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Sonolytic degradation of naphthol blue black at 1700 kHz: Effects of salts, complex matrices and persulfate

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ABSTRACT

In the present work, the effect of salts (NaCl and Na_2SO_4), complex matrices (natural water and seawater) and persulfate ions on the sonochemical degradation of an acidic diazo dye, naphthol blue black (NBB), in water at 1700 kHz was clarified. Additionally, the influence of initial dye concentration and liquid temperature on the extent of NBB sonolytic removal was investigated. It was found that the NBB degradation rate increased with increasing the initial substrate concentration in the range 3–15 mg L^{-1}. Significant enhancements in the NBB degradation rates were observed at high liquid temperatures: 96.5% of NBB was removed at 65 °C after 45 min of sonication while only 51% was reached at 25 °C. The presence of salts (NaCl or Na_2SO_4) has a beneficial effect on NBB degradation. The degradation rate of NBB in the presence of 0.5 and 1 M NaCl was 3–6 times higher than that obtained without salt. Interestingly, faster degradation rates of NBB were observed in complex matrices. The complete destruction of NBB required 40 min in seawater and 60 min in natural water instead of 210 min in distilled water. The addition of persulfate (PS) ions improved substantially the sonochemical degradation of NBB through the ultrasonic (US) generation of sulfate radical ($SO_4^{\bullet-}$), which is more efficient toward the degradation of NBB. The system US/PS resulted in 100% NBB removal within only 60 min of ultrasonic treatment.

1. Introduction

Numerous industries (textile, paper, plastic, food. . .) are major consumer of water and use organic dyes (soluble or pigments) to color their products. These synthetic dyes may be at the same time toxic and responsible for water coloring [1]. Indeed, some dyes are fixed more or less efficient despite the use of fixing agents. The regulations in term of wastewater discharge are also more severe and require manufacturers to treat their effluents. Besides, the majority of synthetic dyes is not biodegradable and can constitute risk factors for our health and environment [1]. Particularly, coloring problems have a significant psychological impact on the population. A colored effluent is perceived by the public as pollutant and dangerous. Approximately half of all known dyes are azo dyes, making them the largest group of synthetic colorants used in industrial scale [2]. These chemicals present a potential human health risk as some of them have been shown to be carcinogenic [2]. Biological and physicochemical treatments and their various combinations are the most used methods for the elimination of dyes from polluted industrial wastewater [1]. The toxicity of azo dyes put, usually, the problem of growth of microorganisms, thereby limiting the effectiveness of the elimination [3]. Physicochemical treatments cannot in most cases suffice to eliminate the organic load contained in these effluents (hard COD) [3]. They are generally based on the adsorption, coagulation/flocculation, precipitation. . .etc [4]. To solve these problems, new techniques have been developed whose aim is to oxidize the non-biodegradable organic matters. Among them, much attention has recently been focused on the so-called advanced oxidation processes (AOPs). In these processes, various techniques (photolysis, photocatalysis, Fenton reaction, UV/H_2O_2, . . .) are applied to produce reactive species, principally, hydroxyl radicals ($^{\bullet}OH$), which are able to induce the degradation and mineralization of water-dissolved organic pollutants [5,6].

A new way of generating $^{\bullet}OH$ radials is the application of ultrasound in which important chemical effects can be observed [7,8]. Application of ultrasound to aqueous solutions induces the formation of vapor and gas-filled microbubbles that grow, undergo a series compression-expansion cycles and then adiabatically collapse causing temperatures of about 5000 K and pressures in excess of 1000 atm therein [9,10]. These extremely conditions inducing,

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inside the bubbles, the pyrolysis of water vapor, oxygen molecules and volatile organic substrates that can be present in the gas phase [8]. Pyrolysis of water vapor yields hydroxyl radicals (•OH) and hydrogen atoms (H•) and with other gases present in the bubble, other active species such as O atoms and HO_2• radicals may create from H_2O and O_2 dissociation and their associate reactions [11,12]. Hydroxyl radicals in particular are very reactive and can transform organic compounds [13,14]. Reactions with free radicals can take place in the gas phase, at the gas-liquid interface and in the bulk solution after transfer of gaseous content into the liquid phase [8]. Accordingly, the sonochemical degradation of an organic compound can be occurred in gas-phase pyrolysis and oxidation for volatile substrates and by reaction with •OH radicals at the gas-liquid interface and in the aqueous phase for nonvolatile substrates [15].

The Mega-range high frequency ultrasonic waves was reputed ineffective for sonochemistry. However, recent papers published by Ghodbane and Hamdaoui [16,17] showed experimentally that 1700 kHz is more effective for sonochemistry than the frequently used low frequency (~20 kHz). Ghodbane and Hamdaoui [16] reported that the degradation rate of acid blue 25 at 1700 kHz was 3.5 times higher than that obtained at 22.5 kHz.

In this study, naphthol blue black (NBB), was taken as substrate model for systematic study of sonochemical process. NBB is an acidic diazo dye, which has a high photo- and thermal stability [18]. It is widely used in the textile industry for dyeing wool, nylon, silk and textile printing [18]. Other industrial use includes coloring of soaps, anodized aluminum and casein, wood stains and writing ink preparation [18]. The objective of this study was to assess the potentiality of ultrasound at 1700 kHz to remove NBB from water and to evaluate the effects of some inorganic species such as salts and persulfate on the sonolytic degradation of the dye. The degradation of NBB in natural water and seawater was investigated because various matrix components may noticeably affect the efficiency of ultrasonic treatment.

2. Materials and methods

2.1. Materials

Naphthol blue black (abbreviation: NBB; CAS 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.

Potassium iodide (KI) was supplied by Riedel-de Haën. Ammonium heptamolybdate $(NH_4)_6Mo_7{\cdot}4H_2O$, potassium persulfate, sodium chloride (NaCl) and sodium sulfate (Na_2SO_4) were supplied by Sigma–Aldrich (analytical grade).

2.2. Reactor

Sonolysis experiments were conducted in cylindrical water-jacketed glass reactor (Fig. 2). The ultrasonic irradiations were emitted from the bottom of the reactor through a piezoelectric disc (diameter 2 cm) operating at 1700 kHz. The temperature of the solution was monitored using a thermocouple immersed in the reacting medium. The reactor operates with only one electric power. Acoustic power dissipated in the reactor (14 W) was estimated using the calorimetric method [19].

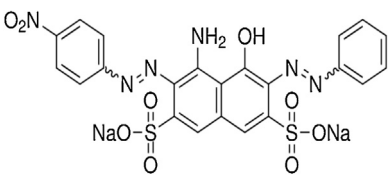

Fig. 1. Chemical structure of naphthol blue black.

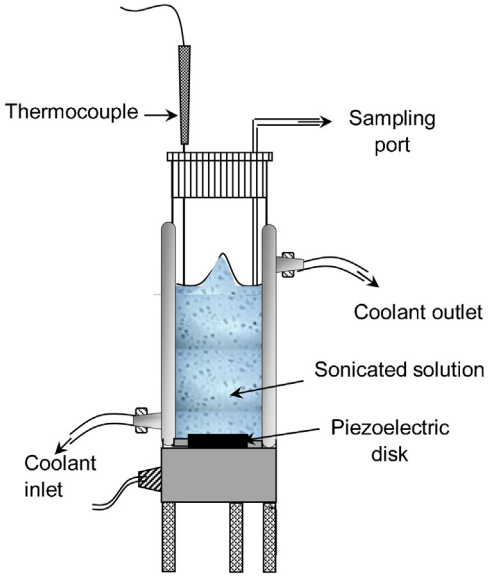

Fig. 2. Scheme of the ultrasonic reactor.

2.3. Procedures

All NBB solutions were prepared with distilled water. Sonochemical experiments involving NBB degradation and H_2O_2 production in pure water were carried out under different conditions using constant solution volume of 100 mL. The temperature of the sonicated solution was kept at desired value by circulating cooling water through a jacket surrounding the cell. The degradation of NBB during the reaction period may be assessed by absorption abatements at two bands [20]: 620 nm that is responsible for the chromophoric group of the dye and 322 nm, which represents the absorption of the aromatic derivatives rings. However, to avoid interference between NBB and its intermediate products, which absorb in the UV region, the degradation of the dye was followed in the visible region at 620 nm.

Hydrogen peroxide concentrations in water were analytically determined using the iodometric method [21].

3. Theoretical procedure

The theoretical procedure used for characterizing the 1700-kHz ultrasonic cavitation field in term of bubble dynamics, bubble temperature, production of oxidizing species and the number of active bubbles is well described in our previous work [22]. The following is only a brief description.

3.1. Single bubble sonochemistry model

This model combines the dynamic of single bubble in acoustic field propagated in water with chemical kinetics occurring inside a bubble during the oscillation. 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 [23]. The radial dynamics of the bubble is described by the Keller–Miksis equation that includes first order