



Removal of carbamazepine in water by electro-activated carbon fiber-peroxydisulfate: Comparison, optimization, recycle, and mechanism study

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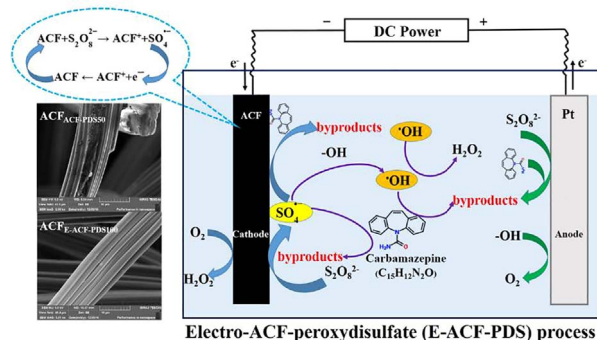
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HIGHLIGHTS

- Electro-ACF-peroxydisulfate process can effectively remove carbamazepine.
- A synergistic effect was found between electrolysis and ACF-peroxydisulfate processes.
- ACF was recycled for 100 times in E-ACF-PDS process with a stable efficiency.
- The electrons on cathode could inhibit the destruction of ACF caused by chemical oxidation.

GRAPHICAL ABSTRACT



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ABSTRACT

Treatment of a persistent organic pollutant, carbamazepine (CBZ), in aqueous solution by a novel and sustainable method, Electro-activated carbon fiber-peroxydisulfate (E-ACF-PDS), was investigated and compared with ACF adsorption, Peroxydisulfate (PDS), ACF-peroxydisulfate (ACF-PDS), E-Pt (Pt-plating titanium as cathode in electrolysis), E-ACF (ACF as cathode in electrolysis), and Electro-Pt-peroxydisulfate (E-Pt-PDS) processes, respectively. The removal rate of CBZ by E-ACF-PDS was 98.78% in 30 min, which was much higher than that of the others. Besides, several tests were performed to investigate the role of operational conditions (pH, peroxydisulfate concentration, electrode potential, temperature, and water matrix) in E-ACF-PDS process. Moreover, it is worth noting that even after recycling ACF for 100 times, the removal rate of CBZ in 30 min decreased only slightly from 98.78% to 97.35% in E-ACF-PDS process. In addition, the surface functional groups, FTIR spectrum, Raman spectrum, BET surface area, micropore volume, and SEM images of ACF before and after long time service were investigated and the results revealed that the ACF could maintain the original features in E-ACF-PDS process even after 100 cycles, while the ACF alone was seriously damaged in ACF-PDS process after 50 cycles. These results suggested that the E-ACF-PDS process could remarkably enhance the removal of CBZ in aqueous solution due to the simultaneous adsorption of contaminants and the increase in the generation of active radicals on ACF. The electrons on cathode could inhibit the destruction of ACF caused by chemical oxidation such as sulfate radicals, hydroxyl radicals, and peroxydisulfate. Therefore, the E-ACF-PDS process may provide

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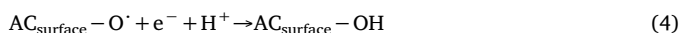
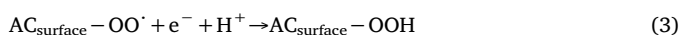
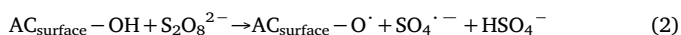
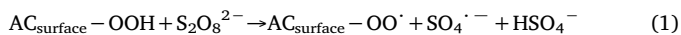
an effective and sustainable approach to water treatment for the elimination of contaminants of emerging concern in water.

1. Introduction

In recent years, the presence of pharmaceuticals and personal care products (PPCPs) in the aquatic environment has raised serious concerns [1]. As one of widely used pharmaceuticals for the treatment of seizure disorders, epilepsy, and relief of neuralgia, carbamazepine (CBZ) is frequently detected in the effluent of wastewater treatment plants (WWTPs) and water bodies at concentrations ranging from 820 to 6300 ng L⁻¹ [2], indicating it can be barely removed through sewage systems. Carbamazepine (C₁₅H₁₂N₂O), which has an olefinic double bond on the central heterocyclic ring, is difficult to be degraded by biodegradation or photodegradation [3,4]. The removal efficiencies of CBZ by traditional WWTPs are typically below 10% as reported [2]. Consequently, CBZ is released into the receiving water bodies of WWTPs, which may cause potential adverse health effects on living beings. Thus, wastewaters containing CBZ should be adequately treated before being discharged into the aqueous environment.

For the purpose of eliminating CBZ completely from wastewater, several advanced oxidation processes (AOPs) have been investigated. These include electrochemical oxidation, Fenton oxidation, and UV photocatalytic degradation [5–7]. The mechanism of AOPs relies on generating radicals, such as sulfate radical and hydroxyl radical. Compared with hydroxyl radical (E⁰ = 2.8 V), sulfate radical (E⁰ = 2.6 V) has more selective oxidation ability but longer half-life. Moreover, sulfate radicals can be activated efficiently from the stable and low cost persulfate by ultraviolet light, transition metal ions, heat, activated carbon (AC), electrolysis, and even gamma radiation in aqueous solution [8–15]. Among these studies, activated carbon-persulfate or activated carbon fiber-persulfate process is considered as one of the promising treatment technologies in water treatment due to the adsorption of various organic pollutants on the AC and their destruction by the generated sulfate radicals at the same time [11,16–18].

Nevertheless, it was reported that the removal rate of contaminants by activated carbon- peroxydisulfate (AC-PDS) was significantly decreased with the increase in reuse cycles of AC due to the reduction of the available surface area of AC and oxidation of oxonium-hydroxyl groups on AC surface, which may act as a catalyst of the electron-transfer mediator for PDS decomposition [16,18]. This suggests that AC may not really act as a catalyst but rather as an initiator for PDS activation. The electron-donating residues on AC (AC_{surface}-OOH and AC_{surface}-OH) decompose PDS into sulfate radicals (Eqs. (1) and (2)). But when this source is exhausted by forming acidic functional groups, the production of sulfate radicals is halted.



To overcome the limits of AC-PDS, we have introduced electric field in the AC-PDS system to inject free electrons onto the surface of the AC (Eqs. (3) and (4)) to maintain the generation of sulfate radicals continuously without exhausting the electron-donating residues of AC. Although the combination of PDS and electrolysis processes (E-PDS process) has been reported in the literature, the cathodes used in these previous studies (platinum) are not as effective as ACF to combine both contaminant adsorption and PDS activation at the same time. Moreover, the durability of ACF in ACF-PDS and E-ACF-PDS (ACF as cathode in E-PDS process) during reuse for long time service, which is very important for application, remains unknown.

The main objective of this study was therefore to investigate E-ACF-PDS treatment of CBZ, which is a widely used model compound in studies using in AOPs. The removal and mineralization of CBZ in the E-ACF-PDS were investigated and compared with PDS, E-ACF, E-Pt, ACF adsorption, E-Pt-PDS (Pt-plating titanium as cathode), and ACF-PDS processes. The effects of the main operating parameters (e.g., voltage potential, PDS concentration, solution pH, temperature, and water matrix) on E-ACF-PDS performance were evaluated systematically. To reveal the mechanism and role of ACF played in the E-ACF-PDS process, *tert*-butyl alcohol (TBA), methanol (MA), and phenol were used as radical scavengers while the pore texture characteristics, surface functional groups, FTIR, and Raman spectrum of virgin and used ACF were analyzed and compared.

2. Materials and methods

2.1. Materials

Commercial ACF was purchased from Zichuan Carbon Fiber limited Company, Qinhuangdao, China. The ACF was soaked in the boiled ultrapure water for 48 h and dried at 378 K for 24 h before use. The target pollutant, Carbamazepine (purity > 98%), was purchased from Sigma-Aldrich Company. Other chemicals were of analytical purity and obtained from Chongqing Dongchuan Chemical Co., Ltd., China. All aqueous solutions were prepared in ultrapure water with a resistivity of 18.2 MΩ.

2.2. Experimental setup

The experimental reactor was a 500 mL glass column (10 cm diameter and 12 cm height), which had a Pt-plating titanium anode and a ACF/Pt-plating titanium cathode at the same size (50 mm × 35 mm). The distance between the anode and cathode was 2 cm. The reactor was placed in a 1 L beaker connected to a constant-temperature water bath for temperature control. A magnetic stirrer was provided at the bottom of the reactor. The electric field was conducted under constant-voltage condition using a DC power supply (Sunjake Co., Shanghai, China).

2.3. Procedures

All the batch experiments were performed in the reactor described above, which was continuously stirred at 800 rpm at a certain temperature. In a typical electrochemical procedure, a fixed amount of PDS was added into 500 mL solution containing 0.042 mM of CBZ, and then, the initial pH was adjusted to the desired value with 1 M NaOH and H₂SO₄. Afterward, the electrodes were inserted into the reactor quickly before turning on the DC power supply to start the electrochemical experiments. Experiments to study the effect of pH on CBZ degradation in E-ACF-PDS were conducted under the same reaction conditions except that PDS concentration decreased to 10 mM and pH was buffered with phosphate (pH 5.0, 7.0 and 9.0, 100 mM phosphate). For the ACF-PDS experiments, the size of ACF used and the experimental procedure was exactly the same as those in the electrochemical experiments except the turning on the DC power supply. All the samples were collected at predetermined time intervals, and filtered using 0.45 μm membranes. Then, the filtered samples were immediately placed in an ice bath (263 K) to quench the oxidation reaction for 20 min before analysis. All the experiments were conducted in triplicate. For the recycle tests, ACF was taken out from the earlier round and put into the fresh solution for the next round without any desorption or treatment.

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