

# On the variations in the size distribution of bulk nanobubbles in response to static pressure increment

静的加圧下のナノバブルのサイズ分布

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## 1. Introduction

Nanometer-sized air bubbles (that is, nanobubbles) have the potential to serve as cavitation nuclei in sonochemistry [1, 2]. In a prior study, Yasui et al. determined the radii of nanobubbles and the acoustic amplitude best suited to sonochemical reactions [3]. Such work has shown that it is possible to generate oxidants such as  $\text{H}_2\text{O}_2$ , atomic O, OH radicals and  $\text{O}_3$  using nanometer-sized bubbles in conjunction with the appropriate frequency and pressure amplitude.

Nanobubbles can be classified into surface and floating bulk nanobubbles [4]. Recently, several studies investigated the effect of increasing the static pressure on bulk nanobubbles, as a means of elucidating the mechanism responsible for their formation and stability [5-7]. To the author's knowledge, size distribution changes in response to static pressure increment during pressurization has not been studied, so far, although the measurements have been made for the size-distribution changes before and after pressurization showing that pressurization at  $5 \times 10^4$  kPa increased the size of bubbles while decreasing the number concentration [5]. The present study mainly represents the first examination of the size distributions of nanobubbles under increasing the static pressure. The role of nanobubbles under sonication is also discussed.

## 2. Experiment

Nanobubbles in pure water (Millipore Essential Elix 5) were prepared using a bubble generator apparatus (YBM Faby-10) based on the impact of bubble cavitation and the shear force imparted by vortex flow, as described in a previous publication [8]. A syringe pump (Teledyne ISCO 260D) was employed to pressurize the nanobubble dispersions. This pump was connected to a rectangular glass cell (inner volume:  $10 \times 5 \times 12$  mm<sup>3</sup>) in a size distribution measurement apparatus (Shimadzu SALD-7500X10) via 0.5 mm inner diameter PEEK tubing. Prior to each trial, a 30 mL sample of the nanobubble water was introduced into

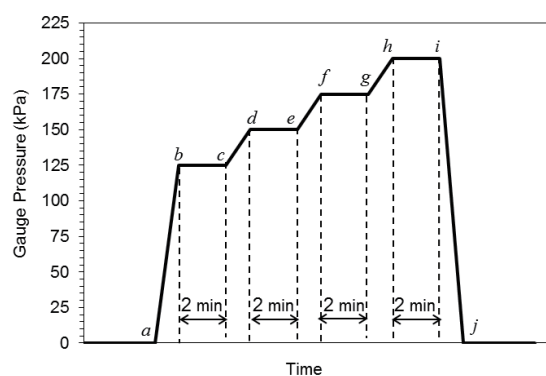


Fig. 1 The time course of the pressurization of nanobubbles in water. The time intervals from *a* to *b*, *c* to *d*, *e* to *f*, *g* to *h* and *i* to *j* are each 1 min. The time intervals from *b* to *c*, *d* to *e*, *f* to *g* and *h* to *i* are each 2 min.

the cell using the syringe pump so as to fill the interior of the cell, while 21-24 mL of the nanobubble water was maintained in the syringe pump and any excess water or air was expelled from the system. In this manner, the measurement system was filled with nanobubble water and could be pressurized by forcing additional water into the cell via the syringe. During size measurements at atmospheric pressure and under pressurization there was no headspace air layer over top of the nanobubble water in the cell. The nanobubble water was maintained at 23 °C using a temperature controller. **Fig. 1** shows the time course of nanobubble water pressurization applied by the syringe pump. Prior to pressurization, the entire system (including the syringe pump, tubing and cell) were at atmospheric pressure. The gauge pressure was subsequently increased over the span of 1 min and then maintained at the new, higher value for 2 min and this process was repeated up to a maximum pressure of 200 kPa. Size distribution measurement was made for each of the following timing before pressurization, during each staying at 125, 150, 175, 200 kPa in the gauge pressure. After that, the static pressure was returned to atmospheric one.

### 3. Results and Discussion

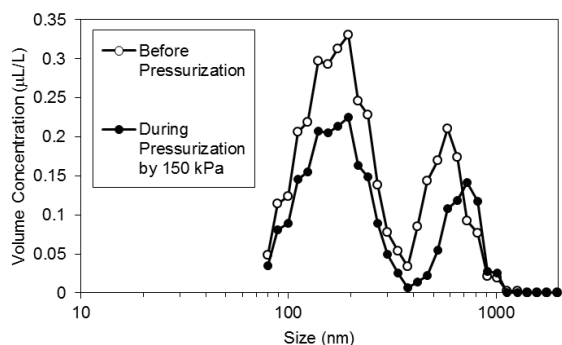


Fig. 2 Comparison in size distribution of nanobubbles between before and during pressurization by 150 kPa as volume concentration representation.

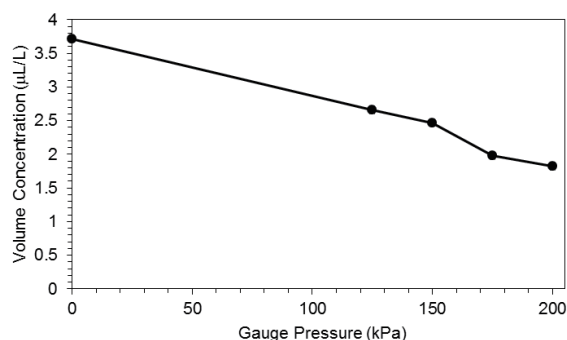


Fig. 3 Total volume concentration of nanobubbles as function of gauge pressure.

**Fig. 2** shows a comparison in size distribution of nanobubbles between before and during pressurization by 150 kPa as volume concentration representation. It was found that at most of the size on the distribution the volume concentration decreased by pressurization. There appeared two maxima of 194 nm and 584 nm in size distribution before pressurization. By pressurization the latter maximum shifted towards the larger size (584 to 728 nm), while the former stayed the same.

**Fig. 3** plots the variation in the total volume concentration against the gauge pressure. The total volume concentration is obtained by integrating volumetric concentration over entire distribution as shown in Fig.2. It was found that as the gauge pressure increased the total volume concentration decreased. Because the total volume of nanobubbles was decreased by pressurization, the present results confirm that at least many of the nanobubbles were gaseous.

The role of nanobubbles under sonication is discussed here through the measurements of the

intensity of sonoluminescence (SL). SL is light emission from a cavitating liquid exposed to intense ultrasound [9]. The intensity of SL from water in the presence/absence of nanobubbles with 54 kHz continuous-wave sinusoidal ultrasound was measured for various acoustic amplitude. It was found that, at excessively high amplitude where a decrease in the SL intensity was often observed, the SL intensity in the presence of nanobubbles was higher than that in the absence. Similar results by the addition of particles [10] were obtained for the measured intensity of luminol sonochemiluminescence [11] generated when oxidizing radicals react with this solute [9].

Thus, the decrease in volume concentration of nanobubbles during static pressure increment indicated that at least many of the nanobubbles were gaseous. Nanobubbles have a potential to provide nucleation sites for sonochemistry.

#### Acknowledgment

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