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Theoretical estimation of the temperature and pressure within collapsing acoustical bubbles

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ABSTRACT

Formation of highly reactive species such as $^{\cdot}$OH, H$^{\cdot}$, $HO_2^{\cdot}$ and H_2O_2 due to transient collapse of cavitation bubbles is the primary mechanism of sonochemical reaction. The crucial parameters influencing the formation of radicals are the temperature and pressure achieved in the bubble during the strong collapse. Experimental determinations estimated a temperature of about 5000 K and pressure of several hundreds of MPa within the collapsing bubble. In this theoretical investigation, computer simulations of chemical reactions occurring in an O_2-bubble oscillating in water irradiated by an ultrasonic wave have been performed for diverse combinations of various parameters such as ultrasound frequency (20–1000 kHz), acoustic amplitude (up to 0.3 MPa), static pressure (0.03–0.3 MPa) and liquid temperature (283–333 K). The aim of this series of computations is to correlate the production of $^{\cdot}$OH radicals to the temperature and pressure achieved in the bubble during the strong collapse. The employed model combines the dynamic of bubble collapse in acoustical field with the chemical kinetics of single bubble. The results of the numerical simulations revealed that the main oxidant created in an O_2 bubble is $^{\cdot}$OH radical. The computer simulations clearly showed the existence of an optimum bubble temperature of about 5200 ± 200 K and pressure of about 250 ± 20 MPa. The predicted value of the bubble temperature for the production of $^{\cdot}$OH radicals is in excellent agreement with that furnished by the experiments. The existence of an optimum bubble temperature and pressure in collapsing bubbles results from the competitions between the reactions of production and those of consumption of $^{\cdot}$OH radicals at high temperatures.

1. Introduction

Acoustic cavitation is the phenomenon observed when ultrasound of sufficient intensity is transmitted through a liquid causing micron-sized gas and vapor bubbles to oscillate, grow and violently implode giving rise to extreme, but localized, conditions within the collapsed cavities (extremely high temperatures and pressures) [1]. Such conditions are primarily responsible for the chemical effects (sonochemistry) associated with acoustic cavitation in liquids [2]. The extremely high temperatures and pressures formed in collapsing cavitation bubbles in aqueous solutions lead to the thermal dissociation of the trapped water vapor into H$^{\cdot}$ and $^{\cdot}$OH radicals, and with other species present, various other reactive species such as $HO_2^{\cdot}$, O and H_2O_2 may form [2,3]. A parallel reaction pathways exist where volatile solutes may evaporate into the bubble and be pyrolysed by the high core temperatures [2,4]. The radical species produced can recombine, react with other gaseous species present in the cavity, or diffuse out of the bubble into the bulk liquid medium to serve as oxidants [5]. Among all the oxidants created in the bubble, $^{\cdot}$OH radical is of primary interest because of its high potential of oxidation. Under certain conditions bubble collapse can also result in light emission, sonoluminescence, originating from the hot core of the bubble during the final stages of collapse [4,6].

Experimental determinations of the temperature within a cavitation bubble have been made by a number of research groups. By fitting the experimentally recorded single bubble sonoluminescence spectra, temperatures in the range of 5000–20,000 K have been estimated [7]. However, experimental estimation of the temperature within the collapsing bubbles based on multibubble sonochemistry and sonoluminescence are reported to be between 750 and 6000 K. Misík et al. [8], using the kinetic isotope effect in an EPR spin-trapping study of the sonolyse of H_2O/D_2O mixtures, found that the cavitation temperature determined was dependent on the specific spin trap used and fall in the range of 1000–4600 K. Misík and Reisz [9] used the kinetic isotope effect in the ultrasound induced production of radicals in organic liquids to estimate the temperature during cavitation. The bubble temperature was found to be in the range 750–6000 K. Suslick et al. [10], using comparative rate thermometry in alkane solutions, postulated that there

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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]
b_f (b_r)	temperature exponent of the forward (reverse) reaction
c	speed of sound in the liquid medium (m s^{-1})
E_{af} (E_{ar})	activation energy of the forward (reverse) reaction (cal mol^{-1})
f	frequency of ultrasonic wave (Hz)
I_a	acoustic intensity of ultrasonic irradiation (W m^{-2})
k_f (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]
p	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)
R_{max}	maximum radius of the bubble (m)
R_0	ambient bubble radius (m)
t	time (s)
T	temperature inside a bubble (K)
T_c	critical temperature of water (374.2 °C)
T_∞	ambient liquid temperature (K)
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})
μ	viscosity of liquid water (N m^{-2} s)

are two regions of sonochemical reactivity: a gas phase zone within the collapsing cavity with an estimated temperature and pressure of 5200 ± 650 K and 50 MPa, respectively, and a thin liquid layer immediately surrounding the collapsing cavity with an estimated temperature of ~1900 K. Henglein and coworkers [11], by means of sonolysis of methane in argon saturated water, estimated the bubble core temperature to be in the range of 1930–2720 K, depending upon the concentration of methane and argon present in water (the temperature decreased with an increase in the percentage of methane). Using *tert*-butanol sonolysis as means, Tauber et al. [12] estimated the bubble temperature to be in the range 2300–3600 K. The higher temperature was obtained for fresh water, whereas the lower temperature was obtained for solution with the highest concentration of *tert*-butanol. Recently, Ashokkumar and coworkers measured the bubble temperature at different conditions using the Methyl Radical Recombination (MRR) method [13–15]. They found that the maximum bubble temperature is obtained in fresh water and is greatly influenced by the frequency of ultrasound. They estimated maximum temperatures of about 3400 ± 200 K at 20 kHz, 4300 ± 200 K at 366 kHz and 3700 ± 200 K (6200 K [15]) at 1056 kHz [13,14].

In the present study, we have theoretically estimated the optimum temperature of collapsing bubble for the production of the oxidants, i.e. $^\bullet$OH radicals. The used model combines the dynamic of bubble collapse in acoustical field propagated in water with a chemical kinetics consisting in nineteen reversible chemical reactions occurring at high temperatures during the strong collapse of the bubble. A series of computations were performed for more than 400 combinations between various parameters including ultrasound frequency in the range 20–1000 kHz, acoustic amplitude up to 0.3 MPa, static pressure from 0.03 to 0.3 MPa and liquid temperature between 283 and 333 K.

2. Model

2.1. Bubble dynamics model

The physical situation of the model is that of a gas and vapor filled spherical bubble isolated in water oscillating under the action of a sinusoidal sound wave. The temperature and pressure in the bubble are assumed to be spatially uniform and the gas content of the bubble behaves as an ideal gas [16]. The radial dynamics of the bubble is described by the Keller–Miksis equation [17,18]:

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

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 $P_A = (2I_a\rho_Lc)^{1/2}$ [19]. Eq. (1) is only accurate to first order in the bubble wall Mach number ($\dot{R}/c$) but, for all acoustic amplitudes in this study, this level of accuracy is sufficient (the speed of the bubble wall ($|\dot{R}|$) at the collapse never exceeds the sound velocity in the liquid c, which is the assumption used in the derivation of the equation [17]).

In the present model, the expansion of the bubble is considered as isothermal and its total compression (implosion phase) is treated as adiabatic [20]. This assumption is very accepted as for high frequencies the lifetime of the bubble is very short and the collapse event occurs rapidly. We assume also 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. There exist in the literature some research studies that include these effects [20–22]. For a comparative point of view, the inclusion of these effects, leading to a realistic situation, might change the absolute values of the predicted collapse temperature and pressure but definitively will not change the predicted trends including the qualitative variation of the maximum radius, the collapse time and the maximum collapse temperature with variation in operational conditions. Additionally, the importance of mass and heat transfer occur over multiple cycles of oscillation changing, in this case, the internal composition of the vapor phase. Consequently, as the present numerical calculations were carried out for one acoustic period, the mass and heat transfer will not also affect significantly the quantitative bubble yield, predicted in one acoustic period. So, 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.

On the basis of the above assumptions, the pressure and temperature inside the bubble at any instant during adiabatic phase can be calculated from the bubble size, using the adiabatic law: