

Effects of operating parameters on sonochemical decomposition of phenol

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Abstract

Ultrasonic removal of phenol under irradiation at 20, 300 and 520 kHz was investigated to assess the impacts of operating parameters on the efficiency of the systems. It was found under our experimental conditions that 20 kHz was the least effective frequency for ultrasonic decomposition of phenol, owing to the low volatility of phenol and the slow rate of OH radical ejection to the bulk solution at this frequency. Assessment of relative rates of destruction and ultrasonic yields showed that maximum efficiency was accomplished with 300 kHz employed in a reactor enclosed with an ultrasonic energy of 14.7 W. The same reactor and frequency was found to provide maximum ejection of hydroxyl radicals to the solution. Impacts of pH and initial concentration on the efficiency of phenol removal were such that acidic pH and high concentrations accelerated the process as related to the increased likelihood of phenol at these conditions to approach the cavity sheath. Separate injection of equivalent volumes of air and argon into the reactors showed that the decomposition was enhanced in the presence of air by virtue of the production of additional reactive species via the reaction of nitrogen with molecular oxygen.

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1. Introduction

Phenols are widely consumed in the industry such as in preservers of paint, leather and textile goods, and in the production of resins, disinfectants, medicine, caprolactam and bisphenol A. Improper handling of these compounds and/or inappropriate disposal of their wastes into water is a major pollution, as many of phenolic compounds are resistant to conventional water treatment processes, and some are recognized as suspected carcinogens [1].

Methods of destroying phenolic wastes in water has been widely investigated and found that advanced oxidation processes (AOPs) are promising alternatives, owing to their potential to generate hydroxyl radicals in solution [2–5]. Among many tools of producing hydroxyl radicals in AOPs (e.g. UV irradiation, ozonation, addition of hydrogen peroxide, Fenton's agent and combinations thereof), ultrasound is a novel method, by which water molecules undergo molecular fragmentation

to release hydroxyl and hydrogen radicals [6]. The underlying mechanism for this phenomenon is acoustic cavitation, which consists of the formation, growth and implosive collapse of gaseous cavity bubbles, resulting in local extremes of temperatures and pressures, at which molecules undergo pyrolysis [7]. As some of these radicals escape into the aqueous phase, they readily attack organic molecules therein for oxidative destruction.

Sonochemical effects can be enhanced by inserting solid particles and/or injecting a soluble gas in solution [6]. However, the effects are more strictly related to the applied frequency, power and the reactor geometry. In general, frequency selection is based on the vapor pressure, solubility and octanol–water partition coefficient of the target chemical. Hydrophobic compounds with high vapor pressures tend to diffuse into the gaseous bubble interior, so that they may easily be destroyed in the bubble–liquid interface and/or the bubble itself. The most suitable frequencies for destroying such compounds lie between 20 and 100 kHz, by which long-lived “stable” cavities are generated [8]. In contrast, hydrophilic compounds particularly at low concentrations tend to remain in the bulk liquid and their destruction is possible only by aqueous phase oxidation caused by the “unstable cavity”

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collapse (at 200–800 kHz), during which the probability of radical escape to the bulk liquid is high [9–10].

There are many studies on sono-degradation of phenol in water, focusing mainly on parametric effects. In principle, short frequency ultrasound was found ineffective, while medium frequencies, particularly 200 and 500 kHz were found to provide sufficiently high decomposition yields [11–15]. In addition, it was found the efficiency of removal at all frequencies could be improved by the addition of catalysts such as Fenton's reagent or CCl_4 , which by reacting with hydrogen radicals increased the availability of uncombined hydroxyl radicals in solution [15,16].

The purpose of this study was to cover a detailed study on ultrasonic decomposition of phenol to fulfill the gaps in the literature and to assess single and interactive effects of operating parameters (e.g. frequency, reactor volume, hydrogen peroxide production, solute concentration and pH, type of saturating gas) on the rate of decomposition and the product yield. The test frequencies were 20, 300 and 520 kHz employed in three different reactors with 80, 150 and 1200 ml, respectively.

2. Experimental

2.1. Material

Phenol was purchased from Riedel Häen (97% pure) in solid form, and was dissolved in deionized water. Potassium ferrocyanide ($\text{K}_3\text{Fe}(\text{CN})_6$), 4-aminoantipyrine, ammonium chloride (NH_4Cl), ammonium hydroxide (NH_4OH), sulfuric acid and all other reagents were obtained from Fluka and used as received.

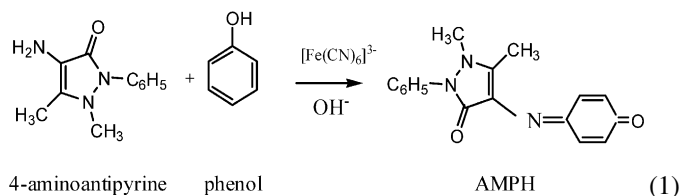
2.2. Apparatus

Three different ultrasonic equipment with three distinct frequencies, power outputs and cell volumes were used throughout the study. The first one was a horn-type sonicator (tip diameter = 12 mm) connected to a 20 kHz Bandelin Sonopuls HD2200 generator with a capacity of 180 W. The tip of the horn was submerged into the liquid from the top of an 80 ml cylindrical glass reactor. The second one was made of a 300 kHz piezo-electric transducer located at the bottom of a 150 ml cell and connected to a generator with a maximum capacity of 25 W (UNDATIM ULTRASONICS). The third equipment consisted of a piezo-electric transducer emitting ultrasonic pressure at 520 kHz and mounted on a titanium plate at the bottom of a cylindrical Pyrex reactor of 1200 ml with a generator capacity of 100 W (UNDATIM ULTRASONICS). In all systems, the cells were equipped with a water-cooling jacket to maintain constant liquid temperature.

2.3. Analysis

Phenol was monitored spectrophotometrically by the aminoantipyrine method [17] using a UNICAM-Helios, alpha/beta double beam spectrophotometer with an optical path-length of 1 cm. The method is based on the reaction of phenol with 4-aminoantipyrine in the presence of potassium ferro-

cyanide at pH 7.9 to form a colored antipyrine complex (AMPH) as shown:



Concentration of phenol in solutions of AMPH was estimated via a calibration curve generated from the absorption of the solutions in the visible band. Hydrogen peroxide was monitored by the analytic procedure described by Klassen et al. [18].

2.4. Procedure

A stock phenol solution of 0.5 M was made in deionized water and stored at 4 °C in the dark. Test samples of 0.1, 0.25, 0.5, 1, 2 and 5 mM were prepared from the stock using deionized water. Sample volumes in 20, 300 and 520 kHz reactors were respectively 80, 100 and 300 ml. The test solutions were bubbled with air or argon for 30 min prior to sonication and the same gas was continually injected into the solution throughout a contact time of 90 min. Samples were withdrawn from the reactors every 10 min for duplicate analysis of the antipyrine complex by spectrometry.

3. Results and discussion

3.1. Effect of initial concentration and the applied frequency

The UV–vis spectrum of AMPH showed that it had two principle absorptions: one at the visible band (508 nm) corresponding to the phenolic chromophore, and the other at near UV (332 nm). Spectral changes in AMPH prepared by reacting the effluents of 300 kHz reactor during 90 min sonication of 5 mM phenol is presented in Fig. 1. Note that sonication not only rendered decolorization (and decomposition) of the phenol complex but also induced UV absorption abatement.

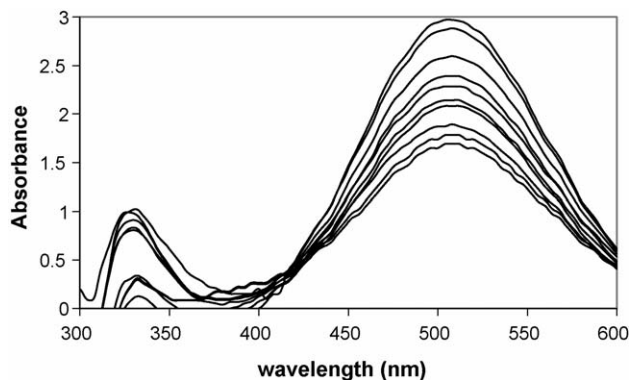


Fig. 1. Changes in the spectrum of AMPH during sonication of 5 mM phenol (300 kHz) for 0, 10, 20, 30, 40, 50, 60, 70, 80 and 90 min at pH 2.0 and in the presence of air flowing at 1.5 l min^{-1} .

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