The effect of liquid temperature on bubble-size distribution in the presence of power ultrasound and carbon tetrachloride

Aissa Dehane1 · Slimane Merouani1 · Oualid Hamdaoui2

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Abstract
Acoustic cavitation-induced sonochemistry is employed for a variety of industrial and laboratory-scale physical and chemical applications, including cleaning, nanomaterial synthesis, and destruction of water contaminants. In acoustic bubbles, CCl4 pyrolysis can totally alter the bubble sonochemistry as well as the active bubble-size population. The present theoretical work provides the unique study on the effect of liquid temperature on the size distribution of acoustically active bubbles in the presence of CCl4 (i.e., precursor of reactive chlorine species, RCS, and scavenger of hydrogen atom in pyrolytic reactions) in the bulk liquid. An updated reaction scheme for CCl4 sonopyrolysis is used. It was found that the sonopyrolysis of CCl4 within the bubble reduces its maximal temperature, but it notably increases its maximal molar yield. For lower CCl4 concentrations (≤ 0.1 mM), the broadness of active bubbles range for the total oxidants yield increased proportionally with the rise of liquid temperature from 20 to 50 °C. Nevertheless, the increase of CCl4 concentration amortizes this width increase over the same range of liquid temperature (20–50 °C). At higher concentrations of CCl4 (> 0.1 mM), the broadness of the active bubbles range becomes approximately constant and independent of the liquid temperature and CCl4 concentration.

Keywords Sonochemistry · Carbon tetrachloride · Active bubbles · Liquid temperature · Reactive chlorine species (RCS)

List of symbols

- $A_f$ ($A_r$): 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)
- $P_{max}$: 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)
- $R$: Radius of the bubble (m)
- $R_{max}$: Maximum radius of the bubble (m)
- $R_b$: Ambient bubble radius (m)
- $R_g$: Ideal gas constant (J mol$^{-1}$ K$^{-1}$)
- $t$: Time (s)
- $T$: Temperature inside a bubble (K)
- $T_{max}$: Maximum temperature inside a bubble (K)
- $T_{∞}$: Bulk liquid temperature (K)
- $X$: Thermal diffusivity inside the bubble (m$^2$ s$^{-1}$)
- $C_p$: Heat capacity concentration inside the bubble (J m$^{-3}$ K$^{-1}$)
- $M$: Molar mass of water (Kg mol$^{-1}$)
- $m$: Evaporation–condensation rate of water (Kg m$^{-2}$ s$^{-1}$)
- $Q$: Energy transferred by heat exchange (J s$^{-1}$)
- $n$: Molar amount (mol)
- $P_B$: Liquid pressure on the external side of the bubble wall (Pa)
- $V$: Volume of the bubble (m$^3$)

Slimane Merouani
s.merouani@yahoo.fr; s.merouani03@gmail.com

1 Laboratory of Environmental Process Engineering, Department of Chemical Engineering, Faculty of Process Engineering, University Salah Boubnider-Constantine 3, P.O. Box 72, 25000 Constantine, Algeria

2 Chemical Engineering Department, College of Engineering, King Saud University, P.O. Box 800, Riyadh 11421, Saudi Arabia
additives have been tested in order to evaluate their effects on the rate of pollutants decomposition, e.g., salts (Mahamuni and Pandit 2006; Guo et al. 2008; Merouani et al. 2010; Ferkous et al. 2016), \( \text{S}_2\text{O}_8^{2-} \) (Ferkous et al. 2017), \( \text{IO}_3^- \) (Hamdaoui and Merouani 2017), \( \text{O}_3 \) (Mahamuni and Pandit 2006; Guo et al. 2008; Merouani and Hamdaoui 2019), \( \text{CCl}_4 \) (Mahamuni and Pandit 2006; Bejarano-Pérez and Suarez-Herrera 2008; Guo et al. 2008; Ghodbane and Hamdaoui 2009; Merouani et al. 2010; Park et al. 2011; Uddin and Okitsu 2016), catalysts (Torres et al. 2007; Bejarano-Pérez and Suarez-Herrera 2008; Guo et al. 2008; Merouani et al. 2010; Boutamine et al. 2017), \( \text{H}_2\text{O}_2 \) (Merouani et al. 2010), \( \text{C}_6\text{F}_{14} \) (Uddin and Okitsu 2016), etc. Of all these additives, \( \text{CCl}_4 \) has shown the most attractive attention due to the huge intensification observed in the removal rate of several nonvolatile organic pollutants (e.g., phenol, Bisphe nol A, 2,4-dinitrophenol, methyl orange, C.I. acid orange 8, etc.) in the presence of \( \text{CCl}_4 \) (Wang et al. 2007; Guo et al. 2008; Gültekin et al. 2009; Merouani et al. 2010).

In sonochemistry, carbon tetrachloride has a double role. First, this substance is considered as a serious water contaminant; therefore, various studies are concentrated on the sonolytic destruction of \( \text{CCl}_4 \) where the different parameters affecting the sonolysis of \( \text{CCl}_4 \) (e.g., frequency, power, temperature, pH, ...) have been widely assessed (Wu et al. 1992; Francony et al. 1996; Hua and Hoffmann 1996; Hung and Hoffmann 1999; Colussi et al. 1999; Lee and Oh 2010). The second role of \( \text{CCl}_4 \) is as an intensification tool for the degradation of nonvolatile contaminants as revealed in the previous section. Thanks to its higher volatility in water, pyrolysis of \( \text{CCl}_4 \) inside acoustic bubbles is the main mechanism that ensures its decomposition (Francony et al. 1996; Pétrier and Francony 1997; Hung and Hoffmann 1999; Colussi et al. 1999; Lim et al. 2011). The pyrolytic decomposition of \( \text{CCl}_4 \) can yield several reactive chlorine species (RCS), e.g. \( \text{Cl}^* \) and \( \text{HOCl} \), which are reactive toward organic substrates (Francony et al. 1996; Pétrier and Francony 1997; Hung and Hoffmann 1999; Colussi et al. 1999; Merouani et al. 2010). Moreover, carbon tetrachloride is considered as a good hydrogen atom scavenger (\( k = 3.8 \times 10^7 \text{M}^{-1} \text{s}^{-1} \) (Weissler et al. 1950)), which limits the ultrafast recombination reaction \( \text{OH}^* + \text{H}^* \rightarrow \text{H}_2\text{O} \), thereby increasing the yield of hydroxyl radical that can achieve the bulk solution.

On the other hand, it is well known that the size of active bubbles in sonocavitating medium is an interval rather than a single value, as demonstrated experimentally and theoretically (Burdin et al. 1999; Tsochatzidis et al. 2001; Labouret and Frohly 2002; Avvaru and Pandit 2009; Brotoch et al. 2009; Iida et al. 2010). After a depth research in the literature reports focusing on the use of \( \text{CCl}_4 \) as intensification technique, it was observed that either RCS generation or \( \text{H}^* \) scavenging (or both) induced by \( \text{CCl}_4 \) during sonolysis is the main mechanism reported for justifying the intensifying role.

\[ \text{Surface tension of water (N m}^{-1}\text{)} \]
\[ \text{Density of liquid water (kg m}^{-3}\text{)} \]
\[ \text{Density inside the bubble (kg m}^{-3}\text{)} \]
\[ \text{Thermal conductivity of the mixture (W m}^{-1}\text{K)} \]
\[ \text{Dynamic viscosity (Pa s)} \]
\[ \text{Thermal conductivity of species } k \text{ (W m}^{-1}\text{K)} \]
\[ \text{Density inside the bubble (kg m}^{-3}\text{)} \]
\[ \text{Density of liquid water (kg m}^{-3}\text{)} \]
\[ \text{Saturated vapor density (Kg m}^{-3}\text{)} \]
\[ \text{Saturated vapor density (Kg m}^{-3}\text{)} \]
\[ \text{Stoichiometric coefficient of the } k \text{th chemical species in the } i \text{th reaction} \]
\[ \text{Production rate of the } k \text{th species (mol s}^{-1}\text{m}^{-3}\text{)} \]
of $\text{CCl}_4$ vis-à-vis the sonolytic degradation of nonvolatile organics (Wang et al. 2007; Guo et al. 2008; Ghodbane and Hamdaoui 2009; Gültekin et al. 2009; Merouani et al. 2010). However, since $\text{CCl}_4$ can act as an oxidants-generator (RCS) from the endothermal dissociation of $\text{CCl}_4$ within the bubble, this means that the bubbles population (active bubbles) in the sonochemical reactor may be greatly modified due to the change of the sonoactivity of individual cavities. Thus, one of the most important characteristics of the cavitation field, which is the size of active bubbles (i.e. expressed in terms of ambient bubble radius, $R_0$), may be directly affected by the $\text{CCl}_4$ sonochemistry. This issue was never studied previously, neither experimentally nor theoretically. The determination of the population of active bubbles has a great importance, because if this range is accurately determined, it will be easy to study the effects produced by these bubbles and their interactions with the ultrasonic irradiation (Labouret and Frohly 2010). In fact, the extreme temperature reached inside the bubble and the chemical bubble yield are both strongly affected by this parameters (i.e., $R_0$) (Yasui et al. 2008; Merouani et al. 2013; Merouani and Hamdaoui 2016). Besides, we believe that $\text{CCl}_4$ sonochemistry may increase the number of active bubbles, which could be then an alternative mechanism for justifying the intensifying role of $\text{CCl}_4$ toward the sonochemical process applied for the degradation of nonvolatile organic pollutants.

The present work gives a new theoretical approach for better understanding the effect of $\text{CCl}_4$ on the size of active bubbles for the production of reactive oxygen and chlorine species, all with relation to the pyrolytic conversion efficiency of $\text{CCl}_4$. A special focus is made on the effect of liquid temperature due to the volatile character of $\text{CCl}_4$, which implies that the evaporated amount of $\text{CCl}_4$ with the bubbles could be controlled by the liquid temperature alone. The adopted mathematical model is accurate and includes all transfer phenomena (i.e., mass and heat) and considers the reaction heat resulted from the endothermic decomposition of water vapor and $\text{CCl}_4$ during the whole period of the strong bubble collapse. To the best of our knowledge, no theoretical study treating this subject is available in the literature. The strong point of our study is the manner by which we are simulating the degradation of $\text{CCl}_4$ inside the bubble, as all the transfer phenomena (mass transport, heat exchange and chemical reactions heat) in addition to bubble dynamic and kinetics are considered during the oscillation of bubble inside the sonicated liquid.

**Theoretical model**

Because acoustic cavitation is the central event causing all actions (physical, chemical and biological) of ultrasound in aqueous solution, the single bubble approach is widely adopted by many researchers in order to understand the most important effects generated within the bubble, which are the bubble sonochemistry and sonoluminescence (Kamath et al. 1993; Toegel and Lohse 2003; Yasui et al. 2003, 2008, 2011; Adewuyi and Khan 2012; Ferkous et al. 2015; Gadi Man and Trujillo 2016; Nazari-Mahroo et al. 2018). In addition, the single bubble is considered as a sample of the whole population of cavities found in solution. Consequently, the main tool for understanding such complicate microscopic problems, which occur very fast inside the acoustic bubble, is via the numerical simulation of single bubble sonochemistry. In that case, the bubble dynamics model could be accurate and includes the evaporation and reactions of $\text{CCl}_4$ inside the bubble, the mass transfer of water vapor across the bubble wall, the heat exchanged between the bubble and the surrounding medium as well the reaction heats resulted from the pyrolytic reactions inside the bubble at the strong collapse.

The mathematical model we are using in this study is detailed is paragraph S1 of the Supplementary Material. It is principally based on the theoretical works of Yasui (Yasui 1995) for the bubble dynamics and energy balance and that of Toegel (Toegel and Lohse 2003) for heat transfer and mass transport. The Yasui’s and Toegel’s models are widely used by many researchers (Adewuyi and Khan 2012; Gadi Man and Trujillo 2016; Nazari-Mahroo et al. 2018) for their precise results and accurate simulations for the bubble dynamics as well as its kinetic process. A kinetics mechanism consisting in 26 reversible chemical reactions (Table S2), involving $H_2O, H^*, O, \cdot OH, HO_2^*, H_2O_2, H_3O^+, CCl_4, CCCI_3; CCl_2, Cl, Cl_2, C_2Cl_4, C_2Cl_2, C_2Cl_6, HCl$ and HOCl, is taken into account for simulating the internal bubble chemistry in the presence of $\text{CCl}_4$. The bubble dynamics model includes the effects of (1) liquid viscosity and compressibility, (2) $\text{CCl}_4$ evaporation and reactions, (3) non-equilibrium evaporation and condensation of water vapor at the bubble wall, (4) thermal conduction both inside and outside a bubble and (5) chemical reactions heat. All numerical simulations have been conducted for an argon bubble containing different carbon tetrachloride concentrations (from 0 to 5.2 mM). The model is able to determine the temperature, pressure, bubble radius evolution, bubble wall velocity and the chemical bubble yield at any instant during the bubble oscillation.

**Results and discussion**

Due to the broadness of the operating conditions that affect directly or indirectly the sonochemical activity at macroscopic (solution, i.e. multibubble system) or microscopic (single bubble) scales (e.g., wave frequency, acoustic intensity, liquid temperature, static pressure, saturation gases), we
are limited our study on the frequency of 355 kHz and argon as saturation atmosphere. The choice of these conditions is supported by the fact that (1) the different experimental works conducted in the field of sonochemistry demonstrated that the maximum sonochemical efficiency (as quantified by chemical dosimetry or pollutant oxidation, i.e. including CCl₄ itself) is located at around the frequency of 355 kHz (Mark et al. 1998; Kang et al. 1999; Beckett and Hua 2000, 2001; Kodá et al. 2003; Kanthale et al. 2008; Yang et al. 2008; Lim et al. 2011; Merouani et al. 2014a) and (2) argon atmosphere is widely used to ensure high bubble temperature thanks to its higher polytropic index and lower thermal conductivity than all other mono- and diatomic gases (i.e. this gas creates an efficient environment for the pyrolysis of CCl₄ inside the bubbles (Lim et al. 2011)). Additionally, to reduce the number of simulations, the acoustic intensity of the sound pressure wave is fixed at Iₐ = 1 W/cm². This value of Iₐ coupled with the selected frequency (355 kHz) is largely sufficient to ensure the production of inertial reactive cavities for sonochemistry (Merouani et al. 2014b, 2015a, b; Merouani and Hamdaoui 2016). Also, in the next sections, the ambient bubble radius (i.e., R₀) indicates the bubble radius in the absence of ultrasound irradiation; an active bubble denotes a bubble which is able to generate oxidants with yield ≥ the production threshold yield (10⁻¹⁷ mol), the range of active bubbles denotes the interval of R₀ for bubbles achieving production yields of oxidants ≥ the production threshold yield (10⁻¹⁷ mol), the optimum ambient bubble radius (optimum R₀) denotes the ambient bubble radius at which the production rate of oxidant is maximal.

**Bubble kinetics**

As it is well established, the sonoactivity of the acoustic bubble is totally located at around the end of the bubble collapse where the maximum bubble compression is attained during the compression cycle of the acoustic wave (Yasui et al. 2008, 2003, 2004, 2005; Merouani et al. 2014b, c, 2015c, 2016, 2020; Merouani and Hamdaoui 2016; Kerboua and Hamdaoui 2017). In this region, the peak temperature is achieved when the minimum bubble radius (Rₘₜₐₜ) is attained. Figure 1a–c shows the evolution of the reactions system and the temperatures inside the bubble as function of time at around the end of the bubble collapse for two ambient bubble radius, R₀ = 3.2 and 6 µm, in the absence [(a) and (b)] and presence [(c) and (d)] of CCl₄ at 5.2 mM in the aqueous liquid. These numerical simulations are established for a frequency of 355 kHz, an acoustic intensity of 1 W/cm² and a liquid temperature of 20 °C.

For an argon bubble (Fig. 1a, b, it is clear that the increase of bubble size from 3.2 to 6 µm causes the maximal bubble temperature (Tₘₜₐₜ) to be decreased from 4036 to 2361 K. This is owing to the fact that when the ambient radius is increased, a huge amount of water vapor enters the bubble during the rarefaction phase; consequently, the bubble collapse is cushioned (mild collapse). As a result, the production of the various species is reduced proportionally to the maximal bubble temperature decrease. The total production of reactive oxygen species (ROS: •OH, HO₂•, O, O₃ and H₂O₂) goes down from 9.33 × 10⁻¹⁸ to 3.01 × 10⁻²¹ mol, when the ambient bubble radius is increased from 3.2 µm (Tₘₜₐₜ = 4036 K) to 6 µm (Tₘₜₐₜ = 2361 K), respectively. On the other hand, the presence of CCl₄ in the liquid phase (5.2 mM) reduces the maximal bubble temperature for both cases of R₀, 3.2 and 6 µm, as clearly shown in Fig. 1c, d. This is due to the endothermal dissociation of carbon tetrachloride and its lower polytropic index cₚ/cᵥ = 1.11, as compared to 1.67 for argon and 1.33 for water vapor. In this case, the increase of ambient bubble radius from 3.2 to 6 µm lowers the maximal bubble temperature from 2699 to 1975 K, respectively. However, in spite of this reduction of temperature for both cases (3.2 and 6 µm, Fig. 1c, d), the overall molar yield of the bubble is increased conversely to the previous case (argon bubble). The total yielding of oxidants [ROS + RCS (•CCl₃, •Cl, Cl₂, HOCl)] increases from 9.1 × 10⁻¹⁸ mol (ROS = 8.98 × 10⁻¹⁷ mol and RCS = 8.21 × 10⁻¹⁶ mol) to 1.17 × 10⁻¹⁵ mol (ROS = 4.13 × 10⁻¹⁷ mol and RCS = 1.13 × 10⁻¹⁵ mol), as the bubble radius goes up from 3.2 µm (Tₘₜₐₜ = 2699 K) to 6 µm (Tₘₜₐₜ = 1975 K), respectively. In addition, the conversion efficiency of CCl₄ goes down from 67.72 to 12.91% for the same increase of bubble size (3.2 to 6 µm, respectively, as calculated from Figs. 1c, d. All these results indicate the important impacts observed when the ambient bubble size is changed especially in the presence of CCl₄ in the bulk liquid. This means that the distribution of active bubbles is certainly affected by the presence of carbon tetrachloride. In the next section, the effect of liquid temperature (20 to 50 °C) on the range of active bubbles for the production of ROS, RCS and ROS + RCS is explored for various CCl₄ concentrations in the bulk solution (0 to 5.2 mM).

**Effect of liquid temperature on the range of active bubbles in the presence of CCl₄**

It should firstly be noted that for the liquid temperatures of 20–50 °C the solubility of CCl₄ is around 55.2 mM (slight difference) (Donald et al. 2006). Therefore, the maximal concentration of CCl₄ is taken as 5.2 mM for the solution temperatures from 20 to 50 °C. Through varying Rₘ Gaussinan curves for the maximum production of ROS, RCS and all the oxidants are obtained for both cases, the absence and presence of CCl₄ in the bulk liquid (i.e., some curves are provided in Fig. 3, which will be discussed later). These trends were already reported by several researches treating the bubble-size distribution in the absence of solutes.
Fig. 1 Evolution of the reactions system inside the bubble as a function of time at around the end of the bubble collapse.

(a) and (b) Argon bubble (0 mM of CCl₄ in solution) for initial radii \( R₀ \) of 3.2 µm and 6 µm, respectively. (c) and (d) Argon bubble in presence of CCl₄ in the solution (5.2 mM) for initial radii \( R₀ \) of 3.2 and 6 µm, respectively. Simulation conditions: frequency: 355 kHz, Acoustic intensity: 1 W cm⁻², Liquid temperature: 20 °C, Static pressure: 1 atm. Vertical axes in (a–d) are in logarithmic scale.

(1) for (a–d)
in the aqueous phase. Table 1 regroups the calculated range of active bubbles for the production of ROS, RCS, and all oxidants (ROS + RCS) for various liquid temperatures (20–50 °C) and CCl₄ concentrations (0 to 5.2 mM). The optimum bubble radii for the maximal production rate of ROS, RCS and all oxidants (ROS + RCS) are given in Fig. 2a–c as a function of CCl₄ concentration (0–5.2 mM) and liquid temperature in the range of 20–50 °C. All these results are calculated for a wave frequency of 355 kHz and Iₙ = 1 W/cm². In Fig. 4a–c, the broadness of active bubbles ranges is given as functions of carbon tetrachloride concentration and liquid temperature. It should be noted that the threshold limit of oxidants production is fixed at 10⁻¹⁷ mol (Table 1, Fig. 3b–d and Fig. 4a–c), conversely to the previous cases (Yasui et al. 2008; Merouani et al. 2013), where this limit is reduced to 10⁻²² mol for water free of solutes. This is due to the presence of CCl₄ inside the bubble, which largely increases its molar yield as well as the threshold limit of production.

Complex behaviors are observed especially when CCl₄ concentration is increased, as clearly seen from Fig. 2a–c and Fig. 4a–c. For an argon bubble (Fig. 2a or c), the optimum bubble size for the maximal production of ROS goes up gradually with the rise of liquid temperature. The increase of the bulk liquid temperature from 20 to 50 °C causes the optimum bubble radius to be raised by 23.1% (2.6–3.2 µm). This interval of optimum radii is in good agreement with the experimental results obtained by Brothie et al. (Brothie et al. 2009), where an optimum of 3.2 µm is obtained at 355 kHz. An opposite behavior has been observed by Merouani et al. (Merouani and Hamdaoui 2016) for the production of hydrogen, where the optimum radius is shifted toward lower radii for the same range of liquid temperature (20 to 50 °C). However, in Merouani et al.’s work transfer phenomena (mass transport, heat exchange and chemical reactions heat) are not included into the bubble internal energy balance conversely to our study. Therefore, a direct comparison of our study to Merouani’s work could not be accurate.

The increase of the optimum bubble radius for an argon bubble (0 mM of CCl₄, Fig. 2a, c) is explained according to the findings of Fig. 3a, b. It should be noted that the production rate of oxidants (ROS or RCS) is presented as normalized quantity (i.e. y-axes of Fig. 3b–d) with respect to the R₀ of the maximum production rate. The use of the normalized scale in Fig. 3a–d is essential for the homogeneity and appearance of curves, because the use of absolute quantities reduces the visibility of some curves (of higher production rates) against those of lower production rates, which could not allow observing (determining) well the optimum radius for the maximum chemical bubble yield. Additionally, the superimposition of the 20 and 50° curves in Fig. 3d did not mean that there is no effect of liquid temperature on the production rate of oxidants (because the normalized production is used); the effect exists but it was hidden by the use the normalized scale.

At 20 °C (Fig. 3a), the maximal bubble temperature achieved at the collapse is increased proportionally with the rise of vapor amount (violent collapse) until the bubble radius of 2.5 µm where more increase of vapor quantity causes the bubble temperature to be reduced. This is due to the molar heat of water vapor and its endothermal dissociation, which reduce the violence of collapse. However, the production rate of ROS continues to go up to a radius of 2.7 µm, where after that the formation rate of these species and bubble temperature decreased with the rapid increase of vapor amount inside the bubble (Fig. 3a, b). The same behavior is observed at 50 °C (Fig. 3a, b, however, at this
temperature more bubbles are activated (see also the range results in Table 1, case of 0 mM CCl₄), due to the increase of vapor amount housed at collapse for this population of bubbles. Consequently, the maximal temperature is shifted toward \( R_0 \) equaling to 1.5 µm. For larger bubbles (Fig. 3a), the bubble temperature is slightly increased with the rise of vapor amount (increase of vapor pressure) at the liquid temperature of 50 °C. This is because at 1 W/cm², the increase of vapor quantity inside the bubble increases the violence of collapse (\( T_{\text{max}} \) goes up). Nevertheless, this increase of cavitation temperature is very dependent on the vapor amount housed inside the bubble at the end of compression phase. Consequently, bubble temperature in this region (larger bubbles) is not highly increased. On the other hand, the production rate of ROS at 50 °C is increased until the bubble radius of 3.2 µm (Fig. 3b), where further increase of vapor amount (Fig. 3a) reduces the production rate of ROS. Therefore, this optimum values of \( R_0 = 3.2 \) µm result from the competition between bubble temperature and the amount of vapor found into the bubble at collapse. In addition, in this case (50 °C), the range of active bubbles (for 0 mM of CCl₄) is broadened (Table 1, Fig. 3b and Fig. 4a) in spite that bubble temperature for this case (50 °C) is lower or slightly higher than the case of 20 °C (Fig. 3a). This is because at the liquid temperature of 50 °C, the vapor pressure increases within the acoustic cavitation; therefore, the overall molar yields of bubbles increases. A similar behavior is observed for the optimum size and the range of active bubbles when CCl₄ concentration goes up (Table 1, Fig. 2a,b and Fig. 4a,b). However, in this case the optimum radius and the width of active bubbles range are hardly increased for higher CCl₄ concentrations compared to the argon bubble case (Table 1). All these modifications in bubble behavior are owing to the presence of CCl₄ within the cavitation. On the other hand, at low concentrations of carbon tetrachloride (≤ 0.01 mM for RCS (Fig. 4b) and ≤ 0.1 mM for ROS (Fig. 4a), the widthness of active bubbles range is rapidly increased, because of the presence of CCl₄ inside the bubble, which increases the production rate of the oxidants in spite of the decrease of the maximum bubble temperature (discussed in the previous section). Consequently, the upper and the lower limits of the activated bubbles range (for both ROS and RCS) are extended proportionally with the rise of the liquid temperature which increases CCl₄ mole fraction inside the bubble (see Table 1 and Fig. 4a,b). This results in a gradual increase of the optimum bubble radius for the maximal production of ROS and RCS (Fig. 2a, b). Approximately, the same response is obtained at the CCl₄ concentration of 0.01 mM compared to the argon bubble case (Fig. 2b). As the concentration of CCl₄ goes up (≥ 0.01 mM for RCS (Fig. 4b) and 0.1 mM for ROS (Fig. 4a)), the widthness of active bubbles range (for oxygen and reactive chlorine species) is hardly increased (Table 1, Fig. 3c, d). For the reactive

| CCl₄ concentration | ROS | RCS | ROS + RCS |
|-------------------|-----|-----|----------|
| 0 mM (Argon-bubble) | 2.1–3.1 | 1.43–4.15 | 0.98–6.0 |
| 0.01 mM | 2.1–3.2 | 1.42–4.5 | 0.98–6.0 |
| 0.1 mM | 1.95–5.5 | 1.22–7.6 | 0.97–7.7 |
| 1 mM | 1.6–6.75 | 1.25–7.26 | 0.91–7.5 |
| 3 mM | 1.45–6.8 | 1.2–7.26 | 0.91–7.5 |
| 5.2 mM | 1.45–6.8 | 1.2–7.26 | 0.91–7.5 |

Simulation conditions: frequency = 355 kHz, Acoustic intensity \( I_n = 1 \) W cm⁻², static pressure \( P_\infty = 1 \) atm.
Fig. 2  Optimum ambient bubble radii ($R_0$) for the maximal production rate of a ROS (•OH, O, O$_2$, HO$_2^*$ and H$_2$O$_2$), b RCS (•CCl$_3$, •CCl$_2$, •Cl, Cl$_2$, HOCl) and c all oxidant (ROS + RCS) as function of liquid temperature (20–50 °C) and CCl$_4$ concentration (0–5.2 mM). Simulation conditions: frequency: 355 kHz, Acoustic intensity: 1 W cm$^{-2}$, Static pressure: 1 atm. Vertical axes in (a)–(d) are in logarithmic scale.
Fig. 3  

(a) Evolution of water vapor amount and the maximum bubble temperature at the collapse for an argon bubble (0 mM of CCl₄ in solution) as a function of ambient bubble radius (R₀) for two liquid temperatures (20 and 50 °C). 

(b) Normalized production rate of ROS (*OH, O, O₃, HO₂* and H₂O₂) inside an argon bubble (0 mM of CCl₄ in solution) as a function of ambient bubble radii for two liquid temperatures (20 and 50 °C). 

(c) Normalized production rate of ROS for CCl₄-Ar-bubble (5.2 mM of CCl₄ in solution) as a function of ambient bubble radii for two liquid temperatures (20 and 50 °C). 

(d) Normalized production rate of RCS (*CCl₃, CCl₂, *Cl, Cl₂, HOCl) for CCl₄-Ar-bubble (5.2 mM of CCl₄ in solution) as a function of ambient bubble radii for two liquid temperatures (20 and 50 °C). 

Simulation conditions: frequency: 355 kHz, Acoustic intensity: 1 W cm⁻², Static pressure: 1 atm. The reported production rates have been normalized with respect to the maximal yielding obtained for each case (reactive oxygen and chlorine species).
chlorine species, this width becomes approximately constant and independent of the liquid temperature and CCl₄ concentration (Fig. 4b and Fig. 3d). This is due to the rise of CCl₄ concentration, which limits the extension of the upper and the lower bounds of oxidants’ production range, where this effect is more intense for the RCS than for ROS (Fig. 4a, b). In fact, as the concentration of CCl₄ inside the bubble goes up, the maximal temperature of cavitation at collapse and the CCl₄ conversion are reduced (see previous section), which implies the generation of the reactive chlorine species as well as the broadness of the active bubbles range could be limited. However, the formation of ROS is not highly affected even at lower bubble temperatures (see previous section) due to the reaction of CCl₄ molecules with hydrogen atoms (reaction 23 of Table S2), which increases the yield of hydroxyl radicals (and so of ROS). Approximately the same trend as in Fig. 4a, b is observed for the optimum bubble radii either for RCS or for ROS, as shown in Fig. 2a, b.

In general, for CCl₄ concentration equal to or lower than 0.1 mM for liquid temperature from 20 to 50 °C, it is observed that the optimum ambient bubble radii for ROS are localized at around ~3 µm, conversely to the RCS, which are situated at around ~6 µm (Fig. 2a, b). This is because at this level ([CCl₄] ≤ 0.1 mM), the effect of CCl₄ concentration on the bubble sonoactivity is not enough to control the range at which the maximal production rate of ROS appears (Fig. 2a, b). For higher CCl₄ concentrations (> 0.1 mM), the effect of CCl₄ on the bubble sonoactivity is increased; consequently, the broadness of active bubbles range is reduced (Fig. 4a, b), due to the lower polytropic index \( \frac{c_p}{c_v} \) of CCl₄ (1.11 compared to 1.67 for argon and 1.33 for water vapor) and its higher endothermal reaction. As a result, the optimum radii for the maximal production rate of ROS appear approximately at around the same range (~4.3–5.1 µm) as that for the ROS (~4.9–5.3 µm) (Fig. 2a, b).

In order to get the overall view of the liquid temperature effect on the range of active bubbles (and optimum radius) in the presence of CCl₄, we refer ourselves to Figs. 2c and 4c. These figures (total oxidants) show the combined impacts of the previous cases of Fig. 2a, b and Fig. 4a, b. Figure 4c shows that when CCl₄ concentration is equal to or lower than 0.1 mM, the broadness of active bubbles range (CCl₄ concentration is fixed) increases proportionally with the rise of liquid temperature. However, the increase of CCl₄ concentration in solution amortizes this width increase over the same range of liquid temperature (20 to 50 °C). All these are explained by the effect of CCl₄ on the active bubble range (already discussed). For the higher concentrations of CCl₄ (> 0.1 mM), the broadness of the active bubbles range becomes approximately constant and independent of the liquid temperature and carbon tetrachloride concentration as it is discussed previously for the case of reactive chlorine species. In Fig. 2c, it is clear that the optimum radius for the maximal production of the total oxidants is RCS yielding dependent. This dominance is obtained for all CCl₄ concentrations (with the exception of the argon bubble and the case of CCl₄ concentration of 0.01 mM, which give the same response). This conclusion reveals the importance of RCS production compared to that of ROS. However, because no experimental works are available in the literature concerning the effect of liquid temperature on the variation of active bubbles population, the comparison cannot be made to support our results. On the other hand, the present results are in line with several experimental works, indicating that the rapid elimination of different nonvolatile organic contaminants (e.g., phenol, bisphenol A, 2,4-dinitrophenol, methyl orange, C.I. acid orange 8, etc.) in the presence of CCl₄.

![Fig. 3 (continued)](image-url)
Fig. 4 Broadness of active bubbles range (according to Table 1) for the maximal production rate of \( \text{a} \) ROS (\( *\text{OH}, \text{O}, \text{O}_2, \text{HO}_2^* \) and \( \text{H}_2\text{O}_2 \)), \( \text{b} \) RCS (\( \text{CCl}_3, \text{CCl}_2, *\text{Cl}, \text{Cl}_2, \text{HOCl} \)) and \( \text{c} \) all oxidant (ROS + RCS) as a function of liquid temperature (20–50 °C) and \( \text{CCl}_4 \) concentration (0–5.2 mM). Simulation conditions: frequency: 355 kHz, Acoustic intensity: 1 W cm\(^{-2}\), Static pressure: 1 atm.

contaminants is due to the co-attack of RCS in addition to hydroxyl radicals (Wang et al. 2007; Guo et al. 2008; Gültekin et al. 2009; Merouani et al. 2010).

In light of the previous findings and discussion, let now answering to the problematic made in the last paragraph of the introduction concerning the third suggested mechanism by which CCl₄ improves the degradation of nonvolatile organic pollutants in aqueous solutions. We have proposed that in addition to the RCS generation and H⁺-scavenger roles played by the CCl₄ sonolysis, CCl₄ can broaden the size of active bubbles for the production of oxidants, which implies a drastic increase of the number of active bubbles in the reacting medium. With a simple regard to the results of Table 1, we can confirm that our suggestion is highly correct and the liquid temperature rise could increase more the wideness of R₀ for the production of the oxidants. Therefore, the increase of the number of active bubbles, imposed by the CCl₄ sonopyrolysis, is without doubt another mechanism that could be imperatively involved for clarifying the intensification aspect of CCl₄ toward the sonochemical degradation of nonvolatile organic contaminants.

**Conclusion**

In this paper, we attempted to reveal the impact of liquid temperature on the range of active bubbles in the presence of carbon tetrachloride in the bulk liquid. First, it has been found that the presence of CCl₄ inside the bubble modifies completely the sonoactivity of this acoustic cavitation, where this behavior is observed for the whole range of bubbles. It was demonstrated that the decomposition of CCl₄ inside the bubble reduces its maximal temperature, conversely to its total production, where it was found that the overall molar yield of bubble increased even when bubble temperature is decreased, especially for larger bubble radii. In addition, this reduction of bubble temperature (T_max) is accompanied by the decrease of CCl₄ conversion (%). On the other hand, it has been demonstrated that, when CCl₄ concentration in the solution is equal to or lower than 0.1 mM, the wideness of active bubbles range for total oxidants yield increased proportionally with the rise of liquid temperature from 20 to 50 °C. However, the increase of CCl₄ concentration amortizes this width increase over the same range of liquid temperature (20–50 °C). At higher concentrations of CCl₄ (>0.1 mM), the broadness of the active bubbles range becomes approximately constant and independent of the liquid temperature and carbon tetrachloride concentration. Moreover, it was found that the optimum radii (~5 µm) for the maximal production of the total oxidants are controlled by the yield of the reactive chlorine species. This behavior is observed for all CCl₄ concentrations with the exception of the argon bubble and the case of CCl₄ concentration of 0.01 mM, which have the same response (2.5–3 µm). This work reveals the importance of RCS production compared to ROS, especially for higher concentrations of CCl₄ in the liquid phase. More interestingly, the most important finding of the present study is that the CCl₄ sonopyrolysis could increase the number of active bubbles, and therefore, this aspect could be without doubt another mechanism that could be implicated for clarifying the intensification aspect of CCl₄ toward the sonochemical degradation of nonvolatile pollutants.

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**Declarations**

**Conflict of interest** All authors declare that they have no conflict of interest.

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