

# Influence of $\text{TiO}_2$ concentration on the synergistic effect between photocatalysis and high-frequency ultrasound for organic pollutant mineralization in water

Ricardo A. Torres<sup>a,b,c</sup>, Jessica I. Nieto<sup>c</sup>, Evelyne Combet<sup>b</sup>,  
Christian Pétrier<sup>b</sup>, Cesar Pulgarin<sup>c,\*</sup>

<sup>a</sup> Grupo de Electroquímica, Instituto de Química, Facultad de Ciencias Exactas y Naturales, Universidad de Antioquia, A. A. 1226 Medellín, Colombia

<sup>b</sup> Université de Savoie, 73376 Le Bourget-du-Lac, France

<sup>c</sup> Groupe de Génie Électrochimique, Institut des Sciences et Ingénierie Chimique, Ecole Polytechnique Fédérale de Lausanne, CH-1015 Lausanne, Switzerland

Received 16 August 2007; received in revised form 5 November 2007; accepted 11 November 2007

Available online 22 November 2007

## Abstract

This work deals with the coupling of sonolysis (300 kHz, 80 W) and solar photocatalysis using titanium dioxide, two advanced oxidation processes for the degradation of a model organic pollutant, bisphenol A (BPA). Initially, the performances of the two separate processes in both the elimination and mineralization of BPA were compared. Even if identical BPA byproducts were formed, the two processes were complementary, while ultrasound was better able to eliminate the target pollutant, photocatalysis proved to be more efficient for reaching mineralization. Using the combined system, an interesting synergistic effect, which depends on the titanium dioxide loading, was observed for BPA mineralization. The best synergistic effect was found at low catalyst loading. After 4 h of combined treatment using  $0.05 \text{ g L}^{-1}$  of titanium dioxide, 62% of dissolved organic carbon (DOC) was eliminated. In contrast, 6 or 12% of DOC was removed by ultrasound alone or photocatalysis alone, respectively. Using  $1 \text{ g L}^{-1}$  of catalyst, 68 or 50% of DOC was removed by ultrasound/photocatalysis or photocatalysis, respectively. The poor synergistic effect at this catalyst loading can be explained by an inhibiting effect of the titanium dioxide on the cavitation activity.

© 2007 Elsevier B.V. All rights reserved.

**Keywords:** Sonochemical degradation; Photocatalysis; Sonophotocatalysis; Endocrine disrupting chemical; Bisphenol A elimination; Water treatment; Advanced oxidation

## 1. Introduction

In recent years, there has been increasing interest in the application of advanced oxidation processes (AOPs) as attractive alternative treatments for contaminated ground, surface, and wastewaters containing anthropogenic toxins with low biodegradability, as well as for the purification and disinfection of drinking water [1–3]. AOPs are all based on the production of  $\bullet\text{OH}$  radicals. These radicals are extraordinarily reactive species that attack most organic molecules, with rate constants usually on the order of  $10^6$  to  $10^9 \text{ mol L}^{-1} \text{ s}^{-1}$  [4].

Interestingly, AOPs offer different routes to  $\bullet\text{OH}$  radical production, allowing easier tailoring of the process for specific treatment requirements.

Although AOPs have been successfully used for the elimination of a huge number of organic compounds, AOPs cannot achieve complete mineralization of pollutants in several cases due to economical or technical reasons. Thus, in recent years efforts have been made to develop systems combining two different AOPs [5,6] or an AOP with a biological process [7,8].

Among the existing AOPs, sonochemical oxidation has received considerable attention because of its particular efficacy toward volatile and/or hydrophobic compounds [9,10]. Ultrasound irradiation of aqueous solutions leads to acoustic cavitation, i.e., the cyclic formation, growth and

\* Corresponding author. Tel.: +41 21 693 47 20; fax: +41 21 693 56 90.

E-mail address: [cesar.pulgarin@epfl.ch](mailto:cesar.pulgarin@epfl.ch) (C. Pulgarin).

adiabatic implosion of microbubbles. Under these conditions, organic substances with an elevated fugacity character are pyrolysed [11], while non-volatile compounds are degraded by hydroxyl radicals coming from water and oxygen dissociation [12]. Hydrophobic substrates are mainly degraded at the interface of bubbles and solution. Because of the short lifetime of  $\bullet\text{OH}$  radicals, a large fraction recombine at the interface of the bubble before reacting with hydrophilic substances. Thus, polar organic compounds are eliminated in the solution bulk, to a much lower extent than volatile and hydrophobic substrates [13].

$\text{TiO}_2$  photocatalysis is another AOP that has gained considerable attention as a means for water remediation [14,15]. In this process, UV light ( $\lambda \leq 387$  nm) is used to excite an electron from the  $\text{TiO}_2$  valence band to the conduction band to produce an electron–hole pair. Oxygen dissolved in solution can scavenge excited electrons, limiting the electron–hole recombination. Organic compounds may suffer oxidation directly at the hole. The hole can also undergo charge transfer with adsorbed water molecules or with surface-bound hydroxide species, forming  $\bullet\text{OH}$  radicals. Those organics that remain near the catalyst surface, i.e., highly polar substances, are more likely to be oxidized by photocatalysis.

Enhancement of the photocatalysis efficiency using low-frequency ultrasound (20–100 kHz) has been widely investigated [16–19]. However, in spite of the notable performance of high-frequency ultrasound (frequencies  $>100$  kHz) for degrading organic compounds, fewer reports have addressed the combination of photocatalysis and high-frequency ultrasound. Moreover, the synergistic effect between these two processes is a controversial matter. Stock et al. [20] recently reported that, after 4 h of treatment using a combined high-frequency ultrasound/photocatalysis system, the mineralization rate of an azo dye was greater than that resulting from an additive

effect of the individual AOPs. In contrast, Théron et al. [16], studying the degradation of phenyltrifluoromethylketone, reported the absence of synergy between the same two AOPs.

The aim of this work is to contribute to the understanding of the synergistic effect between  $\text{TiO}_2$  photocatalysis and high-frequency ultrasound for the degradation and mineralization of organic pollutants. Bisphenol A [BPA: 2,2-bis(4-hydroxyphenyl)propane] was selected as the target compound because it is an endocrine disrupting chemical widely used in the plastic industry as a monomer for production of epoxy resins and polycarbonate. This compound is released into the environment from bottles, packaging, landfill leachates, paper, and plastics plants [21–23]. Previous reports have shown that BPA treatments with classical chemical processes, such as chlorination, lead to products with even higher endocrine disrupting effects and/or toxicity [24–26].

## 2. Experimental

### 2.1. Reagents

Bisphenol A and ammonium heptamolybdate were obtained from Aldrich. Potassium iodide, acetic acid, and sulfuric acid were supplied by Fluka. Potassium hydrogen phthalate and acetonitrile were purchased from Merck and Fisher Chemicals, respectively. All these chemicals were used without any further purification. Throughout the experiments, oxygen was added to the system. Oxygen was selected as saturating gas instead of air in order to avoid the formation of nitrite and nitrate ions during sonication of aqueous solutions [5], and because of its excellent performance in ultrasound water treatments [27,28].

Milli-Q water was used throughout, for the preparation of aqueous solutions and as a component of the mobile phase in HPLC analysis.

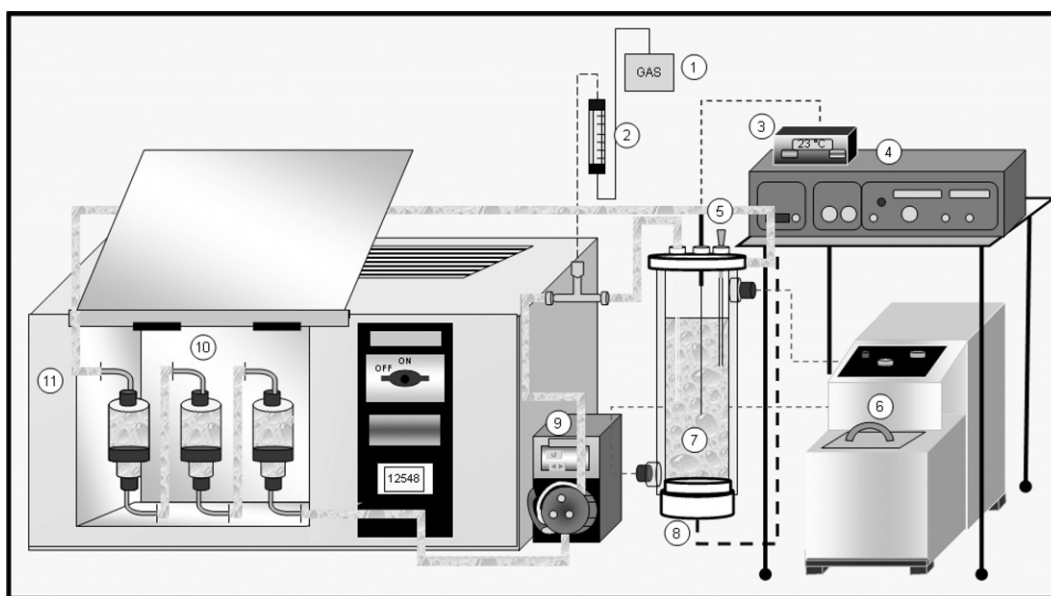


Fig. 1. Sonochemical/photocatalysis reactor used for the BPA treatment. Numbers in circles define the following: (1) Gas container; (2) gas flow control; (3) temperature control; (4) ultrasound generator; (5) sampling; (6) thermostatic bath; (7) sonochemical reactor; (8) piezo-electric disc; (9) peristaltic pump; (10) Pyrex vessels; (11) solar simulator.

Download English Version:

<https://daneshyari.com/en/article/48364>

Download Persian Version:

<https://daneshyari.com/article/48364>

[Daneshyari.com](https://daneshyari.com)