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The effect of ultrasonic waves on the nucleation of pure water and degassed water

Deyang Yu [a,b], Baolin Liu [a,*], Bochun Wang [a]

[a] University of Shanghai for Science and Technology, Shanghai 200093, China
[b] Henan University of Urban Construction, Pingdingshan 467036, China

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ABSTRACT

In order to clarify the mechanism of nucleation of ice induced by ultrasound, ultrasonic waves have been applied to supercooled pure water and degassed water, respectively. For each experiment, water sample is cooled at a constant cooling rate of 0.15 °C/min and the ultrasonic waves are applied from the water temperature of 0 °C until the water in a sample vessel nucleates. This nucleation temperature is measured. The use of ultrasound increased the nucleation temperature of both degassed water and pure water. However, the undercooling temperature for pure water to nucleate is less than that of degassed water. It is concluded that cavitation and fluctuations of density, energy and temperature induced by ultrasound are factors that affect the nucleation of water. Cavitation is a major factor for sonocrystallisation of ice.

1. Introduction

Several studies have been done on ultrasonic-induced nucleation of solid crystals in a variety of liquids. The experimental results have indicated that the presence of an ultrasonic pressure wave can increase the nucleation temperature of liquids (varying from organic fluids to metals) [1–7]. It is usually considered that the cavitation bubbles in supercooled water generated by ultrasonic vibrations can cause the nucleation of ice crystals [8–11]. According to the theoretical model described by Hickling [12,13], the high pressure generated by the collapse of a single cavitation bubble is sufficiently high to increase the equilibrium freezing temperature of liquids, thus causing nucleation of solid crystals. Generally, it is difficult to observe directly the cavitation events in an experiment due to the transient nature of cavitation bubbles, their small dimensions and large population [14,15]. Therefore the exact mechanism of ultrasonic-induced nucleation of ice is still under debate [16,17]. Although Hickling [12] reported that a single ice nucleation can be generated by an isolated stable cavitation bubble in an ultrasonic sound field. The question as to whether cavitation is a necessary condition of ultrasonic-induced nucleation for liquids is still unanswered [2].

The effect of ultrasonic waves on the nucleation of pure water and degassed water was investigated experimentally in this paper. The mechanisms of nucleation of ice induced by ultrasound waves were discussed.

* Corresponding author. Tel.: +86 21 55271180; fax: +86 21 55270695.
E-mail address: blliuk@163.com (B. Liu).

2. Materials and methods

2.1. Experimental apparatus

The experimental apparatus is shown in Fig. 1. It is composed of five main components:

(1) *Ultrasonic bath system:* An ultrasonic bath system (SCQ-600, Shanghai Shengpu Ultrasonic Equipment Plant, China) consists of an ultrasonic generator, 12 ultrasonic transducers and an ultrasonic tank. The 12 piezoelectric transducers are evenly bonded to the bottom of the ultrasonic tank. Ultrasonic waves are delivered from the bottom to the liquid medium in the tank. The output power of the ultrasonic waves can be adjusted in the range 0–600 W. The frequency of the ultrasonic waves is 40 kHz.
(2) *Sample vessel:* The sample vessel is a glass tube with an outer diameter of 11 mm, inner diameter of 10 mm and height of 100 mm. The thin wall of the tube is essentially transparent to the ultrasonic pressure wave.
(3) *Cooling system:* The ultrasonic tank is filled with the ethylene glycol–water solution (50% by volume). The solution is cooled by circulating the mixture through a refrigerated circulator (DL-4020, Ningbo Scientz Biotechnology Co., Ltd., China). The sample vessel filled with sample (approximately 1 cm^3) is immersed in the mixture.
(4) *Temperature acquisition system:* The system is composed of a T-type thermocouple, a set of ADAM modules (ADAM-4018/4520, Advantech Company, China) and a computer. The temperatures both outerside and inside a test tube are

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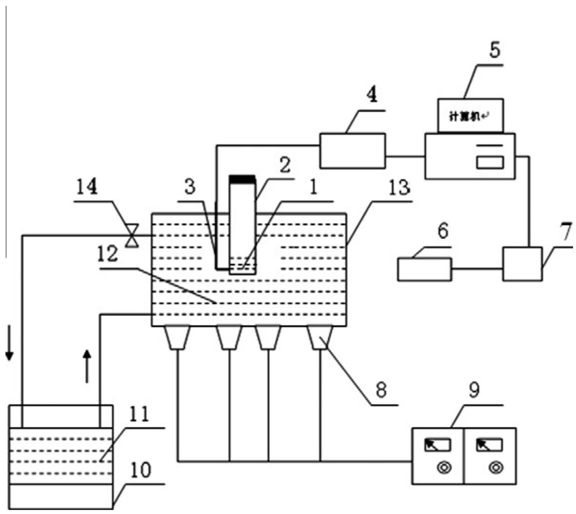

Fig. 1. Schematic diagram of experimental apparatus (1 – water sample; 2 – a test tube; 3 – thermometer probe; 4 – ADAM modules; 5 – computer; 6 – a telescopic lens; 7 – video camera; 8 – ultrasonic transducer; 9 – ultrasonic generator; 10 – refrigerated circulator; 11 and 12 – coolant; 13 – ultrasonic tank; 14 – flow control valve).

monitored. It was confirmed by preliminary experiments that the outside temperature of a test tube is very close to the sample temperature, because the wall of the tube is thin and the cooling rate of the sample is slow to ensure the heat transfer from outside to inside.

(5) *Image collecting system:* During the experiments, a video camera (640 × 480 pixels) equipped with a telescopic lens (MV1.3H/SVM-20, Shanghai WeiTu Optics & Electron Technology Co., Ltd., China) is used to monitor the nucleation of ice. Thirty photographs can be taken by this camera in 1 s. The nucleation of ice and its location in the test tube can be clearly observed.

2.2. Sample preparation

The samples are pure water and degassed water. Pure water was made by filtering tap water through a 5 μm filter, an ion exchanger and an automatical double pure water distillatory. Degassed water was obtained by boiling pure water for 30 min to remove the air and then eliminate the effect of cavitation bubbles on the nucleation of ice [18,19]. The test tube filled with the degassed water (approximately 1 cm^3) is heated by an alcohol burner. Furthermore, the mouth of the tube is upward inclined. When steam erupted from the tube, a rubber stopper should be immediately fit into the mouth of the tube and heating should be stopped.

2.3. Experimental methodology

In order to investigate the mechanism of the effect of ultrasound, two different types of experiments have been conducted. In the first set of experiments, the nucleation of pure water has been performed under the influence of the ultrasonic waves (i.e. in the presence of cavitation). In the second set, the nucleation of degassed water has been carried out in the presence of the ultrasonic field (i.e. in the absence of cavitation). Six intensities of the ultrasonic waves (I_{diss}) are applied, which are 0.0, 0.26, 0.32, 0.37, 0.42 and 0.49 W/cm^2, respectively. The dissipated acoustic intensities (I_{diss}) in the water samples were obtained by a calorimetric method. Measuring the rate of temperature increase due to the conversion of ultrasound energy into heat and calculating I_{diss} according to the following expression:

$$I_{diss} = (dT/dt)_{t=0} C_p \cdot M/A_p \tag{1}$$

where C_p is heat capacity of water (J kg^{-1} $°C^{-1}$), M is mass of water sample (kg), $(dT/dt)_{t=0}$ is the initial slope of the curve of temperature versus time (°C/min) and A_p is the cross-section area of the bottom of the test tube (mm^2).

For each experiment, the temperature of water sample is decreased from room temperature to −20 °C. The cooling rate is approximately 0.15 °C/min and the outside temperature of the test tube is recorded at 1 s intervals (placing a thermocouple close to the outside surface of the test tube). The ultrasonic waves are applied intermittently from the measured temperature of 0 °C until the water in a sample vessel nucleates. The interval for every ultrasonic treatment is 45 s. The time for each ultrasonic treatment is 5 s. As the intensity of ultrasound and the temperature of fluid might vary with the position in the ultrasonic tank, care is taken to keep the sample vessels in the same position during each experimental run. About 20 samples are used for the experiment under the same conditions. A new water sample and test tube are used for each experiment to avoid effects of preceding experiments.

Although thermocouple in supercooled water will initiate nucleation, the inside temperature of the test tube is also monitored (placing a thermocouple directly in the water sample) to confirm that cavitation was produced or not in the water sample.

In order to explore the influence of various acoustic effects on the nucleation of ice (no ice nucleation induced by the thermocouple), a video camera equipped with a telescopic lens is used to record the freezing process of pure water and degassed water in an ultrasonic field (placing a thermocouple close to the outside surface of the test tube).

3. Results and discussion

3.1. Effect of ultrasound on the nucleation temperature

Fig. 2 is a typical freezing curve of pure water with ultrasound being applied at an intensity of 0.37 W/cm^2 (temperature being measured outside the test tube). It shows the point at which the temperature of the pure water sample rapidly increased induced by the latent heat of crystallisation, which was recorded as the nucleation temperature or onset of nucleation of ice. The absolute value of a difference between the nucleation temperature and 0 °C is called the degree of supercooling at freezing, ΔT. The heating effects caused by ultrasound vibrations induced a raise in the temperature of the water sample (Fig. 2). However, the heat induced by ultrasound is much less than the latent heat of crystalli-

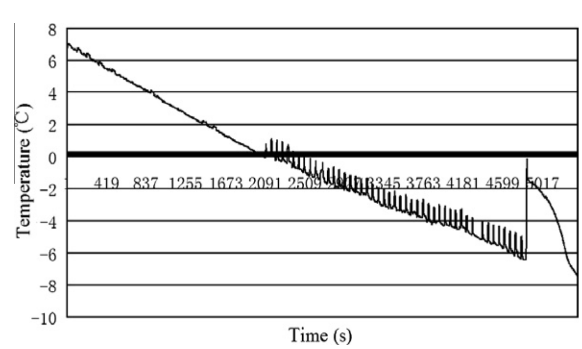

Fig. 2. Typical freezing curve of pure water with ultrasound being applied at an intensity of 0.37 W/cm^2 (temperature being measured outside the test tube).