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Optimization of sono-electrochemical oxidation of ibuprofen in wastewater

Nam Tran[a], Patrick Drogui[a,*], Laurent Nguyen[b], Satinder K. Brar[a]

[a] Institut National de la Recherche Scientifique (INRS-Eau Terre et Environnement), Université du Québec, 490 Rue de la Couronne, Québec, QC, Canada
[b] École Nationale du Génie de L'eau et de L'environnement de Strasbourg, Université de Strasbourg, 1 Quai Koch, BP 61039, Strasbourg 67070, France

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ABSTRACT

The decomposition of ibuprofen in synthetic solution and in municipal wastewater effluent was investigated using an electro-sonochemical reactor. The concept of coupling ultrasonication and electrooxidation (EO–US) treatment for ibuprofen removal was demonstrated. The degree of synergy was 5.26%. Several factors such as current intensity, ultrasound power, treatment time were investigated. A factorial experimental design was used for determining the influent parameters on the ibuprofen degradation. The current intensity and treatment time were the main influent parameters on the degradation rate. Using a 2^3 factorial matrix, the best performance of ibuprofen degradation (77% of removal) was obtained by selecting 40 W of ultrasound power, a current intensity of 5.0 A and a treatment time of 120 min. Subsequently, the optimal experimental parameters for ibuprofen degradation have been investigated by using a central composite methodology. Under these, optimal conditions determined by this method, EO–US can be applied to oxidize ibuprofen in municipal wastewater effluent (up to 90% of degradation) while using a current intensity of 4.09 A for a period of 110 min and by applying 20 W of ultrasound power.

Introduction

The increasing production and intensive use of pharmaceutical products have led to the entry of these products into the environment and to eventual pollutions of soil, ground and surface waters. The pharmaceutical residues are rejected into surface waters, ground water and may enter into drinking water sources [1,2]. Ibuprofen (IBU) is an anti-inflammatory, analgesic and antipyretic in the human treatment of fever and pain often found in several water sources [2,3]. IBU have been found, among other pharmaceutical residues, in sewage wastewaters and surface waters [1–4]. A typical concentration of IBU between 2 ng L^{-1} and 8.77 μg L^{-1} has been detected in US [5] and UK surface waters [6]. Several researchers have been reported that drugs found in aquatic systems may present a potential hazard for human health, especially where no advanced wastewater treatments are used [7–9]. The removal of IBUs from water is a difficult task due to their low concentration and refractory properties [4]. Conventional methods frequently used in water treatment, such as biological and physicochemical treatments have been reported to be sometimes ineffective to remove emerging organic pollutants (EOPs) such as IBU. Therefore, a need exists for efficient treatment technology for the removal of this pollutant from the aquatic environment.

Advanced oxidation processes (AOPs) have been proposed as alternative methods to remove many toxic and bio-recalcitrant compounds in wastewater. Recent studies show that IBU degradation can be achieved through some other AOPs treatment such as electrochemical oxidation. For instance, pharmaceutical IBU was removed from water using an electrochemical oxidation process by means of carbon nanotubes electrodes [10]. After 116 min of electrooxidation, a relatively high degradation rate of 78% and a high mineralization rate of 60.84% were recorded at a potential of +1.25 V versus Ag/AgCl in the presence of 10 mg L^{-1} of IBU. Ambuludi et al. [3] have also studied IBU (C_0 = 0.2 mM) oxidation using an electrochemical advanced oxidation process in the presence of boron-doped diamond (BDD) anode electrode.

Abbreviations: ANOVA, analysis of variance; AOP, advanced oxidation processes; BDD, boron-doped diamond; EOP, emerging organic pollutant; IBU, ibuprofen; CCD, central composite design; EO, electrooxidation; EO–US, sonoelectrooxidation; FD, factorial design; LC, liquid chromatography; MS, mass spectrometry; MWE, municipal waste water; RSM, response surface methodology; RPM, round per minute; SE, synthetic effluent; Ti, titanium; Ti/PbO_2, titanium coated with lead oxide; TOC, total organic carbon; UV, ultraviolet; US, ultrasonication; VIS, visible.

* Corresponding author at: Institut National de la Recherche Scientifique (INRS-Eau Terre et Environnement), Université du Québec, 490 Rue de la Couronne, Québec, QC G1K 9A9, Canada. Tel.: +1 418 654 3119; fax: +1 418 654 2600.
E-mail address: patrick.drogui@ete.inrs.ca (P. Drogui).

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After 8 h of electrolysis, total organic carbon (TOC) removal ranged from 91% to 96% by applying a current intensity in the range of 50–500 mA. Likewise, Ciriaco et al. [4] studied IBU oxidation by using an Ti/Pt/PbO_2/BDD anode electrode. Different current densities were applied (10, 20 and 30 mA cm^{-2}) in the presence of initial concentrations of IBU ranging from 0.22 to 1.75 mM. The COD removal ranged from 60 to 95%, whereas TOC removal varied from 48 to 92%.

In spite of good oxidation rates of IBU, many of electrochemical processes are inefficient to subsequently oxidize by-products formed during electrolysis [11,12]. Indeed, during electrolysis, the electrode surface can be covered with organic substances that are formed during the treatment of organic-containing effluent. The formation of organic substances on the electrode surface reduces its electrode active surface. Since 1990s, the ultrasound process has received considerable interest for the destruction of organic pollutants that are present in wastewater [13–15]. The ultrasonication process has been identified as a successful alternative for the destruction and mineralization of some recalcitrant organic compounds in water and does not require the addition of chemicals [13,16,17]. Acoustic cavitation, derived from the high calorimetric power of a liquid, can provide unusual and unique reaction sites as a result of the extremely transient and small cavitation bubbles that are created with high temperatures and high pressures [11,18]. The beneficial results from exposing electrochemical cells to the effects of power ultrasound include the enhancement of mass transport, the increase of current efficiencies and the continuous activation of the electrode surface [12,18]. These effects can be ascribed to the rapid generation and collapse of microbubbles within the electrolyte medium or nearby the electrode surface. This cleaning effect has been reported to improve electron exchanges by peeling out passivation films on surface electrode or piercing them by micro holes [11,12,19].

The general objective of this study is to investigate the combination of ultrasonication (US) with electrooxidation (EO) to remove IBU from water. The specific objectives of this study are the followings: (i) evaluate the interest of coupling ultrasonication and electrooxidation processes for IBU removal from water; (ii) study the influence of the principal experimental parameters (electrical current, ultrasound power, treatment time, temperature) on the efficiency of the process for IBU degradation; (iii) use a statistical methodology for a rational analysis of the combination of operational factors that led to the best conditions for IBU degradation and; (iv) verify the quality of treated effluent (versus untreated effluent) while treating real municipal wastewater contaminated by IBU.

Materials and methods

Preparation of the synthetic effluent (SE)

The water samples used in the first step study were prepared synthetically by dissolving IBU in de-ionized water. IBU analytical grade reagent (99%) was obtained from Sigma–Aldrich (Canada). Stock solutions of IBU were prepared in a beaker containing 100 mL of methanol and 1.0 g of IBU. To eliminate the effect of methanol on IBU removal, the SE solution of IBU was solubilized by diluting small amount of stock solution (3.5 mL) in the de-ionized water (a beaker volume of 3.5 L) using a magnetic stirrer (500 rpm) at room temperature (25 °C). The resulting mixtures constituted the IBU

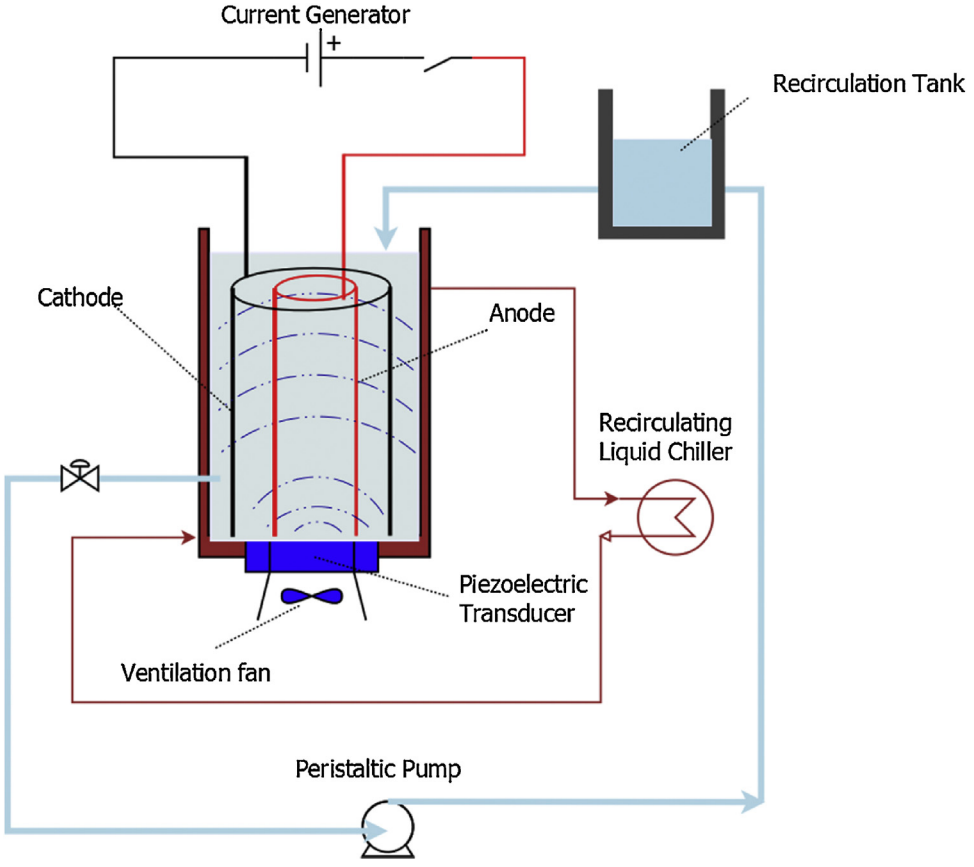

Fig. 1. Schematic view of sono-electrooxidation reactor with recirculation loop.