Cold Gas Traps for Ice Particle Formation

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Water, with the characteristic angular structure of its molecule, is an extraordinary substance for a variety of reasons. For example, its excellent properties as a solvent allowed for the possibility of life on Earth. Likewise, its density anomaly, with maximum density at +4°C, allows ice to float on water, which affects inland waters and arctic oceans. Also, its comparatively high melting and boiling temperatures and large latent heat have strong implications for weather and climate. To date, many details of the basic processes related to water's impact on the weather remain unclear (*1*). These include the formation of different types of precipitation, the radiative properties of the resulting particles, and the chemical behavior of the surfaces of the particles.

Another intriguing property of water is its capability for sonoluminescence: small gas bubbles in water can be positioned in the pressure antinodes of a stationary ultrasonic field (SUSF), and caused to pulsate with the sound frequency and to luminesce (*2*). This phenomenon of single-bubble sonoluminescence, which is not well understood at present, has not been observed with any other liquid.

Here, we describe an effect that can be considered as inversely similar in its experimental conditions: if strong temperature gradients exist in a gaseous environment, water or ice aerosol gathers in the pressure node areas of a SUSF and forms ice and snow particles (*3*). The SUSFs are generated by ultrasonic levitators like the ones used in other disciplines, such as material science, fluid dynamics, and analytical chemistry (*4*). They work at the same frequencies at which sonoluminescence has been seen [20 and 58 kHz (*5*)]. The aerosol particles have roughly the same size and shape as the objects in the sonoluminescence experiments, but are small, dense spheres of ice or supercooled water, not little bubbles floating in a water matrix.

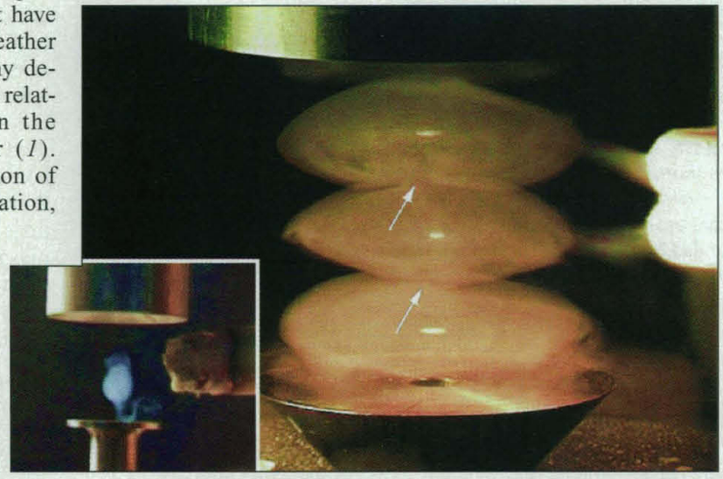

Fig. 1. **(Right)** Cold gas traps, shown as four rotational ellipsoids that form around the pressure nodes of a stationary ultrasonic field (SUSF) at an acoustic frequency of 20 kHz (the uppermost node is hard to see). Cold ice aerosol is sucked into the ellipsoids through two narrow "channels" from the right. Thin "necks" (arrows) allow the exchange of material between zones. In each pressure node, an ice particle has been formed. **(Left)** In contrast, warm aerosol such as cigarette smoke cannot be confined to the pressure nodes in a stable way, but is expelled from the nodes through the little "tongue" that appears at the bottom right. There is evidence that heavy gas can be trapped in an SUSF in a way similar to the trapping of cold ice aerosol (*11*).

The effect can be separated into two different types of phenomena, neither of which has, to our knowledge, been observed previously. First, a fog of cold water and ice near the SUSF region is sucked into the SUSF and distributed there in prolate rotational ellipsoids in such a way that it surrounds the pressure node centers (Fig. 1, right). Adjacent ellipsoids exchange aerosol through thin "necks" (Fig. 1, arrows) and thus tend to equalize the corresponding aerosol volumes. This results in a relative temperature constancy of a few kelvins inside the ellipsoids and a sharp boundary with the surrounding gas, with a temperature gradient of up to 50 K over a few millimeters. The trap effect cannot be produced with a warm aerosol: cigarette smoke, for example, is expelled from the SUSF pressure node areas (Fig. 1, left). Therefore, we propose that the zones of the SUSF pressure nodes be termed "cold gas traps."

The second phenomenon is the formation of larger ice particles from ice aerosol in the pressure node areas, or motion antinodes, of the SUSF at temperatures between –25° and 0°C (Fig. 1, right). Three basic mechanisms for this phenomenon can be distinguished (*3*): (i) the agglomeration of individual aerosol particles in or near vortices; (ii) the agglomeration of several secondary particles, that is, of particles that formed according to process (i) [an example of this process is observed with the larger particles formed from relatively small secondary particles (Fig. 2, A through C; horizontal arrows)]; and (iii) the growth or degradation of an ice particle via the gas phase by deposition or evaporation of water molecules. This process was observed on larger ice and snow particles that were stationary in the SUSF (Fig. 2, D through I). All three processes occur in nature.

The decisive property responsible for the agglomeration is the presence of a quasi-liquid layer (QLL) on the surface of the ice particles (6). In atmospheric research, a lot of attention is focused on the QLL because of its suspected key role in particle agglomeration, in the adsorption of gases, and in surface-catalyzed chemical reactions (1). We note that, compared to experiments in aerosol chambers, the technique used here offers the advantage that all these processes can be studied without the perturbing wall effects of any experimental containment. The particles are levitated and fixed in the SUSF pressure nodes and can easily be investigated with physical (for example, optical) and chemical methods. An example is the process of change of shape of a snowflake (Fig. 2, D through F).

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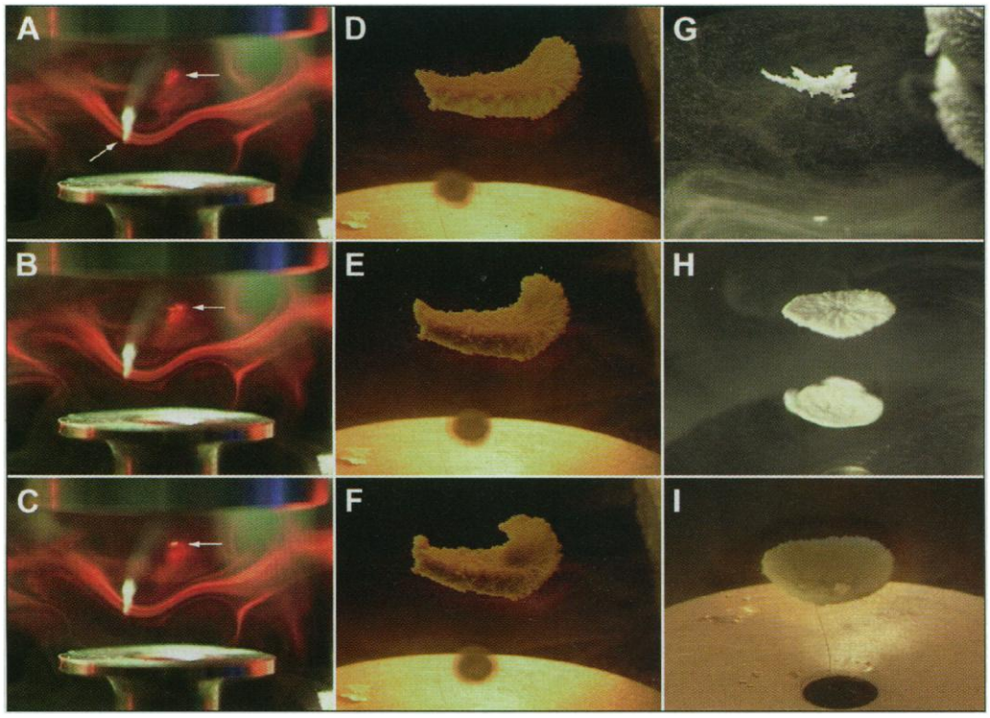

Fig. 2. (**A** through **C**) Process of ice-particle formation from cold ice aerosol in the SUSF at 58 kHz. The center vertical plane is illuminated by a sheet of red helium-neon laser light. The time interval between consecutive frames is 0.16 s. A small cloud of secondary ice particles starts developing [(A) and (B)] and solidifies (C) as an ice particle with a diameter of 0.6 mm (horizontal arrows). The diagonal arrow in (A) marks a mantle thermocouple. (**D** through **F**) Process of shape evolution of a snowflake in the SUSF at 20 kHz (10 mm in length at the beginning). (**G** through **I**) Different manifestations of snow and ice particles generated in the SUSF at 20 kHz. (G) Snowflake, 5 mm in length, surrounded by secondary ice particles with a size of 10 to 100 μm. (H) Prolate snowflake with furrowed surface, 7 mm in diameter, with a similar rotating object below. (I) Shell-shaped rotational ice body, 7 mm in diameter. Apparently, the latter does not exist in nature.

By varying the experimental parameters and perturbing the SUSF symmetry, a great variety of ice and snow particles can be generated and investigated. Their sizes range from 10 μm to >10 mm. Most of these particles show shapes that also occur in nature (Fig. 2, D through H). The shell-shaped rotational ice body (Fig. 2I) demonstrates that shapes that do not exist in nature can also be formed through the use of this technique. Compared with cloud chambers of one or several meters in size (*7*, *8*), our experimental setup (*3*) is much smaller and simpler. A question that remains to be answered is how the SUSF acts on the process for forming ice particles. The occurrence of so many natural shapes among the produced particles suggests that the influence of the SUSF on particle formation is likely to be small.

Local distortion of the SUSF allows the particle to be rotated around arbitrarily oriented axes. Usually, the vertical axis and two horizontal axes will be of practical interest. This feature has applications in the determination of integrated scattering phase functions of small ice or other particles (*9*). These phase functions describe the dependence of the intensity of scattered light such as sunlight on scattering angle, and thus have important implications to Earth's radiation budget (*8*).

An interesting generalization of the technique may be the increase of the temperature difference between sites outside and inside the pressure node areas. This difference is now about 50 K, but can be increased considerably if aerosol that has been brought to liquid nitrogen (–195°C) or liquid helium temperature (–269°C) can be trapped. There are many applications of such a "low-temperature minilab," one of them being optical spectroscopy of supercooled gases (*10*). Moreover, the present 50 K temperature difference could be "shifted" by changing the ambient temperature. Thus, aerosols of substances other than water become amenable to the same kind of investigations, provided the aerosols are some tens of degrees colder than the surroundings.

Still another application could be the investigation and production of granulated material directly from the gas phase. This technique need not be limited to homogeneous material, but could be exploited for the production of layered and coated particles or of particles that, at some stage of their production, undergo a chemical reaction. Instead of a chemical reaction, an additive could also initiate a change in physical state. For example, the addition of a liquid could allow the agglomeration of very fine dust particles that are otherwise hard to produce in granulate form.

Finally, it should be mentioned that our first experiments performed with bromine gas at homogenous temperatures indicate that high-density gas in a low-density environment is also trapped in the SUSF pressure node areas (11).

In summary, a phenomenon has been shown to exist in which cold aerosol gathers in the pressure node areas of a stationary ultrasonic field to form liquid and solid particles of a great variety of sizes, shapes, and properties. Because gravity has little influence on the observed phenomenon and orientation in space is not a limitation, aerosol reactors based on the effects described could be used both in terrestrial and spaceborne facilities. The technique we describe is likely to find applications in basic research, such as atmospheric physics and chemistry, or in industrial processes like the production of granules for medicine and other purposes.

References and Notes

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