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Alternate pulses of ultrasound and electricity enhanced electrochemical process for *p*-nitrophenol degradation ☆

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ABSTRACT

A novel alternated ultrasonic and electric pulse enhanced electrochemical process was developed and used for investigating its effectiveness on the degradation of *p*-nitrophenol (PNP) in an aqueous solution. The impacts of pulse mode, pH, cell voltage, supporting electrolyte concentration, ultrasonic power and the initial concentration of PNP on the performance of PNP degradation were evaluated. Possible pathway of PNP degradation in this system was proposed based on the intermediates identified by GC–MS. Experimental results showed that 94.1% of PNP could be removed at 2 h in the dual-pulse ultrasound enhanced electrochemical (dual-pulse US-EC) process at mild operating conditions (i.e., pulse mode of electrochemical pulse time (T_{EC}) = 50 ms and ultrasonic pulse time (T_{US}) = 100 ms, initial pH of 3.0, cell voltage of 10 V, Na_2SO_4 concentration of 0.05 M, ultrasonic powder of 48.8 W and initial concentration of PNP of 100 mg/L), compared with 89.0%, 58.9%, 2.4% in simultaneous ultrasound enhanced electrochemical (US-EC) process, pulsed electrochemical (EC) process and pulsed ultrasound (US), respectively. Moreover, energy used in the dual-pulse US-EC process was reduced by 50.4% as compared to the US-EC process. The degradation of PNP in the pulsed EC process, US-EC process and dual-pulse process followed pseudo-first-order kinetics. Therefore, the dual-pulse US-EC process was found to be a more effective technique for the degradation of PNP and would have a promising application in wastewater treatment.
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1. Introduction

Advanced oxidation processes (AOPs) based on the generation of very reactive and oxidable free radicals have been used with an increasing prevalence due to their oxidizing power [1] for the treatment of wastewaters containing refractory organic pollutants which caused severe problems for the environment. However, most AOPs faced economic inefficiencies deriving from both equipment capacities and operating costs with the consumption of expensive chemicals such as O_3, H_2O_2, ferrous iron and other expensive unit processes like continuous ultraviolet irradiation [2].

Among AOPs, electrochemical (EC) methods have attracted a great deal of attention basically due to its favorable features such as mild operational condition, efficient functioning at room temperature and no additional requirement of chemicals [3]. Ultrasound (US) could be used to enhance the mass transport of reactants to the electrode surface, which increased the efficiency of EC process and kept the electrode clean and activated [4]. Thus, researchers recently have tested the simultaneous ultrasound enhanced electrochemical (US-EC) process where US and electrochemical process ran simultaneously for the treatment of different types of wastewater. Many attempts have been made to study the improvement of dye EC treatment through the combination of ultrasound and demonstrate the feasibility of this hybrid technique [5–7]. The effects of ultrasound on the degradation of pentachlorophenol by boron-doped diamond electrodes were investigated and the results showed that the greater pentachlorophenol degradation was obtained due to the increase of mass transport, minimization of the electrode fouling and the combined generation of hydroxyl radicals [8]. The kinetics of US-EC oxidation of diuron on boron-doped diamond electrodes was analyzed and the results demonstrated improvements on the mineralization kinetics of diuron closely to 43% when ultrasound was coupled to the EC treatment [9]. According to Kim et al. [10], the application of ultrasound on electrolysis of pentachlorophenol enhanced the electrolysis of pentachlorophenol and the kinetic constant of EC process was 0.0072 min^{-1}, while that of US-EC process was 0.02 min^{-1}, obtained at the frequency of 35 kHz. It was discussed that ultrasound enhanced electrochemical oxidation of phenol and phthalic acid with boron-doped diamond electrodes, and the results showed that ultrasound had remarkable influences

☆ Patent-pending technique.

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on EC oxidation in areas such as degradation efficiency, EC oxidation energy consumption, mass transport and EC reaction, etc. [11].

It was clear that there were many advantages of degradation of refractory compounds by using ultrasound in an electrochemical reactor. However, acoustic streaming and micro jetting produced by ultrasound could disrupt the directional movement of charged ions to the electrodes, resulting in a weak electrode reaction. Moreover, ultrasound cavitation created a large number of bubbles, which reduced the conductivity of electrolytes [12]. In addition, simultaneously using US and EC reactions could greatly increase energy consumption of the treatment. As a result, even though the combination of US might benefit to EC process, the ultrasound enhanced electrochemical treatment was restrained in industrial applications for the disadvantages discussed above.

The coupling of electrochemical processes and ultrasound has found a larger number of applications, especially in the field of materials science and energy technology [13]. Haas et al. [14] have presented a sonoelectrochemical method for the preparation of nanosized copper particles. A research demonstrated that at the same extraction efficiency of myrobalan nuts, pulsed ultrasound could reduce energy consumption by 20% over continuous ultrasound [15]. Meanwhile, the pulsed ultrasound used in an electrochemical reaction could reduce ultrasonic cavitation erosion on electrodes, resulting in a longer service life of the electrodes. Otherwise, the application of the pulsed electrochemical oxidation could not only conserve energy [16], but also promote production of free radicals and improve degradation efficiency [17]. Based on prior discussed facts, though the alternating US-EC method was widely applied in those fields above, it was still a novel method in the field of wastewater treatment, especially for the degradation of refractory organics in wastewater. Thus, it might be possible to use the pulsed US and the pulsed EC process in a way to maximize the enhancement of US to EC process and minimize their negative impacts.

In this work, we used a dual-pulse US-EC technique, synchronizing pulsed US and pulsed EC alternatively, to utilize the best of both to degrade refractory organics. The objective of this study was to investigate the feasibility of the dual-pulse US-EC process and compare its performance with the ones of pulsed US, pulsed EC process and US-EC process. *p*-Nitrophenol (PNP) was chosen as a test sample because it is a widely recognized refractory organic existing in agricultural irrigation effluents and industrial effluents, which were discharged from chemical plants producing pesticides, explosives dyestuffs, etc. [18]. Once released in the effluent, even at low concentrations, PNP might present high risks to ecological environment and human health. Thus, the efficient degradation of PNP is quite important for environmental protection. The percentage of PNP degradation and energy consumption were selected as the major criteria for the comparison in this study. The impacts of process parameters such as initial pH, cell voltage, ultrasonic power, electrolyte concentrations and initial concentration of PNP were studied. Moreover, changes of UV–Vis spectra during the dual-pulse US-EC coupling process and main degradation intermediates were investigated.

2. Materials and methods

2.1. Materials

All reagents used in this study were in analytical grade and used without further purification. All solutions were prepared from distilled water. H_2SO_4 and NaOH were used for initial pH adjustment of PNP solution whenever required, and Na_2SO_4 was employed as a supporting electrolyte in the electrochemical reaction.

2.2. Experimental setup

Fig. 1a presented a schematic of the experimental setup for the experiments carried out in this study. It included an electrochemical cell with a 150 ml beaker, a magnetic stirrer, an ultrasonic generator (Guangzhou KEPU Ultrasonic Electric Technological Ltd., China), a DC power (Shenzhen Atten Instruments Co., China) and a programmable logic controller (PLC) (Mitsubishi Electric., Japan). The cell was installed with Ti/Sb–SnO_2 dimensionally stable anode (DSA) and titanium electrodes (Baoji De Chen industry & trade Ltd., China). Effective area of the electrode and electrode gap was set as 18 cm^2 and 2.5 cm, respectively. The reactor was immersed into a water bath to keep the temperature around 30 °C. A titanium ultrasound horn was immersed into the liquid phase for 2.0 cm with a frequency at 22 kHz. A programmed PLC was used to synchronize electrical power and ultrasonic generation, from which a variation of durations of electric pulses and ultrasonic pulses were produced. The ultrasonic generator produced an ultrasonic pulse, which was triggered immediately after an electric pulse (Fig. 1b). During the electrochemical period, the EC process took place without ultrasound, avoiding the interference of ultrasound on the electrochemical reaction and saving energy. While during the ultrasonic period, the EC power was shut off and the ultrasound was kicked off, which not only activated the electrodes but also effectively eliminated the concentration polarization and electrochemical polarization occurring in the sole EC process. The pulsed US, pulsed EC process, and US-EC process were also performed by this experiment apparatus.

2.3. Analysis

The ultrasonic power (P_{US}) dissipated in the reactor was measured by calorimetric method [19] (the rise in temperature of a fixed quantity of water in an insulated container for given time was measured) using Eq. (1):

$$P_{US} = m \times C \times (dT/dt) \quad (1)$$

where: m is the mass of water (g), C is the specific heat capacity of water (4.186 J g^{-1} K^{-1}), and (dT/dt) is the temperature rise per second at zero time (K s^{-1}), obtained from the tangent to the curve.

The maximum absorbance wavelength (λ_{max}) of PNP was 317 nm when pH was lower than 4.0, while it was 400 nm when the solution pH value was higher than 8.0. The pH value of sample solutions were adjusted to 3.0, and the absorption intensities of the solution were determined at 317 nm on UV–Visible

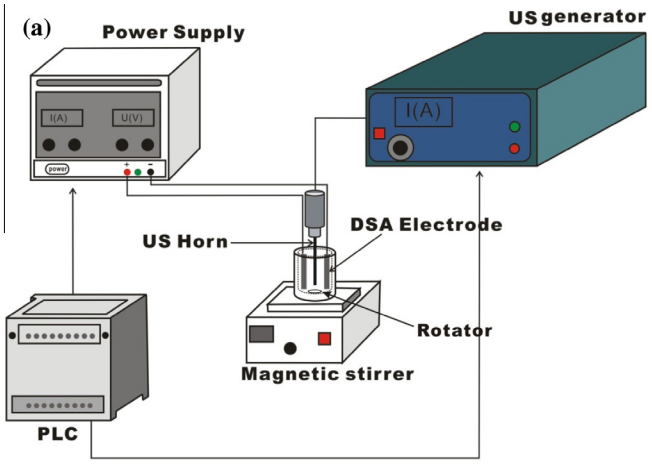

Fig. 1a. Experimental setup for dual-pulse US-EC, US-EC processes.

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