Research of pretreatment for reverse osmosis in landfill leachate treatment process

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ABSTRACT

Landfill leachate treated by reverse osmosis poses significant environmental challenges due to high conductivity and chemical oxygen demand (COD) in southwest China. This study investigates the efficacy of a coagulation + Fenton pretreatment in enhancing the performance of reverse osmosis (RO) systems for landfill leachate treatment. The study monitored the conductivity and osmosis pressure of landfill leachate pre-treated with coagulation and Fenton processes. Coagulation was optimized at pH 6.5 using polyferric sulfate (PFS) as the coagulant, followed by the addition of H_2O_2. Pretreatment reduced leachate conductivity from 18.62 mS/cm to 9.68 mS/cm and decreased the average osmosis pressure by 3.2 bar. The optimal COD removal rate of 64.03% was achieved with a PFS dosage of 1.2 g/L and H_2O_2 dosage of 8 mL/L. The findings suggest that the coagulation and Fenton pretreatment significantly enhances RO efficiency by lowering conductivity and osmosis pressure. The reduction in COD indicates improved organic pollutant removal, crucial for minimizing RO membrane fouling. The pretreatment method substantially improves RO performance in landfill leachate treatment, offering a viable approach for managing high-conductivity waste streams.

Keyword: Conductivity, osmosis pressure, pretreatment, DTRO

1. INTRODUCTION

Conventional treatment technologies for landfill leachate are divided into physical and chemical treatment technologies, biological treatment technologies, and land treatment technologies. Since 2008, these methods are no longer for treating high-concentration landfill leachate due to unable to consistently meet new landfill pollutant control standards[1-8].

Landfill leachate imposes a significant pollution load on RO membranes. The water production rate of DTRO is limited by the membrane's separation capacity. It is necessary to increase physical parameters such as operating pressure to cope with the continuously rising salinity of the leachate. Theoretically, the water production rate will gradually decrease and approach 0 as the net driving pressure approaches its upper limit. Adding antiscalants and performing chemical cleaning can temporarily alleviate membrane fouling, but these measures will accelerate membrane aging. Therefore, it is crucial to explore suitable pretreatment processes to mitigate the pollution load.

Fenton oxidation and coagulating sedimentation are both widely used in landfill leachate treatment. The Fenton oxidation method involves the oxidation and decomposition of organic matter through electron transfer and other mechanisms. During this process, Fe^{2+} is oxidized to Fe^{3+}, resulting in coagulation and precipitation that facilitates the removal of organic matter. Under the catalytic influence of Fe^{2+}, H_2O_2 decomposes to generate hydroxyl radicals $\cdot OH$, which oxidize organic compounds, breaking them down into smaller molecules. This reaction leads to the formation of precipitates that effectively remove organic matter from the solution. Coagulation + Fenton can be used as pretreatment before the DTRO system to improve the efficiency of reverse osmosis membrane. This paper observes coagulation + Fenton oxidation as the pretreatment process in landfill leachate treatment by DTRO. The causes of the influence are analyzed and the corresponding optimization methods are explored[9-13].

2. EXPERIMENT AND EQUIPMENT

2.1 Coagulation

The polymeric iron sulfate (PFS), polymeric aluminum chloride (PAC), and polymeric aluminum ferric chloride (PAFC) were selected as coagulants for comparison in this experiment. The pH value was adjusted to 6.5 with H_2SO_4 and NaOH

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Fifth International Conference on Green Energy, Environment, and Sustainable Development (GEESD 2024),
edited by M. Aghaei, X. Zhang, H. Ren, Proc. of SPIE Vol. 13279, 132793G · © 2024 SPIE · 0277-786X
 · doi: 10.1117/12.3044421

solution. Coagulants were added in a six-link stirrer. Then, the mixture was vigorously coagulated at 300 r/min for 2 minutes, stirred at 100 r/min for 20 minutes, stood for 30 minutes. The supernatant fluid was extracted for detection and analysis.

2.2 Fenton oxidation

Fenton reagent was added to the leachate after coagulation and precipitation treatment. The pH value was adjusted and stirred at 300 r/min and 100 r/min for 1 min and 60 min respectively in a six-link mixer, then stood for 30 minutes. The COD concentration was detected in the supernatant fluid.

2.3 Coagulation + Fenton + DTRO

The leachate which was pretreated by coagulation + Fenton connected into the first stage DTRO. The changes of osmotic pressure were detected.

2.4 Main equipment and reagent

The main experimental equipment includes the 6511 electric mixer and the SC766 experimental mixer.

The reagents include: H_2O_2 (30%), PFS, PAC, PAFC, ferrous sulfate heptahydrate, concentrated sulfuric acid, sodium hydroxide, distilled water, etc.

The main pollutants of leachate are shown in Table 1.

Table 1. Main pollutant concentration of leachate.

Pollutants	SS	TP	COD	NH_3-N	BOD_5	TN
Concentration (mg/L)	118	33.2	2.23×10^3	196.7	440	292.3

3. EXPERIMENT RESULTS AND ANALYSIS

3.1 Comparison of coagulants

The removal efficiency of PFS, PAC, and PAFC was compared. The dosage of the drug is 0.2~1.2 g/L. Fe_2O_3 mass fraction of PFS≥24.40%; Al_2O_3 mass fraction of PAC≥29.05%; Fe_2O_3 mass fraction ≥2.46% and Al_2O_3 mass fraction ≥23.60% in PAFC. The removal efficiency of these three coagulants on SS are shown in Figure 1.

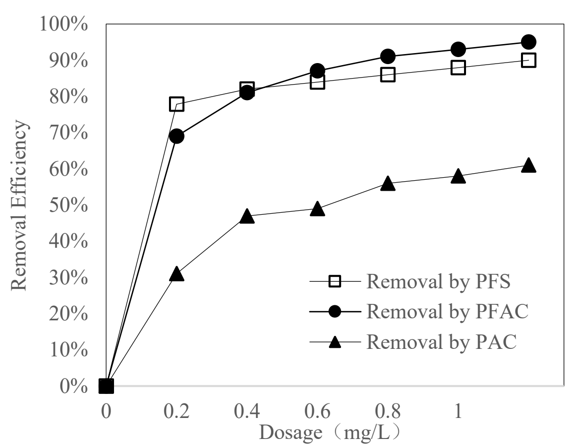

Figure 1. Comparison of coagulants on SS removal.

The removal rate of SS steadily increases with the increase of flocculant dosage. The removal rate of PFAC exceeds PFS's after the dosage reaches 0.4 g/L. The PFAC is a new polymer synthesized by copolymerization of aluminum and iron, which combines the characteristics of traditional aluminum and iron salts. But there will be a destabilization and re stabilization process of the colloid as the dosage of flocculant increases. The removal efficiency of PFS increases more rapidly and more stable. Therefore, the PFS was chosen as the coagulant in this experiment.

3.2 Optimization of dosage

The COD of the raw leachate is 2.23 g/L. The dosage of PFS is 0.2~1.4 g/L. Vigorously coagulate for 2 minutes, stir slowly for 30 minutes, then stand for 30 minutes to extract the supernatant. The COD removal efficiency is shown in Figure 2.

The mass concentration of H_2O_2 is 30%, with a mass density of 1.122 g/L, a molecular weight of 34, and a molar concentration of 9.9 mol/L approximately. The dosage of H_2O_2 is 2-12 mL/L and $FeS0_4 \cdot 7H_2O$ is 2 g/L, the pH value is 4, and the oxidation time is 2 hours. The changes of COD removal efficiency by dosage are shown in Figure 3.

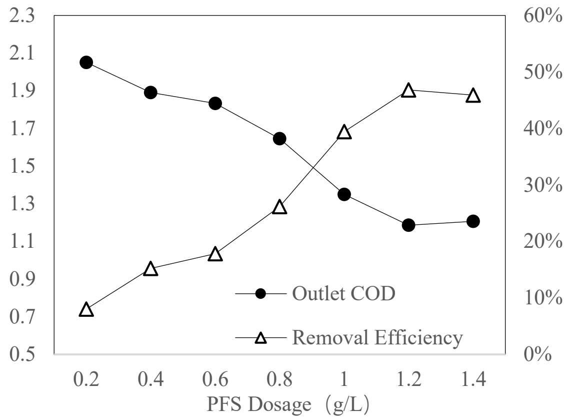

Figure 2. Effect on removal of COD by PFS.

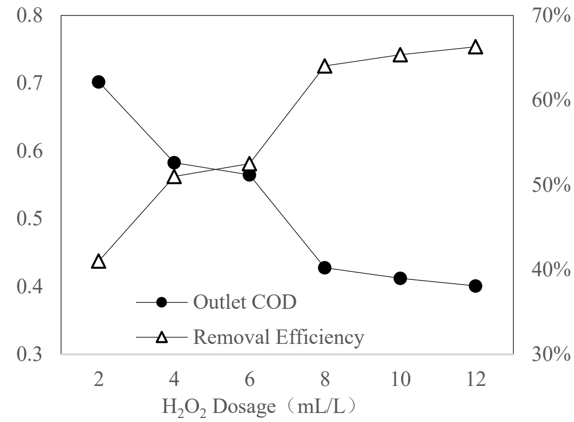

Figure 3. Effect on removal of COD by H_2O_2.

The removal rate reaches 46.82% when the PFS dosage is 1.2 g/L. The colloidal surface will be coated with coagulant particles and losing its ability to re aggregate with more polymeric ferric sulfate. There would be instability and difficulty in generating multi-core hydroxyl polymers. The flocculation effect decreases, and the COD removal efficiency also decreases when the PFS dosage was 1.2 g/L in the experiment.

The removal rate reaches 64.03% when the H_2O_2 dosage is 8 mL/L. The more H_2O_2 added, the more ·OH produced, the stronger oxidized, and the higher removal rate of COD achieved. The change of removal rate slows down after exceeding 8 mg/L. Excessive addition of H_2O_2 will affect the economic efficiency of system operation. In this experiment, the dosage of H_2O_2 was at 8 ml/L.

3.3 Osmosis pressure after pretreatment

The raw leachate pretreated by coagulation + Fenton was connected into a single-stage DTRO. The recovery rate was set to 90%. The osmosis pressure is shown in Figure 4.

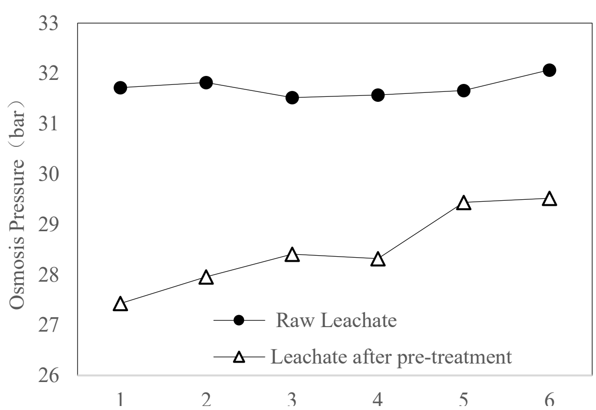

Figure 4. Comparison of the operating pressure.

As shown in experimental results. The conductivity of the leachate pretreated by coagulation + Fenton decreased from 18.62 mS/cm to 9.68 mS/cm under the same recovery rate. The average osmosis pressure was 31.73 bar of untreated leachate. Then the pressure was 28.51 bar under pretreatment. There is a decrease of 3.2 bar compared with untreated. The service life of the membrane is related to the backwash frequency and operating pressure. Therefore, the pretreatment process can reduce the pollution load of RO membranes to improve the operational performance of the DTRO system. And the service life of RO membranes can be further extended.

3.4 Comparison with other pretreatment methods

Due to the large variability in water quality and quantity, complex composition, high concentrations of organic matter and ammonia nitrogen, imbalance in microbial nutrient ratios, and high pollutant shock loads, conventional biological and biochemical methods face challenges in maintaining stable treatment efficiency. Biological reactors like UASB require sophisticated operational management and involve high investment costs. In contrast, coagulation and Fenton oxidation methods only require adjustment of dosage according to water quality changes to achieve relatively stable treatment results. For small to medium-sized municipal landfills with limited availability of skilled personnel and relatively backward management capabilities, using physicochemical methods as pretreatment is more suitable.

3.5 Analysis of pretreatment impact

The more OH^- ions produced, the better the oxidation and degradation of organic compounds. A higher production of OH^- ions by increasing the dosage of H_2O_2. The rate of $\cdot OH$ production slows down when the dosage ratio exceeds a certain point and the concentration of H_2O_2 surpasses that of Fe^{2+}. The higher concentration of Fe^{2+} leads to excess Fe^{2+} being oxidized by H_2O_2 to Fe^{3+}, which consumes $\cdot OH$ excessively and hinders the degradation of organic matter. Pretreatment can slow down the accumulation of salinity in the landfill area. And the growth of osmotic pressure can be controlled. Pretreatment is more conducive to reducing membrane fouling and extending membrane life compared with RO cleaning[14-16].

4. CONCLUSION

The coagulation of traditional iron and aluminum salts is very fast. However, the floc would be unstable for fast hydrolysis process. The products formed by inorganic polymer flocculants have the advantage of relatively stable hydrolysis within a certain stage. And the sedimentation removal efficiency of flocs is higher.

The production rate and amount of hydroxyl radicals $\cdot OH$ will directly affect the oxidation reaction. The concentration of H_2O_2, $FeSO_4$, and pH in the leachate can all affect the production of hydroxyl radicals $\cdot OH$. It should be based on the actual composition of pollutants to decide the dosage of H_2O_2, $FeSO_4$, and pH value, molar ratio.

The experiments show that the combination of the oxidative reduction for COD by Fenton oxidation and flocculating Settling for pollutant particles by coagulation is efficient. The pretreatment of coagulation + Fenton can effectively reduce the operating pressure of DTRO and extend its service life. And this pretreatment process has been running well for up to a year. The operating cost of reverse osmosis with pretreatment does not increase significantly, and the dosage of chemicals can be reduced by using photocatalysis and electrocatalysis. The cost advantage will gradually apparent in the middle and later stages of landfill operation.

REFERENCES

[1] Renou, S., Givaudan, J. G. and Poulain, S., "Landfill leachate treatment: Review and opportunity," Journal of Hazardous Materials 150(3), 468-493 (2008).

[2] Baawain, M. S. and Al-Futaisi, A. M., "Studying groundwater quality affected by barka dumping site: an integrated approach," Arabian Journal for Science and Engineering 39(8), 5943-5957 (2014).

[3] Matosić, M., Terzić, S., Korajlija, J. H., Mijatović, I. and Ahel, M., "Treatment of a landfill leachate containing compounds of pharmaceutical origin," Water Science & Technology 58(3), 597-602 (2008).

[4] Ahel, M., Mikac, N., Cosovic, B., Prohic, E. and Soukup, V., "The impact of contamination from a municipal solid waste landfill (Zagreb, Croatia) on underlying soil," Continuum 37(8), 203-210 (2006).

[5] Ikem, A., Osibanjo, O., Sridhar, M. K. C. and Sobande, A., "Evaluation of groundwater quality characteristics near two waste sites in Ibadan and Lagos, Nigeria," Water, Air, & Soil Pollution 140(1), 307-333 (2002).

[6] Kjeldsen, P., Barlaz, M.A., Rooker, A.P, Baun, A., Ledin, A., et al., "Present and long-term composition of MSW landfill leachate: a review," Critical Reviews in Environmental Science and Technology 32(4), 297-336 (2002).

[7] Luo, C. Y., Hu, Y. Y., Chen, Y. M. and Wu, S. M., "Theoretical study on the effect of leachate recirculation from refuse landfill," China Water & Wastewater 19(2), 5-7 (2003).

[8] Qin, L. and Zhou, G., "Research on and development of ferric-salt composite flocculants," Industrial Water Treatment 23(2), 15-18 (2003).

[9] Linke, X. U., "Study on coagulating treatment of waste leachate by polyaluminum chloride and polyacrylamide," Journal of Anhui Agricultural Sciences 39, 10656928 (2011).

[10] Cheng, L. H., Huang, J. L. and Ni, F. X., "A kinetic study on the generation of ·OH by Fenton reagent," Journal of Environmental Pollution Control Technology and Equipment 4(5), 12-15 (2003).

[11] Zhao, B. Q., Chen, S., Sun, D. Z. and Chung, J. S., "Removal of refractory pollutants in landfill leachate by Fenton process," Journal of Harbin Institute of Technology 39(8), 1285-1288 (2007).

[12] Zhang, H. and Huang, C. P., "Treatment of landfill leachate by Fenton process," China Water & Wastewater 17(3), 1-3 (2001).

[13] Zhu, Z. L., Sun, M. and Wang, H. L., "Research on Fenton oxidation pretreatment of landfill leachate," Journal of Ecology and Environment 19(10), 2484-2488 (2010).

[14] Li, Z., Bai, J., Li, Y. and Wang, F., "Removal of refractory organics from landfill leachate by in situ electrogenerated H_2O_2 combined with an Fe0 Fenton-like process," Waste Manag Res 41(5), 1004-1013 (2023).

[15] Liu, H., Huang, J. and Zhang, H., "Effect and mechanism of Mn^{2+} on urease activity during anaerobic biological treatment of landfill leachate," Environmental Science and Pollution Research 31, 41290-41300 (2024).

[16] Kuppam, C., et al., "Innovative electrocoagulation strategies for landfill leachate treatment: Comparative analysis of aluminum and iron electrodes," Separation Science and Technology. Published online: 07 Mar 2024.