The Aggregation of Ice Particles in Clouds and Fogs at Low Temperatures[1]

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ABSTRACT

A previously derived theoretical expression, supported by experimental observations, for the rate of sintering between two ice spheres is applied to the case of adhesion between ice particles down to temperatures of −40C. Appreciable bonding is shown to take place between the particles within 10 seconds of contact even at −40C. The results predicted by the theory are found to be in quantitative agreement with the degree of sintering observed in aggregates of ice particles from natural ice fogs.

1. Introduction

In natural clouds, conditions are rarely such as to produce supersaturations in excess of a few tenths of 1% with respect to liquid water; but, with respect to ice, these same conditions correspond to supersaturations of 10% and 20% at −10C and −20C, respectively. For this reason the growth of ice crystals in clouds by condensation from the vapor phase is significantly faster than the growth of supercooled droplets by condensation. However, Houghton (1950) and Mason (1953) have shown that even in deep layer clouds, in which crystals may reside for an hour, growth by condensation alone cannot produce ice crystals greater than 1 or 2 mm in size. Crystals of this size, on melting and suffering evaporation in the dry environment between the cloud base and ground, can give rise to little more than drizzle drops at ground level. In order for ice particles in a cloud to achieve dimensions which, on melting, produce drops of sufficient size to reach the ground as large raindrops, the conglomeration of numerous crystals of ice into large aggregates must take place.

Although the aggregation of ice particles in clouds has long been recognized as an important process in the formation of rain, the exact mechanism by which ice adheres to ice, and the conditions under which this may take place, has only recently been elucidated. As a result of this work it is now clear that aggregates of small ice particles in clouds may occur at temperatures of −40C and below, whereas it had previously been thought that aggregation could only occur at temperatures above about −10C. In the present paper the experimental and theoretical facts concerning the adhesion of ice are reviewed first, then the results of Hobbs and Mason (1964) are used to predict the size of the ice bond which will form between ice particles which come into contact in natural clouds and fogs at low temperatures. Finally, the results of these calculations are compared with the recent experimental observations of Kumai (1964) on ice crystal aggregates in ice fogs.

2. The mechanism of ice adhesion

During the last ten years several laboratory investigations have been reported on the adhesion of ice to ice. Prior to this recent activity the most important work on the subject was a series of classic experiments performed by Faraday (1860) more than a hundred years ago. Faraday's experiments, however, were confined to temperatures very close to the melting point and he appears to have not investigated the adhesive properties of ice at lower temperatures. The first investigation into the effect of temperature on the adhesion of ice particles was made by Nakaya and Matsumoto (1954). In these experiments two ice spheres of about $1\frac{1}{2}$ mm diameter were brought lightly into contact, where they remained for about one minute. The force required to separate the spheres was then measured. Although the results showed considerable scatter, the force of adhesion was found to decrease with temperature from about 5 dynes at −2C to 1 dyne at −15C. Similar results were obtained by Hosler *et al.* (1957). In these experiments the ice spheres were contained in an ice saturated chamber and the forces of adhesion were measured from 0C to −30C. It was found that for spheres of diameter $1\frac{1}{4}$ cm the average value of the forces required to separate the spheres increased sharply from a few dynes at −25C to nearly 700 dynes at 0C.

In order to study the factors governing the amount of aggregation that is likely to occur in natural ice clouds, Hosler and Hallgren (1960) measured the number of crystals collected by a small ice sphere placed in the path of a moving cloud of ice crystals. The crystals were from 7 to 18 microns in diameter and were drawn past the sphere at speeds approximating to that of the terminal velocity of the ice sphere. These workers found that below a temperature of −12C the collection effi-

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ciency decreased with decreasing temperature, but above -12C the collection efficiency decreased with increasing temperature. Hosler and Hallgren suggest that the collection efficiency in their experiments reached a maximum value at -12C due to a change in the predominant crystal habit from prisms to plates at this temperature.

To attempt a theoretical explanation of the adhesion of ice at these low temperatures in terms of the long accepted "pressure-melting" theory raises immediate difficulties. The depression of the melting point of ice to -22C, for example, requires a pressure exceeding 1000 atmospheres! It may, of course, be argued that since the initial area of contact is extremely small the slightest applied force may produce large pressures at the point of contact. But pressure alone does not cause melting unless accompanied by a flow of heat from a source at a higher temperature. No such heat source existed in the adhesion experiments of Nakaya and Matsumoto (1954) or of Hosler *et al.* (1957).

An alternative theory for the adhesion of ice to ice, originally suggested by Faraday (1860) and recently reviewed by Weyl (1951) postulates the existence of a "liquid-like" layer on the surface of ice. When two pieces of ice touch, this liquid-layer is assumed to solidify at the point of contact to form a solid ice bond. This explanation for the adhesion of ice particles of low temperatures has been favored by Nakaya and Matsumoto (1954) and by Hosler and Hallgren (1960). The latter, indeed, go so far as to suggest that the liquid-layer might be thicker on the basal plane of ice than on the prism face, thereby causing plates to form aggregates more readily than do prisms. Fletcher (1962, 1963) has considered the effect of a surface liquid-layer on the total free energy of an ice surface and finds that such a layer would be stable down to -12C. Fletcher estimates that the thickness of this liquid-layer should be about 70Å at -3C and that it should decrease rapidly at lower temperatures, disappearing finally at -12C. If a liquid-layer of these dimensions does exist on the surface of ice, it should be possible to detect its presence by direct experimental techniques such as proton spin resonance, for example. Until direct evidence of this nature is obtained the liquid-layer theory must remain in the nature of a postulate.

A new approach to the problem of ice adhesion was initiated by a suggestion from Kingery (1960) that the phenomenon can be explained on the basis of sintering, a process well known in the field of powder metallurgy. Kingery observed that when two small ice spheres are pushed together to touch at a point, the area of contact between the spheres increases with time even when the original force of contact is removed. The reason for this growth is that two spheres in point contact form a system which is not in thermodynamic equilibrium because the total surface free energy is not a minimum. Under sufficient driving force a neck will therefore form between the spheres and thereby decrease the total surface area. The driving force is provided by the gradient of chemical potential existing between the highly stressed region beneath the concave surface of the neck and the remaining parts of the system remote from the neck (Fig. 1).

Kuczynski (1949) has derived expressions for the radius of the neck x, at time t between two spheres of radius r at temperature T. These expressions can be written in the general form

$$\left(\frac{x}{r}\right)^n = \frac{B(T)}{r^m}t, \tag{1}$$

where, $B(T)$ is a temperature-dependent function, and m and n are integers the values of which depend on the mechanism of material transport which is dominant in causing the neck to grow. By measuring the growth rate of the neck between two polycrystalline ice spheres, Kingery (1960) found values for m and n equal to 4 and 7, respectively and, on the basis of Kuczynski's analysis, concluded that the neck grows by the diffusion of molecules over the surface of the ice. However, from similar experiments on ice spheres, Kuroiwa (1961) obtained values of m and n equal to 3 and 5 and concluded that volume diffusion was the dominant process, particularly at temperatures above -10C.

In a recent paper, Hobbs and Mason (1964) have shown that Kuczynski's analysis for the rates of sintering between two spherical particles cannot be applied

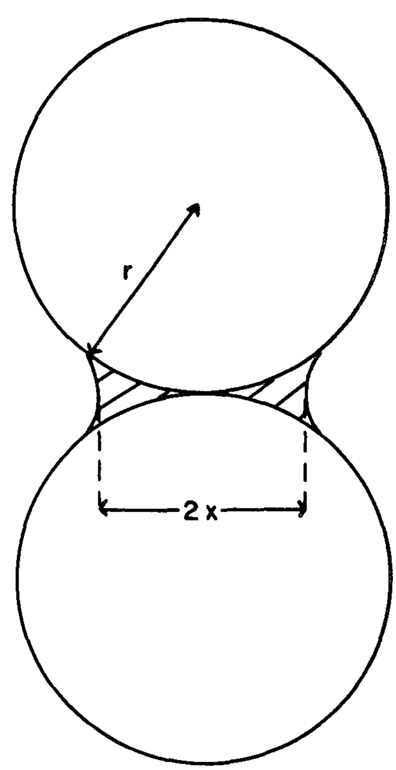


FIG. 1. The geometry of the system.

directly to the case of ice. As a result the conclusions arrived at both by Kingery and by Kuroiwa are probably incorrect. Hobbs and Mason consider that the predominant mechanism for material transport in the sintering of ice is due to the evaporation of molecules from the convex surfaces of the spheres, their diffusion through the environmental gas, and subsequent condensation at the concave neck. They show that the rate of growth of the neck by this mechanism is given by:

$$\left(\frac{x}{r}\right)^5=\frac{20\gamma\delta^5}{kTr^3}F(T)t, \qquad (2)$$

where γ is the surface energy of ice, δ the intermolecular spacing, and k Boltzmann's constant. $F(T)$ is a temperature-dependent term, referred to as the transport coefficient, which is given theoretically by:

$$F(T)=\left[\frac{kT\beta}{p_0 mD_g}+\frac{L_s^2 m\beta}{KkT^2}\right]^{-1}, \qquad (3)$$

where β is the density of ice, p_0 the equilibrium vapor pressure over a plane surface of ice, m the mass of a water molecule, D_g the diffusion coefficient of water vapor in the environmental gas, L_s the latent heat of sublimation, and K the thermal conductivity of the gas.

From measurements made on the rates of growth of the neck formed between both single and polycrystalline ice spheres, Hobbs and Mason verified that Eqs. (2) and (3) give the correct time and particle-size dependence for the growth of the neck, and agree with the absolute size of the neck, x, at time t with reasonable accuracy. On the other hand, the sintering of ice by either volume diffusion or surface diffusion was shown to be slower than that measured experimentally by four orders of magnitude.

Two stages in the growth of the neck between two spheres of ice at −20C are shown in Fig. 2. It is clear

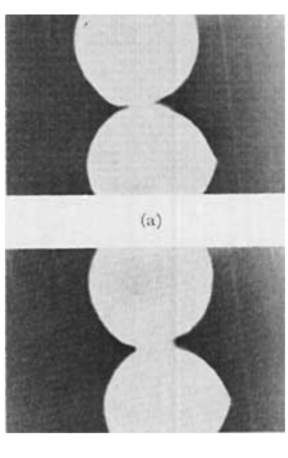


FIG. 2. The formation of an ice bond between two spheres of ice 100 microns in diameter at −20C. (a) 13 min after contact (b) 130 min after contact.

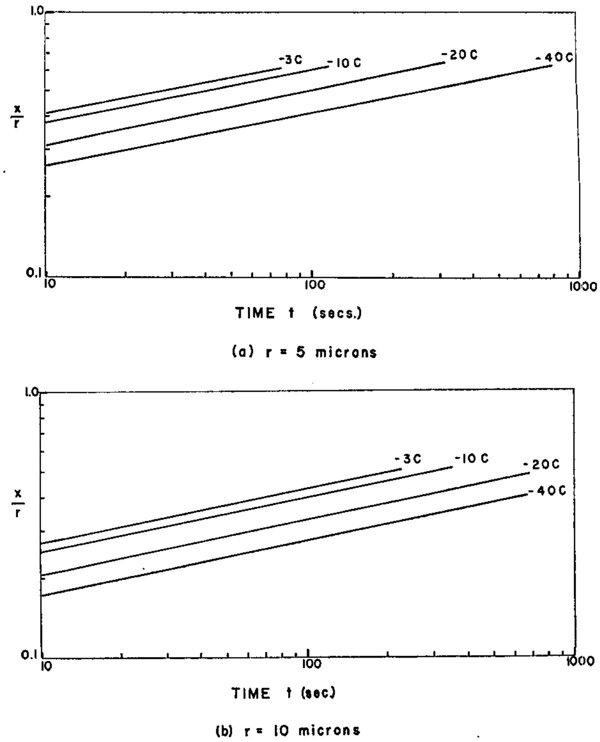


FIG. 3. Radius of the ice neck, x, formed between two spherical particles of radius r as a function of time and temperature.

from these photographs that within a short time of coming into contact the crystals are bonded by an ice neck of considerable size. It is due to the formation of bonds such as this that ice adheres to ice with considerable force.

3. Application of theory to the adhesion of ice particles in clouds

Eq. (2) can now be used, together with the experimental results of Hobbs and Mason (1964), to calculate the size of the ice bond which might be expected to form between small ice particles which come into contact within a cloud. Although the value of the transfer coefficient $F(T)$ may be calculated from expression (3) it is probably preferable to use the experimental values obtained by Hobbs and Mason. In this way any inaccuracies in the values of p_0, D_g, L_s and K are eliminated, and the slightly lower values for $F(T)$ obtained experimentally, compared with those given by expression (3), are allowed for. The experimental values for the transport coefficient $F(T)$ at −3, −10, −20, and −40C are, respectively, 1.0×10^{-7}, 6.5×10^{-8}, 2.5×10^{-8}, and 9.0×10^{-9} cm² sec⁻¹, where the value at −40C has been obtained by extrapolating the experimental results. The results of the calculations are shown in Fig. 3 where the radius of the neck x, expressed as a fraction of the particle radius r, is shown as a function of time and of temperature for two values of r.

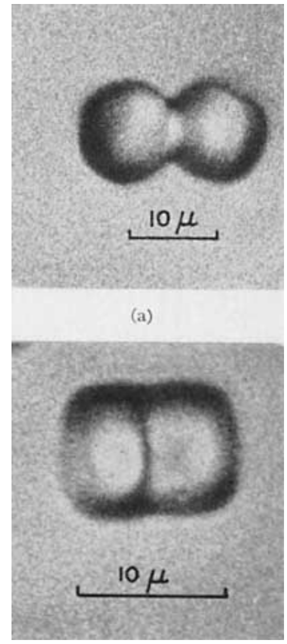
It can be seen from these results that particles of ice with radii of curvature of the order 10 microns need only be in contact for 10 seconds or so in order to develop ice bonds of appreciable size. These bonds are likely to be sufficient to keep the particles in contact despite the buffeting they may received within turbulent clouds. Pairs of particles may then make further collisions with other crystals until aggregates are formed having sufficient fall-speeds to settle out of the cloud and fall as snowflakes, or melt to form raindrops. Even at temperatures as low as −40C the rate of sintering is still rapid enough to cause large ice-necks to form between ice particles within a short period of time. The aggregation of ice particles in clouds, therefore, probably takes place at all temperatures from 0C to well below −40C.

4. Comparison of theory with observations in ice fogs

In a recent paper Kumai (1964) has reported on a study of ice fogs at Fairbanks, Alaska. Ice fogs in this area normally form only when there is a temperature inversion and the ground temperature is below −37C. From observations made on ice particles collected at ground level, Kumai found that the majority of the particles in these fogs were spherical and that 74% of the particles collected consisted of aggregates of two or more spherical ice crystals.

Two examples of the particles collected by Kumai are shown in Fig. 4. The close similarity between the bonding of the crystals shown in Fig. 4(a) and those shown in Fig. 2 suggests that the neck of ice between the particles has formed by the same mechanism in both cases. It needs to be shown, however, that the rate of sintering given by Eq. (2) is sufficiently rapid at −39C to form an ice bond of the size depicted in Fig. 4(a) within the lifetime of the particles in the fog. The ratio of x/r for the crystals shown in Fig. 4(a) is 0.62 and r is approximately 5 microns. From the theoretical curves shown in Fig. 3(a) the time needed for $x/r=0.62$ at −40C, and for particles of 5 micron radius, is 760 seconds. Now, since the fall speed for ice spheres of 5 microns radius is about 1 cm sec^{-1}, the crystals would need to fall a distance of 7.6 meters in order for a neck of ice of the size shown in Fig. 4(a) to form by the evaporation-condensation mechanism. Kumai reports that the ice fog layers at Fairbanks are usually about 10 meters in thickness, a fall-distance of 7.6 meters is therefore quite reasonable. These calculations show that the theory of sintering developed by Hobbs and Mason provides a quantitative explanation of the degree of bonding that is observed to take place in natural ice fogs.

The form of the adhesion between the crystals shown in Fig. 4(b) is obviously quite distinct from that observed to take place between ice crystals in the laboratory (Fig. 2) or that shown in Fig. 4(a). The formation of a neck between two ice spheres by the evaporation-



(b)

FIG. 4. Ice aggregates from natural ice fogs at −39C.

condensation mechanism does not result in a change in the distance between the centers of the two spheres. In Fig. 4(b), however, it is apparent that the two spheres have become distorted and do not have a common tangent at their original point of contact. Further examples of this type are shown by Kumai (1964) in Fig. 8 of his paper. Kumai suggests that this distortion of shape may result from the development of facets on the two crystals after they have become sintered together. The appearance of these aggregates, however, suggests that they have formed by the collision and subsequent rapid freezing of two supercooled droplets. It is possible that while these small droplets remain isolated they do not freeze, even though the temperature is close to the homogeneous nucleation threshold of −41C, but upon colliding with another supercooled droplet, spontaneous freezing takes place during the process of coalesence.

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