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Comprehensive experimental and numerical investigations of the effect of frequency and acoustic intensity on the sonolytic degradation of naphthol blue black in water

Hamza Ferkous [a], Slimane Merouani [a,b,*], Oualid Hamdaoui [a], Yacine Rezgui [c], Miloud Guemini [c]

[a] Laboratory of Environmental Engineering, Department of Process Engineering, Faculty of Engineering, Badji Mokhtar – Annaba University, P.O. Box 12, 23000 Annaba, Algeria
[b] Department of Chemical Engineering, Faculty of Pharmaceutical Engineering Process, University of Constantine 3, Constantine, Algeria
[c] Laboratory of Applied Chemistry and Materials Technology, University of Oum El-Bouaghi, P.O. Box 358, 04000 Oum El Bouaghi, Algeria

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ABSTRACT

In the present work, comprehensive experimental and numerical investigations of the effects of frequency and acoustic intensity on the sonochemical degradation of naphthol blue black (NBB) in water have been carried out. The experiments have been examined at three frequencies (585, 860 and 1140 kHz) and over a wide range of acoustic intensities. The observed experimental results have been discussed using a more realistic approach that combines the single bubble sonochemistry and the number of active bubbles. The single bubble yield has been predicted using a model that combines the bubble dynamics with chemical kinetics consisting of series of chemical reactions (73 reversible reactions) occurring inside an air bubble during the strong collapse. The experimental results showed that the sonochemical degradation rate of NBB increased substantially with increasing acoustic intensity and decreased with increasing ultrasound frequency. The numerical simulations revealed that NBB degraded mainly through the reaction with hydroxyl radical ($^{\cdot}$OH), which is the dominant oxidant detected in the bubble during collapse. The production rate of $^{\cdot}$OH radical inside a single bubble followed the same trend as that of NBB degradation rate. It increased with increasing acoustic intensity and decreased with increasing frequency. The enhancing effect of acoustic intensity toward the degradation of NBB was attributed to the rise of both the individual chemical bubble yield and the number of active bubbles with increasing acoustic intensity. The reducing effect of frequency was attributed to the sharp decrease in the chemical bubble yield with increasing frequency, which would not compensated by the rise of the number of active bubbles with the increase in ultrasound frequency.

1. Introduction

Water pollution due to discharge of colored effluents from textile dye manufacturing is one of the major environmental concerns in today's world. Strong color imparted by dyes to the receiving aquatic ecosystems poses aesthetic problems as well as serious ecological problems, such as carcinogenicity and inhibition of benthic photosynthesis [1]. Azo dyes have been widely used as colorants in a variety of products such as textiles, paper and leather. Approximately half of all known dyes are azo dyes, making them the largest group of synthetic colorants [2]. These chemicals present a potential human health risk as some of them have been shown to be carcinogenic [3]. Traditional methods for dye removal include biological treatment [4,5], coagulation [6,7], filtration [8] and adsorption [9–11]; however, because of high dye concentrations and the increased stability of synthetic dyes, these methods are becoming less effective for the treatment of colored industry effluents. To overcome the problems associated with these traditional methods, much attention has recently been focused on the so-called advanced oxidation processes for water and wastewater decontamination. In these processes various techniques (e.g. photocatalysis, Fenton reaction, UV/H_2O_2) are applied to produce reactive species [12–14], principally, hydroxyl radicals ($^{\cdot}$OH), which are able to induce the degradation and mineralization of organic pollutants [15].

A new way of generating of $^{\cdot}$OH radials is the application of ultrasound in the frequency range 20–1000 kHz, in which case important sonochemical effects can be observed [16,17]. Application of ultrasound to aqueous solutions induces the

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* Corresponding author at: Department of Chemical Engineering, Faculty of Pharmaceutical Engineering Process, University of Constantine 3, Constantine, Algeria. Tel./fax: +213 38876560.
E-mail addresses: s.merouani@yahoo.fr, s.merouani03@gmail.com (S. Merouani).

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Nomenclature

c	speed of sound in the liquid medium (m s^{-1})
f	frequency of ultrasonic wave (Hz)
I_a	acoustic intensity of ultrasonic irradiation (W m^{-2})
I	number of chemical reactions
$k_{NBB\text{-}OH}$	Liquid rate constant of NBB with $^{\cdot}$OH
p	pressure inside a bubble (Pa)
P_A	amplitude of the acoustic pressure (Pa)
P_{g0}	initial gas pressure (Pa)
P_v	vapor pressure of water (Pa)
$p_s(t)$	acoustic pressure
p_∞	ambient static pressure (Pa)
R	radius of the bubble (m)
R_{max}	maximum radius of the bubble (m)
R_{min}	minimum radius of the bubble at the collapse (m)
R_0	ambient bubble radius (m)
t	time (s)
T	temperature inside a bubble (K)
T_∞	ambient liquid temperature (K)
X	symbol of chemical species

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)

formation of vapor and gas-filled microbubbles that grow and then adiabatically collapse causing temperatures of about 5000 K and pressures in excess of 500 atm within the bubbles [18,19]. These extremely conditions inducing the pyrolysis of water vapor, oxygen molecules and volatile organic compounds that can be present in the gas phase [18]. Pyrolysis of water vapor yields hydroxyl radicals ($^{\cdot}$OH) and hydrogen atoms (H$^{\cdot}$) [19]. These active species can recombine, react with other gaseous species present in the cavity to form other active species such as $HO_2^{\cdot}$ and O, or diffuse out of the bubble into the bulk liquid medium [20]. Hydroxyl radicals in particular are very reactive and can transform dissolved organic compounds [21]. Reactions can take place in the gas phase, at the gas–liquid interface and in the solution bulk after transfer of gaseous radicals into the liquid phase [20]. Accordingly, the sonochemical degradation of an organic compound can be influenced by its physicochemical properties. An organic pollutant with high volatility character will be incinerated in the bubble whereas a hydrophilic or hydrophobic compound with low volatility cannot enter the bubble, but will be oxidized in the bulk solution or interfacial area by reaction with $^{\cdot}$OH radicals [16,18].

The most crucial parameters for application of sonolysis are the employed frequency and power. Although the effect of these two parameters on the sonochemical degradation of organic pollutants has been widely examined [22–27], comprehensive studies on the effects of frequency and power are yet to be carried out. The present study provides a comprehensive experimental and numerical investigation of the effects of frequency and acoustic intensity on the sonochemical degradation of organic matters. Naphthol blue black (NBB), an acidic dye of the diazo class, was chosen as substrate model. NBB is widely used in the textile industry for dyeing wool, nylon, silk and textile printing [28]. It is characterized by a high photo- and thermal stability [28]. The experiments have been examined at three frequencies (585, 860 and 1140 kHz) and over a wide range of acoustic intensities. The observed experimental results have been discussed with respect to single bubble sonochemistry and the number of active bubbles. The chemical bubble yield has been predicted using a model that combines the bubble dynamics with chemical kinetics consisting of series of chemical reactions (73 reversible reactions) occurring inside a reactive cavitating bubble.

2. Materials and methods

2.1. Reagents

Naphthol blue black (abbreviation: NBB; Acid Black 1; C.I. number: 1064-48-8; chemical class: azo dye; molecular formula: $C_{22}H_{14}N_6Na_2O_9S_2$, molecular weight: 616.49 g mol^{-1}) was supplied by Sigma–Aldrich and used without any purification. The molecular structure of NBB was shown in Fig. 1.

2.2. Ultrasonic reactor

Sonolysis experiments were conducted in a cylindrical water-jacketed glass reactor (Fig. 2). The ultrasonic waves at 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 generator that aliments the multifrequency transducer operates at various electrical powers that are indicated by a fixed three positions, namely, amp 2, amp 3 and amp 4. The temperature of the solution was monitored using a thermocouple immersed in the reacting medium. Acoustic power dissipated in the reactor was estimated under different conditions using a standard calorimetric method [29,30].

2.3. Procedures

All NBB solutions were prepared with distilled water. Sonochemical experiments were carried out under different conditions using constant solution volume of 300 mL. Aqueous samples were taken periodically from the solution and the concentrations of the dye were determined using a UV–visible spectrophotometer (Lightwave II) at 620 nm. The temperature of the sonicated solution was kept at 25 °C by circulating cooling water through a jacket surrounding the cell. In all the cases, the reactor was open to air and the experiments were carried out at natural pH (pH ~ 6).

3. Theoretical package

The theoretical model used for studying the single bubble sonochemistry at different conditions of frequency and acoustic intensity has been fully described in our previous papers [31,32]. The model combines the dynamic of single bubble with chemical kinetics occurring inside a bubble during the collapse. The following is a brief description of the model.

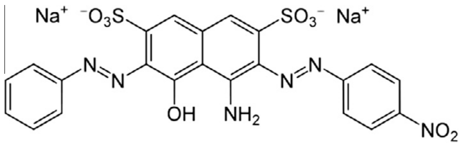

Fig. 1. Molecular structure of NBB.