

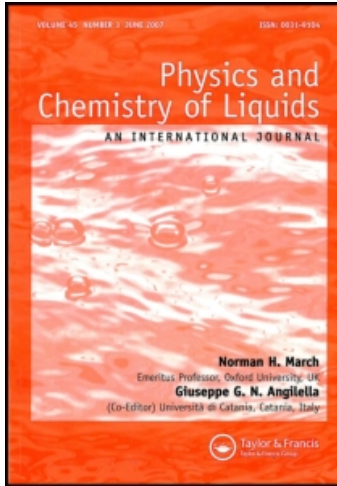
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Physics and Chemistry of Liquids

Publication details, including instructions for authors and subscription information:

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Online publication date: 10 December 2010

To cite this Article Peng, Jianxin and Lu, Yigang(2010) 'Characteristic analysis of cavitation bubbles in carbon dioxide fluid', *Physics and Chemistry of Liquids*, 48: 6, 736 – 746

To link to this Article: DOI: 10.1080/00319100902946437

URL: <http://dx.doi.org/10.1080/00319100902946437>

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Characteristic analysis of cavitation bubbles in carbon dioxide fluid

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(Received 11 February 2008; final version received 3 April 2009)

After analysing the characteristics of bubble cavitation in high-pressure carbon dioxide (CO₂) fluid, cavitation conditions and some correlative physical characteristics are investigated. The results show that the ultrasonic intensity of liquid carbon dioxide to make cavitation occur is affected by the initial radius of the bubbles, hydrostatic pressure, temperature and vapour pressure within the bubbles in liquid CO₂. At the low frequency of ultrasound, the phase-speed of the liquid CO₂ gradually approaches the sound speed of the pure liquid when void fraction increases. At high frequency, the phase-speed is nearly equal to the sound speed in the liquid under different void fractions. The attenuation of ultrasound in liquid carbon dioxide reaches a maximum near the resonance frequency and then decreases when frequency either increases or decreases. At the resonance frequency, the phase-speed and the attenuation increase when the void fraction increases.

Keywords: carbon dioxide; ultrasonic cavitation; cavitation bubble; initial radius; phase speed; attenuation

1. Introduction

Ultrasonic cavitation is a very complex physical phenomenon, which includes the oscillation, growth, collapse and closing of micro-bubbles in the liquid. Not only acousticians and physicists but also chemists are increasingly interested in cavitation [1]. The ultrasound can induce micro-bubbles in the liquid to collapse, and greatly influence chemical reactions. When the ultrasonic intensity is high enough, cavitation may occur in some liquids. The instantaneous cavitation and high temperature in bubbles may cause free radicals to be formed and sonoluminescence to be generated. Shock waves (homogeneous) or the rapid stream (nonhomogeneous) caused by ultrasonic cavitation occur at high pressure. This special energy release modality is able to accelerate chemical reactions or create new reaction channels, so ultrasonic cavitation has been broadly used in chemistry and the chemical industry [2].

As liquid carbon dioxide (CO₂) is cheap, nontoxic, apyrous and non-contaminated, it is used as one kind of nontoxic green solvent in many applications such as chemical reactions, separation and man-made materials. As the energy crisis

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approaches, more and more researchers are investigating the properties of CO_2 , which can not only be used as a reactant, but also as a solvent. For example, dense-phase liquid CO_2 is used to provide an effective means for green chemical industry exploitation. For normal solvents, cavitation does not occur at high pressure, so that research on ultrasonic cavitation is usually focused on normal pressure ranges. In order to let the dense-phase liquid have enough solvency, the dense-phase liquid is normally used at high pressure and high density. Recently, ultrasound cavitation-induced radical formation was used for the polymerisation of methyl methacrylate in CO_2 at high pressure to yield high-molecular-weight polymers. The result shows that sonochemical reactions can be performed in dense-phase fluids, which allows environmentally benign CO_2 to replace conventional organic solvents in many reaction systems [3]. In this article, the characteristics of bubble cavitation in high-pressure CO_2 fluid are analysed, and the conditions for cavitation and some correlative physical characteristics are investigated.

2. A single bubble's cavitation

Consider the bubble of radius R in the liquid CO_2 (see Figure 1 [4]). P_i , P_{out} are the pressures within and outside the bubble, respectively. Then [4],

$$P_i = P_g + P_V \quad (1)$$

$$P_{\text{out}} = P_B + P_\sigma, \quad (2)$$

where P_g , P_V and P_B are the pressures of gas, liquid vapour in the bubble and the liquid pressure at the bubble wall, respectively. P_σ is the surface tension pressure of the bubble, $P_\sigma = 2\sigma/R_0$, σ is the surface tension. If the bubble is initially in a static equilibrium (radius $R = R_0$), the pressure in the liquid outside the bubble P_B equals the hydrostatic pressure P_0 , then the internal pressure of a bubble within the liquid is $P_{i,e} = P_0 + (2\sigma/R_0)$ [4]. If the vapour pressure is P_V , then the pressure of the gas phase within the bubble is [4]

$$P_{g,e} = P_0 + \frac{2\sigma}{R_0} - P_V, \quad (3)$$

where R_0 is the initial radius of the bubble at static equilibrium. When the outside pressure changes from P_0 to P_B , the bubble radius will change from R_0 to R to maintain equilibrium. Assuming the gas inside the bubble behaves as an ideal gas, then the gas pressure within the bubble is [4]

$$P_g = \left(P_0 + \frac{2\sigma}{R_0} - P_V \right) \left(\frac{R_0}{R} \right)^{3k}, \quad (4)$$

where k is the polytropic exponent, ranging from specific heat ratio γ (adiabatic) to 1 (isothermal). From Equations (1), (2) and (4), the pressure in the liquid near the boundary of the bubble wall is [4],

$$P_B = \left(P_0 + \frac{2\sigma}{R_0} - P_V \right) \left(\frac{R_0}{R} \right)^{3k} + P_V - \frac{2\sigma}{R}. \quad (5)$$

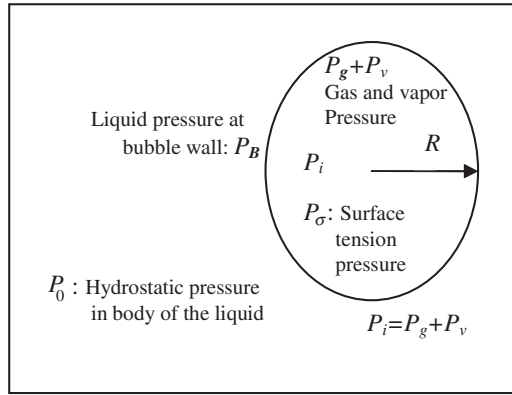


Figure 1. Static pressures of the bubble [4].

Table 1. Physical parameters of water and CO₂ [5].

| Options | Water | CO ₂ |
|--------------------------------------|-----------------------|-----------------------|
| Hydrostatic pressure (Pa) | 1×10^5 | 7.5×10^6 |
| Temperature (K) | 283 | 283 |
| Density (kg m ⁻³) | 999.7 | 899.2 |
| Surface tension (N m ⁻¹) | 0.0742 | 0.00278 |
| Vapor pressure (Pa) | 1216 | 4.486×10^6 |
| Viscosity (Pa s) | 1.31×10^{-3} | 9.15×10^{-5} |
| Sound speed (m s ⁻¹) | 1446.5 | 521.0 |

There is a critical radius, R_{crit} , which a bubble must attain to maintain a stable shape. It can be obtained by differentiating with respect to R and setting the partial differential coefficient of Equation (5) equal to 0. If $R > R_{\text{crit}}$ the bubble becomes unstable. The critical radius is [4]

$$R_{\text{crit}} = \left[\frac{3k}{2\sigma} \left(P_0 + \frac{2\sigma}{R_0} - P_V \right) R_0^{3k} \right]^{1/3k-1}. \quad (6)$$

Correspondingly, the critical liquid pressure $P_{B,\text{crit}}$ is [4]

$$P_{B,\text{crit}} = P_V - \left(1 - \frac{1}{3k} \right) \frac{2\sigma}{R_{\text{crit}}}. \quad (7)$$

Thus, the Blake threshold is [4]

$$P_{\text{Blake}} = P_0 - P_{B,\text{crit}} = P_0 - P_V + \left(1 - \frac{1}{3k} \right) \frac{2\sigma}{R_{\text{crit}}}. \quad (8)$$

In order to further analyse the cavitation characteristics of bubbles in CO₂ fluid, the results are compared with that in water. Some physical parameters are listed in Table 1 [5].

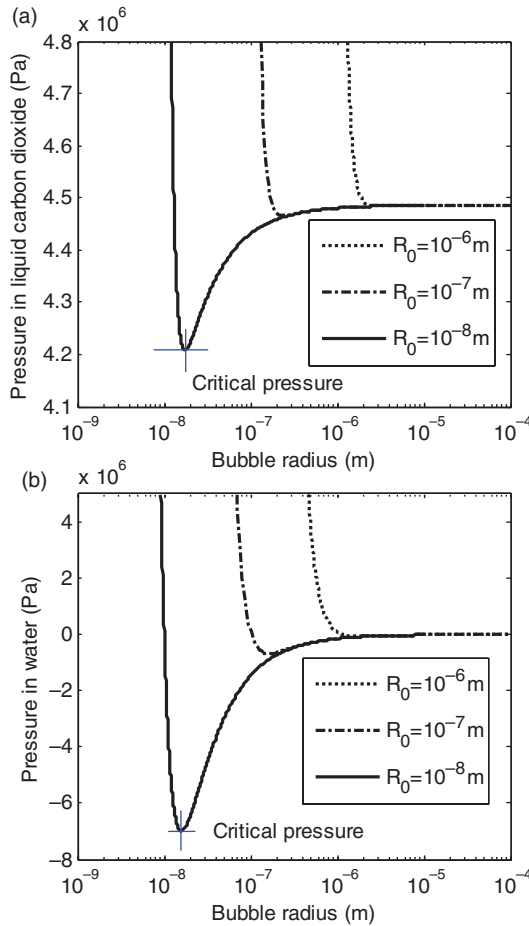


Figure 2. Relationship between the pressure at the bubble wall and initial bubble radius. (a) CO_2 and (b) H_2O .

The relationship between the pressure at the bubble wall and the initial radius of the bubble, which is calculated by Equation (5), is shown in Figure 2. It is assumed that there is no heat exchange between bubbles and liquid, so $k = \gamma$, and that there is only CO_2 and water vapour in the CO_2 bubble and water bubble, respectively. In both water and CO_2 liquid, the volume of the bubble expands and the radius of the bubble increases if the pressure is reduced in the liquid. It becomes unstable if the bubble reaches its critical point. This critical point is the condition for the appearance of cavitation in the liquid. When the ultrasonic frequency is close to or lower than the resonance frequency of the bubble, the bubble will collapse and cavitation will occur. When the initial radius is much bigger (see Figure 2, $R_0 = 10^{-6}$ m), the bubble tends to collapse easily.

From a comparison of the curves in Figure 2, it can be seen that under a given hydrostatic pressure, a negative pressure is needed for cavitation to occur in water, while it is not needed in CO_2 liquid. There are two possible reasons for this phenomenon: one is that the vapour pressure of water is 1216 Pa and it

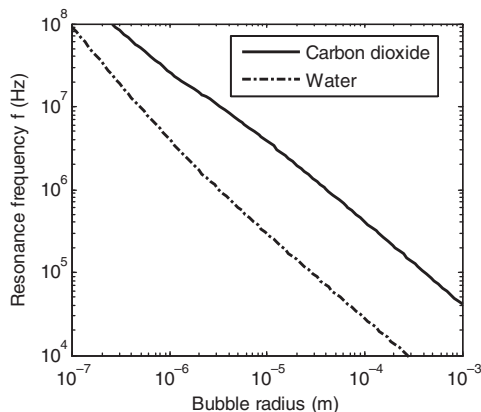


Figure 3. Relationship between the bubble's resonance frequency and its initial radius.

cannot counteract the water's hydrostatic pressure, but the vapour pressure of CO_2 is 4.486 MPa, which is high enough to counteract the hydrostatic pressure, so cavitation can occur under a positive pressure in CO_2 liquid [3]; Another reason is that the surface tension of CO_2 liquid is much smaller than that of water, so the pressure caused by surface tension is smaller in CO_2 liquid than in water.

The relationship between resonance frequency of the bubble and the initial bubble radius in water and CO_2 calculated by Prosperetti's theory [6] is shown in Figure 3 with ultrasonic frequency of 80 kHz. It can be seen that the resonance frequency of the bubble in both water and CO_2 decreases with the initial radius increase. For the same, initial bubble radius the resonance frequency of the bubble in water is lower than that in CO_2 . For example, at 20 kHz resonance frequency, the radius of the bubble is 1×10^{-4} m in water, and 2×10^{-2} m in CO_2 liquid. Assuming the ultrasonic frequency is 20 kHz, the ultrasonic intensity is high, and the pressure generated by ultrasound is greater than the Blake threshold, then cavitation may occur in liquid CO_2 for the bubbles having radii smaller than 2×10^{-2} m, or those bubbles with radii smaller than 1×10^{-4} m in water. The higher the ultrasonic frequency, the smaller the radius of the bubbles for cavitation to be present.

The relationship between the Blake threshold of bubbles and their initial radius at 283 K for water and CO_2 is shown in Figure 4(a). When the bubble's radius is smaller than 10^{-8} m, cavitation occurs more easily in CO_2 at 7.5 MPa hydrostatic pressure than in the water at 0.1 MPa. Cavitation happens when ultrasonic intensity in CO_2 fluid is smaller than in water. When the bubble's radius increases, cavitation occurs more easily in water than in CO_2 . When the hydrostatic pressure in CO_2 is near the vapour pressure in the bubble, for example $P_0 = 4.6$ MPa, $P_V = 4.48$ MPa, the required Blake threshold decreases. Even where the bubble radius is rather large, the required Blake threshold is nearly equal to that required for water at normal atmospheric pressure. In Figure 4(b), it can be seen that, under the same hydrostatic pressure conditions, the required Blake threshold of CO_2 and the ultrasonic intensity decreases when the temperature increases. That is because the vapour pressure increases with temperature.

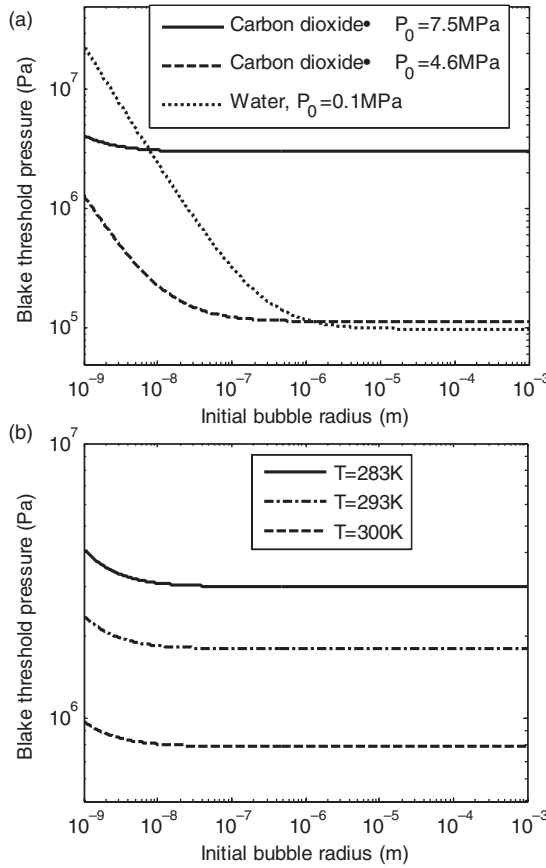


Figure 4. Relationship between the Blake threshold of the bubbles and initial bubble radius. (a) $T=283\text{ K}$ and (b) $P_0=7.5\text{ MPa}$.

3. The liquid’s cavitation

Liquid CO_2 containing bubbles is actually a two-phase mixture: liquid phase and the cavitation bubbles phase. If the bubble radius and the inter-bubble distances are much smaller than the typical length scale of wave propagation and relative motion of the phases is negligible, the mixture can be taken as a continuous medium. It is also assumed that all bubbles remain spherical in shape, the density of the bubbles per unit volume is relatively small and no direct bubble–bubble interactions occur. The wave equation which describes the propagation of ultrasound waves in a bubbly liquid can be obtained [7]:

$$\frac{1}{c_l^2} \frac{\partial^2 p}{\partial t^2} - \nabla^2 p = \rho_l \frac{\partial^2 \beta}{\partial t^2} \tag{9}$$

c_l is the sound speed of the liquid, β is the void fraction of the bubbles [8]

$$\beta = \frac{4\pi}{3} \int_0^\infty R^3(a, x, t) f(a, x) da \tag{10}$$

where $R(a, x, t)$ defines the instantaneous bubble radius at the position x at time t having an equilibrium radius a , $f(a, x)da$ is the number of bubbles per unit volume with equilibrium radii between a and $a + da$ located in the neighbourhood of the point x . The bubble radius $R(a, x, t)$ is described by the Keller–Miksis equation to model the bubble dynamics [9]

$$\left(1 - \frac{\dot{R}}{c}\right) R \ddot{R} + \frac{3}{2} \left(1 - \frac{\dot{R}}{3c}\right) \dot{R}^2 = \frac{1}{\rho_\ell} \left(1 + \frac{\dot{R}}{c}\right) P(\dot{R}, R, t) + \frac{R}{\rho_\ell c} \frac{dp(\dot{R}, R, t)}{dt} \quad (11)$$

$$p(\dot{R}, R, t) = \left(P_0 + \frac{2\sigma}{R_0} - P_V\right) \left(\frac{R_0}{R}\right)^{3k} + P_V - P_0 - \frac{2\sigma}{R_0} - \frac{4\mu\dot{R}}{R} - P_a. \quad (12)$$

P_a is an acoustic driving pressure, c is sound speed and μ is the shear viscosity of the liquid. Assuming a sinusoidal bubble motion, Prosperetti and Commander find that [8]

$$k_m^2 = \frac{\omega^2}{c_l^2} + 4\pi\omega^2 \int_0^\infty \frac{af(a, x)}{\omega_0^2 - \omega^2 + 2ib\omega_0} da, \quad (13)$$

where k_m is the complex wave number, b is the damping constant of a single bubble and ω_0 is the resonance frequency of the bubble which relates to the size of the bubble. Equation (13) can be rewritten as [8]:

$$\frac{c_l^2}{c_m^2} = 1 + 4\pi c_l^2 \int_0^\infty \frac{af(a, x)}{\omega_0^2 - \omega^2 + 2ib\omega_0} da. \quad (14)$$

Let $c_l/c_m = u + iv$, $k_m = \omega/c_m$, then the phase-speed V and attenuation of the liquid A are given by [8]

$$V = c_l/u \quad (15)$$

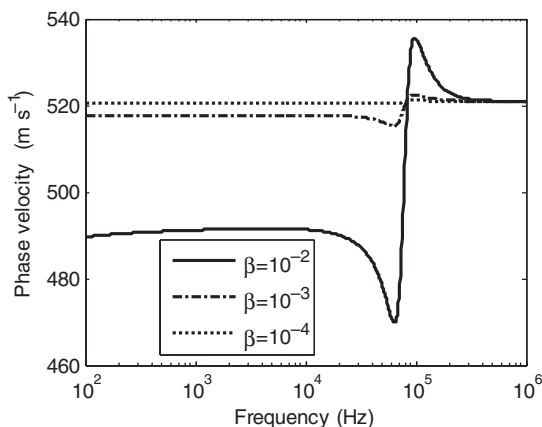
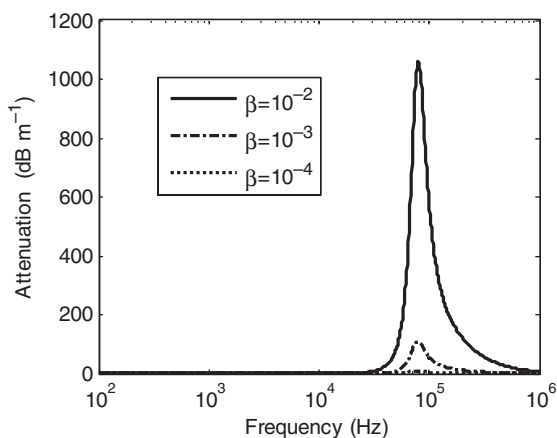
$$A = 20(\log_{10} e)^{(\omega v/c_l)}. \quad (16)$$

For the bubble radius distribution, Prosperetti and Commander [8] assumed a Gaussian distribution

$$f(a) = \begin{cases} C \exp[-(a - a_0)^2/\sigma_a^2] & a_1 < a < a_2 \\ 0 & a \leq a_1, a \geq a_2. \end{cases} \quad (17)$$

C is selected to match the fixed gas void fraction β , a is the bubble's radius. The phase-speed and attenuation of CO_2 at 283 K and 7.5 MPa is calculated for several different void fractions, and are shown in Figures 5 and 6. The following values are chosen for calculation, $a_0 = 0.5 \times 10^{-3}$ m, $a_1 = 0.1 \times 10^{-3}$ m, $a_2 = 0.95 \times 10^{-3}$ m, $\sigma_a = 0.1 \times 10^{-4}$ m, other parameters are obtained from NIST's database [5].

If the CO_2 liquid contains bubbles, its phase-speed decreases to a minimum when the frequency increases to the nearby value of resonance frequency, then rapidly reaches to a maximum when frequency continues to increase above the resonance frequency. After that, the phase-speed decreases and approaches the speed of sound in the liquid without bubbles with increases of frequency.

Figure 5. Phase-speed in CO₂.Figure 6. Attenuation in CO₂.

When the frequency is low, the phase-speed gradually approaches the speed of sound in the liquid (521.0 m s^{-1}) with increases of void fraction. While at high frequency, the phase-speed is nearly equal to the speed of sound in the liquid under different void fractions. At the resonance frequency, the phase-speed increases with the void fraction. On the other hand, ultrasonic attenuation reaches a maximum at the resonance frequency, then attenuation decreases with either increases or decreases of frequency. At the resonance frequency, the attenuation increases when the void fraction increases.

The comparison between the phase-speed and attenuation of CO₂ and that of water under the same bubble distribution is shown in Figures 7 and 8. The void fraction β is 1×10^{-2} , temperature is 283 K, hydrostatic pressure in water is 0.1 MPa, and hydrostatic pressure in CO₂ is 7.5 MPa. The parameters relative to the radius are the same as those of Figures 5 and 6. Other parameters come from NIST's database [5]. The Y value indicates the ratio of phase-speed to the sound speed c_l in pure liquid without bubbles. From Figures 7 and 8, it can be seen that the resonance

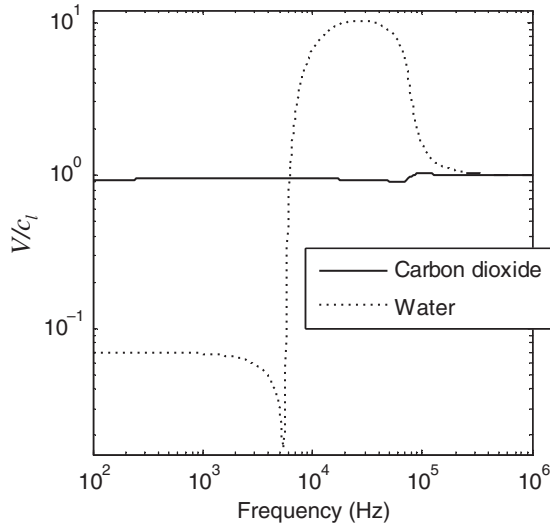


Figure 7. Phase-speed comparison between CO_2 and water.

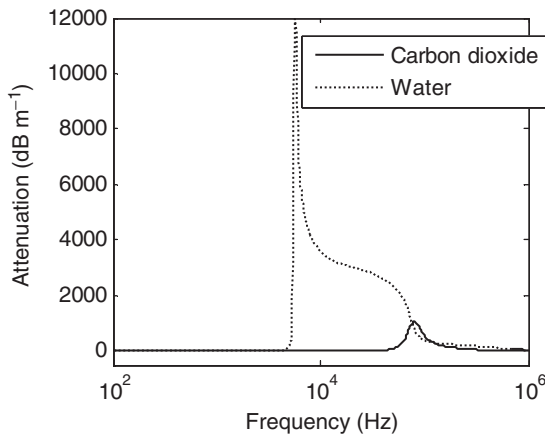


Figure 8. Attenuation comparison between CO_2 and water.

frequency of CO_2 is higher than that of water under the calculated conditions, which is also shown in Figure 3. By comparing the CO_2 with the water, it can be found that the variation of phase-speed and attenuation is much smaller in CO_2 than in the water at the resonance frequency regime. Below the resonance frequency of the bubble, the range of phase-speed is much smaller in CO_2 than in the water. Near the resonance frequency, the frequency range from the minimal phase-speed to the maximal phase-speed is much wider in water than in CO_2 . The phase-speed of both liquids approaches their speed of sound in the pure liquid at the high frequency range. When there are bubbles in the liquid, the ultrasonic attenuation of both liquids increases with frequency, and reaches a maximum at the resonance frequency, and then decreases with the increases of frequency.

4. Discussion

From the above analysis, it can be seen that there is a possibility of ultrasonic cavitation in high-pressure CO₂ under certain conditions and has also been shown by Kuijpers et al. [3] who observed cavitation in CO₂ at 283 K and 7.5 MPa with ultrasound intensity of 125 W cm⁻² [3]. According to formulae $P_A = \sqrt{2\rho cI}$, when $\rho = 899.2 \text{ kg m}^{-3}$, $c = 521.0 \text{ m s}^{-1}$ [5], one can calculate the Blake threshold pressure as 1.08 MPa. This is 2 MPa lower than the theoretical value (3 MPa, Figure 3). This may be because: (1) there are some bubble nuclei in the liquid which cause some weak spots of tension intensity [10]; (2) temperature distribution in the liquid is inhomogeneous and the Blake threshold reduces when temperature rises.

For the supercritical CO₂, no phase boundary exists, which prevents cavitation [3]. However, if CO₂ is not pure or contains other gases, or if the vapour penetrated the bubble, there are nuclei in the supercritical CO₂. These nuclei will lead to cavitation under certain conditions. Raising the liquid's temperature and increasing the vapour pressure in the bubbles may cause the ultrasound to induce cavitation in the supercritical CO₂ fluid. This may need further experimental studies. It was pointed out that the bubble cavitation intensity decreases when the vapour pressure increases [10].

5. Conclusions

For CO₂ liquid with large bubbles, higher ultrasonic intensity is required to induce cavitation when the hydrostatic pressure is greater and the temperature is lower. When the bubble size reduces, the ultrasonic intensity required for cavitation is lower than that in water. When the hydrostatic pressure is much closer to the vapour pressure, cavitation occurs more easily, and the required ultrasonic intensity is relatively lower.

When the frequency is low, the phase-speed of liquid CO₂ gradually approaches the speed of sound in the pure liquid with increases of void fraction. At high frequency, the phase-speed is nearly equal to the speed of sound in the liquid under different void fractions. At the resonance frequency, the phase-speed increases with void fraction.

The attenuation of ultrasound in liquid CO₂ reaches a maximum near the resonance frequency and decreases with increases and decreases of frequency. At the resonance frequency, attenuation increases when the void fraction increases.

Future exploration of the cavitation mechanism in liquid CO₂ will provide theory and methods for substituting CO₂ for traditional solvents. Ultrasonic cavitation in CO₂ fluid will greatly extend the use of ultrasonic chemistry.

Acknowledgements

This work is supported by National Natural Science Foundation of China, Grant No. 10674048 and by Guangdong Provincial Natural Science Foundation, Grant No. 06025714.

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