



Effective removal of contaminants in landfill leachate membrane concentrates by coagulation



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HIGHLIGHTS

- Comparison of iron coagulants for treatment of leachate membrane concentrates.
- Effect of anions (Cl^- , SO_4^{2-} , and NO_3^-) on Fe(III) coagulation.
- Response surface method.
- >80% COD and TOC removals at relatively low FeCl_3 coagulant dose.

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ABSTRACT

Leachate membrane concentrates containing high concentrations of organics and trace toxic compounds pose a major threat to the environment, and their treatment is an urgent issue. In this work, various coagulants were used to treat leachate membrane concentrates. Appropriate pH values for treatments with FeCl_2 , FeSO_4 , polyaluminum chloride, and FeCl_3 were 3, 5, 5, and 4, respectively. FeCl_3 achieved the highest total organic carbon (TOC) removal efficiency. The effect of the various anions in ferric coagulants [FeCl_3 , $\text{Fe}_2(\text{SO}_4)_3$, and $\text{Fe}(\text{NO}_3)_3$] on the TOC removal efficiency was negligible. The main organics remaining in the leachate membrane concentrates after coagulation were humic and fulvic acids. The conditions for coagulation with FeCl_3 were optimized using the response surface method (RSM). The highest TOC, chemical oxygen demand (COD), and chromaticity reduction efficiencies, 81%, 82%, and 97%, respectively, were achieved at pH 4 using FeCl_3 (5 g L^{-1}) and polyacrylamide (PAM; 0.07 g L^{-1}). The COD of leachate membrane concentrates was reduced from 4000 to 718 mg L^{-1} . The mole ratio of removed COD and Fe(III) (2.4 mol) at 5 g L^{-1} FeCl_3 (pH 4, PAM 0.07 g L^{-1}) was lower than that (3.8 mol) at 3 g L^{-1} FeCl_3 (pH 4, PAM 0.07 g L^{-1}); based on the cost and COD removal efficiency, the latter conditions were the best choice. Our work provides guidelines for the treatment of leachate membrane concentrates in engineering.

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1. Introduction

In most developing countries, landfill disposal is considered to be a suitable strategy for waste management (Chong et al., 2005; Rowe and Yu, 2012). Large amounts of leachates are generated as a result of rainfall percolation and degradation of organic wastes (Justin and Zupancic, 2009). Landfill leachate, which is a highly complex wastewater, contains high concentrations of organics,

heavy metals, ammonia, and toxic compounds (Ahmed and Lan, 2012). Some traditional leachate remediation techniques can't meet the Chinese Standard (GB 16889-2008) for Pollution Control on Landfill Sites for Municipal Solid Waste (MEP, 2008). Leachate effluents can almost meet these strict discharge criteria following membrane treatment, which has become increasingly popular for landfill leachate treatment in some western European countries and China (Alvarez-Vazquez et al., 2004; Robinson, 2007; Zheng et al., 2007). However, this process leaves a hidden danger – leachate membrane concentrates, which contain a great amount of refractory pollutants and saline compounds (Calabrò et al., 2010; He et al., 2015). These concentrates typically represent 13–30% of

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the volume of the leachate influent. The predominant fractions of dissolved organic materials inside leachate membrane concentrates are hydrophilic organic and humic substances, which have low biodegradability (Zhang et al., 2013; Clarke et al., 2015). Humic substances readily interact with organic chemicals and metal ions, thus having a significant effect on the environmental fate, mobility, and transport of chemical species (Borggaard et al., 2011; Tang et al., 2014). Humic substances can be classified into three main types, namely humin (insoluble throughout the whole range of pH-values), humic acid (soluble only under alkaline conditions), and fulvic acid (soluble in aqueous media) (Stevenson, 1994).

Some wastewater treatment techniques are used to treat leachate membrane concentrates, e.g., recirculation (Liu et al., 2011), evaporation (Xie et al., 2011), membrane distillation (Qi et al., 2015), adsorption (Wang et al., 2013), electrodialysis (Li et al., 2015), coagulation (Huang et al., 2015), and advanced oxidation processes (Wan et al., 2014). However, the methods which can effectively remove the contaminants in leachate membrane concentrates are few, and some of them are expensive (Li et al., 2015; Qi et al., 2015). Among these, recirculation into a membrane bioreactor (MBR) is the simplest and most widely used technique for treating leachate membrane concentrates, mainly because it is cost-effective (Calabrò et al., 2010; Talalaj, 2014). However, repeated recirculation of leachate membrane concentrates can result in salt accumulation in the MBR, and this adversely affects biological processes (Chen et al., 2008; Fang et al., 2011).

The high concentration of refractory pollutants and saline compounds in leachate membrane concentrates results in low biodegradability. Physicochemical treatments are therefore always the first choices (Singh and Tang, 2013). Coagulation is a simple and cost-effective technique for the treatment of leachate membrane concentrates. It is promising for the removal of dissolved organic materials (Comstock et al., 2011). Also, it could improve the biodegradability of leachate membrane concentrates (Wang and Le, 2016). Zhou et al. (2011) found that coagulation with FeCl_3 removed 26% of dissolved organic carbon (DOC) in reverse osmosis concentrates (ROC) produced in a municipal wastewater reclamation reverse osmosis plant. Dialynas et al. (2008) showed that up to 52% removal of DOC in ROC could be achieved. The type and concentration of coagulant used affects the DOC removal efficiency, therefore selection of an appropriate coagulant, determination of the optimum experimental conditions, assessment of the effect of pH, and determination of the optimum reagent dosage are necessary for performance optimization (Amor et al., 2015).

Previous reports about the treatment of leachate membrane concentrates have focused on contaminant removal by coagulants without a consideration of mechanisms (An and Xu, 2013; Qin and Chen, 2016). We sought to understand coagulation treatments of leachate membrane concentrates more comprehensively. The objectives of this study were (1) to identify suitable coagulants for leachate membrane concentrates and clarify the contaminant removal mechanism; (2) to optimize the performance [chemical oxygen demand (COD), total organic carbon (TOC), and chromaticity reduction efficiencies] of the coagulation process and provide guidelines for the pretreatment of leachate membrane concentrates.

2. Materials and methods

2.1. Leachate membrane concentrates

The leachate membrane concentrates obtained from a municipal solid waste landfill site located in Changzhou, China were stored at 4 °C before treatment process. This landfill entered service in 2003. The raw leachate was first treated in an MBR, and the

resulting effluent was nanofiltered. The MBR effluent was pumped into the nanofiltration device. A spiral curling nanofiltration membrane was used with operational pressure of $(5\text{--}25) \times 10^5$ Pa. The refractory macromolecule organics and some metal ions in leachate were retained while the cleaned water was discharged directly on the opposite side of the nanofiltration membrane. The leachate membrane concentrate typically represents 20% of the volume of the leachate influent. The main leachate characteristics were pH 6.6 ± 0.1 ; conductivity 25.3 ms cm^{-1} ; COD concentration $4135 \pm 140 \text{ mg L}^{-1}$; BOD_5 $124 \pm 5 \text{ mg L}^{-1}$; TOC concentration $1440 \pm 115 \text{ mg L}^{-1}$; and chromaticity 250 ± 10 .

2.2. Coagulation process

All chemicals used were analytical grade and purchased from Shanghai Lingfeng Chemical Reagent Co., China. Coagulation tests were performed in beakers using various Fe amounts added to 100 ml of sample as FeCl_2 , FeSO_4 , FeCl_3 , $\text{Fe}_2(\text{SO}_4)_3$, and $\text{Fe}(\text{NO}_3)_3$, and polyaluminum chloride (PAC) at pHs varying from 3 to 11. The solution pH was adjusted using 2 M H_2SO_4 or 10 M NaOH. After 1 min rapid stirring (180 rpm) in coagulation process, 12 min slow stirring (60 rpm) in flocculation process and 12 h settling (4 h enough actually) at ambient temperature, the supernatant was collected and the TOC concentration was determined. The optimal experimental conditions were obtained on the basis of preliminary experiments. It was found that the best coagulation was achieved with FeCl_3 $1\text{--}5 \text{ g L}^{-1}$, PAM $0.03\text{--}0.07 \text{ g L}^{-1}$, at pH ranging from 3 to 5, in all the cases.

Finally, the most suitable coagulant for the leachate membrane concentrates was identified. The coagulation process was designed and optimized using the response surface method (RSM). Box–Behnken design (BBD) was used to optimize the three main factors i.e., pH, and FeCl_3 and polyacrylamide (PAM) concentrations, in the coagulation process. All experiments were performed in triplicate.

Analysis of variance (ANOVA) was used for graphical analyses of the data to identify the interactions between the process variables and responses (Chafari et al., 2008). The coefficient of determination (R^2) indicated the quality of the polynomial fitting model, and the Fisher's F-test in the same program was used to check the statistical significance. Model terms were evaluated based on the *P*-value (probability), with a 95% confidence level (Chafari et al., 2008). For each response, the square of the correlation coefficient was computed as R^2 . The model calculations agreed well with the experimental data for experiments with a high R^2 value (Bashir et al., 2010); R^2 should be more than 0.8 for a reasonable model. The predicted R^2 (Pred R^2) should be in reasonable agreement with the adjusted R^2 (Adj R^2) (Lee et al., 2010). The coefficient of variation (CV), defined as the percentage ratio of the standard deviation to the mean value, was used as a measure of the reproducibility of the model; a model was considered to be reasonably reproducible if its CV was not greater than 10% (Li et al., 2010).

2.3. Analytical methods

The COD, BOD_5 and chromaticity were measured using standard methods (APHA, 2005). Conductivity was measured using a conductivity meter (S30, SevenEasy, Swiss). The TOC was measured using a TOC analyzer (TOC-5000A, Shimadzu, Japan) after filtration of the samples through a $0.22 \mu\text{m}$ filter membrane (Tianjin Jinteng laboratory equipment Co., China). The samples treated with FeCl_3 , $\text{Fe}_2(\text{SO}_4)_3$, and $\text{Fe}(\text{NO}_3)_3$ under the optimum conditions, and a leachate membrane concentrate sample were dried in an oven (40 °C) for 48 h, mixed with KBr, and pressed into disks for Fourier-transform infrared (FT-IR) analysis (VERTEX 70, Bruker, Germany)

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