



# Ultrasonically enhanced electrochemical oxidation of ibuprofen



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## ABSTRACT

A hybrid advanced oxidation process combining sonochemistry (US) and electrochemistry (EC) for the batch scale degradation of ibuprofen was developed. The performance of this hybrid reactor system was evaluated by quantifying on the degradation of ibuprofen under the variation in electrolytes, frequency, applied voltage, ultrasonic power density and temperature in aqueous solutions with a platinum electrode. Among the methods examined (US, EC and US/EC), the hybrid method US/EC resulted 89.32%, 81.85% and 88.7% degradations while using NaOH, H<sub>2</sub>SO<sub>4</sub> and deionized water (DI), respectively, with a constant electrical voltages of 30 V, an ultrasound frequency of 1000 kHz, and a power density of 100 W L<sup>-1</sup> at 298 K in 1 h. The degradation was established to follow pseudo first order kinetics. In addition, energy consumption and energy efficiencies were also calculated. The probable mechanism for the anodic oxidation of ibuprofen at a platinum electrode was also postulated.

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## 1. Introduction

Pharmaceutical drugs, once introduced into the environment by anthropogenic activities, can act as potent organic pollutants/endocrine-disrupting chemicals. Such chemicals can interfere with the endocrine system in several ways, resulting in an undesired response or disruption, which in turn may affect the health, growth, and reproduction of a wide range of organisms. Numerous studies have shown that pharmaceutical residues are widespread in aquatic environments. Ibuprofen (IBP), with the chemical name, 2-[3-(2-methylpropyl)phenyl] propanoic acid (Fig. 1), used commonly for its analgesic, antipyretic, and anti-inflammatory properties [1], is one such drug. Furthermore, 80% IBP has been detected in the surface water of Korea [2] with a mean average of 0.30 µg/L in plant wastewater influents [3]. In view of this, it is considered worthwhile to initiate the development of a systematic and comprehensive degradation methodology of IBP from aquatic environments.

Various processes for the degradation of IBP have already been reported [4–7]. Among them, advanced oxidation processes (AOPs) generate highly reactive and unstable hydroxyl radicals with a high oxidation potential of 2.80 V [8], which can also oxidize IBP in turn. However, the application of individual AOPs has been reported to be expensive, and generally exhibits lower energy efficiencies as an effective wastewater treatment procedure [9]. On the other hand, various researchers have demonstrated the enhanced disin-

fection rates with amplification of reactive radical productions, as well as their increased interactions with pollutants in combined AOP technologies. Moreover, the destructive mechanisms coupled with interrelated operating parameters of these technologies have resulted in more effective synergistic phenomena encouraging the development of novel combinative hybrid techniques for the degradation of IBP.

Studies performed by Arriaga's group on the sonolytic degradation of IBP reported that ultrasonic (US) irradiation generated long lived intermediates like aliphatic acids [4]. They explained that upon the irradiation of ultrasound, the dissociation of water takes place generating hydroxyl radicals as given below:



The production of these reactive radicals can be attributed to the sonolytic cavitation phenomenon of bubble formation, growth, and implosive collapse where thermal degradation process leads to a hotspot formation with an extreme condition of 2000 °C and 200 atm [10]. Here, mixing time onto the sonochemical reactor can also be considered as an important factor [11].

On the other hand, electrochemistry has also been suggested as an effective method for removing toxic organic pollutants from wastewaters and other aquatic systems [12]. In electrolysis, hydroxyl radical generation takes place through water discharge on the anode surface [13]. Generation of other reactive radicals also takes place in the bulk due to the presence of electrolytes. Ambuludi et al. also investigated about electro-oxidation (EC) of Ibuprofen using platinum or boron doped diamond electrode anode [14]

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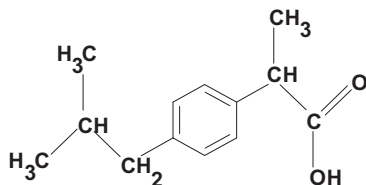
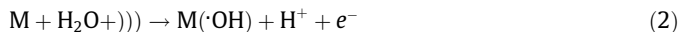
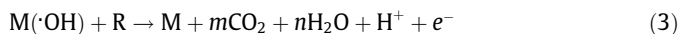


Fig. 1. Molecular structure of IBP.

and mentioned the large production of hydroxyl radicals on the anode surface from water discharge through the reaction:



where M is the electrode material and M(OH $\cdot$ ) is the heterogeneous hydroxyl radicals adsorbed in the anode. These hydroxyl radicals produced are powerful oxidizing agents that they are able to oxidize any organics until the total mineralization.



However, if supporting electrolytes are present, additional catalytic species with powerful oxidizing property are generated. They are capable of degrading the toxic organic pollutants into harmless compounds via “in situ” electrogeneration [15].

When both of these methods are executed separately, efficient environmental remediation can be achieved; however, under many conditions, the organic materials are not completely oxidized due to the generation of hydrophilic intermediates [16,17]. To overcome this limitation, many researchers have developed hybrid techniques encompassing sonication with other oxidation processes, including sono-Fenton and sono-photo-Fenton methods [18], UV/US/H<sub>2</sub>O<sub>2</sub> [19], US/H<sub>2</sub>O<sub>2</sub>/O<sub>3</sub> with zero-valent metals [20] and a combination of ultrasound and electrochemistry [21]. In 2010, Madhavan et al. studied the synergistic degradation of ibuprofen applying sonophotocatalysis in the presence of homogeneous (Fe<sup>3+</sup>) and heterogeneous (TiO<sub>2</sub>) [22]. Other hybrid technique [23] worth mentioning which are already reported includes commercial scale reactor based on combined US and electrochemistry with ozone [24].

The advantages of the combined sonoelectro-oxidation can be seen through the cavitation effects of sonolysis, such as microjet phenomena, where the diffusion-layer thickness of the electrolysis system is decreased to less than 1  $\mu$ m, and activation of the electrode surfaces leads to the enhancement of mass transfer and a more than tenfold increase in electrolytic current [25]. It is already known that organic compounds interact with the surface of solid electrodes (through adsorption processes), leading to electrode fouling and subsequent losses in degradation efficiencies [26,27]. However, the direct application of ultrasound to an electrode surface enhances its performance by cleaning it [28,29] thus, considerably increasing the degradation of toxic compounds [30–33].

Above all, the application of sonoelectrochemistry in wastewater treatment is still under development and is potentially a promising process [34]. To date, no reports have been published for describing the oxidation of ibuprofen using sonoelectrochemistry. Therefore, the main objective of the present research is to examine the effects of ultrasound upon the electro-oxidation of IBP by calculating the kinetic rate constants under the influence of various parameters such as the nature of electrolyte, frequency, applied voltage, ultrasonic power, and temperature. In addition to the above, the development of an efficient degradation system is sought via examination of energy consumption, energy efficiency, and synergistic effects of the applied methodologies.

## 2. Experimental

### 2.1. Materials and reagents

Ibuprofen (98% purity) was obtained from Sigma Aldrich. AR grade NaCl, NaOH, Na<sub>2</sub>SO<sub>4</sub> and H<sub>2</sub>SO<sub>4</sub> were used as electrolytes. NaCl (99.0% purity) and Na<sub>2</sub>SO<sub>4</sub> (90.5% purity) were procured from Samchun Pure Chemical Co., Ltd., Korea. NaOH and H<sub>2</sub>SO<sub>4</sub> were purchased from Showa Chemical Co., Ltd., Japan. For the experiments, a standard solution of 2.0 mg L<sup>−1</sup> IBP was prepared by direct dissolution in de-ionized water. Similarly, standard solutions of different electrolytes were also prepared.

### 2.2. Experimental set up

Sonoelectrochemical degradation experiments were carried out in an undivided 1000 mL cylindrical reactor with a dimension of 15 cm height and 10 cm diameter. The reactor was made of opaque steel designed to prevent exposure to possible photo degradation due to surrounding light during sonoelectrolytic reactions. A water cooling jacket surrounded the reaction vessel (Fig. 2).

The electrolytic apparatus consisted of a platinum anode (6 × 8 cm<sup>2</sup>) and a stainless-steel cathode (7 × 8 cm<sup>2</sup>). Experiments were performed while maintaining a constant voltage of 30 V administered by a regulated DC power supply, EP-3010, PNCYS Co., Ltd. and using various electrolytes, viz., NaOH, NaCl, Na<sub>2</sub>SO<sub>4</sub> and H<sub>2</sub>SO<sub>4</sub> with pH values in the range of 10.75–2.14. The sonoelectrochemical treatment was performed in parallel, operating under the same electrochemical conditions and in the same reactor. The designed electrodes were immersed in the reactor with the ultrasonic transducer placed at the bottom in such a way that the physical mechanisms of ultrasonic waves like microjets, acoustic streaming, microstreaming and turbulence due to cavitation, etc. lead to the mass transfer enhancement and activation of the working electrode surface. According to a report by Y. Son's et al. half of the initial cavitation energy of 35 kHz ultrasound could travel at an irradiation distance of 3.5 m. Hence, in the present case, the ultrasonic transducer was placed within the above said value from the electrodes to make sure that the working electrode receives the impacts of cavitation [35]. The transducer module was a PZT (Tamura Corp.) and ultrasound frequencies of 35, 300, 500 and 1000 kHz were controlled manually (Mirae Ultrasonic Tech. Co., MX-15100). Throughout the experiments, the temperature of the water in the reactor was controlled at 25 °C using a water cooling jacket. Therefore, assuming that there is no heat change, the input power was recorded using a multi-meter (M-4660M, METEX), instead of using a calorimetric method. The calculation of electric consumption for both electrolysis and sonolysis, which are designated as an electrical power density was further extended from that value [35].

Each experimental setup was run for 1 h and repeated three times. The quantitative analysis of US and EC exposed IBP final samples were done using HPLC (1260 Infinity, Agilent Technologies) equipped with a UV (G4212B 1260DAD) detector.

### 2.3. Evaluation methods

#### 2.3.1. Degree of degradation

The degradation of IBP was calculated as shown in Eq. (4)

$$X = \frac{C_0 - C_t}{C_0} \times 100\% \quad (4)$$

where X is the degradation degree, C<sub>0</sub> is the initial concentration of IBP and C<sub>t</sub> is the concentration of IBP at a given reaction time.

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