On the Role of Shock Waves and Adiabatic Cooling in the Nucleation of Ice Crystals by the Lightning Discharge

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ABSTRACT

A cloud of small supercooled water droplets was subjected to shock waves of reproducible intensity in the laboratory. Nucleation of freezing occurred only when the gas driving the shock waves was cooled to −37C or below by adiabatic expansion and subsequently mixed with the droplet-bearing air. Passage of the shock wave did not produce nucleation of the cloud. Results of a numerical model of a lightning discharge show, in the pressure wave from a lightning discharge, that the degree of cooling by adiabatic expansion is probably too small to produce ice crystals by homogeneous nucleation.

1. Introduction

In a previous publication, Goyer (1965a) reviewed the possible effects of the shock wave generated by lightning discharges on hydrometeors. Laboratory studies with a shock tube have demonstrated that bulk supercooled water could be induced to freeze at temperatures above the spontaneous nucleation temperature by shock waves with overpressures of the order of 0.07 atm (Goyer *et al.*, 1965). This effect was tentatively explained by a mechanism involving cavitation. In addition, field experiments in the supercooled plume of Old Faithful Geyser (Goyer, 1965b) showed that supercooled water droplets are indeed shattered and frozen when subjected to shock waves with overpressures as low as 0.007 atm.

Vonnegut and Moore (1965) suggested that adiabatic cooling produced in the expansion wave behind the shock front could lead to homogeneous nucleation of ice crystals, which would seed the larger droplets. This process cannot be responsible for the results of the shock tube and field experiments, because of the low strengths of the shock waves involved. Its plausibility in the case of the presumably much stronger pressure waves generated by lightning discharges will be discussed in this communication.

Further laboratory work has firmly demonstrated that dynamic nucleation of freezing in bulk supercooled water is closely related to the generation and subsequent behavior of bubbles, and that wall effects may have played an important role in the laboratory shock tube results of Goyer *et al.* A report on this further work will be published shortly. In this paper we report the results of experiments in which a cloud of supercooled water droplets, free of wall effects, is subjected to shock waves of known intensity, and discuss the relative effects of mechanical disturbance and adiabatic cooling in triggering of freezing. In addition, the results of a preliminary calculation of the pressure profile behind a cylindrically expanding shock front are used to estimate the magnitude of the adiabatic cooling effect in the pressure wave from a lightning discharge.

2. Experimental procedure

A cloud of supercooled water droplets was subjected to shock waves of reproducible intensity. The cloud was generated by condensation of water vapor from a vessel of warm water placed in the bottom of a domestic deep-freeze box. No attempt was made to characterize the droplet size distribution, but the average droplet size was probably of the order of a few microns, and the size spectrum relatively narrow. Shock waves were produced by rupture of an aluminum foil diaphragm which sealed a 5-cm i.d., 450-cc compression chamber. The shock waves radiated more or less spherically into the cloud, so that the shock strength was a monotonically decreasing function of distance from the source. The bursting strength of the diaphragms was 2.18±0.025 atm, under which conditions the shock wave overpressure was 0.2 atm at 5 cm from the diaphragm. The compression chamber was gradually pressurized with dry nitrogen, after an initial purging period, until spontaneous diaphragm rupture occurred. The initial temperature of the compression chamber could be varied in order to control the final temperature of the nitrogen after expansion to atmospheric pressure. The occurrence of nucleation of the ice phase was detected by visual observation of the cloud, using a collimated light beam in a darkened room; the increase in scattered light from a cloud of liquid droplets upon freezing is well known and very marked. Results of the experiments are shown in Table 1. The table gives the initial temperature of the gas in the compression chamber, its calculated final temperature after an adiabatic expansion to atmospheric pressure, and a rough estimate of the initial temperature of the deep freeze, taken from the reading of a thermometer lying

TABLE 1. Nucleation by adiabatic expansion.

Run no.	Bursting pressure (atm)	Temperature (°C) Freezer	Initial	Final	Extent of nucleation
3	2.26		26.0	-43.0	total
2	2.19		25.0	-41.0	total
5	2.21		27.0	-40.0	total
18	2.16	-19.5	26.5	-39.6	total
6	2.14		28.0	-39.0	total
7	2.22		29.5	-39.0	total
9	2.23	-21	30.0	-39.0	total
12	2.21	-23	30.5	-37.5	incomplete
8	2.18		29.5	-37.0	total
11	2.19	-25	31.25	-36.3	total
10	2.18	-21	30.5	-36.0	little
13	2.19	-22	32.0	-36.0	none
16	2.18	-26	33.0	-35.0	little
14	2.14	-25	32.0	-34.2	little
17	2.16	-24	33.0	-34.0	very little
15	2.16	-25	33.5	-33.3	none

in the bottom of the box. The extent of nucleation reported is a visual estimate of the change in light scattering properties of the cloud as a result of the experiment. The results range from "total" nucleation, in which light scattering from large numbers of ice crystals completely overwhelmed the visual appearance of the water cloud, to "very little," in which case only one or two ice crystals, easily distinguished, were observed in the light beam at any time.

It is seen that there is a very strong correlation between the temperature of the nitrogen after expansion and the nucleation of the ice phase. In all runs in which the final gas temperature was -36C or above, very few if any ice crystals were formed; in all runs but one at lower temperatures, nucleation appeared to be total. These results show that, for shock waves of this intensity and for a cloud of very small water droplets, mechanical disturbance by the shock wave has no nucleating effect. This is perhaps to be expected, since the resistance of droplets to deformation and shattering increases markedly as radius decreases. In addition, nucleation cannot be associated with any cooling occurring in the rarefaction wave that follows the shock through the droplet bearing air, since the amplitude of this wave was also constant. (Some preliminary experiments had been carried out using compressed air, which had not been dried, to drive the shock waves. The change to dry nitrogen was made to avoid the possibility of formation of ice crystals *in situ* in the air, which could have obscured any nucleating effect attributable to the pressure wave. The results of these experiments were indistinguishable from those using dry nitrogen.) It therefore appears that formation of ice crystals was due solely to mixing of the cold, dry nitrogen with the moist, droplet-bearing air in the deep freeze.

Evidence for the efficiency of homogeneous nucleation of ice in adiabatically cooled moist air was obtained from experiments with small plastic bubbles, similar to the toy balloon experiments of Vonnegut (1948). Some packing material widely used in the shipping industry consists of two plastic sheets sandwiching small hemispherical air bubbles about 0.84 cm in diameter. Bursting such hemispheres (simply by pinching them between the thumb and forefinger) in a cloud of supercooled water droplets leads to the rapid transformation (within 4 sec) of the cloud to an ice crystal cloud. Apparently, the air in the bubbles is cooled, when a bubble bursts, to the point that homogeneous nucleation of ice, from the moisture initially present in the bubble, occurs. Vonnegut estimated the concentration of ice crystals thus produced to be about 1.6×10^{10} cm^{-3} of air expanded. Schaefer (1954), in experiments on the breaking of small pressurized glass spheres, also observed ice crystal concentrations in the range 10^{10}–10^{11} cm^{-3}. Maybank and Mason (1959), on the other hand, found a limiting value of about 3×10^{6} ice crystals cm^{-3} from adiabatic expansion of moist air. By sampling and counting the number of ice crystals which settled to the bottom of the freezer in our experiments, assuming a uniform dispersion of ice crystals throughout the box, we obtained an output of 0.4×10^{10} crystals cm^{-3} of air in the bubble, in quite good agreement with Vonnegut and Schaefer but considerably higher than Maybank and Mason's results. Since we did not characterize the contents of the plastic bubbles used in our experiments, it is not possible to account quantitatively for the observed differences. It is of interest to note, however, that in Schaefer's and Vonnegut's experiments, as well as in ours, the adiabatically cooled air parcel was released into an already formed cloud of supercooled droplets. Mason and Maybank, on the other hand, carried out their experiments in the absence of a cloud but in an atmosphere initially saturated with respect to water at -10 or -20C. It is possible that the increase in available water in the experiments carried out in supercooled clouds leads to growth on nascent ice crystals which would be too small to compete for the more restricted quantity of water vapor present in Maybank and Mason's experiments.

In any case, large numbers of ice crystals can be generated by adiabatic cooling of moist air. Bursting of a single small plastic bubble appeared to produce at least as many ice crystals as were formed in our large expansion chamber experiments, and in less time. Typically, the appearance of wholesale nucleation in the expansion chamber experiments took place about 8 sec after diaphragm rupture; this difference, however, could possibly be due to differences in the rate of convective transport of the ice crystals into the path of the light beam in the two experiments.

An interesting observation is that, at final nitrogen temperatures between -33 and -36C, few ice crystals are formed, and icing never propagates through the entire cloud. The crystals which are formed are conspicuous, grow quite large, and settle to the bottom of the freezer compartment without affecting the general appearance of the water cloud. These observations indicate that the seeding mechanism is the rapid growth, by sublimation, of the nascent ice crystals originally formed, and not freezing of supercooled water droplets by collision with nascent ice crystals. If a collision process were operative, the presence of a few crystals should lead in time, through a multiplicative process, to the total icing of the cloud. This was never observed.

3. The pressure wave from a lightning discharge

In general, the sudden release of energy at a point or along a line in a fluid results in the propagation of a radially expanding, steep-fronted compressive wave, followed by a more gradual expansion wave. The expansion reduces the pressure below ambient for a short period before final equilibration of pressure is established. This "negative pressure" phase is a general consequence of the geometry of spherical and cylindrical flow, and is observed in blast waves from explosions, "sonic booms" from supersonic aircraft, exploding wire phenomena, etc. The magnitude of the cooling associated with this overexpansion of the gas in the pressure wave surrounding a lightning discharge is estimated here on the basis of a preliminary numerical integration of the equations of fluid motion for cylindrically symmetric flow. Details of the calculation will be published at a later date.

The equations of motion for compressible fluid flow in cylindrical coordinates (where all quantities are functions only of a radial coordinate and the time) comprise a set of nonlinear partial differential equations [see, for example, Courant and Friedrichs (1948)], for which no general solutions have been obtained. Approximate solutions have been obtained under certain rather limiting assumptions; however, the investigation of such systems over a wide range of conditions generally requires a numerical integration of the equations on a digital computer. We have carried out such a calculation, patterning the work after the computational scheme of Richtmeyer (1957). The calculation bears a strong similarity to that of Brode (1955), who treated the spherical pressure wave problem. The following simplifications are made in the calculation:

1. The lightning discharge column is very long compared with radial distances of interest, and is straight.
2. The atmosphere is a perfect gas with constant specific heats and a specific heat ratio $\gamma = c_p/c_v = 1.4$.
3. The energy of the lightning discharge is instantaneously liberated in a column a few centimeters in radius, which subsequently expands and drives the pressure wave.

The assumption of a straight lightning column is one whose effect is hard to assess. At large distances from the discharge, its effect is probably small, but the random local curvature of the column of actual lightning strokes may give rise to significant deviations at distances comparable to the local radius of curvature. The second assumption is obviously far from reality in and near the discharge column itself. However, Brode (1955) has shown that the inclusion of real gas properties, in the spherical case, only results in a reduction in the efficiency with which the initial excess energy is converted into an acoustic disturbance. In other words, the pressures, as a function of distance and time, calculated using a realistic equation of state for high-temperature air, are essentially no different from those calculated using the simple ideal gas equation of state but with a slightly lower value for the initial energy input. As for the third assumption, the precise nature of the energy input mechanism obviously affects the pressure wave near the axis, where pressures and temperatures are high. However, after the shock wave has propagated a distance about 10 times the radius of the initial column of hot gas, the differences in pressure due to varying initial conditions are largely damped out, and the pressure-vs-distance curves as a function of time can practically be superimposed. Varying the input energy in the calculation has the effect only of changing the radius and time at which given pressure and temperature profiles are observed. Since we are interested in the behavior of the system when the pressure behind the shock front approaches atmospheric conditions, the last two assumptions will thus have little effect on the results.

We therefore feel that the results of this computation give a reasonably close approximation to the behavior of the pressure wave from a lightning discharge in the region of space and time during which adiabatic cooling from the "negative pressure" phase of the pressure wave could be important.

As a specific example, we choose for initial conditions a column of air 3.81 cm in radius (3 inches diameter) whose internal energy has been increased by a factor of 520 over its ambient value. This corresponds, in terms of real air properties, to a temperature of about 24,000K, which is near the average temperature measured spectroscopically in actual lightning discharges (Prueitt, 1963), and to a total energy input of about 6×10^3 joules cm^{-1}. Schonland (1964) gives for the energy input a value of the order of 2×10^4 J cm^{-1} for a lightning flash; since a flash involves an average of 3 or 4 return strokes, our value is a reasonable approximation to the energy per stroke estimated by Schonland. The atmosphere surrounding the discharge column is at 300K and 760 mm Hg.

Figs. 1 and 2 show computed profiles of pressure and temperature, respectively, as a function of distance at selected times in the history of the pressure wave. The evolution of the wave is briefly described here. At first, a very strong compressive wave propagates outward. The strength of the wave decreases rapidly, however, as its frontal area increases directly with the distance from the axis. For the first 1.3 msec, the pressure behind the front is everywhere positive. At about 1.3 msec, the pressure at the axis decreases to the ambient value. The pressure continues to decrease with time at all points behind the shock front, however, until it reaches a minimum value at the axis of about 0.87 atm (0.13 atm below ambient) which persists during the interval from about 3–4 msec after initiation. During this time, the pressure is a monotonically increasing function of the radius, and is substantially constant over the region from the axis out to a point about one-third of the radius of the shock front. As the wave progresses still farther, the pressure everywhere begins to relax back to the ambient value. This relaxation begins at the origin, so that the point of minimum pressure moves away from the axis, as can be seen on Fig. 1.

The temperature of the gas behind the shock front does not return to the ambient value when the pressure does, for reasons to be discussed in the following. Fig. 2 shows that the temperature remains very high near the origin, decreases with radius to a minimum value, and then increases to a lower maximum at the shock front. The minimum in the temperature profile reaches 300K only at 4.4 msec, at which time the pressure minimum has passed its peak and is relaxing back to atmospheric pressure. The lowest temperature attained behind the shock front is about 295K, at about 9 msec after the start of the process. After this time, the minimum value of the temperature also relaxes back to the ambient value, and its location continues to move away from the axis.

The maximum degree of cooling below the ambient temperature, then, appears to be only of the order of −5C. Using the simple adiabatic expansion equation, one would expect an expansion from atmospheric pressure to 0.87 atm to produce a cooling effect of about −12C. The physical reason for this difference lies in the irreversible nature of a shock wave compression. By the second law of thermodynamics, the entropy of a system increases during any irreversible process. If an element of fluid initially at a pressure p_0 is compressed via a shock wave process to a higher pressure p_1, and then allowed to expand adiabatically back to p_0, its final state will be at a higher temperature than its initial state, and the increment in temperature will correspond to the entropy increment resulting from the shock wave compression. The effect is very small for weak shock waves, since in this limit the entropy increment varies as the cube of the overpressure, but by the same criterion it increases rapidly as the shock strength increases. In the case of the cylindrically expanding shock wave from a lightning discharge, the shock strength is very high near the axis, and the gas temperature here remains high after final equilibration of pressure. This is the reason for the minimum in the temperature-vs-radius curves, even though the pressure may be a monotonically increasing function of radius out to the shock front. Adiabatic cooling, therefore, can only be significant in those regions which are not traversed by strong shock waves; namely, those regions relatively distant from the lightning discharge column. Since the pressure excursion below ambient is small

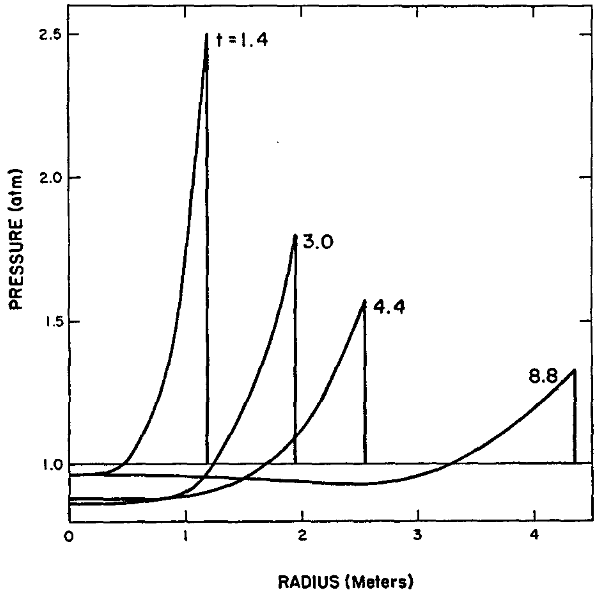

FIG. 1. Radial pressure profiles computed for a model lightning stroke, for times indicated (in milliseconds after energy release).

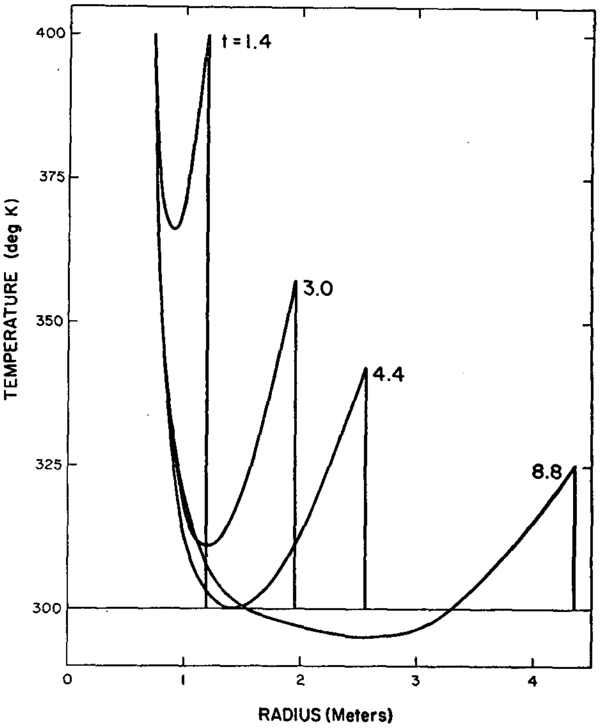

FIG. 2. Radial temperature profiles computed for a model lightning stroke, for times indicated (in milliseconds after energy release).

in these regions, the degree of adiabatic cooling in the pressure wave from the lightning discharge is also small.

4. Discussion

The homogeneous nucleation of ice crystals during rapid adiabatic cooling of moist air is not surprising, and has been observed many times by others. The process is an exceedingly efficient generator of large numbers of ice crystals. Our experiments also show that mixing of cold dry gas which is at or below the Schaefer point (about −37 to −40C) with warmer saturated air is also a relatively efficient ice crystal producer. It is difficult to see, however, the importance of such a process in atmospheric phenomena. Two other aspects of the results of the present work should be briefly discussed; namely, nucleation of freezing by shock waves, and nucleation of freezing due to the temperature field surrounding the lightning discharge.

Nucleation of freezing by shock waves. As noted, we have observed no nucleation attributable to the mechanical disturbance from the shock waves in our experiments. The essential conditions of the experiments, i.e., relatively weak shock waves, and water droplets in the cloud droplet size range or smaller, would both seem to make such an effect unlikely. The results seem to be in contradiction to those of Ives (1941) who reported nucleation in supercooled natural fog banks from the firing of a revolver and the blast from an automobile horn. Although no attempt was made here to characterize our droplet size distribution, our cloud is definitely not comparable to the supercooled plume of Old Faithful Geyser (Goyer, 1965b), where the liquid water concentration, average droplet size, and droplet size variation were all much higher, and where nucleation was induced on a large scale with even weaker shock waves. The mechanism of nucleation in the Yellowstone Park experiments is still unclear. The conditions in the updraft of a convective storm are still different from those of either our laboratory tests or the Yellowstone Park experiments; it is a yet unanswered question whether mechanical disturbances can have an appreciable nucleating effect under such conditions.

Nucleation by adiabatic cooling in the temperature field surrounding a lightning discharge. The numerical model of the lightning stroke and its acoustical consequences reported here indicate that adiabatic cooling is not likely to be great in the vicinity of lightning strokes. Vonnegut and Moore (1965) have estimated that regions of cloud at temperatures as high as −18C could be momentarily cooled to −40C in the pressure wave from a lightning discharge. We feel that this estimate of the magnitude of the effect to be expected is far too large, for two reasons. First, their calculation of the temperature lowering did not take into account the irreversible heating produced by shock wave compression. Second, their estimates of underpressures were based on measurements of blast waves from explosions. The pressure wave from an explosion expands in three dimensions, compared with the two dimensional expansion of a cylindrical wave, and apparently leads to a more pronounced pressure minimum. Brode (1955), in a calculation essentially similar to ours, but for the spherical case, found a minimum pressure of 0.8 atm, compared with 0.87 atm in our results. A further effect arises from the great difference in initial conditions between a high explosive detonation and a simple release of additional energy in the atmosphere, as occurs in a lightning discharge. A high explosive detonation generates a large quantity of excess gas, in addition to the energy released. The impetus from the expansion of this gas apparently leads to a still greater degree of overexpansion in the surrounding atmosphere after passage of the shock wave. In a numerical calculation of the blast wave from a spherical charge of TNT, Brode (1959) found that the pressure could drop below 0.7 atm at points near the explosive charge.

A temperature drop of −5C as predicted by our model is probably too small to produce significant numbers of ice crystals by homogeneous nucleation. This could occur only where the cloud temperature was already near the Schaefer point, at which temperatures it is likely that many natural freezing nuclei would already be active. It is possible that under optimum conditions, heterogeneous freezing nuclei could be activated by a momentary temperature drop. The effect would certainly be limited to small particles. The short duration of the period of temperature lowering (a few milliseconds) would preclude any significant cooling effect on larger particles or droplets.

5. Conclusions

In summary, we draw the following conclusions from these results:

1. A cloud of micron-size supercooled droplets is unaffected by the passage of moderately weak shock waves.
2. Rapid cooling, either by adiabatic expansion or by a mixture of cold gas, is an efficient process for generating ice crystals.
3. It is unlikely that rapid adiabatic expansion will produce significant cooling and homogeneous nucleation of ice in the pressure wave from a lightning discharge.

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