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Electrochemical/peroxydisulfate/Fe^{3+} treatment of landfill leachate nanofiltration concentrate after ultrafiltration

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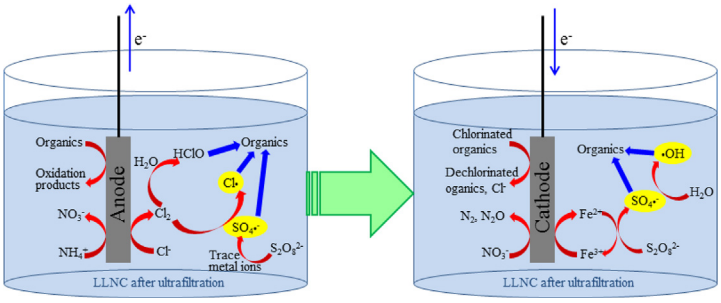
HIGHLIGHTS

- LLNC after ultrafiltration as a practical engineering problem is first studied.
- Anode/PS/Fe^{3+}-Cathode/PS/Fe^{3+} shows good performance on target solution treatment.
- Toxicity does not dramatically increase though Cl^- concentration is high.
- Less PS is consumed in a separated reactor compared to an unseparated reactor.

GRAPHICAL ABSTRACT



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ABSTRACT

Landfill leachate nanofiltration concentrate after ultrafiltration was treated. The experiments were carried out in an electrolytic cell which had an anode chamber and a cathode chamber and could be separated by a proton exchange membrane. Different electrochemical processes combined with peroxydisulfate/Fe^{3+} (PS/Fe^{3+}) were compared and the reactions in anode chamber and cathode chamber were discussed. The Anode/PS/Fe^{3+}-Cathode/PS/Fe^{3+} process shows the best effect on COD removal and color removal. The 3D-EEMFS results indicate that Anode/PS/Fe^{3+}-Cathode/PS/Fe^{3+} process has the best effect on the destruction of organics. In anode chamber, the total COD removal is ascribed to (1) the oxidation by $SO_4^{\cdot -}$ produced from PS activation by original metal ions in the target solution, (2) direct and indirect oxidations by anode, and (3) coagulation caused by Fe^{3+} after pH adjustment. In cathode chamber, the total COD removal is due to (1) the oxidation by $SO_4^{\cdot -}$ produced from PS activation by the regenerated Fe^{2+} from Fe^{3+} reduction on cathode, and (2) coagulation caused by Fe^{3+} after pH adjustment. An optimal experimental condition with current of 80 mA, Fe^{3+} dosage of 15 mM and PS dosage of 37.5 mM is obtained for Anode/PS/Fe^{3+}-Cathode/PS/Fe^{3+} process. The energy consumption between Anode/PS/Fe^{3+}-Cathode/PS/Fe^{3+} (4.42 kWh/kgCOD) and (Anode + Cathode)/PS/Fe^{3+} (4.57 kWh/kgCOD) is similar when 55% of COD is removed. However, the toxicity increase in Anode/PS/Fe^{3+}-Cathode/PS/Fe^{3+} process is obviously less than that in (Anode + Cathode)/PS/Fe^{3+} process.

1. Introduction

Sanitary landfilling is the most common disposal alternative to eliminate municipal solid wastes in many countries [1,2]. In China, more than 80% of urban wastes are disposed at landfill sites. Nevertheless, the leachate generated from landfills contains a variety of contaminants including dissolved organic matter, inorganic compounds, heavy metals and refractory organic substances which must be

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appropriately treated before discharge. To reach the discharge standard, membrane technology combined with other methods has been widely used in leachate treatment process due to its advantages of stable operation, good effect, and small occupancy. Membrane separation processes such as nanofiltration (NF) and reverse osmosis (RO) are increasingly used as a polishing step following biological treatment [3,4]. However, membrane separation processes do not destroy the pollutants in landfill leachate, but merely concentrate them into smaller volume of wastewater, i.e. about 20% to 30% of original leachate [5–7]. Besides, the concentrate is more difficult to be treated due to higher content of recalcitrant organic matters such as humic substances (HS), higher salinity, higher chromaticity and lower biodegradability compared to the original leachate [8]. Therefore, the development of effective methods for landfill leachate concentrates disposal is becoming a necessity.

Currently, the treatment methods for handling leachate concentrates mainly include recharge, membrane distillation, evaporation, and advanced oxidation processes (AOPs) [9]. However, recharge would result in accumulation of pollutants, as the recirculation of concentrates may cause an increase in chemical oxygen demand (COD), ammonia nitrogen (NH_4^+-N) and salinity, which negatively affect the performance of membrane separation process [10]. Membrane distillation brings a high cost, and the corrosion of equipment always exists in evaporation process.

AOPs are attractive methods to remove color, degrade recalcitrant organics and increase the biodegradability of concentrated leachate. Some AOPs have been applied to treat landfill leachate concentrates, such as Fenton [11,12], electro-Fenton [13], O_3 [14,15], O_3/H_2O_2 [15] and O_3/UV [15] (Table S1, Supporting Information (SI)). Other method such as electrocoagulation was also used for the treatment of landfill leachate concentrate [5] (Table S1 (SI)).

Sulfate radical-based AOPs have gained increasing attentions in water and wastewater treatment because of the good effect and stability [16], which are the use of active substances, sulfate radical ($SO_4^{\cdot -}$), generated by the activation of peroxymonosulfate (PMS) or peroxydisulfate (PS) to oxidize organic compounds [17]. Some researchers have studied the effect of sulfate radical-based AOPs on landfill leachate [18-21] (Table S1, (SI)). However, as far as we know, there is no report on the treatment of landfill leachate concentrates by using sulfate radical-based AOPs.

Taken PS as an example, $SO_4^{\cdot -}$ can be generated by the activation of PS using transition metal [17], heat [22], base [23], ultraviolet (UV) [22,24] or ultrasound (US) [25] (Eq. (1)-(2)).

$$S_2O_8^{2-} + e^- \rightarrow SO_4^{2-} + SO_4^{\cdot -} \quad (1)$$

$$S_2O_8^{2-} + \text{heat/UV/US} \rightarrow 2SO_4^{\cdot -} \quad (2)$$

Since it is inexpensive and nontoxic, ferrous ion has been widely used in PS activation (Eq. (3)) [17].

$$S_2O_8^{2-} + Fe^{2+} \rightarrow SO_4^{2-} + SO_4^{\cdot -} + Fe^{3+} \quad (3)$$

In addition, ferrous ion can be regenerated in electrochemical system via cathodic reduction from ferric ion (Eq. (4)) [26-31]. The electrochemically-assisted activation of persulfate by Fe^{2+} or Fe^{3+} has been successfully used in the treatment of 2,4,5-trichlorophenoxyacetic acid [26], acid orange 7 [27,29], pentachlorophenol [28], graywater [30], and phenol [31], etc.

$$Fe^{3+} + e^- \rightarrow Fe^{2+} \quad (4)$$

As aforementioned, landfill leachate concentrate is more resistant to be treated than landfill leachate due to the higher concentration of recalcitrant matters (e.g. HS), the higher salinity, and the lower biodegradability, etc. It is reported that the molecular weights for HS range from a few hundred to millions of daltons [32]. The degradation of HS is uneconomical due to their high aromaticity and high molecular weights. Fortunately, part of the HS could be reclaimed by extraction system (e.g. ultrafiltration and some follow-up processes) from concentrate and could be used as raw material (e.g. for fertilizer production or adsorbent production) or recover energy (e.g. incineration with garbage) [33–38]. Furthermore, the economic cost for HS extraction (about 0.5 \$/m^3 for NF concentrate with a COD reduction of nearly 1800 mg/L and an electricity price of 0.09 \$/kWh in China) should be lower than that for HS degradation. For example, with the same COD reduction (1.8 kg/m^3) and electricity price, the cost for HS degradation in NF concentrate should be at least 3.4 \$/m^3 and 6.0 \$/m^3 in electrochemical process and ozonation process, respectively, since the lowest energy consumption is around 21 kWh/kgCOD for electrochemical process [8,39] and 37 kWh/kgCOD for ozonation process [40]. Thus, HS extraction from leachate concentrate has been implemented in engineering application in recent years and has been recommended in the technical guideline for leachate treatment of municipal solid waste (RISN-TG023-2016) in China [38]. Ultrafiltration is the most commonly-used technology for HS extraction and the extraction ratio on HS depends on their molecular configurations and the operational parameters [37]. Generally, the organics with large molecular weight would be more likely to be extracted. As for the ultrafiltration related to the present study (Fig. S1 (SI)), about 1800 mg/L COD could be separated and the main residues in the concentrate were fulvic acid-like (FA-like) substances (see the 3D-EEMFS results of target solution in the present study). However, Yoon et al. have found that the landfill leachate organics with large molecular weight are more likely to be removed in both Fenton reaction and coagulation [41]. Besides, Wang et al. have found that the humic acid-like (HA-like) substances can be completely removed from landfill leachate NF concentrate (LLNC) in O_3/H_2O_2 or O_3/UV process with the formation of small molecular hydrophilic substances, while the removal of FA-like substances is not significant [15]. Thus, even after ultrafiltration, the concentrate is still difficult to be treated by either oxidation or coagulation.

In the present study, different electrochemical processes combined with PS/Fe^{3+} technique were first employed to deal with a typical LLNC after ultrafiltration. The aims of the current paper are to: (1) develop a feasible method to deal with landfill leachate concentrate after ultrafiltration, which is a practical engineering problem on leachate concentrate treatment and is barely studied so far, (2) reveal the functions of anode and cathode in the electrochemical/PS/Fe^{3+} process and develop an efficient process, and (3) disclose the mechanisms of the treatment process.

2. Material and methods

2.1. Characteristics of target concentrate

The target concentrate (LLNC after ultrafiltration) was collected from a leachate membrane process of solid waste landfill located in Jiangsu Province, China. The age of the landfill is around 8 years. Table 1 lists the main characteristics of the target concentrate. The process of the leachate treatment is shown in Fig. S1 (SI).

2.2. Experimental procedure

The different treatment processes are shown in Fig. S2 (SI). Generally, the treatment of target solution was carried out with oxidation and coagulation in Anode/PS/Fe^{3+}, Cathode/PS/Fe^{3+}, coexistence of Anode and Cathode combined with PS/Fe^{3+} ((Anode + Cathode)/PS/Fe^{3+}), Anode/PS/Fe^{3+} followed by Cathode/PS/Fe^{3+} (Anode/PS/Fe^{3+}-Cathode/PS/Fe^{3+}) and Cathode/PS/Fe^{3+} followed by Anode/PS/Fe^{3+} (Cathode/PS/Fe^{3+}-Anode/PS/Fe^{3+}) processes. For comparison, the electrochemical process with anode and cathode coexistence in one chamber (Anode + Cathode) was also investigated, and only oxidation occurred in this process.

The oxidation experiments were carried out in an H-type reactor