



Sensitivity of free radicals production in acoustically driven bubble to the ultrasonic frequency and nature of dissolved gases



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ABSTRACT

Central events of ultrasonic action are the bubbles of cavitation that can be considered as powered microreactors within which high-energy chemistry occurs. This work presents the results of a comprehensive numerical assessment of frequency and saturating gases effects on single bubble sonochemistry. Computer simulations of chemical reactions occurring inside a bubble oscillating in liquid water irradiated by an ultrasonic wave have been performed for a wide range of ultrasonic frequencies (213–1100 kHz) under different saturating gases (O₂, air, N₂ and H₂). For O₂ and H₂ bubbles, reactions mechanism consisting in 25 reversible chemical reactions were proposed for studying the internal bubble-chemistry whereas 73 reversible reactions were taken into account for air and N₂ bubbles. The numerical simulations have indicated that radicals such as ·OH, H·, HO₂· and O are created in the bubble during the strong collapse. In all cases, hydroxyl radical (·OH) is the main oxidant created in the bubble. The production rate of the oxidants decreases as the driving ultrasonic frequency increases. The production rate of ·OH radical followed the order O₂ > air > N₂ > H₂ and the order becomes more remarkable at higher ultrasonic frequencies. The effect of ultrasonic frequency on single bubble sonochemistry was attributed to its significant impact on the cavitation process whereas the effects of gases were attributed to the nature of the chemistry produced in the bubble at the strong collapse. It was concluded that, in addition to the gas solubility, the nature of the internal bubble chemistry is another parameter of a paramount importance that controls the overall sonochemical activity in aqueous solutions.

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1. Introduction

The chemical effects of ultrasound (sonochemistry) originate from acoustic cavitation, that is, ultrasound-induced formation, growth and violent collapse of microbubbles in a liquid medium [1]. The rapid collapse of cavitation bubbles is nearly adiabatic, rendering each individual bubble a microreactor, inside which temperatures of the order of 5000 K and pressures of hundreds of atmospheres have been shown to exist [2,3]. As a results, water vapor entrapped inside a bubble is dissociated into H· and ·OH radicals, and with other species present, various other reactive species such as HO₂·, O and H₂O₂ may form [4]. Parallel reaction pathways exist where volatile solutes may evaporate into the bubble and be pyrolysed by the high core temperatures [4]. The radical species

produced can recombine, react with other gaseous species present in the cavity or diffuse out of the bubble into the surrounding liquid to serve as oxidants [5]. All chemical reactions promoted by ultrasound are known as sonochemical reactions. Under certain conditions, bubble collapse is also accompanied by the emission of light, called sonoluminescence, originating from the hot core of the bubble during the final stages of collapse [6,7].

A number of factors influence sonochemical activity (radicals' production) and luminescence intensity. These include frequency of ultrasound, dissolved gas, acoustic power, bulk liquid temperature, etc. [8–17]. Among all these parameters, the ultrasonic frequency and the nature of the dissolved gases have shown spectacular effects. The frequency of ultrasound has a significant impact on the critical size of the bubble (resonance size), which decreases as ultrasound frequency increases [18,19] and the lower frequency ultrasound would produce more violent collapse leading to higher localized temperatures and pressures in the bubble. However, recent published papers [8–10,14,15] have suggest a higher sonochemical activity at higher frequency. As a general idea,

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Nomenclature

A_f (A_r)	pre-exponential factor of the forward (reverse) reaction, [(cm ³ mol ⁻¹ s ⁻¹) for two body reaction and (cm ⁶ mol ⁻² s ⁻¹) for three body reaction]	R_{\max}	maximum radius of the bubble (m)
b_f (b_r)	temperature exponent of the forward (reverse) reaction	R_0	ambient bubble radius (m)
c	speed of sound in the liquid medium, (m s ⁻¹)	t	time (s)
E_{af} (E_{ar})	activation energy of the forward (reverse) reaction (cal mol ⁻¹)	T	temperature inside a bubble (K)
f	frequency of ultrasonic wave (Hz)	T_{\max}	maximum temperature inside a bubble (K)
I_a	acoustic intensity of ultrasonic irradiation (W m ⁻²)	T_{∞}	bulk liquid temperature (K)
k_{f9} (k_r)	forward (reverse) reaction constant, [(cm ³ mol ⁻¹ s ⁻¹) for two body reaction and (cm ⁶ mol ⁻² s ⁻¹) for three body reaction]	x_i	solubility (in mole fraction) of the gas i in water
p	pressure inside a bubble (Pa)	Y_{H_2O}	mole fraction of water vapor trapped at the collapse
p_{\max}	maximum pressure inside a bubble (Pa)		
p_{∞}	ambient static pressure (Pa)		
P_A	amplitude of the acoustic pressure (Pa)		
P_v	vapor pressure of water (Pa)		
P_{g0}	initial gas pressure (Pa)		
R	radius of the bubble (m)		

Greek letters

γ	specific heat ratio (c_p/c_v) of the gas mixture
σ	surface tension of liquid water (N m ⁻¹)
ρ	density of liquid water (kg m ⁻³)
λ	gas thermal conductivity (W m ⁻² K)
τ_c	collapse time of the bubble (s)
τ_{exp}	expansion time of the bubble (s)

a gas with high polytropic index ($\gamma = c_p/c_v$) and low thermal conductivity converts more energy upon collapse and gives a greater sonochemical effect than one with low polytropic index. However, recent investigations [12–14,16,17] showed that polyatomic gases can provide a more sonochemical activity than monatomic gases because gases such as oxygen can provide additional sources for the production of active species through the self thermal dissociation inside a bubble and, thus, can compensate for the lower internal cavitation temperatures. On the other hand, Okitsu et al. [20] showed experimental evidence that the bubble temperature induced by high frequency ultrasound is almost the same among different gases and the overall chemical efficiency is in proportion to the gas solubility, which would be closely related to the number of active bubbles. Therefore, the mechanism of saturating gases and ultrasound frequency on the sonochemical activity remained poorly understood. This is in fact because the effects of these two parameters are not well established in microscopic scale (scale of single bubble). In this paper, the results of a comprehensive numerical assessment of frequency and saturating gases effects on sonochemical activity of single cavitation bubble were described. The numerical simulations of the bubble oscillation and chemical reactions therein have been performed for various ultrasonic frequencies (in the range of 213–1100 kHz) under different saturating gases (O₂, air, N₂ and H₂). The employed model combines the dynamic of bubble collapse in acoustic field with the chemical kinetics of single cavitating bubble.

2. Model and computational methods

The theoretical model used in the present numerical simulations has been fully described in our previous works [21–24]. The following is a brief description of the model.

2.1. Bubble dynamics model

A gas and vapor filled spherical bubble isolated in water oscillates under the action of a sinusoidal sound wave. The temperature and pressure in the bubble are assumed spatially uniform and the gas content of the bubble behaves as an ideal gas [25]. The radial dynamics of the bubble is described by the Keller–Miksis equation

that includes first order terms in the Mach number $M = \dot{R}/c$ [26,27]:

$$\left(1 - \frac{\dot{R}}{c}\right)R\ddot{R} + \frac{3}{2}\left(1 - \frac{\dot{R}}{3c}\right)\dot{R}^2 = \frac{1}{\rho_L}\left(1 + \frac{\dot{R}}{c}\right)\left[p - p_{\infty} - \frac{2\sigma}{R} - 4\mu\frac{\dot{R}}{R} + P_A \sin(2\pi ft)\right] + \frac{R}{\rho_L c} \frac{d}{dt}\left[p - p_{\infty} - \frac{2\sigma}{R} - 4\mu\frac{\dot{R}}{R} + P_A \sin(2\pi ft)\right] \quad (1)$$

in this equation dots denote time derivatives (d/dt), R is the radius of the bubble, c is the speed of sound in the liquid, ρ_L is the density of the liquid, σ is the surface tension, μ is the liquid viscosity, p is the pressure inside the bubble, p_{∞} is the ambient static pressure, P_A is the acoustic amplitude and f is the sound frequency. The acoustic amplitude P_A is correlated with the acoustic intensity I_a , or power per unit area, as $P_A = (2I_a\rho_L c)^{1/2}$ [28].

The expansion of the bubble is assumed as isothermal and its total compression is considered as adiabatic [29]. These assumptions, which are widely accepted since the lifetime of an oscillation at high frequency is relatively short with a very rapidly occurring collapse event, were pointed out by Yasui et al. [30] using a more detailed model. We also assume that the vapor pressure in the bubble remains constant during the bubble expansion phase and there is no gas diffusion during expansion and no mass and heat transfer of any kind during collapse. We note here that Storey and Szeri [31] demonstrated that the inclusion of mass transfer on the bubble dynamics has practically no effect on the maximum bubble temperature attained in the bubble at the collapse when the compression ratio of the bubble (R_{\max}/R_{\min}) is less than 20 (R_{\max} is the maximum radius of the bubble and R_{\min} is the minimum bubble radius at the collapse). This level of R_{\max}/R_{\min} was never attained in the present numerical study (see Table 4). Therefore, in order to reduce computational parameters, the current model takes, as input, initial bubble vapor content and neglects mass and heat transfer during bubble expansion and collapse.

Because of the above assumptions, the pressure and temperature inside the bubble at any instant during adiabatic phase can be calculated from the bubble size as

$$p = \left[P_v + P_{g0} \left(\frac{R_0}{R_{\max}} \right)^3 \right] \left(\frac{R_{\max}}{R} \right)^{3\gamma} \quad (2)$$

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