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Theoretical model of ice nucleation induced by inertial acoustic cavitation. Part 2: Number of ice nuclei generated by a single bubble

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ABSTRACT

In the preceding paper (part 1), the pressure and temperature fields close to a bubble undergoing inertial acoustic cavitation were presented. It was shown that extremely high liquid water pressures but quite moderate temperatures were attained near the bubble wall just after the collapse providing the necessary conditions for ice nucleation. In this paper (part 2), the nucleation rate and the nuclei number generated by a single collapsing bubble were determined. The calculations were performed for different driving acoustic pressures, liquid ambient temperatures and bubble initial radius. An optimal acoustic pressure range and a nucleation temperature threshold as function of bubble radius were determined. The capability of moderate power ultrasound to trigger ice nucleation at low undercooling level and for a wide distribution of bubble sizes has thus been assessed on the theoretical ground.

1. Introduction

1.1. Context and aim

Controlling the distribution of ice crystal sizes is a key issue in industrial freezing and freeze-drying processes. Ultrasound is already known to be able to initiate the nucleation of ice in undercooled aqueous solutions and to make the freezing reproducible at the desired temperature. But ice crystals' size predictive tools are still missing for design and optimization of ultrasound assisted freezing processes.

As concerns sono-crystallization from solutions, quantitative theoretical prediction of nucleation was pioneered by Virone et al. [1] and applied to ammonium sulphate crystals. However as concerns crystallization from melts and to our best knowledge, Saclier et al. [2] were for the time being the only authors proposing a fully predictive model of ice nucleation triggered by inertial acoustic cavitation. Their model was applied to a 1 ml pure water sample with a known (measured) bubbles' size distribution and the total number of nuclei was calculated as function of the acoustic pressure and water temperature. The model involved however several simplifications and approximations, especially concerning heat balance of the bubble, heat transfer in the water around the bubble and water thermo-physical properties at very high pressures. The goal of this study was to develop a finer and more comprehensive modeling and to propose nucleation thresholds for bubbles of different initial radii.

The authors of the present study have chosen to continue working with the 'pressure rise effect' [3] (see next section) as the nucleation mechanism and set up a comprehensive theoretical model starting from the bubble wall motion induced by inertial cavitation (see part I) and finishing with the number of nuclei generated by the collapse of a single bubble. In the previous paper [4] (part 1), the pressure and temperature fields close to a bubble undergoing inertial acoustic cavitation were simulated and it was shown that extremely strong liquid water pressures but quite moderate temperatures were reached near the bubble wall just after the collapse.

The next step described in this paper (part 2) was to develop a model of ice nucleation and integrate the nucleation volume rate equation over space and time, using the pressure and temperature profiles induced by the collapse of the bubble in order to obtain finally the number of generated nuclei.

As concerns nucleation kinetics, the classical equation for primary stationary homogeneous nucleation was applied [5], but the parameters of this equation had to be made pressure and temperature dependent.

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1.2. Bibliographical review

In this section a short literature review concerning first the experimental evidence of the effect of ultrasound on ice crystallization and second the theories about the ice nucleation mechanism by ultrasound, will be presented.

Inada et al. [6] and Zhang et al. [7] studied the potential application of ultrasonic waves to produce ice slurries. They found out that applying ultrasound greatly increased the probability of phase change from undercooled water to ice at a given initial water temperature. The probability of phase change was calculated as the ratio of the number of successful freezing tests to the total number of tests in given conditions. The occurrence of phase change induced by ultrasound increased with the total number of gas bubble nuclei in undercooled water, independently of other experimental conditions.

Chow and co-authors [8,9] evaluated the effect of ultrasounds on the primary and secondary nucleation of ice in sucrose solutions. They presented direct optical observations of crystals formed around an immersed sonication probe, around a single levitating and cavitating bubble and in a special ultrasonic cold stage. They pointed out that the primary nucleation of ice in sucrose solutions can be achieved at higher nucleation temperatures in the presence of ultrasound. It was also shown that the nucleation temperatures of ice increase with increasing ultrasonic power. It was observed that pre-existing ice crystals could be fragmented by ultrasound, which modifies the size distribution and generates nucleation sites.

Nakagawa et al. [10] and Hottot et al. [11] carried out freezing and freeze-drying experiments with an aqueous solution of mannitol in a small glass tube (vial). The vials were cooled down and sonicated by means of a cold vibrating plate. Ice crystals were observed using reflected-light optical microscopy over frozen samples in transversal and longitudinal sections. It was shown that the nucleation could be readily triggered at selected sample temperature values below the equilibrium freezing temperature and that small and numerous ice crystals were obtained at lower nucleation temperature (higher undercooling level), while large and directional ice crystals (dendrite type) were obtained at a higher nucleation temperature (lower undercooling level). Saclier et al. [12] continued the work on mannitol solution sono-freezing in vials. Using the same apparatus and methodology, they confirmed the above qualitative results but further adopted a second order experimental design and quantitatively assessed the effect of both the nucleation temperature and the acoustic power on the final crystal size and shape. The ice crystal size was found to decrease with both the level of undercooling and the acoustic power level, whereas their circularity was found to increase with these parameters.

Kiani et al. [13] studied ultrasound-assisted nucleation of pure water, sucrose solution, and agar gel samples inserted in tubing vials. The vials were immersed in an ultrasonic bath. Ultrasound was applied continuously for different durations and at different sample temperatures in the range up to 5 °C below the freezing point. They observed that ultrasound can trigger ice nucleation with high repeatability at the targeted temperatures and hence can be used to control the onset of nucleation. In a continuation of this study [14], the effect of ultrasound intensity on the nucleation of ice in agar gel samples was additionally studied. It was observed that ultrasound irradiation was able to initiate nucleation at different undercooling levels of the gel if optimum intensity and duration of ultrasound application were chosen.

Although ultrasound has long been used to initiate nucleation in undercooled aqueous solutions, the exact mechanism that explains this effect is not yet well known. Acoustic cavitation (the sudden formation and collapse of gas bubbles in liquids by means of ultrasound) appears to cause the nucleation of ice. There is a distinction between stable cavitation when a bubble pulsates about an equilibrium radius over many acoustic cycles and inertial cavitation when the bubble grows extensively and finally collapses [15].

According to the theoretical study by Hickling [3] very high positive pressures occurring during the final stage of the collapse of a bubble in inertial cavitation increase the equilibrium temperature of water and ice (VI or VII, denser than liquid water), thus enhancing nucleation of a specific ice solid phase. The weak point of this scenario is the uncertainty about a subsequent transformation of the nucleated high pressure ice phase into low pressure regular ice Ih. According to another model [16], nucleation is caused by negative pressures that follow the collapse of the cavitation bubble. This effect will produce low pressure ice Ih, but this idea was questioned by the experiments of Ohsaka and Trinh [17].

On the ground of the Hickling's hypothesis, Inada et al. [6] made an attempt to deduce the probability of phase change from undercooled water to ice from the probability of inertial acoustic cavitation of a given bubble population. But in order to fit their experimental results, they needed to adjust two parameters and their model was not predictive.

According to some other experimental results [9], the moderate oscillation of a bubble in stable cavitation may also induce ice nucleation. These authors suggest micro-streamings as a factor promoting nucleation. The other possible nucleation mechanism is the concentration and agglomeration of ice clusters near the bubble due to pressure diffusion (transfer mechanism driving the densest species toward high-pressure zones). On the basis of theoretical considerations, Grossier and co-workers [18,19] argued that the very high pressure gradients that are needed for pressure diffusion to be effective are attainable only for collapsing bubbles. They thus indirectly refuted stable cavitation as a nucleation initiator.

As suggested by Kordylla et al. [20] and Yasui et al. [21], the occurrence of gas/liquid interface (at the bubble wall) in a supersaturated sonicated solution may induce a kind of heterogeneous nucleation, by reducing interfacial solid–liquid tension and thus reducing the nucleation energy barrier (ΔG_c) according to the classical nucleation theory (CNT). Kordylla et al. [20] has in fact identified the contact angle minimizing a least-square error between experimental and theoretical values. This is however rather a 'curve fitting' approach than a real physical model of sono-nucleation. Some studies have demonstrated that single bubbles do not exhibit this effect [8].

More importantly, applying CNT in a global manner for sono-crystallization is not straightforward, since this theory relies on a (metastable) equilibrium hypothesis. At the scale of a bubble, there exists huge pressure gradients (and to a lesser extent temperature gradients) near the bubble, so that the supersaturation itself is distributed over space and time. Thus, considering the nucleation work (ΔG_c) as constant is a somewhat rough approximation in this situation. Possibly, one could invoke a local equilibrium hypothesis, calculate a spatio-temporal profile $\Delta G_c(r,t)$ around the bubble, and deduce a local instantaneous nucleation rate. This is the idea underlying the 'pressure effect' tentatively quantified in the literature [1,2] and developed in this study.

However, even when accounting for such refinement, the hypothesis of stationary nucleation is always done. In fact, local supersaturation varies on the same time scale as the bubble radius (or shorter in the case of shock waves). If this time scale is of the same order of magnitude as the so-called nucleation time lag, nucleation may be in fact transient. Thus, estimations based on quasi-stationary nucleation may be largely overestimated [22].

A quite novel idea about the nucleation mechanism has arisen very recently [23]. The physics of inertial cavitation states that the bubble interior heats up due to a quasi-adiabatic compression just before the collapse, but then cools down due to a