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Effect of the structure of stacked electro-Fenton reactor on treating nanofiltration concentrate of landfill leachate

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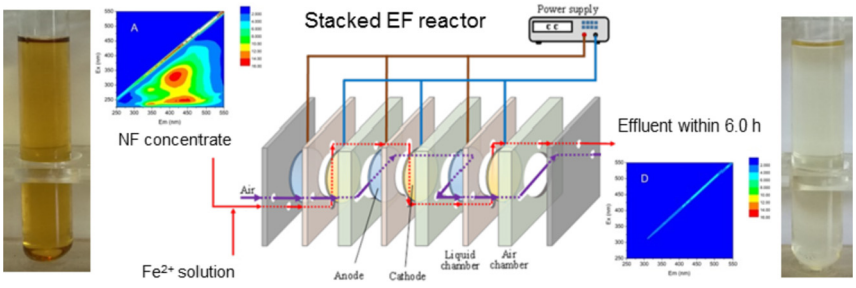
HIGHLIGHTS

- The structure effect of stacked EF reactor was studied.
- More than 70% of COD in NF concentrate was removed in the reactor.
- Specific energy consumption was lower in the stacked EF reactor than in other reactors.
- The COD removal was kept stable within several cycles of operation.
- Humic acids and aromatic proteins were degraded in the stacked EF reactor.

GRAPHICAL ABSTRACT



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ABSTRACT

The membrane concentrate from landfill leachate has great potential risks of the environmental pollution. The aim of this study was to investigate the structure effect of stacked electro-Fenton (EF) reactor on the nanofiltration (NF) concentrate treatment from landfill leachate. The stacked EF reactor was constructed with a carbon-PTFE gas diffusion cathode and an IrO_2-Ta_2O_5 anode with different electrode spacings (i.e., 2, 5, 10, and 40 mm) and electrode pairs (i.e., 1, 3, 6, and 9). Results showed that smaller electrode spacing and more electrode pairs in the stacked EF reactor improved the COD removal in the NF concentrate treatment. The specific energy consumption decreased with smaller electrode spacing but increased with more electrode pairs. Under the current density of 15 mA cm^{-2}, Fe^{2+} dosage of 560 mg L^{-1}, the stacked EF reactor with 9 electrode pairs and the electrode spacing of 2 mm removed 71 ± 6% of the total COD in the NF concentrate within 6 h and the specific energy consumption was 207 ± 20 kWh · kg COD^{-1}. The COD removal was kept stable in the stacked EF reactor within 3 cycles of operation. Three-dimensional fluorescence spectroscopic and gas chromatographic mass spectrometric analysis showed that humic acids and aromatic proteins were efficiently degraded in the EF process and large amount of aromatic hydrocarbons was detected in the treated NF concentrate. Our stacked EF reactor could be used to treat leachate concentrates with effectively degradation of the refractory organic pollutants.

1. Introduction

Landfill leachate is a highly complex wastewater and contains many refractory organics, ammonia, heavy metals, inorganic

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compounds and others (Eggen et al., 2010). Concentrations of the chemical oxygen demand (COD) can reach as high as 70900 mg L^{-1} in the landfill leachate, while the ratio of biological oxygen demand (BOD) to COD can be low as 0.04 (Renou et al., 2008). The conventional biological treatment methods cannot efficiently remove the organics in the landfill leachate to match the standard for discharge into the environment (Wiszniowski et al., 2006; Zhang et al., 2013). With the development of membrane technology, NF and reverse osmosis (RO) have been emerged as an advanced treatment technique of landfill leachate (Trebouet et al., 2001; Ahn et al., 2002; Amaral et al., 2016). The integration of the air-stripping, membrane bioreactor and NF process can remove 88% of COD, 95% of ammonia, and 100% of toxicity in the landfill leachate in a pilot scale (Amaral et al., 2016). After the pretreatment of pH modification, prefiltration, and coagulation with $FeCl_3$, the NF membrane can produce permeate satisfying the local discharge requirement (Trebouet et al., 2001). However, one of major limitations for the NF and RO processes is to generate large amount of concentrate in the landfill leachate treatment (Renou et al., 2008). The membrane concentrate contains almost all the pollutants in the influent of NF or RO process, but the volume of the concentrate was only 13%–40% of the initial volume (He et al., 2015; Fernandes et al., 2017). Thus, the pollutants concentrations were highly improved in the membrane concentrate. The COD in the membrane concentrate can reach 1000–3000 mg L^{-1} with high concentrations of inorganic salts (Zhang et al., 2013; He et al., 2015). Refractory organics, such as ethylbenzene and chlorobenzene, are also found in the membrane concentrates (Zhang et al., 2013). The characteristics of membrane (NF or RO) concentrate from the landfill leachate treatment are greatly different from the raw leachate (Zhang et al., 2013; He et al., 2015; Wang et al., 2016), resulting in difficult to efficiently remove the COD from the membrane concentrate. Thus, in recent years, much attention has been paid on the treatment of membrane concentrates from the landfill leachate (He et al., 2015; Huang et al., 2015; Zhou et al., 2016; Xu et al., 2017). Various methods have been used to treat the membrane concentrate, including a coupled process of coagulation and internal micro-elecrolysis adding hydrogen peroxide (Huang et al., 2015), Fenton oxidative-coagulation combined with photo-Fenton process (Li et al., 2016), and electro-Fenton (EF) process (Labiadh et al., 2016; Fernandes et al., 2017).

The EF process is a promising method with continuous in-situ electro-generation of H_2O_2 at a cathode via the electro-reduction of oxygen gas (Brillas et al., 2009; Nidheesh and Gandhimathi, 2012). With Fe^{2+} as catalyst, hydroxyl radicals can be efficiently produced in the EF process (Brillas et al., 2009). Compared with the conventional Fenton process, the Fenton reaction can be better controlled and the potential risk of H_2O_2 transportation and storage can be eliminated in the EF process (Nidheesh and Gandhimathi, 2012; Yu et al., 2015a). Although the EF process has been widely used to treat landfill leachate (Deng and Englehardt, 2007; Atmaca, 2009; Bashir et al., 2009; Mohajeri et al., 2010; Umar et al., 2010), only a few experiments are carried out for the membrane concentrate treatment of the landfill leachate (Mohajeri et al., 2010; Wang et al., 2012; Labiadh et al., 2016; Fernandes et al., 2017).

The reactor structure in the EF process plays an important role in the EF performance (Nidheesh and Gandhimathi, 2012; Gao et al., 2015; Yu et al., 2015b; Ma et al., 2016). Different configurations of EF reactor have been developed, such as a system with dual gas diffusion electrodes (Yu et al., 2015b), vertical flow reactor, and the sequential flow-through reactor with a 4-layer carbon nanotubes-based membrane stack (Gao et al., 2015; Ma et al., 2016). Our previous results also demonstrated that the stacked reactor with small electrode spacing was highly valuable for enhancing H_2O_2 production in the EF process. The H_2O_2 production could increase from 89.5 ± 2.1 to 663 ± 15 mg L^{-1} with increase of electrode pairs from 1 to 9 in the stacked reactor with the electrode spacing of 2 mm within less than 1 min operation (Lu et al., 2017). The maximum H_2O_2 production rate in the stacked reactor reached 1929 ± 51 mg $L^{-1}\cdot min^{-1}$, showing great potential of the stacked reactor in practice applications (Lu et al., 2017). However, the effect of the stacked EF reactor structure on treating the membrane concentrate from landfill leachate has not been reported so far.

The objective of this study was to investigate the effect of the stacked EF reactor structure on treating the concentrate of landfill leachate. Effects of electrode spacing, number of electrode pairs, and current density in the EF reactor on the COD removal in the NF concentrate were tested.

2. Materials and methods

2.1. NF concentrate

The NFNF concentrate was collected from a landfill leachate treatment plant in Guangdong province, China. An integrated process of anaerobic process, membrane bioreactor treatment (MBR) and NF was used to treat the landfill leachate. Samples from the NF concentrate were adjusted with 10% H_2SO_4 (v/v) solution to keep the pH in the samples at 3.0 for the Fenton reaction. To avoid clogging of the reactor, 0.45 μm cellulose acetate membrane was used to filtrate the acidified NF concentrate (Decarolis et al., 2001; Lu et al., 2009; Dasgupta et al., 2015; Oturan et al., 2015). The filtrated NF concentrate was preserved in refrigerator at 4.0 ± 0.2 °C before experiment. The characteristics of the pretreated NF concentrate were as follows: COD 3100 ± 400 mg L^{-1}, conductivity 30 ± 3 mS cm^{-1}, Cl^- 8580 ± 600 mg L^{-1}, SO_4^{2-} 1350 ± 150 mg L^{-1}, Na^+ 10320 ± 500 mg L^{-1}, K^+ 1940 ± 200 mg L^{-1}, Ca^{2+} 590 ± 60 mg L^{-1}, NH_3-N 60 ± 6 mg L^{-1}, NO_3-N 420 ± 40 mg L^{-1}, NO_2-N 18 ± 2 mg L^{-1}, and pH 7.5 ± 0.5.

2.2. Reactor construction and operation

The dual-chamber reactor was composed of a liquid chamber and a gas chamber. The spacing between the anode and cathode was set at 2, 5, 10, and 40 mm, respectively. Correspondingly, the liquid chamber volumes were 1.4, 3.5, 7, and 28 mL, respectively. The gas chamber was controlled at the effective volume of 3.5 mL throughout the tests. A carbon-PTFE gas diffusion electrode (GDE) was used as the cathode. A titanium plate coated with mixed metal oxides of IrO_2 and Ta_2O_5 was used as anode. The cathodes and anodes were connected in parallel to the negative and positive leads of the power supply (IT6700, ITECH Electronic Co., Ltd, Nanjing, China), respectively. The solution was stored in a glass tank and circulated using a peristaltic pump. To ensure complete effluent homogenization throughout the tests, flow rates were kept at 30 and 150 mL min^{-1}, respectively, in the liquid and gas chambers of the EF reactors. Different dosages of $FeSO_4$ were added to provide Fe^{2+} as catalyst in the EF reaction. The current density was kept at 15 mA cm^{-2} in the EF reactors throughout the tests. Each experiment was conducted in duplicate at the room temperature (28 ± 5 °C).

2.3. Analysis and calculation

Chemical oxygen demand (COD) was measured using the standard method (Clesceri et al., 1998). The pH value was determined using a pH meter (FE 20, Mettler Toledo, Swiss). The ferrous ion concentration was measured using 1,10-orthophenanthroline colorimetric method (Wang et al., 2012). The total iron concentration was determined via reducing the ferric to ferrous ions using