



A method for predicting the number of active bubbles in sonochemical reactors



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ABSTRACT

Knowledge of the number of active bubbles in acoustic cavitation field is very important for the prediction of the performance of ultrasonic reactors toward most chemical processes induced by ultrasound. The literature in this field is scarce, probably due to the complicated nature of the phenomena. We introduce here a relatively simple semi-empirical method for predicting the number of active bubbles in an acoustic cavitation field. By coupling the bubble dynamics in an acoustical field with chemical kinetics occurring in the bubble during oscillation, the amount of the radical species $\cdot\text{OH}$ and $\text{HO}_2\cdot$ and molecular H_2O_2 released by a single bubble was estimated. Knowing that the H_2O_2 measured experimentally during sonication of water comes from the recombination of hydroxyl ($\cdot\text{OH}$) and perhydroxyl ($\text{HO}_2\cdot$) radicals in the liquid phase and assuming that in sonochemistry applications, the cavitation is transient and the bubble fragments at the first collapse, the number of bubbles formed per unit time per unit volume is then easily determined using material balances for H_2O_2 , $\cdot\text{OH}$ and $\text{HO}_2\cdot$ in the liquid phase. The effect of ultrasonic frequency on the number of active bubbles was examined. It was shown that increasing ultrasonic frequency leads to a substantial increase in the number of bubbles formed in the reactor.

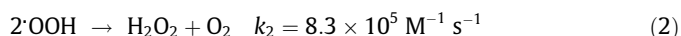
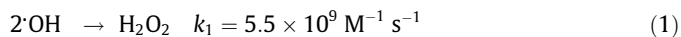
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1. Introduction

Ultrasound has a wide variety of applications, ranging from the degradation of pollutants [1–3], polymerization reactions [4] and fabrication of nano-particles [5,6] to food science [7,8] and biomedical applications [9,10]. Most of these processes arise from acoustic cavitation, that is, the formation, growth and violent collapse of microscopic bubbles as the alternate compressions and rarefactions of the sound waves propagate through the liquid [11]. Acoustic cavitation is also responsible for the emission of broad wavelength light, which is called sonoluminescence (SL) [12,13]. The microbubbles can be either stable, oscillating about their average or equilibrium size for many acoustic cycles or transient when they grow to a certain size in one or at most a few acoustic cycles and violently collapse during the compression part of the wave [14]. Sonochemistry and sonoluminescence are attributed to the transient cavitation bubbles [14].

The chemical effects of acoustic cavitation are a direct result of the very high temperatures, of the order of 5000 K, and pressures,

in the range of hundreds of bars, which are reached in the bubbles when they collapse [15,16]. The extremely high conditions formed in collapsing bubbles in aqueous solutions lead to the thermal dissociation of the trapped water vapor into reactive hydroxyl radicals ($\cdot\text{OH}$) and hydrogen atoms ($\text{H}\cdot$) [17]. These active species can recombine, react with other gaseous species present in the cavity to form other active species such as $\text{HO}_2\cdot$ and O , or diffuse out of the bubble into the bulk liquid medium where they are able to induce chemical transformation [18]. In the absence of any solutes in the liquid medium, these primary active species of sonolysis mostly recombine at the bubble solution interface to form hydrogen peroxide (H_2O_2) that is released in the medium according to the following reactions [19]:



Due to their high reactivity and their short lifetime, the total number of primary active species produced by the acoustic bubbles cannot be directly measured. It is generally accepted that the yield of H_2O_2 can be considered as an indicator for quantifying the overall chemical yield of ultrasound in aqueous media [20,21].

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Nomenclature

c	speed of sound in the liquid medium (m s^{-1})	R	radius of the bubble (m)
f	frequency of ultrasonic wave (Hz)	R_{max}	maximum radius of the bubble (m)
I_a	acoustic intensity of ultrasonic irradiation (W m^{-2})	R_{min}	minimum radius of the bubble at the collapse (m)
I	number of chemical reactions	R_0	ambient bubble radius (m)
K	number of all species in the bubble	$r_{\text{H}_2\text{O}_2}$	production rate of H_2O_2 (mol s^{-1})
N	number of bubbles collapsing per unit volume per unit time ($\text{L}^{-1} \text{s}^{-1}$)	r_{OH}	production rate of $\cdot\text{OH}$ (mol s^{-1})
$n_{\text{H}_2\text{O}}$	number of moles of H_2O in the bubble (mol)	r_{HO_2}	production rate of HO_2 (mol s^{-1})
nt	Number of moles of all species in the bubble (mol)	t	time (s)
$n_{\text{H}_2\text{O}_2}$	number of moles of H_2O_2 released by the collapse of single bubble (mol)	T	temperature inside a bubble (K)
n_{OH}	number of moles of $\cdot\text{OH}$ released by the collapse of single bubble (mol)	T_∞	ambient liquid temperature (K)
n_{HO_2}	number of moles of HO_2 released by the collapse of single bubble (mol)	V	volume of the bubble (m^3)
p	pressure inside a bubble (Pa)	X	symbol of chemical species
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)		

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 ($\text{N m}^{-2} \text{s}$)
ε	molar extinction coefficient of I_3^- ($\text{L mol}^{-1} \text{cm}^{-1}$)

The light emission from acoustic bubbles (SL) arises essentially from vibronically excited states of species, i.e. $\cdot\text{OH}$ radicals, produced as a result of the high temperatures and pressures that are generated within the core of bubbles during strong collapse [22].

The overall efficiency of sonochemical processes depends on the bubble population (bubbles number and sizes), which in turn depend upon various parameters, such as the ultrasonic frequency and acoustic amplitude [23]. Knowledge of the bubbles number and sizes is very important for the prediction of the performance of ultrasonic reactors. The literature in this field is scarce, probably due to complicated nature of the phenomena. In our previous work [24], we have studied theoretically the effect of ultrasonic frequency and acoustic amplitude on the size of sonochemically active bubbles using a model that combines the dynamics of bubble oscillation in acoustical field with chemical kinetics occurring in the bubble during its oscillation. In this work, basing on our theoretical model developed earlier for a single bubble [24], we introduce a relatively simple semi-empirical method for predicting the number of active bubbles in a sonochemical reactor. The effect of ultrasonic frequency on the number of bubbles formed in the sonochemical reactor was examined.

2. Materials and methods

2.1. Ultrasonic reactor

Sonolysis experiments were conducted in cylindrical water-jacketed glass reactors (Fig. 1). The ultrasonic waves of 300 kHz were emitted from the bottom of the reactor through a piezoelectric disc (diameter 4 cm) fixed on Pyrex plate (diameter 5 cm). The ultrasonic waves of 585, 860 and 1140 kHz were delivered from the bottom through a Meinhardt multifrequency transducer (model E/805/T/M, diameter of the active area 5.3 cm). The temperature of the solution was monitored using a thermocouple immersed in the reacting medium. Acoustic power dissipated in the reactor was estimated using a standard calorimetric method [25,26].

2.2. Procedures

Sonochemical experiments involving H_2O_2 production were performed by sonicating 300 mL of distilled water. The tempera-

ture of the aerated solution was maintained at 25 °C by circulating cooling water through a jacket surrounding the cell. The acoustic power delivered to the reactor was adjusted calorimetrically to obtain the same value of acoustic intensity, 2 W cm^{-2} (corresponding to an acoustic amplitude of 1.73 atm), for all frequencies.

Hydrogen peroxide concentrations were analytically determined using the iodometric method [27]. Sample aliquots taken periodically from the reactor during sonolysis were added in the quartz cell of the spectrophotometer containing 1 mL of potassium iodide (0.1 M) and 20 μL of ammonium heptamolybdate (0.01 M). The iodide ion (I^-) reacts with H_2O_2 to form the triiodide ion (I_3^-). The mixed solutions were allowed to stand for 5 min before absorbance was measured. The absorbance was recorded with a UV-visible spectrophotometer (Lightwave II) at the maximum wavelength of the formed triiodide (I_3^-) (352 nm; the molar absorptivity $\varepsilon = 26300 \text{ L mol}^{-1} \text{cm}^{-1}$).

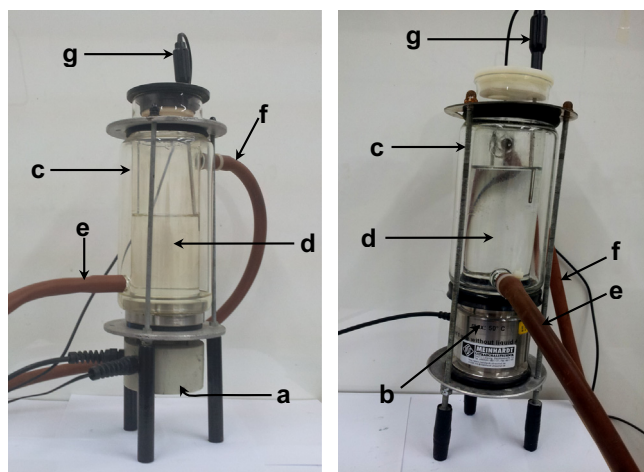


Fig. 1. Sonochemical reactors used for quantifying the amount of H_2O_2 formed in the liquid water during sonication. (a) 300 kHz ultrasonic transducer, (b) Meinhardt multi-frequency transducer operating at 585, 860 and 1140 kHz, (c) cylindrical water-jacketed glass cells, (d) sonicated water, (e) inlet cooling water, (f) outlet cooling water, (g) thermocouples.

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