Progress in Ultrasonic Cleaning Research*

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Abstract  Physical cleaning based on underwater ultrasound is widely used in industry and its cleaning efficiency is known, from recent studies, to be augmented by promoting the mechanical activity of cavitation bubbles. As a side effect, ultrasound cleaning may give rise to material damage from violent collapse of cavitation bubbles. Traditionally, degassed water is favored as cleaning solution in order to reduce the probability of having cavitation (and the resulting erosion); however, there is no chance to promote cleaning efficiency with this approach. In this review, we introduce our recent effort toward the development of an erosion-free ultrasonic cleaning technique using aerated water. Under dissolved gas supersaturation in aerated water, bubbles can be created easily with low-intensity ultrasound and the resulting bubble dynamics are expected to be mild enough to avoid erosive collapse. To demonstrate this conjecture, we run a series of ultrasound cleaning tests with glass samples on which submicron SiO₂ particles are spin-coated; we use a transparent cleaning bath for visualization of acoustic phenomena by a high-speed camera. Dissolved oxygen (DO) supersaturation in water (aerated by oxygen microbubbles) and ultrasound frequency (at 28 kHz or 200 kHz) are varied as parameters, while the input power to drive the ultrasound transducer is fixed and the ultrasound amplitude at the pressure antinode is set at \( P_a = 1.0 \text{ atm} \) (root-mean-square value) for the case of degassed water. Particle removal efficiency (PRE) is defined based on image analysis of light scattering from the residual particles (i.e., the so-called Haze method). We see that there exists an optimal DO supersaturation to maximize the PRE. We also see that the PRE is higher in the case of lower frequency (28 kHz), for its cavitation inception threshold is reduced and the number of activated bubbles is thus increased.

Keywords: Ultrasonic cleaning, Cavitation, Dissolved gas supersaturation, Particle removal efficiency

1. Introduction

Ultrasonic cleaning is one of physical cleaning techniques and widely employed in industry [1]. This procedure is often combined with toxic chemicals in cleaning solutions to enhance its efficiency [2]; however, physical cleaning that does not rely on any chemicals has practical significance from an environmental point of view.

The driving frequency of ultrasound is carefully chosen, depending on its application targets [3]. To degrease glass surfaces and metal components, low-frequency ultrasound (several tens or hundreds of kHz) is mainly used [4]. In this case, the cleaning effect is achieved primarily from dynamics of cavitation bubbles that appear in cleaning solution (i.e., water) under ultrasound irradiation. On the other hand, to remove submicron particles attached on delicate surfaces such as silicon wafers, higher-frequency ultrasound (of order MHz) is favored (i.e., megasonic cleaning) [5]. It was concluded in past studies that the cleaning effect arises from direct action of ultrasound propagation such as instantaneous fluid acceleration and period-averaged acoustic streaming in water (with no nucleated...
bubbles) [6]. However, recent studies [7, 8] suggested that cavitation bubbles can be nucleated under high-intensity megasonic waves and their dynamics play a dominant role in particle removal.

Since micron-sized (or even smaller) gas bubble nuclei are ubiquitous in water, cavitation bubbles are nucleated heterogeneously from the pre-existing nuclei [9, 10]. Cavitation bubbles oscillate with larger amplitude as their equilibrium size approaches the resonant radius $R_r$ (112.3 µm for 28 kHz and 14.6 µm for 200 kHz) [11, 12] through rectified mass diffusion [13]. The dynamics of acoustic cavitation bubbles are expected to create strong shear flow at surfaces to be cleaned [14–18]. Particle removal will be achieved when the shear-induced force defeats particle adhesion such as van der Waals force [19].

Although cavitation bubbles play an essential role in ultrasonic cleaning, their violent collapse may give rise to material damage, which is the so-called cavitation erosion [20–22]. Traditionally, degassed water is favored as cleaning liquid to minimize cavitation erosion by reducing the probability of having cavitation bubble nucleation in the liquid. However, we can no longer promote cleaning efficiency with this approach. Recently, we propose to use aerated water, instead, toward the development of an erosion-free ultrasonic cleaning technique [23]. Under dissolved gas supersaturation in aerated water, bubbles can be created even with low-intensity ultrasound [24, 25] and the resulting bubble dynamics are expected to be mild enough to avoid erosive collapse [26].

In this review, we introduce our recent work [27], toward the development of an erosion-free ultrasonic cleaning technique, where particle removal efficiency (PRE) is evaluated using glass samples spin-coated with SiO$_2$ particles (0.1 µm, 0.5 µm and 1 µm in diameter) under 28 kHz or 200 kHz ultrasound irradiation of a fixed input power. Tap water aerated with oxygen microbubbles is used as cleaning solution; the concentration of dissolved oxygen (DO) is varied as a key parameter. Based on the visualization of acoustic phenomena in the cleaning bath and on the cleaned samples, we examine the PRE dependence on DO supersaturation in the water.

![Schematic of the ultrasound cleaning test.](image)

**Fig. 1** Schematic of the ultrasound cleaning test.

| $f$ (kHz) | $H_f$ (mm) | $h_f$ (mm) |
|----------|-----------|-----------|
| 28 | 66 (≈ $5\lambda_f/4$) | 27 (≈ $\lambda_f/2$) |
| 200 | 68 (≈ $37\lambda_f/4$) | 37 (≈ $5\lambda_f$) |

and the driving frequency of the ultrasound.

### 2. Experimental method

#### 2.1 Generation of DO-supersaturated water

DO-supersaturated water is generated by a fine bubble generator of spiral flow type (Japan patent JP2011-088079). Initially the tank is filled with saturated tap water at room temperature (18-20 °C) and one atmosphere. Oxygen gas is sucked into the generator [28, 29] at a constant volume flow rate of 0.2 L/min and the water in the tank circulates through the generator at 12 L/min. Tap water is aerated with the generated oxygen microbubbles. The dimensionless DO supersaturation in the aerated water is defined by

$$\zeta_{DO} = (c_{DO} - c_s)/c_s.$$  

Here, $c_{DO}$ is the DO concentration in the aerated water, which is measured by a DO meter with a fluorometric sensor (SG9, METTLER TOLEDO) and $c_s$ is the saturated DO concentration under the atmosphere.
This system is operated for 30 minutes, and then a maximum value of DO supersaturation ($\zeta_{\text{DO}} \approx 6.0$) is obtained. Note that other gas species such as nitrogen are effectively purged by oxygen aeration and their concentration is expected to be much lower than the DO concentration in the aerated water. DO supersaturation is controlled by leaving it open to the atmosphere. These operation conditions are the same as in [30].

2.2 Ultrasonic cleaning test

The setup of our ultrasonic cleaning test is illustrated in Fig. 1. The (transparent) acrylic cleaning bath has (inner) lateral dimensions 90 mm × 90 mm and 100 mm × 100 mm, respectively, for ultrasonic transducers driven at $f = 28$ kHz (0.028Z45I, JAPAN PROBE) and at $f = 200$ kHz (B0.2K30I, JAPAN PROBE). The cylindrical-shaped piezo-electric transducer whose ultrasound-emitting surface is planer is inserted into a hole created at the bath bottom. The diameters of the transducers driven at $f = 28$ kHz and $f = 200$ kHz are 65 mm and 38 mm, respectively. The DO-supersaturated water is filled in the bath and the water level is set at $y = H_f$ (where $y$ is measured upward from the transducer's surface) to form a standing-wave-like pressure field that results from superposition of reflected waves inside the bath. Circular glass plates (30 mm in diameter and 1 mm in thickness) on which SiO$_2$ particles (0.1 µm, 0.5 µm, and 1 µm in diameter) are spin-coated on one side are used as cleaning samples. Their spin-coated surface is aligned with the central axis of the transducer. The sample's center is set at $y = h_f$. The values of $H_f$ and $h_f$ are summarized in Table 1.

The transducer is driven by a multi-function generator (WF1973, NF) through a power amplifier (HSA4014, NF). The effective irradiation pressure, $P_e$, is measured at $y = h_f$ by a needle hydrophone (HCT0310, ONDA). In this measurement, degassed water (in which the cleaning sample is not inserted) is used, allowing us to minimize pressure contamination due to acoustic cavitation. The input power supplied to the transducer is tuned to obtain $P_e = 1.0$ atm (corresponding to $\sqrt{2}$ atm in the pressure amplitude of the ultrasound); the same input power used for all the cleaning tests. The duration of the ultrasound irradiation to each cleaning sample is fixed at 30 s.

The acoustic phenomena in the cleaning bath are visualized by a video camera (EX-100, CASIO) with a LED side lighting (SLG-150V, REVOX). For the purpose of better visualization, we do not insert the cleaning sample in the bath.

2.3 Evaluation of the cleaning efficiency

In order to evaluate particles concentration on the glass surface, we use the Haze method [31] in which illumination at 45° from a metal halide lamp (LS-LHA, SIGMAKOKI) is shed on the glass surface and the light scattering due to the residual particles is captured by a digital camera (ILCE-6000, SONY) with a macro lens (SEL30M35, SONY). The average intensity of the light scattering is calculated by image analysis using ImageJ (U.S. National Institutes of
Health, Bethesda, Maryland, USA). Under the assumption that there exists a proportionality between the light scattering intensity and the residual particle density, we define PRE by

$$\text{PRE} = (\sigma_0 - \sigma)/\sigma_0 \times 100 \%$$ (2)

where $\sigma_0$ and $\sigma$ denote the intensity of the light scattering, respectively, before and after the cleaning test.

3. Results and discussion

3.1 Cavitation under DO supersaturation

First, we examine acoustic phenomena (without the cleaning sample) by varying the DO supersaturation. In Fig. 2, we show the visualization of acoustic phenomena in the bath ($f = 28$ kHz) under different DO supersaturation. Even under the low-intensity ultrasound irradiation, visible-sized acoustic bubbles appear more densely under higher DO supersaturation, indicating that the population of active bubble nuclei increases as DO supersaturation increases [32, 33]. For the lower DO-supersaturation cases ($\zeta_{DO} = 0.1$ and 1.0), bubbles are entrapped via the primary Bjerknes force [34] that arises from a standing-wave pressure field. On the other hand, for the higher DO-supersaturation cases ($\zeta_{DO} = 3.0$ and 4.0), acoustic bubbles migrate mainly in the direction of the incident ultrasound propagation toward the water surface. This means that the standing-wave structure in the pressure field is impaired by dissipative effects in the dynamics of the densely populated bubbles [35].

3.2 Cavitation under different frequency

Next, we examine acoustic phenomena by varying the driving frequency of the ultrasound. In Fig. 3, we show the visualization of acoustic phenomena in the bath filled with the DO-supersaturated water, for example, at $\zeta_{DO} = 3.0$. Under 28kHz ultrasound irradiation (Fig. 3(a)), according to the primary Bjerknes force from a standing-wave pressure field, the oscillating bubbles translate along the pressure gradient in the bath. Although, the bubbles' size is not fully resolved in the image, we see that relatively smaller bubbles (namely, sub-resonant-sized bubbles where $R < R_r = 112.3 \mu m$) appear in the region of pressure antinode and super-resonant-sized bubbles ($R > R_r = 112.3 \mu m$) are trapped at the pressure node. The intervals between the antinode and the node are confirmed to be quarter wavelength of the ultrasound ($\lambda_f/4 = 13 \text{ mm}$).

Under 200kHz ultrasound irradiation (Fig. 3(b)), two structures are clearly observed [36, 37]: super-resonant-sized bubbles ($R > R_r = 14.6 \mu m$)
visual in the image are trapped as horizontal streaks whose separation is equal to one half of the ultrasound wavelength ($\lambda / 2 = 3.7$ mm). The entrapment of these bubbles suggests the formation of a standing-wave structure in the vertical direction [38, 39]. And, resonant-sized bubbles rapidly translating upward from the transducer's surface, forming the vertical streaks in the image. Such trapped and translating bubbles are caused, respectively, by the primary Bjerknes force from a standing-wave component in the pressure field and the acoustic radiation force [40, 41] from the traveling wave component. The latter is observed mainly in the central region above the transducer's surface: densely nucleated bubbles can attenuate the acoustic energy from the transducer and the traveling wave component will thus be dominant in this region [42].

3.3 Cleaning test and PRE evaluation

Finally, we examine the PRE dependence on the DO supersaturation and the ultrasound frequency from the cleaning tests (with the fixed power input to the ultrasound transducer). Fig. 4 shows typical Haze images of the cleaning sample before and after the sonication. The dark area corresponds to the cleaned area.

It is interesting to point out that the cleaning effect is higher in the lower frequency case, which may be counterintuitive from the traditional understanding of ultrasonic cleaning; in the case of single-phase underwater ultrasound where cavitation does not occur, the mechanical action including instantaneous fluid acceleration and streaming are emphasized by steeper pressure gradients, thereby leading to more efficient cleaning under higher-frequency ultrasound irradiation. However, the trend is opposite in the present cleaning tests. This is because the particle removal is achieved mainly by the action of cavitation bubbles. As the ultrasound frequency increases, the shorter period of ultrasound suppresses the nucleation [43]. Thus, under a fixed power supplied to the ultrasound transducer, the probability of having cavitation and its cleaning force are thus larger in the lower-frequency case.

For the case of 200kHz ultrasound irradiation, the cleaned area (Fig. 4(b)) has horizontal stripes that are expected to result from the dynamics of bubbles trapped at either nodes or antinodes in the standing-wave-like pressure field (Fig. 3(b)).

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Fig. 5 SEM image of the cleaned area having stripes in the vertical direction along the y axis in Fig. 1. The ultrasound frequency is $f = 200$ kHz.

Fig. 6 Particle removal efficiency (PRE) as a function of DO supersaturation $\zeta_{\text{DO}}$. 
interval between these stripes corresponds to one half of the ultrasound wavelength ($\lambda_f/2 = 3.7$ mm). Furthermore, using a scanning electron microscope (VE-8800, KEYENCE), we visualize the cleaned sample more carefully (Fig. 5). The stripes in the vertical direction have width from 40 µm to 70 µm, which is a bit larger than the diameter of the resonant bubbles ($2R_f = 29.2$ µm). This indicates that wall shear flow induced by (large-amplitude) oscillations of the resonant bubbles can appear in a broader area at the cleaning surface (in comparison to the equilibrium bubble size) [44].

In Fig. 6, we summarize PRE as a function of DO supersaturation from a series of the ultrasound cleaning tests (at $f = 28$ kHz or $f = 200$ kHz) with varying the particle size and DO supersaturation. Regarding the particle size, we confirm that it is harder to remove smaller particles from the surface. We note that when it comes to modeling flow around the smallest particles (0.1 µm in diameter), continuum assumptions may fail. When steep velocity gradients exist above solid surfaces, flow can slip, giving rise to a reduction in wall shear stress [45].

Finally, we discuss the effect of DO supersaturation on the PRE. Under the DO saturation ($\zeta_{DO} = 0$), the value of PRE is very low, for cavitation bubbles appear only sporadically and their cleaning effect is limited. On the other hand, as the DO supersaturation increases, the cavitation inception pressure is reduced so that cavitation bubbles appear more densely. As a result, regardless of the value of the ultrasound frequency, the PRE is augmented by dynamics of these bubbles (see Fig. 2). As pointed out in Fig. 4, the PRE is higher in the lower-frequency case (Fig. 6(a)) in which the cavitation occurs more easily because of the lower cavitation inception pressure [46, 47]. However, as the DO supersaturation increases further, the PRE starts to decrease. We speculate that the population of cavitation bubbles becomes so dense that much of the acoustic energy of the ultrasound emitted from the transducer is damped by bubble dynamics and is not transferred to the cleaning spot [48]; in other words, the acoustic intensity is expected to decrease significantly from that in the case without cavitation bubbles. This suggests that we need to explore the optimal dissolved gas supersaturation to maximize the cleaning efficiency.

### 4. Summary and perspective

In this review, we report on our recent effort toward the development of an erosion-free cleaning with low-intensity ultrasound. Ultrasonic cleaning tests have been conducted with parameters of DO supersaturation and driving frequency of ultrasound. The DO-supersaturated water was generated by aeration with oxygen microbubbles. The PRE was evaluated by using the samples that were glass plates spin-coated with submicron-sized SiO$_2$ particles. From the optical observation, the particle removal was found to be achieved mainly by the action of cavitation bubbles; the PRE is reduced for the cleaning test for the case of the higher ultrasound frequency at which it is harder to obtain bubble nucleation. There exists the optimal supersaturation to maximize the cleaning efficiency.

In our future work [49], we plan to study the trade-off relation between cleaning efficiency and cavitation erosion with varying dissolved-gas supersaturation in ultrasonic cleaning solution. To more clearly see liquid flow and bubbles’ translation in the cleaning bath, we also plan to perform a particle image velocimetry (PIV) analysis.

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