layers. It was concluded that under wet growth the freezing of the water is a continuous process maintaining a thin liquid water film above the ice; particles had sufficient time to produce small aggregates in this water layer before they were trapped in the advancing ice.

c. Spongy growth

The particle deposits seen on the microradiographs of the spongy growth show a remarkable difference from those of the dry and simple wet conditions. During spongy growth the deposit temperature $T_d$ was kept at $0^\circ C$; $T_a$ was $-15^\circ C$ (Fig. 3). In all accretions grown by this spongy condition certain characteristic features could be seen (Fig. 4). Very large aggregates and adjacent areas which are
relatively free of particles are frequently noted. Often these dense aggregates are arranged as lines of concentric rings within a short radial segment. One often sees emerging from these large aggregates lines of particles aligned radially toward the center. Lines of particles oriented with the lines of bubbles were observed (they are unresolvable from the photographs). There are also areas where these definite patterns are interrupted in the presence of lobes (Fig. 3a); here the structure is broken and one can note an appearance of converging lines indicating the direction in which liquid water is channeled outward, a mechanism which might be the origin of some of the spongy growth lobes (Morgan and Prodi, 1969).

The radial cell-like configuration of the trace particle aggregates indicates that during spongy growth the "dendrites" grow outward in the ice-water surface. The heat dissipation from the growing spongy ice to the environment, which is proportional to the heat conductivity, is nonuniform, being greater for the ice \( [2.24 \times 10^{-2} \text{ W cm}^{-1} \text{C}^{-1}] \text{ at 0°C} \) than for the liquid water \( [5.54 \times 10^{-3} \text{ W cm}^{-1} \text{C}^{-1}] \text{ at 0°C} \) (Dorsey, 1940, pp. 273, 482). The faster growing tip of the ice column in the subsurface of the liquid water starts to bulge. The nearby bulges eventually join themselves at some distance from the original solid-ice surface, incorporating the liquid water into cells in a sponge-like structure. These cells contain the particles rejected by the growing ice. During the final freezing stage the tracer particles present within each liquid cell are pushed inward by the ice growing into the interior of the cell; tracer particles are simply concentrated and form the very large aggregates observed in the x-rays. This suggested mechanism of spongy growth explains experimental observations.

d. Transition zone between different growth conditions

Particle distribution at the boundary between sections of ice grown under different ice growth conditions could yield some information which could be applied to studies of natural hailstones. It seems obvious that there must be a transition period from one growth to another and this period is determined by how abruptly or gradually a change in conditions occurs. Some general observations were made of the tracer distribution within the boundary layers, and these are illustrated in Fig. 5.

Fig. 5a shows tracer-contaminated dry growth on clear preformed ice. The tracer shows a sharp boundary and there is no evidence of particle migration at the surface or into the preformed ice.
Fig. 5b shows the transition zone between tracer-contaminated dry growth and simple wet growth. It can be seen that the boundary is still fairly sharp but that some tracer particles are within the wet-growth region. This is very likely due to the migration of particles into the still freezing liquid water when dry accretion began.

When tracer-contaminated simple wet growth is deposited on tracer-free dry growth (Fig. 5c), the transition zone is slightly better defined than in the reverse case (Fig. 5b). Only shallow penetration of the water into the dry region was observed.

The transition zone between spongy growth and dry growth is shown in Fig. 5d. In this test the tracer-free dry accretion was interrupted temporarily by stopping the spray and the air flow in the tunnel, and allowing the surface temperature of the dry deposit to warm up to near 0°C. It can be seen that tracer particles penetrated along the channels into the dry regions, most likely with the excess of water from the spongy accretion. In natural hailstones, changes in icing conditions from dry to wet and finally to spongy are probably more gradual than in this experiment.

4. Conclusions and recommendations

The degree of migration of tracer particles during freezing of ice grown by accretion of droplets defines the growth condition as dry, simple wet or spongy wet. In dry growth no migration can occur as the contaminated droplets freeze immediately upon capture. Simple wet growth results in periodic particle migration which is seen as concentric layers of small aggregates. Formation of large aggregates in liquid water cells present in a sponge-like structure was observed during spongy growth. The formation of liquid water cells was explained by the difference in heat conductivity of water and ice, the latter being larger. The fastest growing dendrites simply entrapped liquid water forming cells with tracer particles.

This work should be treated as an introduction to a different approach for studying hailstone growth. The simultaneous use of water-insoluble and water-soluble tracers should make interpretation of results less speculative. The tracer technique should be used in conjunction with other methods of analysis, including observation of crystal structures (grain size and orientations) and bubble
distribution, as well as with further theoretical studies. The freezing rates and sponginess should be determined in the future, and the relation between them and the size and mass of displaced tracer particles should be established for hailstones grown by accretion of water droplets. The use of natural tracer particles, especially ones which can act as freezing nuclei, should be considered.

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