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Analysis of focused indirect ultrasound via high-speed spatially localized pressure sensing and its consequences on nucleation

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ABSTRACT

Ultrasonication is frequently used to promote crystallization and chemical reactions, with many papers reporting the effects of ultrasonic parameters. High-speed pressure inducers (aka pinducers) and signal processing are used here to gain insights into the spatial localization of energy dissipation that occurs during ultrasonication, facilitating a design that allows ultrasonication to be spatially localized inside a tube, without requiring that the probe directly contact the solution. The fluid pressure measured inside of a flexible silicone tube pressed against an ultrasonic probe is observed to move between nearly constant amplitude oscillations to variable shape and amplitude waveforms that deviate significantly from being periodic. Ultrasonic power is transferred to the fluid inside the tube, generating a wide frequency range including via bubble oscillations. Discrete Fourier Transform analysis indicates complex interactions within the experimental system, in addition to the energy transferred inside the tubing at the expected ultrasonic source frequency. The total acoustic intensity decays exponentially with distance from the zone, dropping by more than two orders-of-magnitude for each 1 cm increase in distance. The pinducer data analyses guide the design of an experimental setup in which crystals nucleate within fluid inside the zone in the tube, right under the ultrasonic probe.

1. Introduction

Ultrasonication has been widely used in daily life and research laboratories, such as glass surface cleaning, pigment dispersion, metal casting, cell disruption, and facilitation of crystallizations and chemical/polymeric reactions [1–13], related to the phenomenon of acoustic cavitation (growth and collapse of microbubbles under an ultrasonic field) [1,14–16]. Ultrasonication has also been used to aid crystallization, such as accelerating nucleation, reducing crystal size, reducing aggregation, or changing polymorphic ratios [4,8–11,13]. Although the exact sonocrystallization mechanism remains to be established [4,15,17,18], several ultrasonic parameters (e.g., ultrasonic intensity or power, time, and volume [1,4,5,19]) have been identified effective in enhancing the control of crystallization processes.

Most bench-scale continuous sonocrystallization researches are based on configurations including an ultrasonic probe or bath. The advantages of probes (most directly inserted into/towards the solution as in Refs. [11–13,20,21]) include that (1) an ultrasonic probe can deliver much higher ultrasound intensity than the ultrasonic bath, with convenient adjustment of power [11–13,20,21]; and (2) there are usually more options of commercially available probes with different shapes and sizes [22]. Ultrasonic baths (where the container was placed such as in Refs. [17,23]) also have advantages, such as (1) lower requirement on sample thermal stability and temperature control, with less contamination probability due to no direct contact with solution; and (2) flexible container shape [17]. A recent multiphase millifluidic tubular crystallizer design combined the advantage of probe (focused energy) and bath (indirect ultrasonication) for its continuous nucleation process, tuning nuclei size with the ultrasonic amplitude [5].

With existing studies as a good starting point, the below questions remain to be answered for future ultrasonication-based nucleation designs: (1) how to narrow the nuclei size distribution, which directly affects the size distribution of product crystals [5,17,24]; (2) how to increase the scalability of the ultrasonication process without having to acquire new equipment [14,25]; and (3) will the ultrasound intensity remain the same if running continuously instead of as short pulses [26]. These questions cannot be addressed easily during crystallizations, which would require many trial-and-error experiments, costing time and materials.

To efficiently approach the above questions and better understand the process, this article measures and analyzes the “primary effect” (this term is used as in Ref. [14]) of ultrasonication, which can directly affect the commonly studied crystallization outcome (the “secondary effect”). The pressure is chosen for measurement, as the direct consequence of

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ultrasonication into liquid is a pressure wave, which causes while being affected by other phenomena such as the generation of micro-bubbles (cavities) and high temperature [4]. Hydrophones are sensors that measure liquid pressures based on a piezoelectric transducer that generates electricity when subjected to a pressure change (e.g., at pulse-mode ultrasonication [6,14]). This article employs a pinducer [6,27], which is a hydrophone of small geometric size (to minimize possible effects on the acoustic field), to measure pressures at multiple spatial locations during continuous ultrasonication.

This article is the first to use a pinducer to facilitate the design and optimization of a crystallization process, specifically, a nucleation subsystem based on a focused indirect ultrasonication zone to enhance reproducibility and the control of nuclei size distribution. Compared to most past ultrasound-aided crystallization studies that focused on the effect and final outcome of the ultrasonication generator's parameters (e.g., power input, frequency) or additive conditions on crystallization [1–4,10,12,13], we quantify the direct pressure consequence of ultrasonication at different configurations for guiding the nucleation process design, which adds value to past studies. With intensified learning of physical phenomena (e.g., bubble dynamics) based on the straightforward signal data analysis (not a "black-box"), this article also provides deeper insights and optimization suggestions for ultrasonication-involved crystallization designs, using tubular nucleation as an example (e.g., with more localized ultrasonic region than in [5]).

2. Materials and methods

2.1. Sonication and pinducer equipment

The experimental setup for pinducer measurement (Fig. 1) consists of an ultrasonic processor (Sonics® VCX750, frequency 20 kHz, input electric power 750 W), an ultrasonic probe (Sonics® 25 mm solid full wave probe, made of Titanium alloy Ti-6Al–4 V) to induce mechanical vibrations, a pressure transducer (pinducer) as a signal receiver (Belden 8216 RG-174/U Type 1C26), and a tube (Masterflex BioPharm Plus platinum-cured silicone tubing, ~3 mm inner diameter, ~6 mm outer diameter) connected front-to-end and filled with deionized water, all inside a plastic tray container of water. The ultrasonic processor and probe, and silicone tube used for pinducer measurement were the same models/operation conditions (e.g., ultrasonic amplitude at 50% of max amplitude) as in a recent continuous crystallizer design with ultrasound-aided nucleation [5]. In this article, only relatively low-frequency ultrasonication (20 kHz) is used, in comparison to high-frequency ultrasonication used for attenuation spectroscopy (e.g., 1–150 MHz as in Ref. [28]).

A needle-shaped pinducer with a tip diameter of 0.28 cm was inserted into the tubing (with the tip inside the fluid inside the tubing) to measure the pressure voltage inside the tubing without significantly changing the tubing geometry. The measurement part of pinducer is its tip only (according to the vendor). The signal from pinducer is sent to a data acquisition board (NI PXI-5105 12-bit 60 MS/s digitizer) installed in a personal computer running Microsoft Windows running LabVIEW, version 2011 that stores time-stamped data onto a hard drive. The data acquisition setup and operator is the same for rock physics [27], except that neither physical amplifier nor filter was used, based on the signal strength. After a warmup time of 1 min (with ultrasonication on), a total number of 82 continuous time signals of pressure voltage (referred to as "traces" [27], in unit of voltage, see Fig. 2 for examples) were collected within 5 min of continuous ultrasonication, meaning an interval of ~3.7 s. For each trace, the first 0.1 s was recorded without reducing the speed of collection (these data rates fill the computer's random access memory, with the data saved to a disk drive after each experiment). The temperature of water inside the plastic tray container was measured and confirmed to show no evident heating before and after the continuous ultrasonication. After the experiments, the data were analyzed and plotted (e.g., Figs. 2–4) using Matlab (version 2013b).

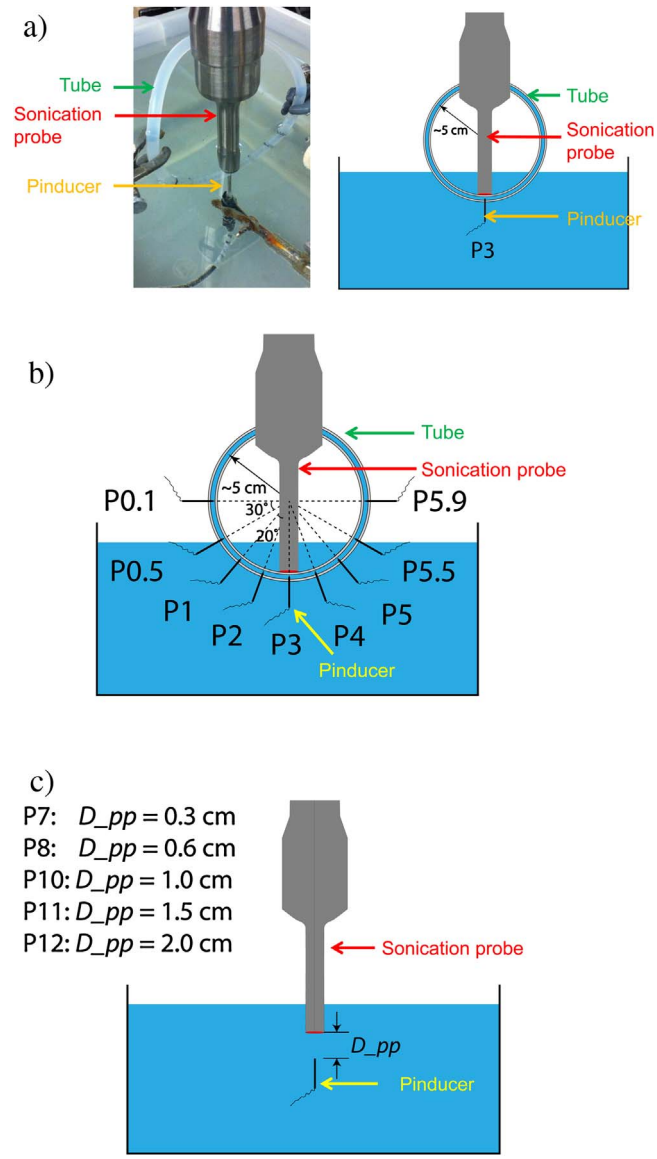

Fig. 1. (a) (Left) Photograph and (right) diagram of one measurement location (P3 in Fig. 1b) of the pinducer setup for pressure measurement inside the tubing. The ultrasonic probe was pressed against the outer wall of tubing, to which the pinducer was inserted perpendicularly. (b) Diagrams of various measurement locations (P0.1, P0.5, P1, P2, P3, P4, P5, P5.5, P5.9) of the pinducer setup for pressure measurement inside the tubing. The pinducer was inserted into the tube at different distances (1/18 or 1/12 of a circle of ~5 cm diameter between adjacent measurement points as labelled 20° or 30°, respectively) from the ultrasonic probe (e.g., "P3" whose photo is Fig. 1a). (c) Diagrams of various measurement locations (P7, P8, P10, P11, P12) of the pinducer setup for pressure measurement along the centerline of the ultrasonic probe (*D_pp* is the distance from measurement location to probe top along the centerline). No tubing was used in (c). For (a) to (c), experimental details are described in Subsection 2.1 and Table 1.

2.2. Nucleation validation and materials

Based on the learnings from the pinducer experiments described above, an ultrasound-aided cooling nucleation validation experiment was designed, with detailed justification in Subsection 3.2. The nucleation setup (configuration in Fig. 5) used the same ultrasonic equipment and conditions (e.g., focused indirect ultrasonic configuration, ultrasonic amplitude, media choice of water, and tubing choice) as for the pinducer measurement. The nucleation occurs inside of a solution of fluid inside of the tube. L-asparagine monohydrate (purity

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