CAVITATION INTENSITY IN GLASS BEAD-WATER MIXTURES BASED ON IODOMETRIC TITRATION

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ABSTRACT

Since the cavitation intensity in solid-liquid mixtures is difficult to predict, it is of necessity to investigate the influence of suspended solids on sonochemical reactions. Tests were carried out using an ultrasonic apparatus and glass beads were fully suspended in water, considering three volume fractions (0.05, 0.075, and 0.10 v/v) and two particle sizes (0.28 and 0.45 mm). Based on iodometry, the I_3^- concentration was measured, which is used to investigate the production of free radicals (·OH) whose quantities indicate cavitation intensity. Results reveal that, with the addition of solid particles, the I_3^- concentrations are lower than the solids-free case. When volume fraction increases or particle diameter decreases, the I_3^- concentration decreases accordingly, which indicates lower intensity of cavitation. The attenuation of ultrasonic waves propagating in solids suspensions is theoretically investigated, and may act as a reasonable explanation for the reduction in sonochemical yields.

Keywords: Cavitation intensity, sonochemical yields, particle size, particle concentration, acoustic attenuation

1. INTRODUCTION

The factors influencing cavitation damage in hydraulic structures have been studied from varied aspects such as the mechanical properties of materials, flow characteristics and liquid properties (Wu, 2004). However, due to the presence of air bubbles, sand particles and micro-organisms in water, growing concern has been expressed over the damage caused by the synergistic effect of different additives, and much research has been conducted on this topic in recent years (Xia et al., 2019; Wang et al., 2018; Gou et al., 2018). For example, sand particles modify liquid properties and thus cavitation intensity (used to character the activity and strength of cavitation), on which cavitation erosion is originally dependent. Apart from the mechanical method involving the erosion, several other methods such as chemistry method (Tuziuti et al., 2004), acoustic method (Miyoshi et al., 2005) and optical method has been developed to measure cavitation intensity. Therefore, it is necessary and feasible to research the effects of solid particles on cavitation intensity by combining these methods, which in turn may shed some light on their impact on cavitation erosion.

Table 1 provides a brief summary of what previous investigators have reported on the effect of solids addition on cavitation intensity based on chemical methods, and all of them noticed a reduction in cavitation intensity as particles were added. However, most of them simply attributed the reduction to the attenuation of acoustic waves (scattering and absorption) without further explanation. There is a large body of literature on the propagation of acoustic waves in solids suspensions (Gibson and Toksöz, 1989; Hay and Mercer, 1982; Allegra and Hawley, 1972; Urick, 1948), from which it is known that the attenuation increases with frequency (Harker and Temple, 1988), but the dependence of acoustic attenuation on the properties of suspended solids (i.e., particle size d_p and particle concentration φ_p) is less known. Hampton (1967) experimentally found that the attenuation of high-frequency sound in particle suspensions was non-monotonic with respect to φ_p, leading to an apparent maximum in attenuation at φ_p near 0.2 v/v.

In this paper, to find out the acoustic attenuation caused by oscillating rigid spheres in viscous fluids, equations fluid mechanics are solved with the general solution generalized to accommodate the oscillatory fluid motion and include the description of the acoustic behaviour of suspensions. The prediction of sound attenuation varying with d_p and φ_p is compared to the experimental results based on the iodometric titration method, and is used to explain the effect of solids addtion on cavitation intensity.

Table 1. Literature reporting the influence of solids addition on sonochemical reactions

Sonochemical system	Particle type (d_p, φ_p)	Remarks	Reference
Degradation of Bromo-benzene (C_6H_5Br)	Organic resin (35 μm, 0.05-5 g/L)	Degradation rate of C_6H_5Br decreased with increasing φ_p	Zhang and Hua (2002)
Dosimeter measuring ·OH radical production	Silica (2-130 μm, 2-200 g/L)	Production of radicals decreased with increasing φ_p	Lu et al. (2002)
Hydrogen peroxide (H_2O_2) formation in water	Quartz (2-16 μm, 1-25 g/L)	Rates of H_2O_2 formation decreased with increasing φ_p	Keck et al. (2002)
Degradation of phenol (C_6H_5OH)	TiO_2 (powder, 100-500 ppm)	Degradation rate of C_6H_5OH decreased with increasing φ_p	Gogate et al. (2004)
Oxidation of I^- ions in aqueous KI solutions	Alumina (1-80 μm, 10-100 g/L)	Concentration of I_3^- decreased with increasing φ_p	Tuziuti et al. (2005)
Hydrogen peroxide (H_2O_2) formation in water	Glass beads (50-5000 μm, 10-200 g/L)	Rates of H_2O_2 formation decreased with increasing φ_p	Her et al. (2011)
Degradation of phenol (C_6H_5OH)	ZnO (powder, 1-4 g/L)	Degradation first decreased and then increased with increasing φ_p	Doltade and Gole (2017)

2. METHODS AND MATERIALS

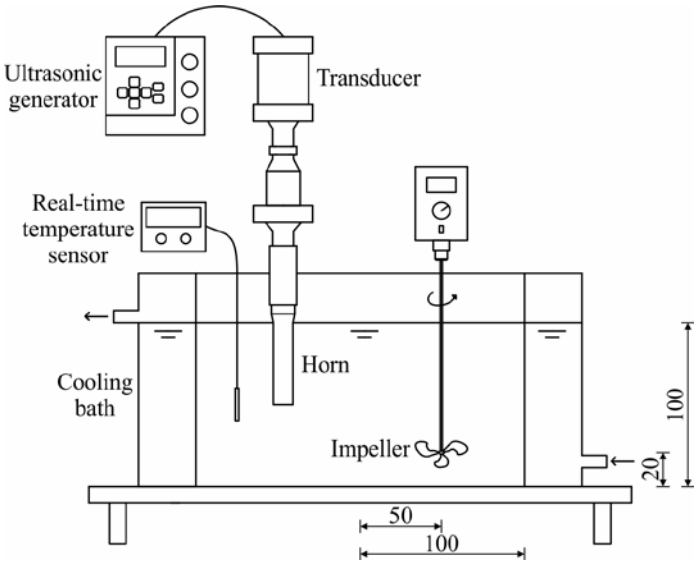

Figure 1. Schematic diagram of the experimental set-up (mm)

As is shown in Fig. 1, the tests were conducted in a cylindrical PVC vessel containing particle suspensions. A transducer was powered by an ultrasonic generator with a vibration frequency of 20±0.5 kHz. The energy was introduced into the suspension by the horn at the end of the transducer, and the input power was set as 300 W. The suspension was agitated to keep the particles fully suspended using a four-bladed impeller, with which the horn was placed symmetrically with regard to the vessel's axis. All tests were conducted beyond the minimum impeller speed at which no particles remain stationary at the bottom of the vessel for more than 1 s, according to the empirical correlation by Zwietering (1958). The suspension temperature was maintained at 25±1 °C by virtue of a cooling bath, and was monitored using a real-time temperature sensor.

The potassium iodide (KI) solution (0.1 mol/L) was prepared just before each test to avoid oxidation, and kept with a pH of 8±0.2 for chemical stability. Submillimeter-sized glass beads were used as solid particles with a sphericity of 85% and densities of 2.55×10^3 kg/m^3. Fig. 2 illustrates the particle-size distribution (PSD) of the glass beads, in which a narrow diameter distribution of these glass beads is shown so that the particle size can be represented by the median diameter (d_{50} = 0.28 and 0.45 mm). The glass beads were washed before the test with deionised water at least three times until the wastewater became no longer turbid, and was then dried at 200 °C within the drying oven for no less than 5 h. The effect of φ_p and d_p on cavitation intensity was investigated with φ_p = 0.05, 0.075, and 0.1 v/v for both particle sizes, and one test without suspending glass beads was also conducted as a control case.

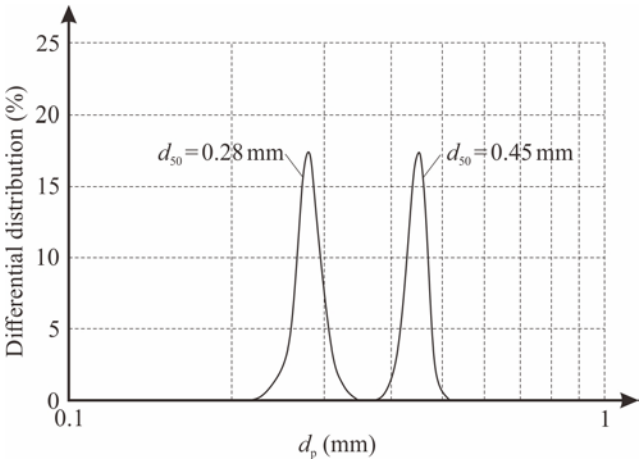

Figure 2. The particle-size distribution (PSD) of glass beads used in this investigation

Prior to the test, according to the Lambert-Beer law, the standard absorbance curve of the triiodide anion (I_3^-) was obtained to build the relationship between the concentration and the absorbance of I_3^-, and the amount of I_3^- formed is an indication of the extent of cavitation intensity. During the test, 6-7 mL sample was taken from supernatant and a centrifugal separator was used to separate the solid particles from the solution. The remaining solution was moved into the glassware, and the absorbance of I_3^- was obtained by a UV-vis spectrophotometer at 350 nm, which was then converted to the concentration of I_3^- based on the standard absorbance curve. This step was repeated for three times to avoid the influence of residual solid particles. Also, the absorbance under the conditions of no particles and no ultrasonic irradiation was tested, and it should be smaller than 0.005 Abs in case of oxidation in air. Each test lasted for 1 h and was repeated three times.

3. RESULTS AND DISCUSSION

3.1 Influence of suspended solids on I_3^- formation

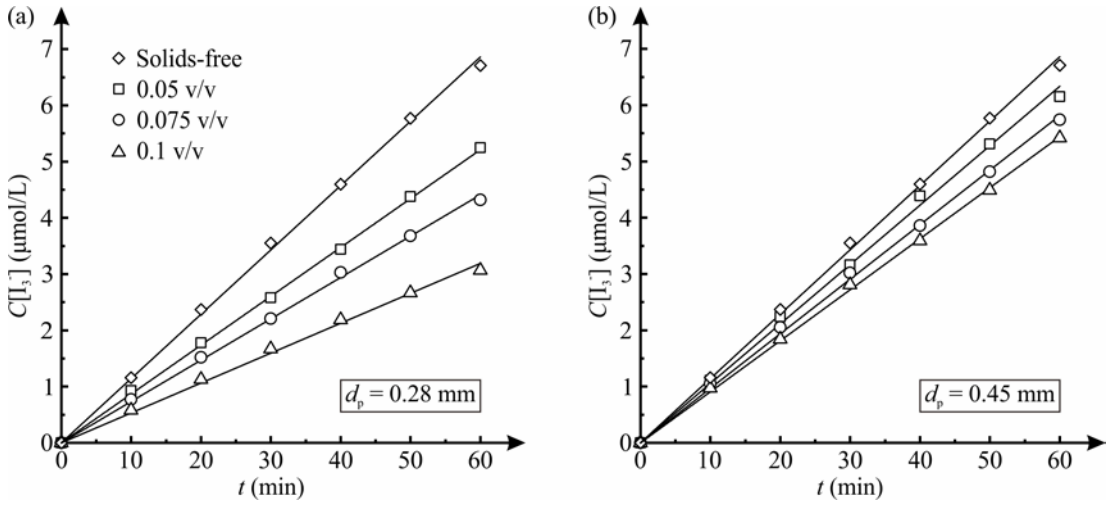

Figure 3. Evolution of $C[I_3^-]$ during sonication for two sizes: (a) d_{50} = 0.28 mm, and (b) d_{50} = 0.45 mm at different φ_p

Fig. 3 reports the aqueous concentration of the oxidation product I_3^- ($C[I_3^-]$) as a function of time (t) with φ_p as a parameter for both particle sizes. $C[I_3^-]$ increases with sonication time for all φ_p, similar to the trends given by other researchers (Koda et al., 2003). It is apparent that the addition of glass beads decreases $C[I_3^-]$ at all φ_p for the two sizes, which indicates that the sonochemical yields are reduced and cavitation intensity is getting lower under the influence of suspended particles. The similarity between the two sizes is that higher φ_p contributes to less I_3^- being formed, while the difference between Fig. 3a and Fig. 3b lies in that, at the same φ_p, bigger size leads to higher sonochemical yields. Take, for example, $C[I_3^-]$ at $\varphi_p = 0.05$ v/v for $d_p = 0.28$ mm and $C[I_3^-]$ at $\varphi_p = 0.1$ v/v for $d_p = 0.45$ mm, although the solid concentration in the latter case is the double of the former, there is not much difference in the I_3^- concentration value between the two cases. Now that the introduction of solids into the liquid phase may provide additional nucleation sites for the formation of cavities and lower the cavitation threshold, the attenuation effects of solid particles seem to surpass the influence of nuclei addition under the test conditions in this investigation. As indicated by $C[I_3^-]$, the attenuation of acoustic waves is very much likely to be dependent on the particle size and concentration; as a result, it is meaningful to find the relationship of acoustic attenuation with φ_p and d_p theoretically.

3.2 Attenuation of ultrasonic waves in solids suspension

Let us assume that, as the ultrasonic wave propagates through the solid particle suspension, six state variables are perturbed by tiny amounts (superscript ') from steady state values (superscript 0): oscillation velocities of the fluid and the solid particle (v_f and v_p), densities of the fluid and the solid particle (ρ_f and ρ_p), the liquid static pressure (p), and the particle concentration (φ_p). The time-averaged velocity of the fluid and the particle as well as the pressure can be taken to be zero. Based on perturbation theory, linear equations (Eqs. (1) and (2): liquid and solid phase continuity equations; Eqs. (3) and (4): momentum conservation equations of the liquid and solids; Eqs. (5) and (6): equations of state with respect to density variation due to pressure perturbation) are as follows:

$$(1-\varphi_p^0)\frac{\partial \rho_f^{'}}{\partial t}+\rho_f^0\frac{\partial(1-\varphi_p^{'})}{\partial t}+(1-\varphi_p^0)\rho_f^0\frac{\partial v_f^{'}}{\partial x}=0 \tag{1}$$

$$\varphi_p^0\frac{\partial \rho_p^{'}}{\partial t}+\rho_p^0\frac{\partial \varphi_p^{'}}{\partial t}+\varphi_p^0\rho_p^0\frac{\partial v_p^{'}}{\partial x}=0 \tag{2}$$

$$(1-\varphi_p^0)\rho_f^0\frac{\partial v_f^{'}}{\partial t}=-\frac{\partial p^{'}}{\partial x}+(\frac{\varphi_p^0}{\frac{4}{3}\pi R_p^3}f_d+\varphi_p^0\frac{\partial p^{'}}{\partial x}) \tag{3}$$

$$\varphi_p^0\rho_p^0\frac{\partial v_p^{'}}{\partial t}=-(\frac{\varphi_p^0}{\frac{4}{3}\pi R_p^3}f_d+\varphi_p^0\frac{\partial p^{'}}{\partial x}) \tag{4}$$

$$\rho_f^{'}=\rho_f^0\frac{p^{'}}{\kappa_f} \tag{5}$$

$$\rho_p^{'}=\rho_p^0\frac{p^{'}}{\kappa_p} \tag{6}$$

in which R_p is the radius of the solid particles; κ_f and κ_p are the bulk modulus of the fluid and the solid particle, respectively; f_d represents the unsteady drag force on merely one single sphere, and was derived by Atkinson and Kytömaa (1991) as:

$$f_d=6\pi\mu R_p(1+R_p\sqrt{\frac{\rho_f^0\omega}{2\mu}})(v_p^{'}-v_f^{'})+\frac{4}{3}\pi R_p^3\rho_f^0(\zeta+\frac{9}{4R_p}\sqrt{\frac{2\mu}{\rho_f^0\omega}})(\frac{\partial v_p^{'}}{\partial t}-\frac{\partial v_f^{'}}{\partial t}) \tag{7}$$

in which μ is the fluid viscosity; ω is the angular frequency; ζ is the added mass coefficient, which depends on the solids concentration, and according to Wallis (1989) and Maxwell (1881), ζ takes the form $(1-\varphi_p)/2$.

Generally speaking, these governing equations (Eqs. (1)-(6)) have wave-like solutions:

$$\left[v_f^{'},v_p^{'},p^{'},\varphi_p^{'},\rho_f^{'},\rho_p^{'}\right]^T=\left[v_{f0},v_{p0},p_0,\varphi_{p0},\rho_{f0},\rho_{p0}\right]^T\exp\left[(\omega t+kx)i\right] \tag{8}$$

in which $k=\omega/c+\alpha i$ is the complex-valued wave number, whose imaginary part (coefficient α) is attenuation per unit distance, also useful in the study of exponentially-decaying evanescent fields; c is the wave speed.

Substituting these perturbed variables into Eqs. (1)-(6), and neglecting these higher-than-first-order terms, one can obtain the following matrix equation:

$$\begin{bmatrix}
k\rho_f^0(1-\varphi_p^0) & 0 & 0 & -\omega\rho_f^0 & \omega(1-\varphi_p^0) & 0\\
0 & k\rho_p^0\varphi_p^0 & 0 & \omega\rho_p^0 & 0 & \omega\varphi_p^0\\
-(A-1)+Bi & \left[(A-1)+\frac{\rho_p^0}{\rho^*(1-\varphi_p^0)}\right]-Bi & \frac{k}{\rho^*\omega(1-\varphi_p^0)} & 0 & 0 & 0\\
\left[(A-1)+\frac{\rho_f^0}{\rho^*\varphi_p^0}\right]-Bi & -(A-1)+Bi & \frac{k}{\rho^*\omega\varphi_p^0} & 0 & 0 & 0\\
0 & 0 & -\frac{\rho_f^0}{\kappa_f} & 0 & 1 & 0\\
0 & 0 & -\frac{\rho_p^0}{\kappa_p} & 0 & 0 & 1
\end{bmatrix}\begin{bmatrix}v_{f0}\\v_{p0}\\p_0\\\varphi_{p0}\\\rho_{f0}\\\rho_{p0}\end{bmatrix}=0 \tag{9}$$

in which

$$A = \frac{9\eta\rho_f}{4(1-\varphi_p)\rho^*}\left(1+\frac{2}{9}\frac{1-\varphi_p}{\eta}\right)+1 \tag{10}$$

$$B = \frac{9\eta\rho_f}{4(1-\varphi_p)\rho^*}(1+\eta) \tag{11}$$

$$\eta = \frac{1}{R_p}\sqrt{\frac{2\mu}{\rho_f\omega}} \tag{12}$$

$$\bar{\rho} = (1-\varphi_p)\rho_f + \varphi_p\rho_p \tag{13}$$

$$\rho^* = (1-\varphi_p)\rho_p + \varphi_p\rho_f\,. \tag{14}$$

The column vector has a non-trivial solution when (and only when) the determinant of a matrix equals nil, so:

$$k^2 = \frac{\bar{\rho}\omega^2}{\bar{\kappa}}\frac{\left(\frac{\rho_p\rho_f}{\bar{\rho}\rho^*}-1\right)+A-Bi}{A-Bi} \tag{15}$$

in which

$$\frac{1}{\bar{\kappa}} = \frac{\varphi_p}{\kappa_p}+\frac{1-\varphi_p}{\kappa_f}\,. \tag{16}$$

Denote the real part and the imaginary part of the complex value k by Re(k) and Im(k), respectively. Since the real part of k is typically larger than the imaginary part by two or more orders of magnitude, then

$$\alpha = \mathrm{Im}(k) \approx \frac{\mathrm{Im}(k^2)}{2\sqrt{\mathrm{Re}(k^2)}} = -\frac{\omega}{2}\sqrt{\frac{\bar{\rho}}{\bar{\kappa}}}\frac{B}{A\sqrt{(C+1)C}} \tag{17}$$

in which

$$C = \frac{A^2+B^2}{\left(\frac{\rho_p\rho_f}{\bar{\rho}\rho^*}-1\right)A}\,. \tag{18}$$

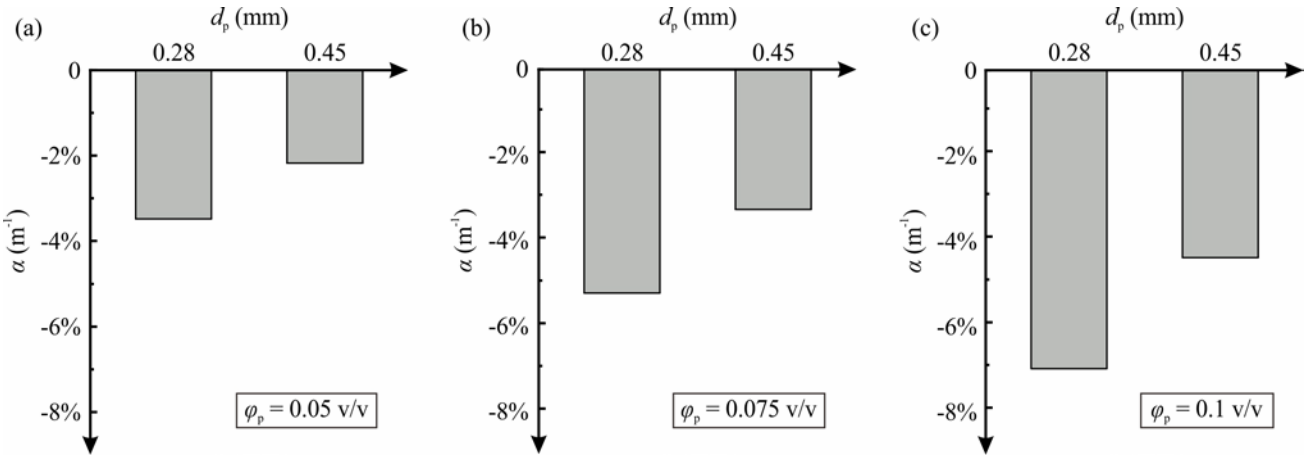

Figure 4. Variation of calculated α with particle concentration φ_p for both sizes

Based on Eq. (17), we can solve out the attenuation coefficient α at φ_p = 0.05, 0.075, and 0.1 v/v for both sizes, as is shown in Fig. 4, from which it is evident that the absolute value of α increases with increasing φ_p, and the smaller the glass beads, the bigger the attenuation effect. This demonstrates that the acoustic attenuation caused by solids addition is closely related to the amount and size of these rigid additives, and that the effect of φ_p and d_p on sonochemical yields can be explained using this hypothesis. The presence of solid particles attenuates the energy of ultrasonic waves, thereby dampening the acoustic intensity propagating through the solid suspension and suppressing the formation of vapour cavities. Higher solids concentration and smaller particle size give rise to greater acoustic attenuation and thus lower intensity of cavitational activity.

4. CONCLUSIONS

The influence of particle concentration and size on sonochemical yields was experimentally investigated using the iodometric titration method, and results indicate that the formation rate of the oxidation product increases with decreasing particle concentration and increasing particle size. The finding can be reasonably corroborated by a theoretically-obtained acoustic attenuation coefficient resulting from suspending solid particles.

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