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Short Communication

A novel integration of three-dimensional electro-Fenton and biological activated carbon and its application in the advanced treatment of biologically pretreated Lurgi coal gasification wastewater

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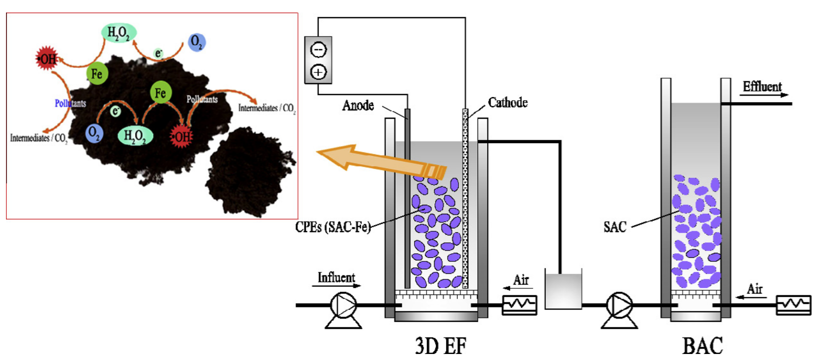
HIGHLIGHTS

- SAC-Fe developed from sewage/iron sludge served as CPEs and catalyst in 3D EF.
- 3D EF exhibited excellent capacity in abating COLOR and toxicity and improving biodegradability.
- The enhancement of pollutants removal in 3D EF attributed to generating more H_2O_2 and ·OH.
- The total operating cost of the integrated process was 1.1 CNY/t.
- The integration of 3D EF and BAC was more efficient at shorter retention time.

GRAPHICAL ABSTRACT



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ABSTRACT

A novel integrated process with three-dimensional electro-Fenton (3D EF) and biological activated carbon (BAC) was employed in advanced treatment of biologically pretreated Lurgi coal gasification wastewater. SAC-Fe (sludge deserved activated carbon from sewage and iron sludge) and SAC (sludge deserved activated carbon) were used in 3D EF as catalytic particle electrodes (CPEs) and in BAC as carriers respectively. Results indicated that 3D EF with SAC-Fe as CPEs represented excellent pollutants and COLOR removals as well as biodegradability improvement. The efficiency enhancement attributed to generating more H_2O_2 and ·OH. The integrated process exhibited efficient performance of COD, BOD_5, total phenols, TOC, TN and COLOR removals at a much shorter retention time, with the corresponding concentrations in effluent of 31.18, 6.69, 4.29, 17.82, 13.88 mg/L and <20 times, allowing discharge criteria to be met. The integrated system was efficient, cost-effective and ecological sustainable and could be a promising technology for engineering applications.

1. Introduction

Lurgi coal gasification wastewater (LCGW) is discharged in the process of coal gas purification, the composition of which is very complex. Although a series of options are employed, a large number of toxic and refractory compounds as well as their derivatives are still residual in the effluent of biologically pretreated LCGW (Zhuang et al., 2014a). However, with the implementation of the increasingly stringent environmental regulations, the quality of secondary effluent is unable to satisfy the discharge standards, especially the requirement of zero liquid discharge, due to the high concentrations of organic matter, ammonia and COLOR. Hitherto,

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more efforts are essential to be focused on advanced treatment of biologically pretreated LCGW.

Recently, advanced oxidation processes (AOPs) have drawn amazing attention for organic pollutants treatment and many efforts have been studied on the treatment of this wastewater (Jia et al., 2015; Xu et al., 2015). As one of AOPs, EF has drawn considerable attention as a promissory alternative technology by overcoming some drawbacks of traditional Fenton process. EF employs electrochemical reactions to generate in situ Fenton reagents from two-electron reduction of oxygen. Since ·OH production does not involve the use of harmful chemicals, this process is environmentally friendly. 3D EF significantly improved treatment efficiency and current efficiency by involving catalytic particle electrodes (CPEs) (Kong et al., 2006).

Meanwhile, an increasing amount of sewage sludge generated from wastewater treatment plant has become an issue of particular concern. Recently, sludge derived carbon has been widely used as an adsorbent for organic pollutants or heavy metals (Phuengprasop et al., 2011). Previous studies have demonstrated that the sewage sludge based activated carbon could serve as an efficient and stable catalyst support for Fenton and catalytic ozonation (Zhuang et al., 2014b). Iron sludge generated in traditional Fenton process, Fe/C micro-electrolysis process and other physico-chemical methods, which mainly constituted of iron and organic compounds. The concept "using waste to treat waste" nowadays becomes more attractive in the field of environmental engineering, offering a promising strategy for the utilization of sewage sludge and iron sludge. Iron sludge can serve as the iron source for Fenton catalyst preparation.

Additionally, although 3D EF oxidation has been proven to be an alternative method for the treatment of toxic and recalcitrant organic pollutions, it consumes a lot of energy and the general current efficiency is low (Anotai et al., 2006). Pretreatment of wastewater with poor biodegradability and high toxicity using 3D EF to improve biodegradability for further biological process may be more reasonable to avoid high energy consumption. Therefore, it is of great advantages to integrate 3D EF with biological process as a more efficient and cost-effective process. BAC involved activated carbon in biological reactor to combine adsorption and biodegradation (Reungoat et al., 2012). Hitherto, the integrated 3D EF–BAC system for the advanced treatment of biologically pretreated LCGW has not been reported in the literature yet. In the present study, a novel CPEs (SAC-Fe) developed for sewage sludge and iron sludge was applied in 3D EF oxidation of biologically pretreated LCGW and its electrocatalytic performance was investigated. The pollutants removal and biodegradability enhancement in 3D EF were evaluated. Meanwhile, the possible mechanism of SAC-Fe in 3D EF was discussed. Furthermore, the performance of the integrated process with 3D EF and BAC was evaluated and this novel integrated system exhibited substantial advantages in eliminating pollutants and shortening the hydraulic retention time.

2. Methods

2.1. Materials

The real biologically pretreated LCGW used in this study was collected from the effluent of secondary settling tank in the full-scale wastewater treatment facility in Harbin, China. The main characteristics of the wastewater were as follows: COD 140–190 mg/L, BOD_5/COD 0.05–0.09, COLOR 300–400 times, total phenols 80–120 mg/L, total organic carbon (TOC) 90–125 mg/L, NH_4^+-N 10–15 mg/L, total nitrogen (TN) 45–70 mg/L. The pH ranged between 7.0 and 8.0.

The dewatered sewage sludge sample used in this study was collected from the Wenchang wastewater treatment plant in Harbin, China. Iron sludge was obtained from a pilot industrial wastewater treatment plant (Harbin, China) setup with the combined process of Fe/C micro-electrolysis and Fenton oxidation. The CPEs of sludge deserved carbon with iron species (SAC-Fe) were synthesized from the pre-mixture of sewage sludge and iron sludge, followed by pyrolysis. The sludge deserved carbon (SAC) was prepared without the addition of iron sludge. The preparation processes were according to the methods in the previous reports (Gu et al., 2012; Zhuang et al., 2014b). Fe_3O_4 was the main component in SAC-Fe to be acted as catalyst (catalytic site) and chemical bonds were formed between Fe species and carbon matrix, according to the analytical results of XRD and FTIR (Figs. S1 and S2). The main characteristics of SAC-Fe were as follows: 351.6 m^2/g of BET area, 0.258 cm^3/g of macro and mesopores volumes, 3.614 nm of average pore size, 15.43% of Fe, 5.99% of Si and 2.86% of Al. The detail properties of the CPEs were listed in Table S1.

2.2. Experimental procedures

3D EF reaction was conducted in a one-compartment electrochemical cell (1.0 L) at room temperature. Ti/SnO_2 and active carbon fiber (4×5 cm) were the anode and cathode, both electrodes were fixed on two plastic brackets with the distance between the electrodes of 5 cm. CPEs were filled into real biologically pretreated LCGW and shaken for 48 h to achieve adsorption equilibrium. Then the adsorption saturated CPEs were transferred into electrolysis cell between anode and cathode. The amount of CPEs varied from 2.5 to 10 g/L to ascertain the optimal CPEs dosage. The degradation reaction was initialed by switching on the DC current. Current was adjusted with the increasing current density of 5, 10, 15 and 20 mA/cm^2. Air was bubbled from the bottom of the reactor (4 L/min) to provide oxygen and generate stirring in the solution. The supernatant of 3D EF was subsequently fed to the BAC system for the further purification. The prepared SAC was filled in BAC as carriers. The start-up and operational strategies of BAC system were described by previous literatures (Yapsakli and Çeçen, 2010; Kalkan et al., 2011) and has been operated for one month. The schematic diagram of the integrated process was shown in Fig. S3. The pH was adjusted with H_2SO_4 (1 mol/L) and NaOH (1 mol/L).

2.3. Analytical methods

BET area was determined using Micromeritics ASAP 2020 via nitrogen adsorption. Microspores volume was calculated by *t*-plot method and the macro(meso)pores volume as well as pore diameter were calculated using the Barrett–Joyner–Halenda method. Elemental analysis was carried out on an X-ray fluorescence spectra (XRF) (1800, Shimadzu) and an Elemental Analyzer (Elementar Vario EL III). COD, BOD_5, NH_4^+-N and total phenols were measured according to Standard Methods (APHA, 1998). COLOR was measured by dilution multiple method (Zhang et al., 2014). TOC and TN were measured with a TOC Analyzer (TOC-CPN, Shimadzu, Japan). The generation of hydroxyl radicals (·OH) was monitored by means of terephthalic acid fluorescent probe method on RF-6500 fluorescence spectrometer. Hydrogen peroxide generated in the solution was measured with iodide method. The results were average of at least three measurements with an accuracy of ±5%.