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Treatment of coking wastewater by a novel electric assisted micro-electrolysis filter

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ABSTRACT

A newly designed electric assisted micro-electrolysis filter (E-ME) was developed to investigate its degradation efficiency for coking wastewater and correlated characteristics. The performance of the E-ME system was compared with separate electrolysis (SE) and micro-electrolysis (ME) systems. The results showed a prominent synergistic effect on COD removal in E-ME systems. Gas chromatography/mass spectrometry (GC-MS) analysis confirmed that the applied electric field enhanced the degradation of phenolic compounds. Meanwhile, more biodegradable oxygen-bearing compounds were detected. SEM images of granular activated carbon (GAC) showed that inactivation and blocking were inhibited during the E-ME process. The effects of applied voltage and initial pH in E-ME systems were also studied. The best voltage value was 1 V, but synergistic effects existed even with lower applied voltage. E-ME systems exhibited some pH buffering capacity and attained the best efficiency in neutral media, which means that there is no need to adjust pH prior to or during the treatment process. Therefore, E-ME systems were confirmed as a promising technology for treatment of coking wastewater and other refractory wastewater.

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Introduction

Coking wastewater, generated from coke plants and coal gasification plants, contains high concentrations of ammonia, phenol, cyanide, thiocyanate and other aromatic hydrocarbons, as well as a variety of nitrogen-, oxygen- and sulfur-containing heterocyclic compounds (Maranon et al., 2008; Li et al., 2003; Neufeld and Spinola, 1978). Most of these compounds are considered hazardous to the environment and a genotoxic risk to human beings (Zhu et al., 2013). Therefore, removing these toxic pollutants before discharging them into the surrounding environment is of great importance. Yet traditional wastewater treatment technologies are markedly ineffective at this task due to some compounds in coking wastewater being toxic and refractory (Ning et al., 2005).

Micro-electrolysis (ME), involving zero-valent iron and granular activated carbon (ZVI/GAC), has been successfully applied in the treatment of wastewater (Yang, 2009; Lv et al., 2011; Jiao et al., 2015; Zhang et al., 2015). In ME systems, numerous microscopic galvanic cells are formed spontaneously between ZVI and carbon, releasing Fe(II), hydroxyl and [H] (Yang et al., 2009). Contaminants are removed by a combination of chemical and physical processes, as has been summarized by many reports (Cheng et al., 2007; Yang, 2009). Briefly, these processes include (1) reduction by [H] and electrons at the cathode; (2) oxidation by radicals and oxidants produced during ME; (3) coagulation by the ferrous and ferric hydroxides formed from oxidation and precipitation of Fe(II), that are released from the anodes; (4) precipitation of large particulates under the effect of electromigration and electrophoresis; and

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(5) adsorption of organic contaminants by GAC. The ME system has some particular advantages such as simplicity, easy operation, low cost and high efficiency. Nonetheless, ME still suffers from some shortcomings that constrain its wide application. First of all, the reaction sites tend to gradually decrease and result in the deterioration of treatment performance after the precipitation of particulate substances by ferrous and ferric hydroxides on the metal scrap surface and/or adsorption by GAC (Zhang et al., 2014; Han et al., 2016). Secondly, ME processes tend to consume a large amount of acid to adjust the pH, because they operate most efficiently in acidic solutions (Cheng et al., 2007), hence increasing the treatment cost significantly. Therefore, enhancing the treatment performance of ME in a neutral or alkaline medium may be a good solution to overcome the shortcomings mentioned above.

Recent studies suggest that O_3 and H_2O_2 integrated with the ME process could obtain excellent treatment performance (Deng and Englehardt, 2009; Ruan et al., 2010). However, such external added oxidation reagents would lead to operation problems. In addition, some highly toxic intermediates may be formed during the treatment.

Electrolysis treatment was first proposed by Eugene Hermite in 1889, and has gradually become a promising process focusing on treating wastewaters containing refractory pollutants (Mohan et al., 2007; Liu et al., 2012). In the electrolysis process, the degradation of pollutants is attributed to either direct or indirect oxidation processes (Zhu et al., 2009; He et al., 2013; Yavuz and Koparal, 2006). In the direct anodic oxidation process, the pollutants are first adsorbed on the anode surface and then destroyed by the anodic electron transfer reaction. In the indirect oxidation process, strong oxidants such as hydroxyl radicals, hydrogen peroxide and ozone are electrochemically generated, which destroy the pollutants in the solution by oxidation reactions.

In order to improve the treatment performance of the ME process in neutral medium and solve inactivation and blocking problems, a self-designed electric filter reactor was applied to investigate the feasibility of the combination of an applied electric field with micro-electrolysis (E-ME). The electric filter reactor was mainly made of a stainless-steel net cathode and ZVI/GAC bed anode. To understand the performance of the combination of applied electric field with micro-electrolysis, coking wastewater was treated, with COD as the main target parameter. The optimal operation conditions were achieved for the process in laboratory-scale experiments. Furthermore, the characteristics of effluent and performance of E-ME processes were investigated by gas chromatography mass spectroscopy.

1. Materials and methods

1.1. Materials

The chemical reagents used in the experiment were all analytical grade. A commercial and cheap GAC with particle size of 4 mm was used. Iron filings were used as the iron source, with mean diameter of 0.5 mm and length ranging from 1 to 2 mm. A 20 mesh stainless steel net was used as the electrode.

The raw wastewater used in this investigation was obtained from the outlet of a coke plant near Kunming, Yunnan, China. The main characteristics of the raw wastewater were as follows: the initial pH was in the range 6.1–6.4; the values of COD and $NH_3\text{-H}$ were 5265–5865 mg/L and 684–704 mg/L, respectively.

1.2. Experimental system and procedure

The diagram of the experimental apparatus is shown in Fig. 1. A glass column was filled with a well-distributed mixture of GAC and ZVI. The mass ratio of GAC to iron filings was 2:1. In order to eliminate as much as possible the influence of adsorption from GAC on the ME process, a 0.5 hr immersion test of the GAC in a large amount of raw coking wastewater was conducted. Stainless steel net served as the cathode, and the ZVI/GAC bed as the anode, which was divided into two parts by stainless steel net and connected to a power source. The detailed structure of the ZVI/GAC bed is also shown in Fig. 1. The related parameters were designed as follows: diameter of the column 10 cm, height 30 cm, height of the solid filler 4 cm. A DC potentiostat was employed as the power supply with a voltage output up to 30 V and an air pump as the aerator. 600 mL raw coking wastewater was loaded into the experimental reactor.

1.3. Analytical methods

Samples were taken directly from the device without pH adjustment to further flocculate. The COD of the samples was analyzed by the potassium dichromate method (Gao et al., 2011). $NH_3\text{-H}$ was determined according to the Chinese SEPA standard method by Nessler's reagent spectrophotometry. For the gas chromatography–mass spectrometry (GC–MS) analysis, effluent samples pretreated by trichloromethane liquid–liquid extraction were used for GC–MS analysis using the Agilent 7890 series GC system supplied with a 5975 mass selective detector. The GC program was as follows: injector temperature, 280°C; column temperature, 40–150–280°C (temperature program during run); carrier gas, helium; split ratio, 10:1; total flow rate, 1.0 mL/min. The relative content of organic compounds in the effluent was evaluated by the peak area from the results of GC–MS analysis.

2. Results and discussion

2.1. Synergetic effect within E-ME

2.1.1. Degradation performance

The performance of the coupled technique combining electrolysis (applied electric field) with micro-electrolysis (E-ME) was investigated by comparing with separate electrolysis (SE) and micro-electrolysis (ME). The removal ratios of COD and $NH_3\text{-N}$ in SE, ME, and E-ME systems after 5 hr reaction at pH 6.4 are displayed in Fig. 2.

In contrast to the $NH_3\text{-N}$ removal rate of 29% in SE, more than 59% and 60% $NH_3\text{-N}$ removal rate could be achieved in ME and E-ME, respectively. It is easy to see that the ZVI/GAC bed played the dominant role in the removal of $NH_3\text{-N}$ and

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