



# Activated carbon electrodes: Electrochemical oxidation coupled with desalination for wastewater treatment



Feng Duan<sup>a,b</sup>, Yuping Li<sup>a,\*</sup>, Hongbin Cao<sup>a,\*</sup>, Yi Wang<sup>a</sup>, John C. Crittenden<sup>c</sup>, Yi Zhang<sup>a</sup>

<sup>a</sup> Key Laboratory of Green Process and Engineering, Institute of Process Engineering, Chinese Academy of Sciences, Beijing 100190, China

<sup>b</sup> University of Chinese Academy of Sciences, Beijing 100049, China

<sup>c</sup> Brook Byers Institute for Sustainable Systems, Georgia Institute of Technology, Atlanta, GA 30332, United States

## HIGHLIGHTS

- Simultaneous organics degradation and salt removal were realized.
- Salt and phenol removal efficiency were enhanced by bubbling O<sub>2</sub> gas.
- Phenol degradation was related to the electrogenerated chlorine oxidants.

## ARTICLE INFO

### Article history:

Received 21 October 2014

Received in revised form 17 December 2014

Accepted 18 December 2014

Available online 10 January 2015

Handling Editor: E. Brillas

### Keywords:

Electrosorption

Desalination

Capacitive deionization

Electrochemical oxidation

Phenol

Wastewater treatment

## ABSTRACT

The wastewater usually contains low-concentration organic pollutants and some inorganic salts after biological treatment. In the present work, the possibility of simultaneous removal of them by combining electrochemical oxidation and electrosorption was investigated. Phenol and sodium chloride were chosen as representative of organic pollutants and inorganic salts and a pair of activated carbon plate electrodes were used as anode and cathode. Some important working conditions such as oxygen concentration, applied potential and temperature were evaluated to reach both efficient phenol removal and desalination. Under optimized 2.0 V of applied potential, 38 °C of temperature, and 500 mL min<sup>-1</sup> of oxygen flow, over 90% of phenol, 60% of TOC and 20% of salinity were removed during 300 min of electrolysis time. Phenol was removed by both adsorption and electrochemical oxidation, which may proceed directly or indirectly by chlorine and hypochlorite oxidation. Chlorophenols were detected as degradation intermediates, but they were finally transformed to carboxylic acids. Desalination was possibly attributed to electrosorption of ions in the pores of activated carbon electrodes. The charging/regeneration cycling experiment showed good stability of the electrodes. This provides a new strategy for wastewater treatment and recycling.

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## 1. Introduction

Water shortage poses a great threat to the sustainability of human beings, and water recycling has been an effective way to solve the problem. Municipal wastewater, industrial wastewater, or wastewater that comes from household sources can be treated and recycled for non-potable and potable purposes. The treatment methods vary with the wastewater characteristics and end uses (Chen et al., 2013). For example, coking wastewater is common in steel industry, which contains many pollutants such as ammonia and phenolic compounds (Li et al., 2010). Even after the biological

treatment, inorganic ions (like Cl<sup>-</sup> ion) and refractory organic compounds still exist (Li et al., 2011) which needs to be removed if the wastewater will be reused in the production process.

Electrochemical advanced oxidation processes (EAOPs) have been developed as a promising technology for removing persistent organic contaminants, as they use electron as reagent and is environmentally clean. The main drawback is relatively low current efficiency and high energy consumption (Brillas et al., 2009). The organic pollutants can be destroyed by direct oxidation or indirect oxidation (such as chlorine, S<sub>2</sub>O<sub>8</sub><sup>2-</sup>) (Martinez-Huitle and Ferro, 2006). The anode materials are usually DSA-type electrodes (Chatzisyneon et al., 2010; Li et al., 2013), or boron doped diamond (BDD) electrodes (Daghrir et al., 2014). Carbon anodes are not often reported due to their low O<sub>2</sub> evolution overpotential and less efficient production of oxidants like hydroxyl radical

\* Corresponding authors at: No. 1 North Second Street, Zhongguancun, Haidian District, Beijing 100190, China. Tel.: +86 10 82544844; fax: +86 10 82544844 816.

E-mail addresses: [ypli@ipe.ac.cn](mailto:ypli@ipe.ac.cn) (Y. Li), [hbcao@ipe.ac.cn](mailto:hbcao@ipe.ac.cn) (H. Cao).

(Ghernaout et al., 2011). Fan et al. (2008) investigated Amaranth azo dye degradation with activated carbon fiber (ACF) as anode. Electro-oxidation or electro-reduction of the pollutant was achieved by applying positive and negative potential, respectively.

In addition to organic pollutants, the salts need to be removed for water reuse. Recently capacitive deionization (CDI) has received great interest as a desalination technology due to advantages such as low energy-consumption and no secondary pollution (Oren, 2008; Porada et al., 2013). The ions are electrosorbed by applied a potential to the porous electrodes and desorbed by shorting the electrodes or reversing the potential. The most frequently used electrodes are porous carbon materials with high surface areas, abundant pore structures and some surface groups (Avraham et al., 2011; Gao et al., 2013, 2014; Wang et al., 2014).

In this work, the decontamination of wastewater containing phenol and NaCl was investigated by electrochemical oxidation coupled with desalination using activated carbon (AC) electrodes, which has been rarely reported. The target is to develop a technology that is efficient in removal of organic pollutants and inorganic salts simultaneously and applicable in the advanced treatment of industrial wastewater. We examined the effects of oxygen concentration, applied voltage and temperature on the removal efficiency of phenol and salt. The phenol degradation pathway and intermediates were carefully studied.

## 2. Materials and methods

### 2.1. Preparation of carbon electrode

To prepare the carbon plate electrode, 1.6 g AC was first mixed with 0.67 g Polytetrafluoroethylene (PTFE) emulsion (60 wt%) to obtain weight percentage of 80%:20%. Ethanol was added and the mixture was stirred at 70 °C to form a carbon paste. Then it was rolled, treated at 240 °C for 40 min and 360 °C for 1 h. The thermal treatment may increase the physical stability of electrode (Wang et al., 2013) and surface oxygen-containing groups which affects the electrosorption of ions. The carbon flake (size of about 25 mm × 25 mm × 0.5 mm) was pressed on graphite paper which was used as current collector.

### 2.2. Electrochemical setup

The electrochemical setup used here is similar to that reported previously (Duan et al., 2014), as shown in Fig. SM-1. The undivided cell consists of two AC electrodes that are placed parallel and separated by a rubber gasket. Batch mode experiments were conducted using the continuously recycling system equipped with an electrochemical cell, a peristaltic pump, a power supply and a conductivity/pH meter. Typically the mixed solution of 100 mL of 17.1 mM NaCl and 1.1 mM phenol was employed as feed solution. The flow rate was kept at 40 mL min<sup>-1</sup>. The applied potential and temperature were optimized in the range of 1.5–3.0 V and 28–48 °C, respectively. Prior to the experiment, the solution was purged with O<sub>2</sub> or N<sub>2</sub> at 500 mL min<sup>-1</sup> for 30 min. The gas purging continued during the polarization process.

### 2.3. Electrode characterization

The morphology of the electrode was characterized by scanning electron microscopy (SEM, Quanta 250, FEI). Nitrogen sorption isotherm was measured at 77 K with an automated gas sorption analyzer (autosorb iQ, Quantachrome). Before measurement, the sample was degassed under vacuum at 120 °C for 12 h. Cyclic voltammetry (CV) was performed in the electrochemical workstation (Autolab PGSTAT302 N, Metrohm). The AC electrode (size:

10 mm × 10 mm, weight: about 10 mg) was used as the working electrode, with platinum sheet as counter electrode and saturated calomel electrode (SCE) as reference electrode. CV was measured in 17.1 mM NaCl at scan rate of 1–50 mV s<sup>-1</sup> and potential of –1.0 to 1.5 V (vs. SCE) under N<sub>2</sub> or O<sub>2</sub> bubbling condition. In some cases, 1.1 mM or 5.5 mM phenol was added to evaluate whether phenol could be oxidized directly.

### 2.4. Analytical methods

The conductivity of the effluent was constantly monitored using a conductivity meter (Orion 3-Star, Thermo Fisher Scientific Inc.). The relationship between the conductivity and concentration was calibrated before the experiments. The concentration of phenol and its oxidation intermediates was analyzed by high performance liquid chromatography (HPLC, 1200 infinity series, Agilent) equipped with Zorbax SB-C18 column. The mobile phase was a mixture of methanol and water containing 10 mM H<sub>3</sub>PO<sub>4</sub> (25%/75%, V/V). Total organic carbon (TOC) was measured by a TOC analyzer (TOC-V<sub>CPH</sub>, Shimadzu). The H<sub>2</sub>O<sub>2</sub> concentration was determined by spectrophotometric analysis with potassium titanium (IV) oxalate reagent at 400 nm (Sellers, 1980), while the total concentration of possible oxidants (such as H<sub>2</sub>O<sub>2</sub> and chlorine) was determined by the iodide method at 350 nm (Kormann et al., 1988).

The specific energy consumption (EC) of TOC and NaCl removal was calculated by the following equation (Santos et al., 2010):

$$EC_{\text{TOC}}(\text{kW h}(\text{kg TOC})^{-1}) = \frac{1000 \times U \int_0^t I dt}{\Delta\text{TOC} \times V} \quad (1)$$

$$EC_{\text{NaCl}}(\text{kW h}(\text{kg NaCl})^{-1}) = \frac{1000 \times U \int_0^t I dt}{\Delta C \times V} \quad (2)$$

where  $U$  is the applied potential (V),  $I$  is the current (A),  $t$  is the polarization time (h),  $\Delta\text{TOC}$  is the difference in TOC excluding that removed by electrode adsorption (mg L<sup>-1</sup>),  $\Delta C$  is the change in salt concentration (mg L<sup>-1</sup>),  $V$  is the solution volume (L).

## 3. Results and discussion

### 3.1. Characterization of AC electrode

The SEM image, nitrogen adsorption-desorption isotherm and pore size distribution (PSD) of the electrode are shown in Fig. SM-2. The SEM image displays that the addition of the PTFE polymer to AC powder yields excellent physical stability. Note that the isotherm exhibits obvious hysteresis at high relative pressure, suggesting the presence of mesopores. The BET specific surface area is 300 m<sup>2</sup> g<sup>-1</sup>, while the micropore surface area is 98 m<sup>2</sup> g<sup>-1</sup>. The PSD curve displays that most of the pores are above 1 nm, which is beneficial to ion adsorption.

The CV curves of AC electrode are presented in Fig. 1. Fig. 1a and b shows the CV curves under O<sub>2</sub> or N<sub>2</sub> atmosphere. The anodic or cathodic peak is not seen at sweep rate of 10 mV s<sup>-1</sup> or 50 mV s<sup>-1</sup>, while they are observed at a low sweep rate of 1 mV s<sup>-1</sup> (the blue<sup>1</sup> line). The anodic current increases quickly at a potential higher than 1.2 V, which may due to Cl<sup>-</sup> oxidation. A cathodic peak is also observed at 1.0 V. The CV curves under O<sub>2</sub> or N<sub>2</sub> atmosphere at 1 mV s<sup>-1</sup> are compared in Fig. 1c. The main difference is that the cathodic current is higher under O<sub>2</sub> atmosphere, as O<sub>2</sub> will be reduced at a negative potential.

Fig. 1d displays that a new oxidation peak appears at about 0.8 V in the presence of phenol, and it increases with the phenol

<sup>1</sup> For interpretation of color in Fig. 1, the reader is referred to the web version of this article.

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