Ice Crystal Agglomeration: T Formation

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ABSTRACT

Numerous agglomerates shaped somewhat like the letter T were observed among replicas of ice crystals collected from laboratory ice clouds and replicated using the resin vapor technique. In fact, it was found that about 55% and 60% of all aggregates were T formations when the crystals involved were columns and platelets, respectively. Several processes, including an electrical and a hydrodynamic mechanism, are considered as possible explanations for the formation of T's. The temperature gradients existing in growing ice particles were calculated, and the resulting charge separation due to the thermoelectric effect in ice is found to be far too small to account for the relative abundance of T's experimentally observed. A hydrodynamic process, however, offers a reasonable explanation for T formation.

1. Introduction

Odencrantz *et al.* (1968a) have suggested that pairs of ice crystals agglomerated into configurations resembling the letter "T" result from an electrical attraction between the crystals. They offer the hypothesis that there is a charge separation within individual ice crystals such that the basal plane faces carry an excess charge of one sign while the prism faces carry an excess charge of the opposite sign. Qualitatively, such a charge distribution would favor the formation of T agglomerates. Odencrantz *et al.* do not mention a mechanism for the charge separation within the ice crystals, but rather rely on experimental data which, they feel, indicates the presence of this charge separation. An example of a platelet T, replicated and photographed by Odencrantz *et al.*, appears on the cover of the 21 June 1968 issue of *Science.* Cheng (1967) earlier proposed a similar theory for T formation. He went on to suggest that the thermoelectric effect in ice (Latham and Mason, 1961; Takahashi, 1966) is responsible for the charge separation. Magono and Tazawa (1968) discussed various hypotheses for T formation and concluded that coulomb forces of the type Cheng suggested might be responsible for this right-angle agglomeration.

This writer has made a study of T crystals and has considered a number of hypotheses as possible explanations for their occurrence. In the case of the electrical hypothesis, it is assumed that the charge separation is due to the thermoelectric effect in ice (as Cheng assumed), and on this basis the electrical mechanism is found to be insufficient to account for the experimental observations on T formation. It is still possible, however, that another, more powerful, charge-separation mechanism is operating, but if such a process exists, it is not clear exactly what it is. Another hypothesis to account for T crystals would involve the hydrodynamic effects of the air flow around falling and interacting crystals. This explanation is favored by the present writer. Other hypotheses considered were that the collision between a supercooled water droplet and a crystal could result in a T crystal, and that by purely sublimational growth from an appropriate site on a crystal a T could develop.

2. Experimental procedure

Ice clouds were produced in a 5-ft^3 freezer by seeding supercooled fogs in the chamber with dry ice or expansion devices. Crystals were collected on glass slides placed on the bottom of the freezer and subsequently replicated using the resin vapor technique (Smith-Johannsen, 1965; Odencrantz *et al.*, 1968b). As this technique does not involve exposing the crystals to a mass replicating liquid, but only to the vapors of methyl 2-cyanoacrylate monomer which condenses and polymerizes on each particle, rafting due to surface tension was not encountered. Furthermore, the glass slides were allowed to collect the crystals for only short periods of time (~30 sec) to reduce the possibility that crystals would form aggregates by falling on top of ice particles already present on the slide.

Based on data taken from samples of 20 ice clouds produced in the experimental cold chamber, it was found that 55% and 60% of all composite crystals were T formations, when the crystals involved were columns and platelets, respectively. It was also possible to estimate the fraction of the entire population of particles in an ice aerosol that were T formations, typically 15% for platelets, 10% for columns. These last figures were very sensitive to crystal concentration. For platelets, at concentrations nearly an order of magnitude smaller than the usual concentrations obtained (about 10^3 particles cm^{-3}), the fraction of all crystals that were

T's was less than 5%, whereas at concentrations an order of magnitude larger than usual, this percentage rose to 35–40%. However, the portion of all multicrystalline forms that were T's remained relatively constant during these changes in crystal concentration.

3. First considerations

A description of T crystals will clearly indicate that T's cannot develop by growth, sublimational or from a frozen droplet, on a crystal face but must result from the agglomeration of two independently nucleated crystals. In the case of columns, a T is formed when the tip of one column becomes attached to the middle portion of another (see Fig. 1). For platelets, the periphery of one crystal will become attached to the central region of another. In both cases, orthogonality of the *c* axes of the crystals involved is characteristic. Actually, about three-quarters of all T formations exhibit exact orthogonality, the other quarter having angles more or less randomly distributed within 45° of the vertical. (There is, of course, a certain degree of arbitrariness in the definition of a T crystal). In addition, the point of juncture of T crystals is often somewhat asymmetrically located. Composite crystals resembling more the letter L than T were quite frequently observed. The ratio of T's to L's is about 5 for the platelet case.

Now, if T crystals resulted from a collision between a crystal and a supercooled water droplet, one would expect that the deviation of the *c* axes of the two crystals would be accurately restricted to one of several well-defined angles for the temperatures encountered experimentally (>-15C; see Hallett, 1964). Secondly, a crystal such as was shown on the cover of *Science*, namely a platelet T, could not develop from a frozen droplet on the basal plane face of a platelet because of disruption of the normal pattern of vapor pressure gradients that must exist to produce a hexagonally symmetrical platelet. The two crystals composing the T must have developed independently to maintain their symmetrical form. Furthermore, T's have been observed where each of the two crystals was a perfectly formed dendrite.

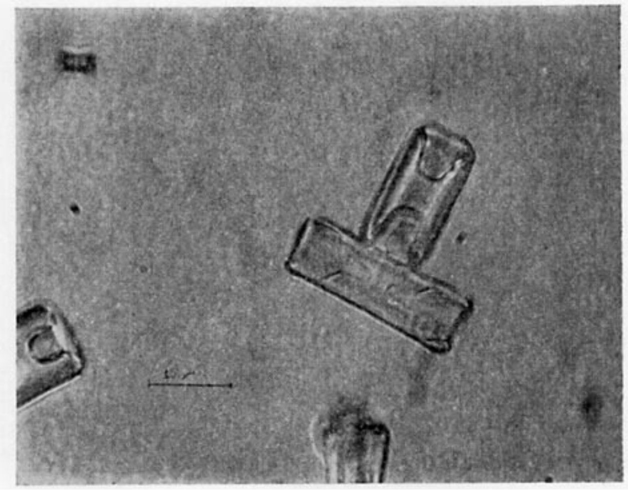

FIG. 1. Replica of a column T formed at -8C.

Thus, it is concluded that T's must result from collisions between ice crystals. Simple calculations of the expected rate of crystal collisions based on typical particle concentrations and a reasonable range of fall-speeds give results consistent wtih the observed rate of aggregate formation.

4. Electrical model

The electrical mechanism is now considered as a possible model to explain the generation of the observed large number of T's. In the case of a growing columnar crystal, because of its habit, the rate of water vapor deposition per unit area is greater on the ends than on the side of the column. Hence, the tips of the column would become slightly warmer than the central region of the particle. According to Takahashi, at any given temperature, a warm zone on a crystal will gain a charge of one sign, while the colder zone will gain a charge of the opposite sign. Two crystals possessing such quadrupole (and higher multipole) charge distributions could interact electrically. Qualitatively this model looks good; one of the most energetically stable aggregates that could form would be a T. Quantitatively, however, it is found that the forces arising from this temperature-gradient, charge-separation mechanism would not be large enough to account for the observations.

For the electrical force between two crystals to be non-negligible, it is necessary that it be at least of the same order of magnitude as the weight of the particles when the crystals are in close approach. One can easily calculate that a temperature gradient of over 10,000C cm^{-1} would be required for the electrical force between, say, two columns 30μ long and separated by a distance of the columnar radius to approach the force of gravity.

However, by standard methods of heat conduction theory, it can be shown that the temperature gradients developed in growing ice crystals, columnar or platelets, for typical laboratory conditions are only 1–5C cm^{-1}. It is concluded that the role of a temperature-gradient, charge-separation mechanism in the production of T's is negligible.

5. Hydrodynamic model

By considering the hydrodynamic forces exerted on single ice particles settling through the air and those between two particles in close approach, one can arrive at a seemingly good description of T formation in ice clouds.

From the theory of hydrodynamics, it is known that discs and needles can fall stably at any orientation through a fluid medium at sufficiently small Reynolds numbers. For larger Reynolds numbers these particles will fall such that they offer maximum resistance to the fluid. By using corn syrup and thin aluminum discs to simulate the behavior of ice platelets in air, it was found that the transition between these two regimes occurs at a Reynolds number of approximately 0.8, with the Reynolds number defined in terms of the particle diameter. This corresponds to an ice disc of radius 65μ falling through air. According to this result, ice discs whose radii are smaller than 65μ can settle through the air stably in any arbitrary orientation. This is certainly the case for the platelets formed in the experimental cold chamber.

There is some contention, however, over the value of the Reynolds number marking the upper limit of the range in which the discs can fall stably in any attitude. Jayaweera and Mason (1966) found the transition point to be less than 0.07, whereas Willmarth *et al.* (1964) feel a Reynolds number of the order of 1 is more appropriate.

The platelets encountered in the laboratory typically had radii of about 15μ, with a corresponding Reynolds number of 0.012. The present simulation experiments indicate that these particles would be randomly oriented in space in the ice clouds. They also predict that a 15μ radius ice disc, 2μ thick, would fall approximately twice as fast edgewise as broadside. Thus, the faster vertically oriented platelets would have a relatively good chance to collide with the more slowly moving horizontally oriented crystals, and T formations would be the result of such encounters. This effect is to some extent, of course, counteracted by the fact that the collision cross section is reduced compared with that for a collision between platelets falling broadside.

In the case that both interacting platelets are falling broadside and are not directly in line with one another, there is a torque on the upper particle which tends to twist it into perpendicular orientation with respect to the lower crystal. This effect has already been noted by Jayaweera and Mason (1965). They found that when the discs are displaced horizontally by less than a radius, the upper disc accelerates toward the leader and comes to rest on it at an angle, which for large Reynolds numbers is small but may approach 90° at a Reynolds number of about 5. During this interaction, the leader remains horizontal. In the present experiments, where Reynolds numbers of only a few hundredths were used, it was found that the lower disc changed orientation as the upper particle came in close approach, particularly if the centers of the discs were displaced by a radius. However, the angle formed by the two discs at contact was usually about 90°. After initial contact between the two discs had taken place, it was commonly observed that a shifting in orientation occurred, often resulting in the collapse of the T formation. In the ice crystal case, this tendency would be inhibited by the ice bond formed between the crystals (Hobbs, 1965).

Simulation experiments using columns were not carried out, but it is expected that mechanisms similar to those observed in the platelet case are involved in the formation of column T's.

Even though the particles in an ice cloud (columns or platelets), if sufficiently small, are randomly oriented in space, there are two factors which tend to favor the formation of T's; namely, the hydrodynamic torque often present in the crystal interactions which favors orthogonality of the crystal *c* axes and the relatively frequent collisions of vertically oriented crystals with those horizontally oriented. Thus, it is not unreasonable that 55–60% of all aggregates were observed to be T's. The variation in numbers of T's with crystal concentration is also consistent with the hydrodynamic hypothesis.

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