



Enhanced sonochemical degradation of bisphenol-A by bicarbonate ions

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ABSTRACT

Sonochemical elimination of organic pollutants can take place through two degradation pathways. Molecules with relatively large Henry's law constants will be incinerated inside the cavitation bubble, while nonvolatile molecules with low Henry's law constants will be oxidised by the OH[•] ejected from the bubble of cavitation. Taking bisphenol-A as a model pollutant, this study points out an alternate degradation route, mediated by bicarbonate ions, which is significant for the elimination of micro-pollutants at concentrations present in natural waters. In this process, OH[•] radicals react with bicarbonate ions to produce the carbonate radical, which, unlike the OH[•] radical, can migrate towards the bulk of the solution and therefore induce the degradation of the micro-pollutants present in the bulk solution. As a consequence, initial degradation rate is increased by a factor 3.2 at low concentration of bisphenol-A (0.022 μmol l⁻¹) in presence of bicarbonate in water.

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1. Introduction

Since 1954, experimental studies have shown that the propagation of an ultrasonic wave in water can lead to the elimination of organic pollution [1–3].

Ultrasonic treatment is considered as an emerging Advanced Oxidation Process (AOP) because it generates OH[•] radicals through-out acoustic cavitation that induces the homolytic scission of a molecule of water [4].

Acoustic cavitation is defined as the cyclical formation, growth, and collapse of micro-bubbles [5]. The rapid collapse of the bubbles adiabatically compresses gas and entrapped vapor, producing small local hot spots [6,7]. At the final step of the bubble collapse, the temperature inside the residual bubble is thought to exceed 2000 K [8]. The chemistry involved in the destruction of organic pollutants is not identical for all organic contaminants. The chemical pathway and the rate of elimination depend on the volatility of the pollutant molecule. A molecule with a large Henry's constant will be incinerated inside the bubble of cavitation, while a nonvolatile molecule with a low Henry's constant will be oxidised by the OH[•] ejected from the cavitation bubble [9–11].

Even if the elimination of organic pollutants upon ultrasonic action occurs with high efficiency, the application of this technology to real natural and waste waters would pose the additional problem of the potential effects of inorganic ions. Several investigations have studied sonochemical degradation processes in the presence of inorganic species have recently reported that sonochemical degradation of dyes and bisphenol-A can be enhanced in the presence of inorganic ions [12,13]. In contrast, in the case of *p*-nitrophenol [14] and also in the case of bisphenol-A [15], the studies have reported that degradation rates are not significantly affected by inorganic ions. Therefore, the influence of inorganic species on the sonochemical degradation of organic compounds remains controversial.

Because of their usual presence in industrial and natural waters, the impact of carbonate and/or bicarbonates ions on the performance of water treatments has been widely investigated. The negative effect of carbonate/bicarbonate ions on several AOPs: TiO₂ photocatalysis [16,17], photo-Fenton [18] and UV/O₃ [19] is well known.

The aim of this work is to examine the effect of bicarbonate ions on the sonochemical treatment of organic pollutants in water. Bisphenol-A (BPA) was selected as the pollutant model because BPA, a compound of the plastics industry, exhibits an endocrine disrupting effect (EDC) at very low (μg-ng) levels [20,21].

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2. Experimental

2.1. Reagents

Bisphenol-A and sodium hydroxide were obtained from Sigma-Aldrich (St. Louis, MO). Sulfuric acid, sodium sulfate, sodium chloride, sodium phosphate, sodium bicarbonate were supplied by Acros Organics (New Jersey, USA). Acetonitrile (HPLC gradient grade), was purchased from Fisher Scientific (Loughborough, UK). All chemicals were used without further purification. Deionized water, obtained with activated carbon and ion exchange resins from Fisher Bioblock Scientific (Illkirch, France), was used throughout the study for the preparation of aqueous solutions, and as a component of the mobile phase in analysis by high performance liquid chromatography (HPLC).

2.2. Apparatus

Experiments were performed in a cylindrical water-jacketed glass reactor with a Teflon holder. Ultrasonic waves (300 kHz, 80 W) were emitted from the bottom of the reactor through a piezo-electric disc (diameter 4 cm) fixed on a Pyrex plate (diameter 5 cm) [22]. Energy dissipated in the reactor was measured by the calorimetric method [23]. The reactor was sampled periodically (1 ml aliquots) for analysis.

2.3. Analyses

Quantitative analysis of the parent compounds was performed by HPLC using a Waters Associates 590 instrument equipped with a Supelcosil LC-18 column (ID = 4.6 mm, length = 250 mm). Sample injections were achieved with a Rheodyne injection system equipped with a 20 μ l sample loop for BPA concentrations above 1 mg/l and a 200 μ l loop for the lowest concentrations. Detection of absorbing organic compounds was realised with a UV detector (model 440) set at 190 nm. The mobile phase, water/acetonitrile (50/50, v/v), was run in an isocratic mode. Using this methodology, a detection limit of 0.002 μ mol l⁻¹ of BPA was reached. Identification of primary BPA intermediates was made by HPLC/MS through a Hewlett Packard series 1100 MSD with electrospray ionization.

3. Results and discussion

3.1. Influence of bicarbonate ion concentration on BPA sonolysis at two concentrations of substrate

Application of 300 kHz and 80 W ultrasound irradiation to 300 ml of deionised water BPA solution (300 μ M) caused an exponential decrease of the BPA concentration with time. Substrate elimination reached 55% of the initial concentration after 15 min irradiation (Fig. 1).

Addition of bicarbonate ion, in the range 12–500 mg l⁻¹, for an identical pH (pH 8.3) had no significant effect on the degradation rate of BPA. At lower BPA concentrations (0.12 μ M), however, the addition of bicarbonate had a significant concentration dependant effect on the rate of degradation of BPA (Fig. 2). Unexpectedly, higher bicarbonate concentration produced higher initial rates of

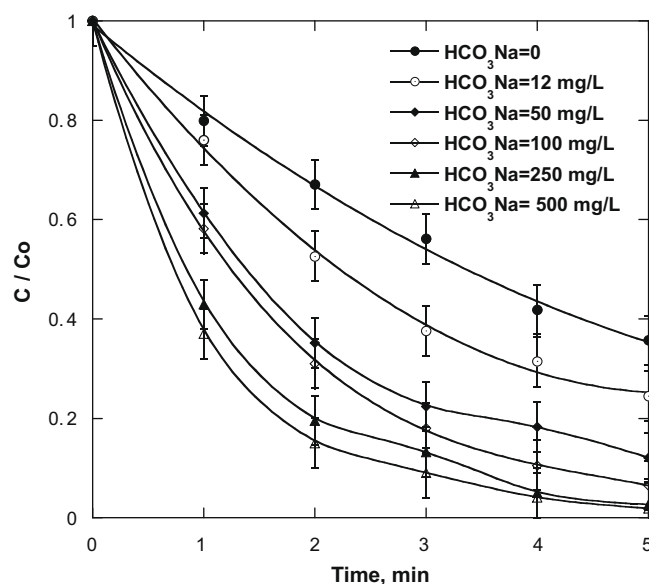


Fig. 2. The effect of bicarbonate concentration on the initial rate of BPA sonochemical degradation. (initial BPA concentration 0.12 μ M; 300 kHz; 300 ml; 80 W; pH 8.3; $T = 21^\circ\text{C}$).

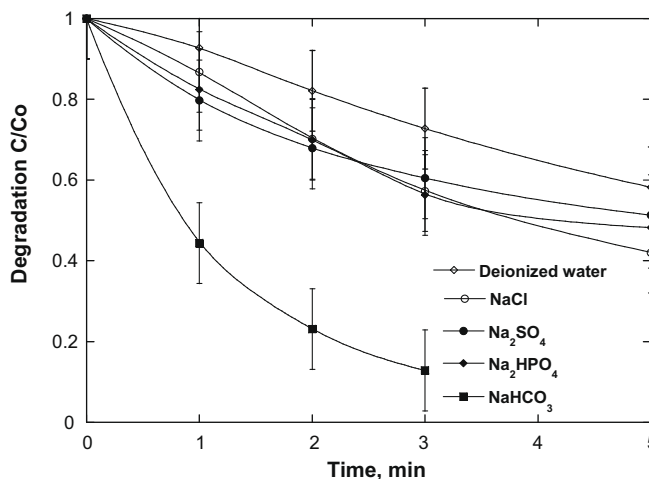


Fig. 3. The effect of inorganic ions on the rate of BPA sonochemical degradation. (initial BPA concentration 0.12 μ M; 300 kHz; 300 ml; 80 W; pH 8.3; $T = 21^\circ\text{C}$ in absence or presence of 6 mM inorganic salts NaCl, Na_2SO_4 , Na_2HPO_4 , or NaHCO_3).

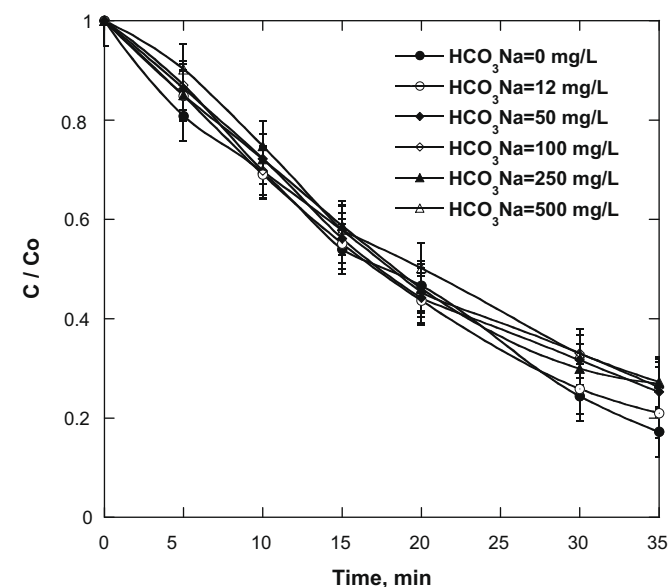


Fig. 1. The effect of sodium bicarbonate on the sonochemical degradation of BPA over time. (initial BPA concentration 300 μ M; 300 kHz, 300 ml, 80 W, pH 8.3, $T = 21^\circ\text{C}$).

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