Environmental Science
Advances

ROYAL SOCIETY
OF CHEMISTRY

PAPER

View Article Online
View Journal

Cite this: DOI: 10.1039/d6va00153j

Advanced treatment of landfill leachate reverse osmosis concentrate *via* an integrated coagulation–sedimentation–electrochemical oxidation–biological aerated filter process

Hao Zheng,[a] Salma Tabassum, *[bc] Jun Li,*[a] Guanglei Li,[a] Hüseyin Altundag[bc]
and Ningwen Zhang[a]

Reverse osmosis membrane filtration concentrate is a type of organic wastewater from landfill leachate treatment that has a high concentration of organic matter, refractory macromolecules, and low biodegradability. The reverse osmosis concentrate was treated in this study using a combination of physicochemical and biological processes. Single-factor test analysis for coagulation and sedimentation indicated that 2000 mg L^{-1} of polymeric ferric sulphate (PFS), a pH of 7, and a precipitation time of 40 min yielded the best removal effect. The COD, NH_3–N, and UV_{254} removal efficiencies were 74%, 34%, and 64%, respectively. The influent of the electrochemical oxidation is the effluent pretreated by coagulation and precipitation. The anode material was a ruthenium–iridium–titanium plate electrode, and the cathode material was a stainless-steel plate electrode. The removal efficiencies of COD, NH_3–N, and UV_{254} in the electrochemical oxidation process were 67%, 83%, and 80%, respectively. The BOD_5/COD ratio increased to 0.5, and the unit energy consumption of COD was 39.4038 kW h kg^{-1}. The electrochemical oxidation effluent was then subjected to advanced treatment in an aerated biological filter. The influent flow rate was 0.3 L h^{-1}, the air-to-water ratio was 4 : 1, and the temperature was 20 °C–30 °C. No glucose was added to the subsequent influent. The COD, NH_3–N, and UV_{254} removal efficiencies were 71%, 73%, and 69%, respectively. Following biological treatment in the aerated biological filter, the B/C ratio of the reverse osmosis membrane concentrate improved. The operational results of the combined processes indicated removal efficiencies of 97%, 98%, and 97% for COD, UV_{254}, and NH_3–N, respectively. The effluent quality attained the Class A standard of the Pollutant Discharge Standard for Urban Sewage Treatment Plants (GB18918-2002).

Received 31st March 2026
Accepted 25th May 2026

DOI: 10.1039/d6va00153j

rsc.li/esadvances

Environmental significance

A combined coagulation–sedimentation–electrochemical oxidation–biological aerated filter process was employed for the advanced treatment of reverse osmosis membrane filtration concentrate, which is a type of organic wastewater from landfill leachate treatment. The integrated process presents a clear synergistic mechanism: coagulation removes macromolecular humus *via* charge neutralization and adsorption bridging. Coagulation removed the suspended solids and a portion of humic acids (64% UV_{254} removal), thereby reducing organic load on the electrooxidation unit and mitigating electrode fouling. Electrochemical oxidation, dominated by active chlorine, improves biodegradability and removes ammonia nitrogen; BAF mineralizes small-molecular-weight organics. Combined treatment removal efficiencies of COD, UV_{254}, and NH_3–N are 97.46%, 98%, and 96.8%, respectively. The effluent quality attained the Class A standard of the Pollutant Discharge Standard for Urban Sewage Treatment Plants.

1. Introduction

According to statistical data, the daily waste output per person is 400–450 kg per year.[1] Currently, the treatment of landfill leachate has become a key issue.[2] Reverse osmosis is frequently used in advanced treatment due to the increasingly stringent regulations on landfill leachate discharge. The reverse osmosis membrane treatment technique provides a superior initial treatment effect by removing concentrated solutions. However, it produces effluents with high concentrations of organic pollutants and poor biochemical properties, which makes it unsuitable for the direct adoption of biological treatment technology.[3,4] The membrane system is the final stage of treating landfill leachate, and improper disposal can lead to secondary pollution. In practical engineering applications,

---

[a] *School of Municipal and Environmental Engineering, Shenyang Jianzhu University, Shenyang, 110168, China. E-mail: junlee@sjzu.edu.cn*

[b] *Department of Chemistry, Faculty of Science, Sakarya University, Sakarya, 54187, Turkey. E-mail: tsalma@sakarya.edu.tr; salmazenith@gmail.com*

[c] *Biomedical, Magnetic and Semiconductor Materials Research Center (BIMAS-RC), Sakarya University, Sakarya, 54187, Turkey*

concentrated liquids are treated using advanced oxidation processes (AOPs), recharge methods, and evaporation methods.[5,6] Recharge technology is a simple, economical, and effective method for processing concentrates; however, several issues remain.[7,8] There are some contamination risks to nearby groundwater when using the recharge method to treat landfill leachate concentrate. Although this pollution risk may not be apparent in the short term, it will ultimately lead to salt accumulation and reduced water production efficiency. It poses a risk of unbalanced osmotic pressure across the entire combined process system due to the excessively high salt content that accumulates during treatment. Ultimately, if it is not managed promptly, it harms the ecological environment.[4,9] Although the standard high-temperature evaporation process does not pose the dangers mentioned above, it can still cause problems, such as equipment corrosion, in practical engineering. Before this study, the water quality index of the concentrated liquid treated with the landfill leachate membrane was analyzed, and it was found that advanced oxidation technology is a more suitable choice for treating this concentrated liquid.[10–13] However, advanced oxidation technology is essentially a physical and chemical reaction. When it is applied to membrane filtration concentrate, it must be combined with other processes to achieve a combined treatment.

In an experimental study by Xiaoyun *et al.*,[14] the two coagulation precipitations effectively degrade a large number of high-molecular-weight organic compounds in the membrane rate concentrate, converting them into lower-molecular-weight organic compounds, which is beneficial for the subsequent photoelectric oxidation treatment. According to Yunhai's test results,[15] COD and chromaticity can be effectively removed from water samples using coagulation and sedimentation. Studies have shown that the type of coagulant has a strong relationship with effluent quality and economic costs in the treatment of concentrated liquid from landfill leachate after membrane filtration. The concentrated solution contains a high concentration of negatively charged humic acid colloidal particles. Adding a positively charged coagulant to the water sample is a reasonable approach for treating the concentrate.[16] Thus, a coagulation–sedimentation–electrochemical oxidation–biological aerated filter (BAF) is employed in this study to treat the concentrate resulting from reverse osmosis membrane treatment of landfill leachate. Pretreatment is necessary before the electrochemical advanced oxidation technology to treat the membrane filtration concentrate. In real-world applications, the concentrate can be efficiently treated to remove high levels of organic pollutants and control costs through rational process design. Adding a specific coagulant to the water allows the suspended colloidal ions or tiny biological particles to adsorb and combine, which is the fundamental mechanism of the coagulation–precipitation reaction.

The mechanisms of coagulation and precipitation primarily involve compression of the electric double layer,[17] charge neutralization, adsorption bridging, and entrapment/sweeping. The coagulation and sedimentation process is commonly employed as a pretreatment step in the advanced treatment flow of the membrane filtration concentrate stage or in the majority of landfill leachate treatment processes.[18,19] The operating mechanism of electrochemical oxidation involves placing a solution or suspension of organic wastewater into the reaction device and applying a DC power supply to oxidize organic matter or to oxidize low-valence metal ions into high-valence metal ions. The organic matter in the solution are then oxidized by high-valence metal ions.[11,20] According to the different oxidation mechanisms, electrochemical oxidation treatment technology can be divided into two treatment processes: direct oxidation and indirect oxidation. In this study, the anode adopted is a $Ti/RuO_2–IrO_2$ active anode, which is significantly different from non-active anodes (*e.g.*, BDD, PbO_2) that generate a large number of hydroxyl radicals ($\cdot OH$). This active anode has extremely weak $\cdot OH$ production capacity, and its surface mainly forms high-valent metal oxide active sites ($MO_x(\cdot O)$). In the presence of high Cl^- concentration, it preferentially catalyzes the conversion of Cl^- into active chlorine species (Cl_2, HOCl, and ClO^-) for indirect oxidation of pollutants. In the two oxidation processes, organic pollutants in the solution are degraded and removed together. Electrochemical oxidation reactions can be conducted at room temperature, with low investment costs, simple-to-operate equipment, and optimum reaction conditions. This results in more effective removal in a short time, with controllable energy input and without the need for external chemical oxidants. Compared to other biological treatment methods, the biological aerated filter (BAF) offers higher benefits for removing organic pollutants from solution. This device can facilitate contact oxidation between organisms and achieve solid–liquid separation as a fast filter.[21] The operating principle of a biological aerated filter can be explained in three aspects: filtration-retention, biodegradation, and biological flocculation.[22,23]

In this study, a small-scale laboratory test was conducted to verify the feasibility of the combined process. The aim is to explore a new, efficient, low-cost, and environmentally friendly combined "coagulation–sedimentation–electrochemical oxidation–biological aerated filter" treatment process for the concentrate after reverse osmosis membrane treatment of landfill leachate. Recently, extensive studies have been conducted on membrane-based treatment and nitrogen removal for landfill leachate.[24–26] Although the coagulation–advanced oxidation–BAF combined process has been applied to leachate treatment, most studies have focused on conventional leachate rather than reverse osmosis concentrate, which has extremely low biodegradability and high Cl^- content. Compared with existing literature, this study provides three fundamental novelties: (1) targeting high-chloride RO leachate concentrate; (2) using a $Ti/RuO_2–IrO_2$ electrode with active chlorine as the dominant oxidant; (3) achieving synergistic optimization and high-efficiency deep-purification *via* the three-stage integrated process. The main objectives are: (i) to explore the removal effect of different types of coagulants on the concentrate after reverse osmosis membrane treatment of landfill leachate. The coagulation and sedimentation process is used to remove colloids and suspended solids from the concentrate, thereby reducing COD and UV_{254}. The most suitable coagulant is determined through static tests. The impact of external factors on the treatment effect of the coagulation and sedimentation process was investigated. The impact of the initial pH and sedimentation time on the removal of organic pollutants in the concentrate was examined, and the optimal operating parameters of the coagulation and sedimentation test were determined. (ii) The coagulation effluent under the optimal test conditions was used as the test water for electrochemical oxidation. Based on the IrO_2–RuO_2/Ti electrode material, the removal effects on COD, UV_{254}, BOD_5/COD, and NH_3–N were investigated at different current densities, electrode spacings, and reaction times. The orthogonal experimental method was used to analyze the results and obtain the optimal experimental conditions for electrochemical oxidation. (iii) The effluent from the first two process flows under the optimal test conditions was used as the test influent for the biological aerated filter to explore the removal effect of COD and NH_3–N in the effluent. Stable and dynamic operational experiments were carried out. The test study used a small-scale device for a coagulation–electrochemical oxidation–BAF combined process for advanced treatment of the reverse osmosis membrane filtrate concentrate from landfill leachate. Continuous and stable operation was achieved under the optimal test conditions, and the removal effects of the entire process flow on COD, UV_{254}, and NH_3–N were investigated.

2. Materials and methods

2.1 Concentration of landfill leachate after reverse osmosis membrane treatment

The test water samples used in this study were collected from the reverse osmosis effluent (landfill leachate concentrate obtained after reverse osmosis membrane filtration) of the Daxin Municipal Solid Waste Treatment Plant in Shenyang, Liaoning Province, China. The waste treatment plant employed the UASB + MBR + DTRO process to treat the raw landfill leachate. The objective was to treat the samples to meet the discharge standards of a municipal wastewater treatment plant, and the test water quality indicators were COD (1325.425 mg L^{-1}), BOD_5 (25.9 mg L^{-1}), BOD_5/COD (0.019), pH (6.7), UV_{254} (36.736 cm^{-1}), ammonia nitrogen (52.733 mg L^{-1}), Cl^- (3628.837 mg L^{-1}), electrical conductivity (41.7 mS cm^{-1}) and TN (310.394 mg L^{-1}).

2.2. Operating device

2.2.1. Coagulation and sedimentation. The coagulation and sedimentation test utilized a six-unit synchronous automatic lifting mixer. The running time and rotating speed of this device during operation can be set according to the test requirements (Fig. SI1).

2.2.2 Electrochemical oxidation. The reactor material for the electrochemical oxidation test was plexiglass, and the effective volume of the device was 1.5*L* (length: 15 cm, width: 5 cm, and height: 20 cm). The anode used in the electrochemical oxidation unit is a Ti/RuO_2–IrO_2 coated electrode (active anode), which differs from non-active anodes (*e.g.*, BDD, PbO_2) that produce a large amount of ·OH. Many published studies have confirmed that this electrode has an extremely weak ability to generate ·OH *via* water oxidation, and that its surface mainly forms high-valent metal oxide active sites ($MO_x(\cdot O)$).[10,11,16] In the high Cl^- matrix of leachate RO concentrate, this electrode preferentially catalyzes the oxidation of Cl^- to generate active chlorine species (Cl_2, HOCl, ClO^-), which are the dominant oxidants for degrading organics and ammonia nitrogen. The two electrode plates are placed vertically inside the reactor of the electrochemical oxidation apparatus, with 3 cm separating them. During the entire electrochemical oxidation test, a constant current was provided by an adjustable DC regulated power supply (0–10 A, 0–60 V), which could independently adjust the current according to the test requirements. A magnetic stirrer was placed beneath the electrochemical device to ensure uniform mixing of the water sample during electrochemical oxidation (Fig. 1).

2.2.3 Biological aerated filter (BAF) treatment of membrane filtration concentrate. The reactor had an inner diameter of 100 mm, a height of 1200 mm, and was made of plexiglass columns. During operation, the filter media consisted of spherical ceramic particles with a diameter of about 3 mm. These ceramic particles were packed to a height of 0.6 m within the entire reactor. The influent to the biological aeration filter was designed to flow upward. The aeration pipe, backwash inlet pipe, and influent pipe were all located at the bottom of the biological aeration filter. A peristaltic pump controlled the influent flow. An outlet was located at the top, and the water was discharged through a top overflow outlet.

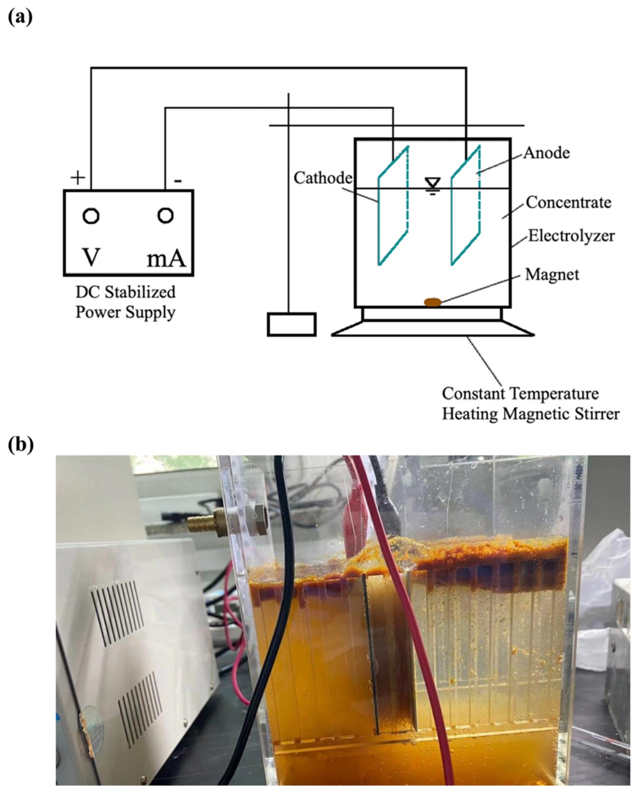

Fig. 1 Experimental setup. (a) Schematic and (b) photograph of the electrochemical reaction test device.

2.3 Operating method

2.3.1 Coagulation and sedimentation test. Since the coagulation–sedimentation test does not require temperature adjustment, this test can be conducted at room temperature. A 100 mL sample of landfill leachate reverse osmosis membrane filtration concentrate was diluted 5 times to 500 mL and placed in a 1 L beaker. According to the test requirements, the pH of the water samples was adjusted using diluted sulfuric acid and a sodium hydroxide solution. Different coagulant doses were added to each test water sample, which were then placed on the six-unit mixer. The mixer speed was set to 200 rpm for 5 minutes, then to 50 rpm for 30 minutes. Finally, it was allowed to settle for 60 minutes, and the supernatant was extracted with a coarse syringe and filtered with quantitative filter paper to determine COD, UV_{254}, and ammonia nitrogen.

2.3.1.1Optimal coagulant selection. Most landfill leachate reverse osmosis concentrate pollutants are anions, so cationic coagulants were selected. The type and amount of coagulants added are significant, as different coagulants have different effects on treating pollutants. The three coagulants, polyferric sulfate (PFS), polyaluminum chloride (PAC) and ferric chloride ($FeCl_3$), were added at different dosages to investigate the influence of COD, NH_3–N, and UV_{254} removal in coagulation–sedimentation; the most effective coagulant was then selected for subsequent experiments.

2.3.1.2Coagulation and sedimentation effect. A 5-fold diluted landfill leachate reverse osmosis membrane filtration concentrate (500 mL) was placed in a 1 L beaker without adjusting the initial pH, and then different doses of PAC were added to each beaker. 0.8, 1, 1.2, 1.4, 1.6, 1.8, 2, 2.2, 2.4, 2.6, 2.8, and 3 g L^{-1}. Under these conditions, the rotation speed of the six-joint agitator should be adjusted to 200 rpm, stirring for 5 min, then stirring at a slow speed of 50 rpm for 30 min. The landfill leachate was left to settle for 60 min, and the supernatant liquid was extracted with a thick needle to prevent the lower layer of flocs from affecting the test results. A UV spectrophotometer was used to determine COD and ammonia nitrogen. The same procedure was followed for PFS and $FeCl_3$.

2.3.2 Electrochemical oxidation test. The coagulation and sedimentation water was used as the test water for the electrochemical oxidation. Since precipitated flocs form during the coagulation and precipitation, even after 60 minutes of settling, many precipitates still floated in the test water, so the coagulation–sedimentation effluent was transferred to the electrochemical oxidation device using a coarse syringe. Different current density and reaction time factors were investigated to determine their effects on ammonia nitrogen, COD, and UV_{254}.

2.3.3 Start-up and operation of the BAF reactor. The microorganisms in the aerated biological filter utilize the granular filler as a biological carrier for growth. The interception, adsorption, and microbial oxidative degradation can be achieved simultaneously in this reactor due to the unique structure of the aerated biological filter.[27] Additionally, within the aerated biological filter, dissolved oxygen diffuses, allowing the biofilm to form under both aerobic and anaerobic conditions and to progress from the inside to the outside. This reactor can integrate nitrification and denitrification to achieve efficient nitrogen removal.[27] This section utilized simulated water inflow and simulated coagulation precipitation–electrochemical oxidation to achieve the effect of a simulated combination. The water ratio for the simulation test is shown in Tables 1 and 2.

The biological aerated filter test was mainly divided into two stages: the sludge inoculation stage and the continuous water inflow stage. In this experimental study, the aerated biological filter was started up using sludge inoculation. To the MBR return sludge from a landfill, sodium acetate, ammonium sulfate, and potassium dihydrogen phosphate were added at a C : N : P ratio of 100 : 5 : 1 to meet the sludge's nutritional needs. The sludge is then aerated for 24 h. The next day, the mixed liquor from the sludge in the container was discharged, and the above process was repeated three times. The sludge was then placed in the reactor, and the influent flow rate was adjusted to 0.3 L h^{-1} with an air-to-water ratio of 4 : 1. During operation, air and water were continuously introduced into the reactor. When the COD removal efficiency was stable, it indicated that the biofilm formation on the aerated biological filter was complete. Subsequently, the proportion of simulated wastewater concentrate in the influent was increased sequentially (25%, 50%, 75%, and 100%, respectively), ensuring that the influent flow rate and air-to-water ratio remained consistent with those during the biofilm formation period. After all the simulated wastewater was used as influent, the biological aerated filter startup performance test was confirmed to be complete.

2.4 Analytical methods

The analysis methods were based on the water monitoring specifications and method standards of the Ministry of Environmental Protection.[28] All experiments and measurements were performed in triplicate ($n = 3$). The data presented in this study are expressed as means. The following parameters were determined: NH_4^+–N (Nessler's reagent spectrophotometry: UV spectrophotometer), COD (UV spectrophotometer), UV_{254} (UV spectrophotometer), electrical conductivity (glass electrode method), BOD_5 (dilution culture method), TN (potassium persulfate oxidation-UV spectrophotometer), and pH (portable pH meter; instrument: PHS-29A type).

The power consumption of the entire process system is expressed in terms of unit energy consumption and energy consumption over a given period. Unit energy consumption is

Table 1 Composition of the simulated water distribution

	Concentration (mg L^{-1})
Glucose	380–420
Total nitrogen	310.39
Ammonium chloride	25–35
Potassium nitrate	65–75
Dihydrogen phosphate	5–10
Trace elements	0.2 mL L^{-1}

Table 2 Concentration of trace elements

Trace elements I	Concentration (mg L^{-1})	Trace elements II	Concentration (mg L^{-1})
EDTA	5000	EDTA	5000
$FeSO_4 \cdot 7H_2O$	5000	$Mn_2 \cdot 4H_2O$	990
		$ZnSO_4 \cdot 7H_2O$	430
		$NiCl_2 \cdot 6H_2O$	190
		$CuSO_4 \cdot 5H_2O$	250
		$CoCl_2$	240
		$NaMoO_4 \cdot 2H_2O$	220
		$Na_2SeO_4 \cdot 2H_2O$	210
		H_3BO_4	14

an important evaluation index for the electrochemical oxidation reaction process. The formula for calculating unit energy consumption is as follows (in COD):[17]

$$Q = \frac{1000 U I \Delta t}{(\mathrm{COD_o} - \mathrm{COD_t}) V},$$

where Q = energy consumption required in a specific period, kW h $\mathrm{kg_{COD}}^{-1}$; U = plate voltage, V; I = current intensity through the plate, A; Δt = reaction time interval, h; and V = volume of liquid to be treated, L.

3. Results and discussion

3.1. Treatment of membrane filtration concentrate by coagulation and sedimentation

3.1.1 Optimal coagulant selection test. All data in the results section are presented as the average of three independent replicates, with error bars indicating standard deviation. When the PAC dosage was 0.8–2 g L^{-1}, COD, NH_3–N, and UV_{254} removal efficiencies increased with the dosage (Fig. 2a). For a PAC dosage of 2 g L^{-1}, the removal effect of COD and UV_{254} in the concentrate was the best, with removal efficiencies of 60% and 63%, respectively, and the concentrations of the two were 523.557 and 12.328 mg L^{-1}, respectively. For PAC, an increased dosage of 2 g L^{-1} to 3 g L^{-1}, NH_3–N removal efficiency reached a stable state (28%). The highest NH_3–N removal efficiency (30%) was achieved at a PAC dosage of 2.5 g L^{-1}, with a concentration of 36.935 mg L^{-1}. In most cases, PAC is converted into aluminum hydroxide or other large particles that are suspended in the RO concentrate, causing turbidity in the effluent and requiring a prolonged sedimentation time to obtain the supernatant. The COD removal effect also improved slightly with increased PAC dosage. Therefore, an acidic environment is beneficial for impurity removal.

When the PFS dosage was 0.8–2 g L^{-1}, COD, NH_3–N, and UV_{254} removal efficiencies in the concentrate gradually increased with increasing dosage (Fig. 2b). When the dosage of PFS was 2 g L^{-1}, the removal efficiency of both indicators was optimal, with COD and UV_{254} removal efficiencies reaching 73% and 67%, respectively, and their concentrations at this point being 363.226 mg L^{-1} and 12.187 mg L^{-1}, respectively. When the PFS dosage exceeded 2 g L^{-1}, the removal efficiency decreased. When PFS was added, the NH_3–N removal efficiency remained stable, fluctuating around 30%. The best NH_3–N removal effect was at a PFS dosage of 2200 mg L^{-1} (removal efficiency 34%; concentration 34.974 mg L^{-1}).

The reason for this phenomenon is that the coagulant PFS has a positive charge, while the colloidal particles suspended in the water sample have a negative charge. When negatively charged particles react with the positively charged coagulant, they form large, tightly bound flocs that settle quickly to the bottom of the solution, making it easy to separate the lower-layer flocs from the supernatant. However, with increasing coagulant concentration, the number of positively charged, highly active groups increases, resulting in fewer available adsorption sites for colloidal particles suspended in the solution to adsorb onto the cationic coagulant. This state destabilized the colloidal particles suspended in the solution, thereby affecting the coagulant's removal efficiency.[29] At this time, if a larger dose of coagulant is added to the solution, it will not only fail to improve the removal efficiency of coagulation and sedimentation for organic pollutants but also waste coagulant and increase test costs. Based on the above analysis, the best PFS dosage was 2000 mg L^{-1}. When excessive coagulant was added, colloids with the same positive charge in the wastewater repel one another, increasing SS and turbidity in the concentrate.

As shown in Fig. 2c, at a $FeCl_3$ dosage of 2.2 g L^{-1}, the COD and UV_{254} removals were the highest, with removal efficiencies of 58% and 59%, respectively. The concentrations of the two were 551.589 and 15.123 mg L^{-1}, respectively. When the $FeCl_3$ dosage exceeded 2.2 g L^{-1}, the COD and NH_3–N removal effects decreased gradually. When the ferric chloride dosage ranged from 0.8 g L^{-1} to 2.2 g L^{-1}, the NH_3–N removal efficiency gradually increased to 26.836% and then stabilized at around 27%. Iron-based compounds can react with hydroxide ions to form ferric hydroxide. Under acidic conditions, iron-containing substances can hydrolyze, forming positively charged ions with multiple nuclei. The abundance of polynuclear positive ions is due to ferric hydroxide, as the suspended colloidal particles in the water samples are negatively charged. As the dosage increased, these positively charged colloids repel each other.

Comparing the effects of the above three coagulants (PAC, PFS, $FeCl_3$), it was found that polyferric sulfate and ferric chloride were more effective than PAC under the same dosage conditions in treating COD. When the dosage of coagulants exceeded 2 g L^{-1}, the three coagulants exhibited distinct effects on removing COD, UV_{254}, and NH_3–N in the concentrate. With increasing PFS and $FeCl_3$ dosages, COD removal in the reverse osmosis concentrates initially increased, then decreased. PFS was always better than $FeCl_3$. The reason for this discrepancy might be that, when the adsorption sites on the suspended colloidal particles in the concentrate reached saturation, further increasing the iron dosage would enhance bridging and trapping. However, excessive iron addition would cause iron ions to adsorb onto the surfaces of suspended colloidal particles, leading to a reversal of surface charge and reducing the neutralization effect. In this case, increasing the dosage would not improve coagulation and sedimentation effects but would waste coagulants, thereby increasing experimental costs.

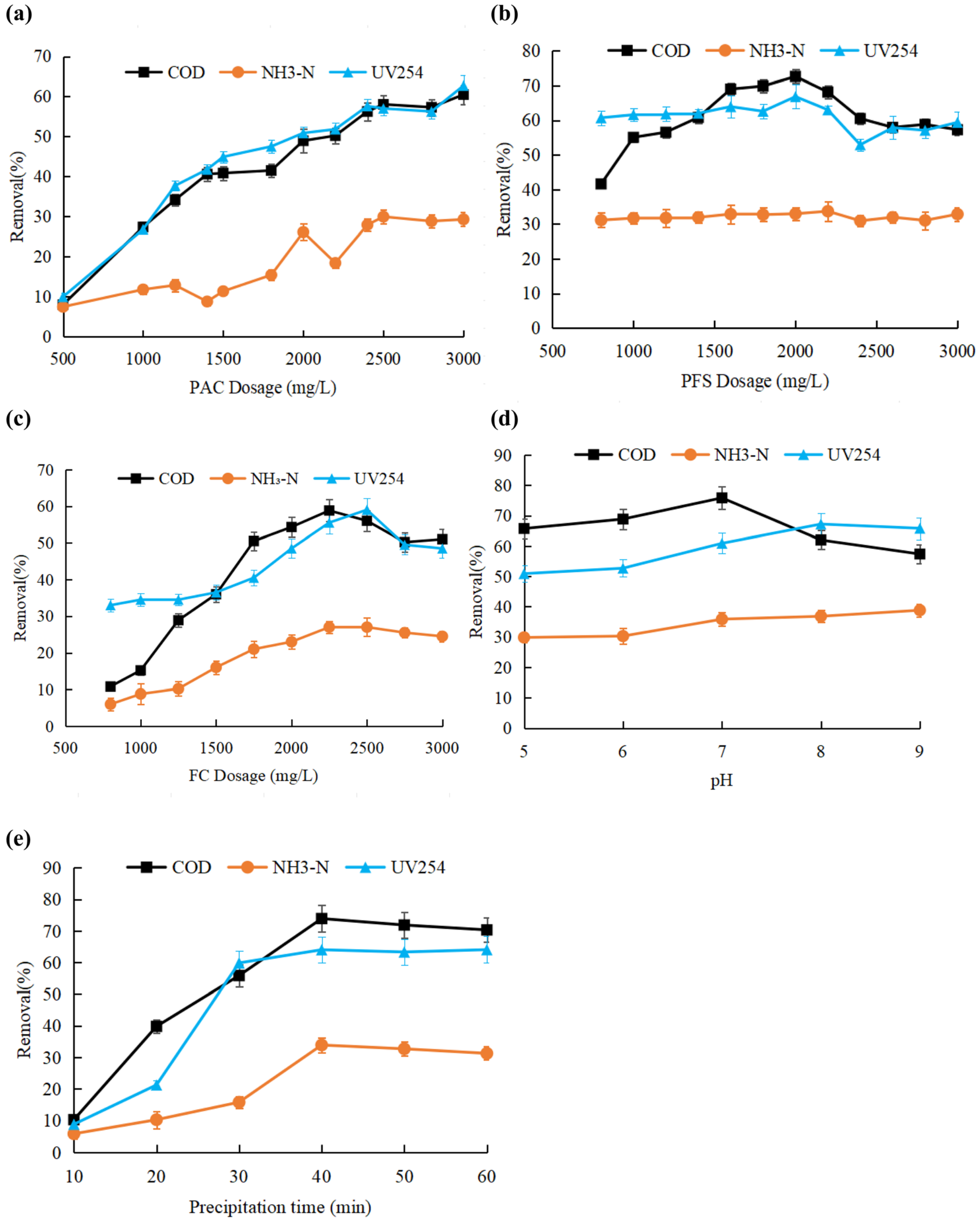

Fig. 2 Effect of the (a) PAC, (b) PFS, and (c) $FeCl_3$ dosage, (d) initial pH (PFS dosage = 2000 mg L^{-1}), and (e) precipitation time on the removal of COD, NH_3–N and UV_{254}.

Compared with PAC, the COD and UV_{254} water quality indicators in the concentrate gradually increased with increasing PAC dosage. However, the removal effect was significantly higher than that of PFS at the same dosage. This indicates that iron salts are significantly more effective than aluminum salts. Iron coagulants are more effective than aluminum-based coagulants for wastewater treatment.[30] Long-term reliance on aluminum salts has been suspected of increasing carcinogenic and mutagenic risks. Therefore, this further confirms that PFS is highly efficient and effective in the coagulation process. PFS was selected as the coagulant for coagulation and sedimentation pretreatment in this study, and the optimal conditions for pretreating reverse osmosis concentrate with PFS were determined through experimental testing. The coagulation pretreatment in this study does not involve hydroxyl radical (·OH) oxidation. It mainly removes colloidal humic substances and macromolecular organics from landfill leachate reverse osmosis concentrate by charge neutralization, adsorption, bridging, and sweeping with PFS, resulting in significant reductions in COD and UV_{254} during the pretreatment stage.

3.1.2. Effect of initial pH value on the coagulation effect. A 5-fold diluted RO concentrate of landfill leachate (500 mL) was placed in a 1 L beaker, and dilute sulfuric acid and sodium hydroxide solutions were used to adjust the pH to 5, 6, 7, 8, and 9, respectively. The same coagulant PFS dosage of 2000 mg L^{-1} was added to each beaker. The six-unit stirrer was used to stir at 200 rpm for 5 min, then at 50 rpm for 30 min. The landfill leachate was then left to settle for 60 minutes, and the supernatant was extracted with a coarse syringe to prevent the lower flocs from affecting the test results. After filtration with quantitative filter paper, COD, NH_3–N, and UV_{254} in the water sample were determined. When the initial pH was 5–7, COD, NH_3–N, and UV_{254} concentrations in the concentrate increased with the increase in pH. For pH 7, the COD, NH_3–N, and UV_{254} removal efficiencies were 76%, 36%, and 61%, respectively, and the concentrations were 320.27, 33.82 and 14.364 mg L^{-1}, respectively (Fig. 2d). When the initial pH was 7–9, although the removal effects of NH_3–N and UV_{254} gradually increased with the increase in pH value, the COD removal efficiency slowly decreased. The UV_{254} removal efficiency reached a maximum of 67% at an initial pH of 8.

Under strong acidic conditions, PFS primarily removes macromolecular humic substances from the reverse osmosis concentrate by charge neutralization. When the pH was too low, many positively charged hydrogen ions surrounded the macromolecular humic organic matter, preventing contact between iron-containing ions and the macromolecular humic organic matter, resulting in poor coagulation and sedimentation. As the pH increases, ferric hydroxide precipitates form, exerting a net-like sweeping effect on the humic colloids in the concentrate. This net-sweeping process promotes the precipitation of flocs and their separation from the solution. When the pH is greater than 7.0, the solution is alkaline. Under these conditions, the ferric hydroxide precipitate will dissolve, leading to reduced removal efficiency.

Additionally, the mass of negative charges per unit volume increases, leading the suspended colloidal particles to resume a stable state that is not conducive to forming a stable system.[3] Based on the above analysis, it is evident that the initial pH of the reaction significantly affects the coagulation effect of PFS. Adjusting the initial pH value of the coagulation to 7 is thus recommended.

3.1.3. Influence of settling time on coagulation effect. The same procedure used in Section 3.1.2 was followed for sampling preparation, except that the landfill leachate was allowed to settle for 10, 20, 30, 40, 50, and 60 minutes. The supernatant was then extracted with a coarse syringe to prevent the lower flocs from affecting the test results. After filtration with quantitative filter paper, the COD, NH_3–N, and UV_{254} in the water sample were determined (Fig. 2e). Following pretreatment of the reverse osmosis concentrate by coagulation and sedimentation, the effluent quality was significantly improved. A comparison of the influent and effluent water quality is shown in Table 3.

When the settling time was increased from 10 to 40 minutes, the removal efficiencies of COD, NH_3–N, and UV_{254} increased rapidly. At a settling time of 40 minutes, the removal effect was close to optimal. COD, NH_3–N, and UV_{254} removal efficiencies were 74%, 34% and 64%, respectively. The concentrations were 346.4, 34.842 and 13.225 mg L^{-1}. When the time exceeded 40 minutes, the COD, NH_3–N, and UV_{254} removal efficiencies tended to stabilize with increasing sedimentation time, indicating that further increases in time had little effect on their removal. Forty minutes was set as the optimal treatment condition. This trend is consistent with the optimal conditions for coagulation of humic-rich leachate reported by Chen *et al.*,[2] in which charge neutralization and adsorption bridging achieve the highest efficiency at neutral pH. The primary purpose of the sedimentation process is to separate the precipitated flocs from the supernatant in the water, and to investigate the influence of settling time on coagulation performance. In the coagulation process, sedimentation time is one of the important factors affecting the quality of the treated supernatant. Under short sedimentation time, fine solid particles remain in the supernatant, affecting the experimental study of the supernatant. If sedimentation time is prolonged, achieving better water quality indicators may be more beneficial, but it also increases costs. The coagulation process with PFS will produce a certain amount of iron-containing chemical sludge, a typical solid waste secondary pollutant that requires centralized harmless treatment in practical engineering.

3.2 Electrochemical oxidation treatment of membrane filtration concentrate

The test in Section 3.1 demonstrated that coagulation–sedimentation is an effective method for pretreating RO concentrates. The coagulation–sedimentation process can remove 70% of COD and 60% of UV_{254} from the concentrate. However, it has not achieved optimal NH_3–N removal, as the reagents were introduced to adjust the reaction's initial pH in Section 3.1. Chloride was introduced to the reverse osmosis concentrate, which increased the conductivity of the coagulation–sedimented effluent from 41.7 to 53.2 mS cm^{-1}. In an electrochemical oxidation reaction, increased conductivity leads to a more complete reaction. In advanced wastewater oxidation technologies, electrochemical oxidation primarily converts Cl^- ions in wastewater into strong oxidizing intermediates, such as ClO^- ions. These intermediates can degrade a large number of high-molecular-weight organic pollutants in the reverse osmosis concentrate, breaking them down into carbon dioxide and water, which can then be volatilized, thereby improving the biodegradability of the reverse osmosis concentrate.

To explore the optimal test conditions for treating the test water samples after coagulation and sedimentation, we utilized ruthenium–iridium–titanium electrode plates as the anode material. The investigation focused on two main factors (electrolysis density and reaction time) influencing the removal

Table 3 Effluent quality of the coagulation pretreatment concentrates

	COD/mg L^{-1}	NH_3–N/mg L^{-1}	UV_{254}/mg L^{-1}	BOD_5/COD
RO concentrate	1325.425	52.733	36.736	0.019
Coagulation pretreatment effluent	346.4	34.842	13.225	0.11
Removal efficiency %	74	34	64	—

effect of various water quality indicators, aiming to determine the optimal parameter conditions. The water sample used was the reverse osmosis concentrate of landfill leachate after coagulation and sedimentation pretreatment, as described in Section 3.1. The anode material was an IrO_2–RuO_2/Ti plate electrode, and the cathode material was a stainless-steel plate electrode (100 mm × 80 mm). The electrochemical device is shown in Fig. 1. The landfill leachate reverse osmosis concentrate, after coagulation and sedimentation pretreatment under the optimal conditions, was injected into the electrochemical device using a coarse-coated syringe. The electrode spacing was set at 3 mm, and the current density was controlled at 5, 7.5, 10, 12.5, and 15 A dm^{-2}, with electrolytic oxidation times of 20, 40, 60, 80, 100, and 120 min. After the reaction, a thick flocculent substance floated on the surface of the wastewater. The clear liquid at the bottom was extracted with a coarse syringe, and its UV absorbance at 254 nm was measured with a spectrophotometer.

3.2.1. Effect of current density on UV_{254} removal. As shown in Fig. 3a, the UV_{254} removal effect in the water samples improved with increasing reaction time. After oxidation for 120 minutes at current densities of 5, 7.5, 10, 12.5, and 15 A dm^{-2}, the UV_{254} removal efficiency in the reverse osmosis membrane rate concentrate was 45%, 55%, 63%, 77%, and 80%, respectively. At a maximum current density of 15 A dm^{-2} (oxidation time 120 min), the UV_{254} removal efficiency reached its highest value (concentration 2.606 mg L^{-1}). A large portion of the humic acid in the concentrate can be removed by oxidation, as the hydroxyl radicals generated during oxidation react chemically with the macromolecular organic matter in the concentrate, leading to the disappearance of the hydroxyl radicals and the generation of new reactive free radicals, thereby sustaining the reaction. Simultaneously, the strong oxidizing substances produced during the reaction react with the newly formed free radicals. This reaction breaks the chemical bonds of aromatic hydrocarbons within the concentrate's humic substances, oxidizing and degrading them into products such as carbon dioxide and water.

The removal efficiency of humic acid at a current density of 15 A dm^{-2} is significantly higher than that at current densities of 5 A dm^{-2}, 7.5 A dm^{-2}, and 10 A dm^{-2}, but not substantially different from that at a current density of 12.5 A dm^{-2}. This is because as the current density increases, more hydroxyl radicals and hypochlorous acid are generated per unit time, leading to the oxidation and degradation of more large-molecule humic acid molecules in a shorter time. However, the increased current density causes the formation of a dense oxide film on the anode surface, hindering further metal oxidation. This phenomenon, known as passivation and polarization of the electrode surface, shortens the lifespan of the electrode plate and reduces current efficiency. The removal efficiency of UV_{254} at a current density of 12.5 A dm^{-2} is not significantly different from that at 15 A dm^{-2}, indicating that increasing the current density does not achieve the desired removal effect.

3.2.2 Effect of current densities on the NH_3–N removal. As shown in Fig. 3b, the effluent NH_3–N concentration after coagulation and sedimentation at different current densities increases to varying degrees over the oxidation time of 0–20 min. This is because the organic nitrogen in the coagulation and sedimentation effluent is degraded by electrochemical oxidation, breaking the molecular chains of large organic pollutants in the test water sample and converting some of them into NH_3–N, thereby increasing the ammonia nitrogen concentration in the effluent.

However, the NH_3–N concentration decreased with increasing oxidation time. Hydroxyl radicals and hypochlorous acid generated during the oxidation process can oxidize and degrade NH_3–N, generating nitrogen gas and water, which can then volatilize. The removal efficiency of NH_3–N varies with current density. At a reaction time of 120 min and a current density of 15 A dm^{-2}, the NH_3–N removal efficiency in coagulation–sedimentation water reached 82%. At this time, NH_3–N concentration dropped to 6.028 mg L^{-1}. To further investigate the reasons for the increase in NH_3–N at the early stage of the reaction, TN concentrations at different current densities were measured to determine whether the total nitrogen removal effect is associated with this increase.

TN concentration in the coagulation and sedimentation effluent decreased significantly within 20 minutes of the reaction, which indicated that during the electrochemical oxidation process, organic nitrogen in the effluent was oxidized by hydroxyl radicals and hypochlorous acid in the water, resulting in the release of ammonia (Fig. 3c), which led to an inevitable increase in the NH_3–N concentration in the test water sample. With increasing current density, the degradation rate of total nitrogen in the test effluent accelerated. For TN removal, a current density of 15 A dm^{-2} was significantly better than current densities of 5 A dm^{-2}, 7.5 A dm^{-2}, and 10 A dm^{-2}, but not substantially different from the removal effect at a current density of 12.5 A dm^{-2}. At the five other current densities and a reaction time of 120 minutes, most of the TN in the coagulation–sedimentation effluent was removed, indicating that most of the nitrogen-containing substances in the effluent were converted into N_2.

3.2.3 Effect of current density on the reduction of chloride ion concentration. As shown in Fig. 3d, due to the addition of pH-adjusting agents to the solution during the coagulation and sedimentation process, and the presence of a large amount of

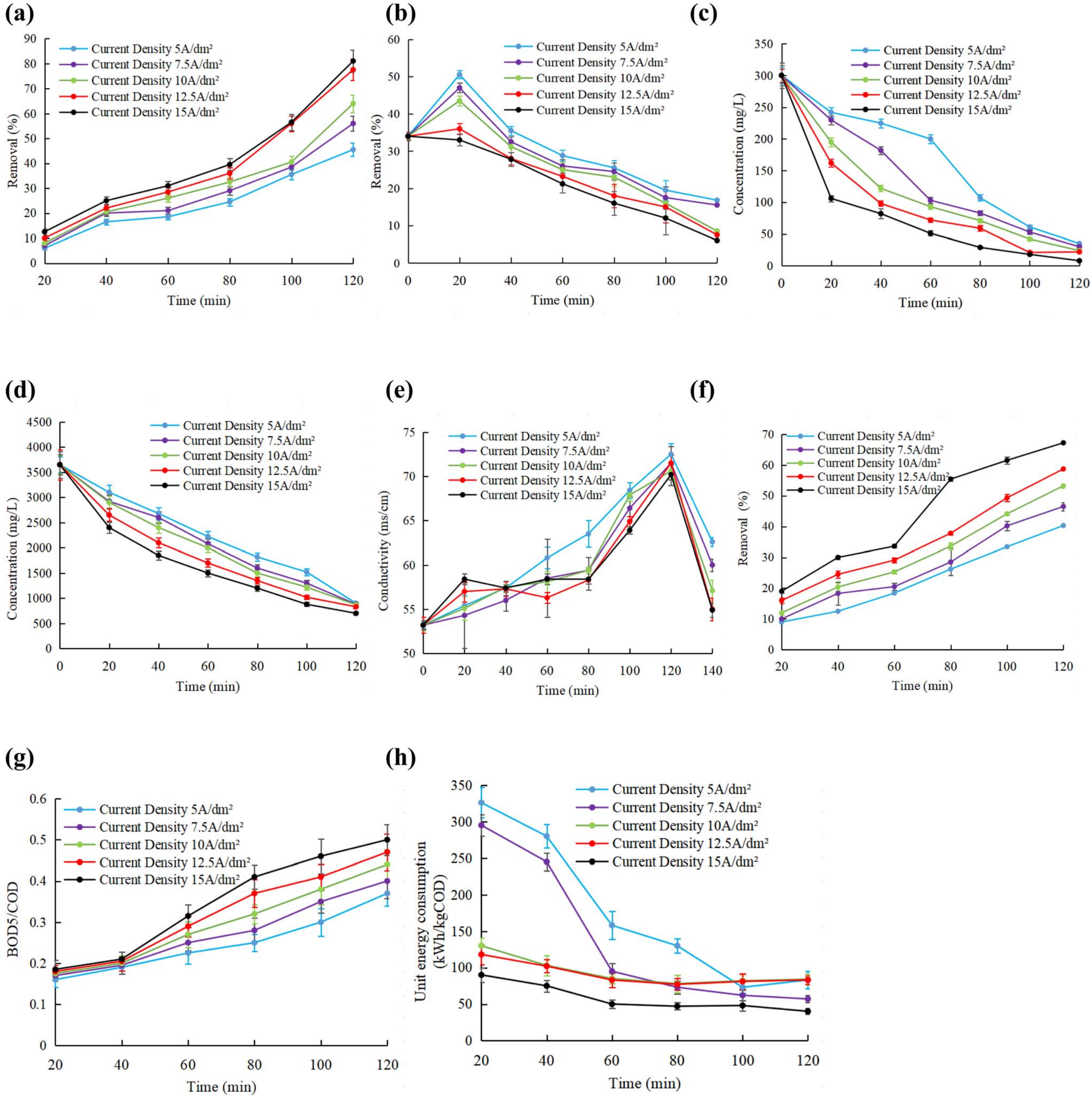

Fig. 3 Effect of different current densities on (a) UV_{254}, (b) NH_3–N, (c) TN, and (d) Cl^- removal, (e) conductivity, (f) COD removal, (g) biodegradability, and (h) variation of the unit energy consumption with time.

chloride ions in the reverse osmosis membrane concentrate, the effluent after coagulation and sedimentation contains a significant amount of chloride ions during the electrochemical oxidation reaction. The chloride ion concentration in the test water sample gradually decreases with increasing reaction time. This phenomenon is attributed to the following: during the electrochemical oxidation reaction, some chloride ions in the test water sample are oxidized to hypochlorite (ClO^-) ions, which participate in indirect electrochemical oxidation and remove organic matter from the concentrate. The remaining chloride ions are converted into chlorine gas at the anode, which then volatilizes. This explains the pungent odour detected after 60 minutes, as the chlorine gas has volatilized. The reaction equations are as follows:

$$\text{Reactions at the anode: } 2Cl^- \rightarrow Cl_2 + 2e,$$

$$2H_2O \rightarrow O_2 + 4H^+ + 4e.$$

$$\text{Reactions in solution: } Cl_2 + 2OH^- \rightarrow ClO^- + Cl^- + H_2O,$$

$$HClO \rightarrow H^+ + ClO^-.$$

$$\text{Reactions at the cathode: } 2H_2O + 2e \rightarrow 2OH^- + H_2,$$

$$ClO^- + H_2O + 2e \rightarrow Cl^- + 2OH^-.$$

As shown in the formula, the presence of Cl_2 generated at the anode and the formation of ClO^- from OH^- consume most of the Cl^- in the solution, thereby decreasing the pH. At this point, the residual chlorine in the concentrate mainly exists in the form of hypochlorite (ClO^-). A decline in chloride ion concentration in the concentrate may affect the conductivity. It should be noted that the conversion of Cl^- into active chlorine may result in the formation of trace chlorinated disinfection byproducts (DBPs), such as trihalomethanes (THMs) and haloacetic acids (HAAs), which are potential organic secondary pollutants in the effluent.

3.2.4 Effect of current density on the conductivity. The conductivity value increases with time during the reaction period 0–120 min but decreases with time during the period of 120–140 min (Fig. 3e). The highest conductivity, 72.32 mS cm^{-1}, is achieved at 120 min under a current density of 5 A dm^{-2}. In the initial stage of the reaction, most of the organic matter in the test water sample was degraded during electrochemical oxidation. This organic matter was oxidized and degraded into smaller organic molecules by hypochlorite ions, increasing the ion concentration in the test water sample and thereby improving conductivity. However, as the reaction time increases, a large amount of organic matter is converted into CO_2 and H_2O, and chloride ions in the concentrate are oxidized to chlorine gas, which is released into the air. This reduces the number of ions in the concentrate, resulting in lower conductivity values. During electrochemical oxidation, the strength of the electron transfer in the concentrate determines the conductivity value. Fluctuations in conductivity indicate changes in the concentrations of cations and anions during electrochemical oxidation. Therefore, measuring conductivity is essential during experiments.

3.2.5 Effects of current density on COD removal. As shown in Fig. 3f, when the current density is 15 A dm^{-2}, the COD removal effect of the concentrate on the test water sample during the reaction process is better at all measurement times than when the current density is 5 A dm^{-2}, 7.5 A dm^{-2}, 10 A dm^{-2}, and 12.5 A dm^{-2}. During operation, the COD removal efficiency increases with increasing electrochemical oxidation time. At a reaction time of 120 min, the COD removal efficiency reaches 67%, and the COD content in the test water sample is 113.283 mg L^{-1}. The reason for this phenomenon is that the increased current density provides more energy for electrochemical oxidation, thus degrading and removing more organic pollutants from the reverse osmosis membrane concentrate. However, the experimental study shows that higher current density does not necessarily yield better results. On the one hand, as the current density increases, the energy consumed in the electrochemical oxidation process increases, and the amount of organic pollutants removed per unit time decreases. Furthermore, the higher the current density, the higher the solution temperature, requiring more sophisticated equipment for the electrochemical oxidation reaction. A current density of 15 A dm^{-2} showed relatively good COD removal efficiency in the concentrate; at a reaction time of 120 min, the COD removal efficiency was 58%, and the COD content in the concentrate was 144.12 mg L^{-1}.

3.2.6 Effects of current density on biodegradability. The current density has a significant impact on the BOD_5/COD ratio of the concentrate during the effluent treatment process, following coagulation and sedimentation pretreatment (Fig. 3g). At a reaction time of 120 min, the BOD_5/COD values at current densities of 5 A dm^{-2}, 7.5 A dm^{-2}, 10 A dm^{-2}, 12.5 A dm^{-2}, and 15 A dm^{-2} are 0.37, 0.4, 0.44, 0.47, and 0.5, respectively. When the current density is 15 A dm^{-2} and the reaction time is 120 min, the BOD_5/COD ratio reaches 0.5, which is the best for biodegradability under the same time conditions compared to the other four current densities. After coagulation pretreatment, the effluent water quality was significantly improved after electrochemical oxidation. A comparison of the results of influent and effluent water quality is shown in Table 4.

3.2.7. Energy consumption analysis. When the electrochemical oxidation method is applied to the effluent after coagulation and sedimentation pretreatment, the energy consumption during the oxidation process should be analyzed. For the effluent treatment process after coagulation and sedimentation pretreatment, the current density was set to 5, 7.5, 10, 12.5, and 15 A dm^{-2}, corresponding to 4 A, 6 A, 8 A, 10 A, and 12 A, respectively. As shown in Fig. 3h, within the first 40 minutes of electrochemical oxidation, the unit energy consumption for the entire reaction was relatively high. During the reaction, the DC power supply converts some of its electrical energy into energy within the test water sample. In contrast, the remaining electrical energy is used throughout the reaction to remove humic substances from the water sample. The energy consumption per unit volume at a current density of 15 A dm^{-2} is significantly lower than that at other current densities. At a reaction time of 120 min, the energy consumption per unit volume at a current density of 15 A dm^{-2} is 39.4038 kW h $kg_{COD}{}^{-1}$. Therefore, a current density of 15 A dm^{-2} was used in this test. The unit energy consumption of this system (39.40 kW h $kg_{COD}{}^{-1}$) falls within the reasonable range for electrochemical oxidation processes of landfill leachate concentrate. Compared with other AOPs, it is higher than Fenton oxidation (15–30 kW h $kg_{COD}{}^{-1}$) but lower than BDD electrochemical oxidation (50–80 kW h $kg_{COD}{}^{-1}$) and ozone oxidation (40–60 kW h $kg_{COD}{}^{-1}$) under similar treatment conditions reported in literature.[31]

3.2.8 Mechanism of electrochemical oxidation. The electrochemical oxidation process in this study is dominated by Cl^--mediated active chlorine oxidation rather than $^{\bullet}OH$ oxidation. The $Ti/RuO_2–IrO_2$ active anode catalyzes the oxidation of Cl^- in the leachate concentrate to Cl_2, which further disproportionates to HOCl and ClO^- (active chlorine species). These active chlorine species are the core oxidants for degrading refractory organics and removing NH_3–N. On one hand, active chlorine destroys the aromatic ring structure and macromolecular chains of humic substances (reflected by the sharp decrease in UV_{254}), converts non-biodegradable

Table 4 Effluent quality of electrochemical oxidation treatment

	COD/mg L^{-1}	NH_3–N/mg L^{-1}	UV_{254}/mg L^{-1}	BOD_5/COD
Coagulation pretreatment water	346.4	34.842	13.225	0.11
Electrochemical oxidation of water	113.28	6.028	2.595	0.5
Removal efficiency	67%	83%	80%	—

macromolecules into small molecular organics, and significantly improves the BOD_5/COD ratio from 0.11 to 0.5. On the other hand, active chlorine rapidly oxidizes NH_3–N to N_2 through breakpoint chlorination, achieving high-efficiency denitrification. The continuous decrease in Cl^- concentration and the volatilization of Cl_2 odor observed during the experiment directly confirm the occurrence of Cl^--mediated indirect oxidation. It should be emphasized that the contribution of $^{\bullet}OH$ to pollutant degradation in this system is negligible due to the low $^{\bullet}OH$ production capacity of the Ti/RuO_2–IrO_2 electrode. Different from the $^{\bullet}OH$-dominated oxidation by the BDD electrode reported by Wang *et al.*[10] and Song *et al.*,[16] our Ti/RuO_2–IrO_2 system mainly relies on active chlorine, which is more suitable for high-Cl^- leachate concentrates. It should be noted that this study evaluated only the removal performance of RO leachate concentrate using conventional bulk water quality indicators (COD, NH_3–N, UV_{254}), without further identification or quantification of specific organic contaminants or their transformation products.

3.3. BAF treatment of membrane filtration concentrate: BAF start-up

During the biofilm-formation stage (Fig. 4a), the influent COD concentration ranged from 129.3948 to 140.2384 mg L^{-1}, and the effluent concentration ranged from 30.4662 to 63.8418 mg L^{-1}. The removal efficiency fluctuated between 51% and 77%. Recent studies show that COD removal is due to the interception and adsorption by the filter media, as well as to the oxidation effect of microorganisms attached to the media.[24–26] During operation of the aerated biological filter bioreactor, the biofilm on the filter media gradually matures, thereby enhancing the reactor's overall COD removal efficiency. Backwashing was performed between days 16 and 18. After the backwashing, the aerated biological filter failed to recover its treatment capacity to its pre-backwashing level in a timely manner, resulting in a decrease in COD removal efficiency during this short period. However, within a specific timeframe, the volume of biological growth on the filter media increases, so even after backwashing, the reactor can recover its capacity in a very short time. Therefore, the removal rate increased rapidly within 19 to 22 days, reaching over 80%. From the 23rd day onward, the COD removal rate stabilized, resulting in final effluent COD concentrations consistently below 25 mg L^{-1} (influent COD fluctuated around130 mg L^{-1}). During biofilm formation, sludge samples were collected daily at the same time to observe growth morphology. The influent and effluent COD concentrations were measured, and changes in COD were observed. During the initial startup phase, the microorganisms attached to the filter media were still adapting to the influent, leading to unstable COD removal efficiency and fluctuating removal efficiency. However, on the 23rd day of reactor operation, after acclimatization and cultivation, the biofilm attached to the filter media matured. Sampling at this time revealed that the sludge had turned black and exhibited excellent settling properties.

After 23 days of operation, COD removal efficiency was approximately 80%, with minimal fluctuation, indicating stable COD removal and confirming successful biofilm formation in the aerated biological filter. After biofilm formation in the BAF reactor was initiated, various indicators in the effluent from the coagulation–sedimentation–electrochemical oxidation treatment were analyzed. Simulated wastewater was used as the influent to replace the effluent from the first two treatment stages, and the influent-to-reactor ratio was gradually increased. A certain proportion of glucose was added to the influent, as glucose promotes the growth of microorganisms attached to the filter media. During reactor operation, no temperature adjustment was required; maintaining the room temperature at 20 °C–30 °C was sufficient. The influent flow rate was controlled at 0.3 L h^{-1}, maintaining an air-to-water ratio of 4 : 1. The initial reaction pH was adjusted to approximately 6.5–7.5. Nitrogen and phosphorus were supplemented at a C : N : P ratio of 100 : 5 : 1. The BOD_5/COD ratio of the influent was monitored and fluctuated around 0.5. The entire operation of the aerated biological filter can be divided into four stages.

Stage I (8 days), COD (simulated influent) : COD (glucose) ratio ≈ 1 : 4, and simulated influent COD concentration was controlled within 103.3972–123.283 mg L^{-1}. Stage II (8 days), COD (simulated influent) : COD (glucose) ≈ 1 : 2, and the simulated influent COD concentration was controlled within 106.927–121.119 mg L^{-1}. Stage III (8 days), the COD (simulated influent) : COD (glucose) ≈ 3 : 4, and the simulated influent COD concentration was controlled within 101.003–118.293 mg L^{-1}. Stage IV (10 days), the COD (simulated influent) : COD (glucose) ≈ 3 : 4, and the COD concentration of the simulated influent was controlled within 110.927–115.273 mg L^{-1}.

3.3.1. Removal effect of BAF on COD. During stage I (Fig. 4b), influent COD, effluent COD and COD removal efficiency were 103.397–123.283 mg L^{-1}, 41.394–52.009 mg L^{-1}, and 50–65%, respectively. For stage II, influent COD, effluent COD and COD removal efficiency were 106.927–121.119 mg L^{-1}, 34.98–55.344 mg L^{-1}, and 52–69%, respectively. During stage III operation, influent COD concentration was 101.003–108.298 mg L^{-1}, effluent COD concentration was 19.454–48.423 mg L^{-1}, and the removal efficiency reached 59–73%. In stage IV, the COD concentration in the influent was 110.927–

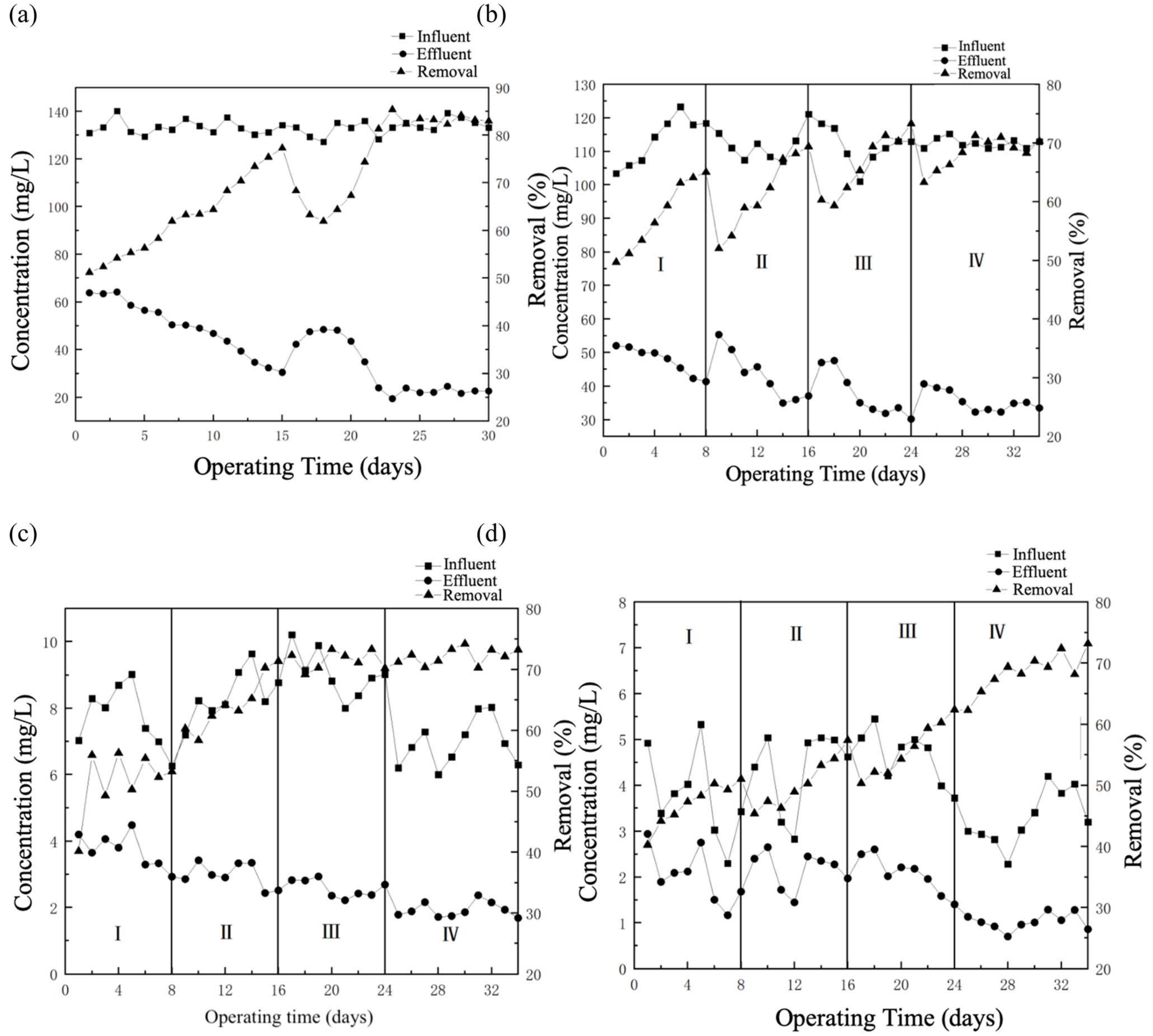

Fig. 4 Removal effect of (a) COD during start-up, (b) BAF on COD, (c) BAF on NH_3–N, and (d) BAF on UV_{254}.

115.273 mg L^{-1}, the effluent was 32.254–40.717 mg L^{-1}, and the removal efficiency was 63–71%. The COD removal was better throughout the BAF reactor's operation. In the initial stage of the reaction, the simulated influent, after coagulation, sedimentation, and electrochemical oxidation, initially contains some recalcitrant organic matter. Microorganisms attached to the reactor's filter media were not adapted to the simulated influent within a short period, resulting in low COD removal efficiency in the initial stage, indicating that the removal effect on the influent COD was not significant. As the reactor operated, the microorganisms gradually adapted to the influent water quality. Therefore, increasing the proportion of simulated wastewater in the influent from 25% to 50% resulted in a very low COD removal rate in the initial stage of the second stage. However, as the reactor operated, the COD removal effect gradually improved. After 34 days of operation, the microorganisms in the reactor were cultivated and acclimatized, and the aerated biological filter reactor achieved stability. The BOD_5/COD increased from 0.5 to 0.55, indicating improved biodegradability. The COD removal efficiency of BAF remained around 70%, and the effluent COD stabilized at 33 mg L^{-1} (Table 5).

3.3.2 Removal effect of BAF on NH_3–N. During the operation of the BAF reactor, under the premise of maintaining a roughly constant influent NH_3–N concentration (influent NH_3–N $\approx$ 6.028 mg L^{-1}), the NH_3–N removal was investigated when the proportion of concentrated solution in the simulated feedwater was 25%, 50%, 75%, and 100% of the influent. As seen in Fig. 4c, after 34 days of acclimation. During the four stages of BAF reactor operation, the NH_3–N removal efficiency gradually increased and then stabilized. During stage I, influent NH_3–N, effluent NH_3–N and NH_3–N removal efficiency were

Table 5 Coagulation pretreatment–electrochemical oxidation in the BAF reactor

	COD/mg L^{-1}	NH_3–N/mg L^{-1}	UV_{254}/mg L^{-1}	BOD_5/COD
Electrochemical oxidation of water	113.28	6.028	2.595	0.5
BAF-treated effluent	32.264	1.686	0.7	0.55
Removal efficiency	71%	73%	69%	—

6.263–9.0238 mg L^{-1}, 2.93–4.49 mg L^{-1}, and 40–56%, respectively. For stage II, influent NH_3–N, effluent NH_3–N, and NH_3–N removal efficiency were 7.203–9.64 mg L^{-1}, 2.44–3.42 mg L^{-1}, and 58–71%, respectively. During the stage III operation, the influent NH_3–N concentration ranged from 8.001 to 10.208 mg L^{-1}, the effluent NH_3–N concentration ranged from 2.22 to 2.94 mg L^{-1}, and the removal efficiency ranged from 69% to 73%. In stage IV, influent NH_3–N concentration ranged from 6.0023 to 8.028 mg L^{-1}, while effluent concentration ranged from 1.69 to 2.369 mg L^{-1}, resulting in removal efficiencies of 70% to 74%. As the BAF reactor operated, the NH_3–N removal efficiency fluctuated during the initial stages (I and II), but as the operation progressed, it stabilized at approximately 73% (Table 5).

3.3.3 Removal effect of BAF on UV_{254}. During the operation of the BAF reactor, under the premise of maintaining a roughly the same influent UV_{254} concentration (influent $UV_{254} \approx$ 2.595 mg L^{-1}), the removal effect of UV_{254} was observed when the proportion of the concentrated solution in the simulated wastewater was 25%, 50%, 75%, and 100% in the influent. As shown in Fig. 4d, during the BAF test, humic acid was added to the simulated wastewater at a concentration based on the effluent obtained after coagulation, sedimentation, and electrochemical oxidation. During stage I, influent UV_{254}, effluent UV_{254} and removal efficiency were 2.302–5.329 mg L^{-1}, 1.168–2.942 mg L^{-1}, and 40–51%, respectively. For stage II, influent UV_{254}, effluent UV_{254}, and removal efficiency were 2.830–5.0395 mg L^{-1}, 1.445–2.65 mg L^{-1}, and 45–57%, respectively. During stage III operation, the influent UV_{254} concentration ranged from 3.723 to 5.452 mg L^{-1}, the effluent UV_{254} concentration ranged from 1.4 to 2.6 mg L^{-1}, and the removal efficiency ranged from 50% to 62%. In stage IV, the UV_{254} concentration in the influent was 2.287–4.198 mg L^{-1}, while the effluent concentration was 0.7–1.287 mg L^{-1}, resulting in a removal efficiency of 62–73%. As the BAF reactor operated, the UV_{254} removal efficiency fluctuated during the initial stages (I and II) but stabilized at around 69% as the experiment progressed (Table 5). The subsequent BAF process further mineralizes small molecular organics through biodegradation, forming a complete "macromolecular removal → biodegradability improvement → deep mineralization" synergistic treatment pathway. The stable removal performance of BAF in this study is consistent with the findings of Li,[19] which verified the feasibility of BAF for advanced treatment of pre-oxidized leachate.

4. Conclusion

Most studies in the literature have focused on raw leachate or biologically treated effluent. In contrast, the water sample treated in this study was RO concentrate, characterized by high concentrations of refractory organic matter (*e.g.*, humic substances) and high COD content. The treatment difficulty and operating costs of RO concentrate are considerably higher than those of raw leachate. Therefore, a low-cost coagulation–sedimentation process was employed as a pretreatment prior to electrochemical oxidation, and a biological aerated filter was used to conduct experimental research on landfill leachate RO concentrate. During the pretreatment process, a large amount of high-concentration humic macromolecular organic pollutants in the concentrate after reverse osmosis membrane treatment can be removed, along with high COD levels. Coagulation–sedimentation can reduce treatment costs for landfill leachate concentrate, improve biodegradability and macromolecular organic matter removal, and decrease COD, UV_{254}, and NH_3–N. According to our results, coagulation removed 74% of the COD, thereby reducing the overpotential requirement of the subsequent electrooxidation unit. Electrooxidation can break down macromolecular organic compounds into small-chain carboxylic acids, thereby enhancing the biodegradability of the effluent for the subsequent BAF. The integrated process presents a clear synergistic mechanism: coagulation removes macromolecular humus *via* charge neutralization and adsorption bridging. Coagulation removed suspended solids and a portion of humic acids (achieving 64% UV_{254} removal), thereby reducing the organic load on the electrooxidation unit and mitigating electrode fouling. Electrochemical oxidation dominated by active chlorine improves biodegradability and removes ammonia nitrogen; BAF further mineralizes small-molecular-weight organics. Electrooxidation → BAF synergy: electrooxidation increased the BOD_5/COD ratio from 0.11 to 0.5, a prerequisite for effective BAF performance—the BAF alone was virtually incapable of removing COD (removal efficiency < 10%). Without coagulation, the electrooxidation unit would suffer from an excessively high organic load (leading to increased energy consumption); without electrooxidation, the BAF would be ineffective. The effluent quality attained the Class A standard of the Pollutant Discharge Standard for Urban Sewage Treatment Plants (GB18918-2002). Therefore, despite the lack of a rigorous quantitative comparison, the integrated process demonstrates clear system-level advantages from a functional complementarity perspective.

The unit energy consumption for electrochemical oxidation in this study was 39.40 kW h $kg_{COD}{}^{-1}$, comparable to those of common AOPs for leachate treatment. However, this study focused only on lab-scale performance. It did not estimate total capital expenditure (CAPEX), operating expenditure (OPEX), or large-scale economic feasibility, which will be further evaluated in future pilot applications. The units in this study were operated independently without process integration. Therefore, it is recommended to achieve physical connection and sequential operation of the three processes at the pilot scale, and to conduct systematic control experiments to validate their synergistic effects. It should be noted that quenching experiments for active species identification and intermediate byproduct detection (*e.g.*, GC-MS and LC-MS) were not performed in this study. Future research will conduct relevant experimental verification to quantify the contributions of active chlorine and $^{\bullet}$OH and to clarify the complete degradation pathway of refractory organics. More useful outcomes might be gained if this research were implemented in a pilot-scale trial that connected the coagulation–sedimentation tank, the electrochemical oxidation tank, and the aerated biological filter to replace the simulated wastewater. In addition, potential secondary pollution risks, including chlorinated byproducts from electrochemical oxidation and chemical sludge from coagulation, should be fully considered in engineering applications. Relevant control and disposal measures should be implemented to ensure environmentally friendly operation of the integrated process.

Conflicts of interest

There are no conflicts to declare.

Data availability

The data supporting this article have been included as part of the supplementary information (SI). Supplementary information is available. See DOI: https://doi.org/10.1039/d6va00153j.

Acknowledgements

The authors thank the School of Municipal and Environmental Engineering at the Shenyang Jianzhu University for providing the research facilities. The Liaoning Innovative Research Team at the University-LNIRT supports this research. Dr Tabassum is also grateful to the Department of Chemistry at the Sakarya University.

References

1 J. Tao, Current Status and Application Prospects of Urban Solid Waste Composting Treatment, *Tianjin Sci. Technol.*, 2008, **05**, 46–47.
2 H. Chen, *et al.*, Treatment of landfill leachate by coagulation: A review, *Sci. Total Environ.*, 2024, **912**, 169294.
3 Y. Yanlei, *Polyferric coagulation-Fenton-BAF Treatment of Landfill Leachate Reverse Osmosis Concentrate*, South China University of Technology, Guangzhou, China, 2015.
4 H. Hasar, *et al.*, Stripping/flocculation/membrane bioreactor/reverse osmosis treatment of municipal landfill leachate, *J. Hazard. Mater.*, 2009, **171**(1), 309–317.
5 Z. Jian, A review of membrane filtration concentrate treatment technology for landfill leachate, *Guangdong Chem. Ind.*, 2012, **38**(8), 264–265.
6 R. Keyikoglu, *et al.*, A review on treatment of membrane concentrates generated from landfill leachate treatment processes, *Sep. Purif. Technol.*, 2021, **259**, 118182.
7 R. Mehta, *et al.*, Refuse decomposition in the presence and absence of leachate recirculation, *J. Environ. Eng.*, 2002, **128**(3), 228–236.
8 D. Wang, D. Liu and L. Zhi, Study on the variation law of water quality of reverse osmosis concentrate recharge water, *Environ. Sci. Res.*, 2014, **35**(7), 2822–2828.
9 X. Ntampou, A. I. Zouboulis and P. Samaras, Appropriate combination of physico-chemical methods (coagulation/flocculation and ozonation) for the efficient treatment of landfill leachates, *Chemosphere*, 2006, **62**(5), 722–730.
10 F. Wang, *et al.*, Treatment of aged leachate by a three-dimensional electrochemical system with biochar as particle electrodes, *Environ. Technol.*, 2025, **46**(19), 3904–3917.
11 T. Junchao, *Electrochemical Oxidation Method for Treating Landfill Leachate Membrane Filtration Concentrate*, Chongqing University, Chongqing, China, 2015.
12 N. Amaral-Silva, *et al.*, Ozonation and perozonation on the biodegradability improvement of a landfill leachate, *J. Environ. Chem. Eng.*, 2016, **4**(1), 527–533.
13 L. Xudong, *Enhanced Electrochemical Oxidation Treatment of Landfill Leachate Membrane Deconcentrate*, Xi'an University of Engineering, Xi'an, China, 2019.
14 W. Xiaoyun, *et al.*, Advanced treatment of landfill leachate membrane filtrate concentrate using enhanced coagulation-photoelectric oxidation combined process, *J. Environ. Eng.*, 2012, **9**, 3040–3046.
15 W. Yunhai, *Coagulation-Advanced Oxidation Treatment Technology of Landfill Leachate Nanofiltration Concentrate*, Shanghai Jiao Tong University, Shanghai, China, 2017.
16 Z. Song, *et al.*, Performance enhancement strategies for electrooxidation degradation of landfill leachate: A review, *Chin. Chem. Lett.*, 2025, **36**(12), 111008.
17 J. Hailong, *Development and Effect Study of Coagulant for Mixed Fruit Peel*, Jinan University, Shandong, China, 2019.
18 M. S. Bilgili, *et al.*, COD fractions of leachate from aerobic and anaerobic pilot scale landfill reactors, *J. Hazard. Mater.*, 2008, **158**(1), 157–163.
19 M. Li, *Advanced Treatment of Landfill Leachate Using a Combined Coagulation-Catalytic Ozonation-Biological Aerated Filter Process*, Hunan University, Changsha, China, 2023.
20 Z. Luo, *et al.*, Research progress on electrochemical treatment of heavy metal wastewater, *China Water Wastewater*, 2009, **25**(16), 6–10.
21 Z. Jun, H. Wu, and C. Hanfei, *New Technologies and Engineering Examples of Wastewater Treatment Using Aerated Biological Filters*, Chemical Industry Press, Beijing, China, 2002.
22 F. Shaobin, *Denitrification Performance and Microbial Distribution Characteristics of Aerated Biological Filter*, South China University of Technology, Guangzhou, China, 2014.
23 L. Jingshi and W. Xiaojun, Pilot-scale study on deep treatment of landfill leachate using a combined biological

fluidized bed-Fenton-aerated biological filter process, *Water Treat. Technol.*, 2012, **38**(1), 4.

24 J. Zhu, *et al.*, Quorum quenching driven microbial community to biofouling control in membrane bioreactor for landfill leachate treatment, *J. Membr. Sci.*, 2025, **722**, 123899.

25 X. Xiao, *et al.*, Enhancing operational stability and pollutants removal in dynamic membrane bioreactors via granular activated carbon for landfill leachate treatment, *J. Membr. Sci.*, 2025, **736**, 124726.

26 L. Xu, *et al.*, Single-stage bioreactor integrating anammox and sulfur-driven autotrophic denitrification for nitrogen removal from landfill leachate, *J. Hazard. Mater.*, 2026, **505**, 141444.

27 J. Yang, S. Jiang and L. Zhaoli, Low-temperature start-up test of upflow aerated biological filter, *Water Treat. Technol.*, 2011, **37**(1), 112–115.

28 Water and Wastewater Monitoring and Analysis Methods, The editorial board of State Bureau of Environmental Protection, in *Water and Wastewater Monitoring Analysis Method (4th Edition)*, China Environmental Science Press, Beijing, 2002, pp. 88–439, 448.

29 J. Xue, *Study on the Factors and Mechanisms Affecting FSS and Polyferric Sulfate (PFS) Coagulation*, Huazhong University of Science and Technology, Wuhan, China, 2016.

30 R. M. El-taweel, *et al.*, A review of coagulation explaining its definition, mechanism, coagulant types, and optimization models; RSM, and ANN, *Curr. Res. Green Sustain. Chem.*, 2023, **6**, 100358.

31 Y. Zhu, *et al.*, Electrocatalytic Oxidation of Nanofiltration Concentrated Landfill Leachate with High-efficiency Ti/RuO2-IrO2-Pt 8% Anode, *ChemistrySelect*, 2025, **10**(6), e202405054.