

Simultaneous Removal of Nitrogen and Refractory Organics from a Biologically Treated Leachate by Pulse Electrochemical Oxidation in a Multi-channel Flow Reactor

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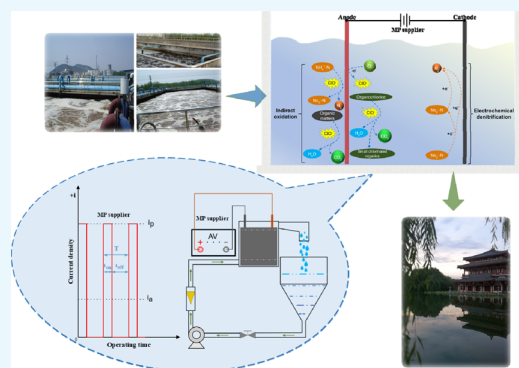


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ABSTRACT: Electrochemical oxidation (EO) is often used in the advanced treatment of refractory wastewater. However, in a conventional EO process of direct-current (DC) power supply, oxide layers often form on the anodes, which not only hinder the oxidation reaction on them but also cause higher energy consumption. In this paper, a biologically treated leachate (BTL) of municipal solid waste (MSW) was comparably treated by EO with DC (DC–EO), monopulse (MP–EO), and double pulse (DP–EO) power source models in a home-made multi-channel flow reactor. The effects of process parameters of current density (I_A), superficial liquid velocity (U_L), pulse frequency (f_p), duty ratio (R_D), and so forth on the removal efficiency of chemical oxygen demand (COD) (RE_{COD}), total organic carbon (TOC) (RE_{TOC}), and total nitrogen (TN) (RE_{TN}) were investigated simultaneously. Average energy consumption (\overline{EC}) and organic composition of the treated effluent of DC–EO and MP–EO were also compared comprehensively, and a new mechanism of MP–EO has been proposed accordingly. Under optimal conditions, 2 L of BTL was treated by MP–EO for 180 min, and the RE_{COD} , RE_{TOC} , and RE_{TN} could reach as high as 80, 30, and 80%, respectively. Compared with DC–EO, the \overline{EC} of MP–EO is reduced by 69.27%. Besides, the kinds of organic matter in the treated effluent of MP–EO are reduced from 53 in the BTL to 11, which is much less than in the DC–EO process of 29 kinds. Therefore, the MP–EO process exhibits excellent removal performance of organics and TN and economic prospects in the treatment of refractory organic wastewater.



1. INTRODUCTION

With the rapid increase in municipal solid waste (MSW), the garbage siege phenomenon has become one of the most urgent global environmental problems, which may become a stumbling block to the global economic and urbanization process. Compared with the MSW of developed countries, the MSW in China exhibits higher moisture and organic contents but lower average calorific value and recyclable components.¹ Therefore, before the harmless disposal by landfill or incineration, a large amount of toxic and harmful leachate can be produced in the process of accumulation and storage of the MSW. Many organic pollutants in the MSW leachate show transgenic effects (carcinogenic, disabling, and mutagenic),² which not only affect the expected growth of plants and animals but also cause irreversible damage to the nervous and immune systems of the human being and other organisms.² Improper disposal of the leachate can cause serious pollution of surface water and groundwater, which may lead to serious damages to the ecosystem. However, the MSW leachate is difficult to meet the water quality of discharge standard only through biological treatment due to the high

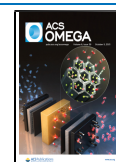
concentration of refractory organics and nitrogen-containing compounds.^{3,4}

Many advanced oxidation processes (AOPs) like electrochemical oxidation (EO), electrocoagulation, photocatalysis, ozonation, Fenton/Fenton-like oxidation, and photo-Fenton oxidation, and so forth were applied for the treatment of refractory organics.^{5,6} Compared with other AOPs, EO exhibits advantages in higher degradation efficiency, lower treatment cost, no addition of chemical oxidants, smaller footprint and simple operation of treatment equipment, less production of sludge, and no secondary pollution. As a result, the EO process has been widely used in the treatment of aquaculture wastewater,⁷ olive pomace leachate,⁸ and toilet wastewater.⁹

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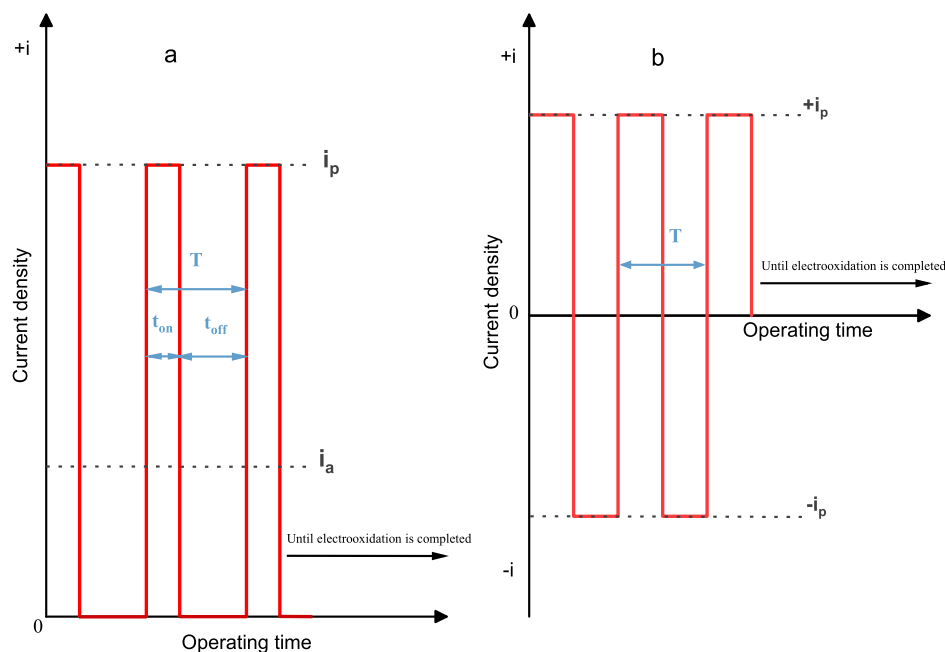


Figure 1. General concept of the pulsating current technology of MP power supply (a) and DP power supply (b).

In addition, EO was also used to remove synthetic dyes,¹⁰ humus/humus-like acids,¹¹ drug residues,¹² and pesticides.¹³

Due to the up-mentioned advantages, many EO and coupling EO processes of electro-flocculation,^{14–16} electro-Fenton,^{14,17} direct and indirect EO,^{18–20} electrochemical reduction,^{21–23} ion-exchange membrane electrochemistry,^{24–26} three-dimensional electrode system,^{27–29} and electrochemical integration process^{30–35} were applied for the treatment of MSW leachate. However, oxidation films are often formed on the anodes,³⁶ which results in fouling of the electrodes and seriously hinders the degradation of organic pollutants on the metal anodes during the traditional EO treatment process with direct-current (DC–EO). However, the formation of oxidation films and fouling of electrodes, anodes loss, and energy consumption are significantly reduced when the pulse power supply is applied to the EO process (PS–EO).³⁷ As a result, PS–EO reduces the total current intensity but improves the current efficiency (CE), which is more suitable for industrial treatment of refractory wastewater.³⁸

Unlike the continuous electrolysis of DC–EO, the PS–EO process is characterized as existing pulse intervals with pulse-on time (t_{on}) and pulse-off time (t_{off}).³⁶ The sum of t_{on} and t_{off} is the pulse period (T), and the reciprocal of T is defined as pulse frequency (f_p). Therefore, duty ratio (R_D) is the ratio of t_{on} to T , which is an important parameter that is often adjusted to achieve an energy-saving effect.³⁹ The average current (i_a) is the current during pulse-on (i_p) multiplied by the ratio of t_{on} to T . Also, it is worth noting that the difference between the monopulse (MP) and double pulse (DP) in the latter process will change the current direction cyclically and constantly, while the former one will not (Figure 1). The concept of PS–EO-related parameters is shown in the following eqs 1–3 and Figure 1.

$$f_p = \frac{1}{t_{on} + t_{off}} = \frac{1}{T} \quad (1)$$

$$R_D = \frac{t_{on}}{t_{on} + t_{off}} = \frac{t_{on}}{T} \quad (2)$$

$$i_a = i_p \times \frac{t_{on}}{t_{on} + t_{off}} = i_p \times \frac{t_{on}}{T} \quad (3)$$

As summarized above, the EO, especially the PS–EO process, is an efficient and cost-effective wastewater treatment technology, which shows great potential for the pretreatment or advanced treatment of the MSW leachate in practical industrial applications. Besides, the MSW leachate also shows extremely high conductivity and concentration of chloride ion (Cl^-), which could be used by EO for indirect oxidation to reduce the treatment cost.^{39,40} However, seldom does work focus on the comparative study with the DC–EO and PS–EO processes comprehensively for the treatment of real wastewater of the MSW leachate. Moreover, many researchers emphasize the preparation of various electrodes but ignore the design of electrochemical reactors.^{41–44}

In this work, different EO processes of DC–EO, EO with MP (MP–EO), and EO with a DP (DP–EO) in a home-made multi-channel flow electrochemical reactor (MCFER) for the advanced treatment of a biologically treated leachate (BTL) were investigated comparatively. The effects of operating parameters of current density (I_A), superficial liquid velocity (U_L), initial concentration of Cl^- (C_{Cl^-}), f_p , and R_D on the removal efficiency of chemical oxygen demand (COD) (RE_{COD}), total organic carbon (TOC) (RE_{TOC}), and total nitrogen (TN) (RE_{TN}) were studied simultaneously. Average energy consumption ($\overline{\text{EC}}$) and organic composition of the treated effluent of DC–EO and PS–EO were also compared comprehensively, and a new mechanism of the PS–EO process was proposed correspondingly.

2. RESULTS AND DISCUSSION

2.1. Comparison of Different EO Processes. The effects of different sources of power supply of the EO process on RE_{COD} and RE_{TN} of BTL were investigated, and the results are

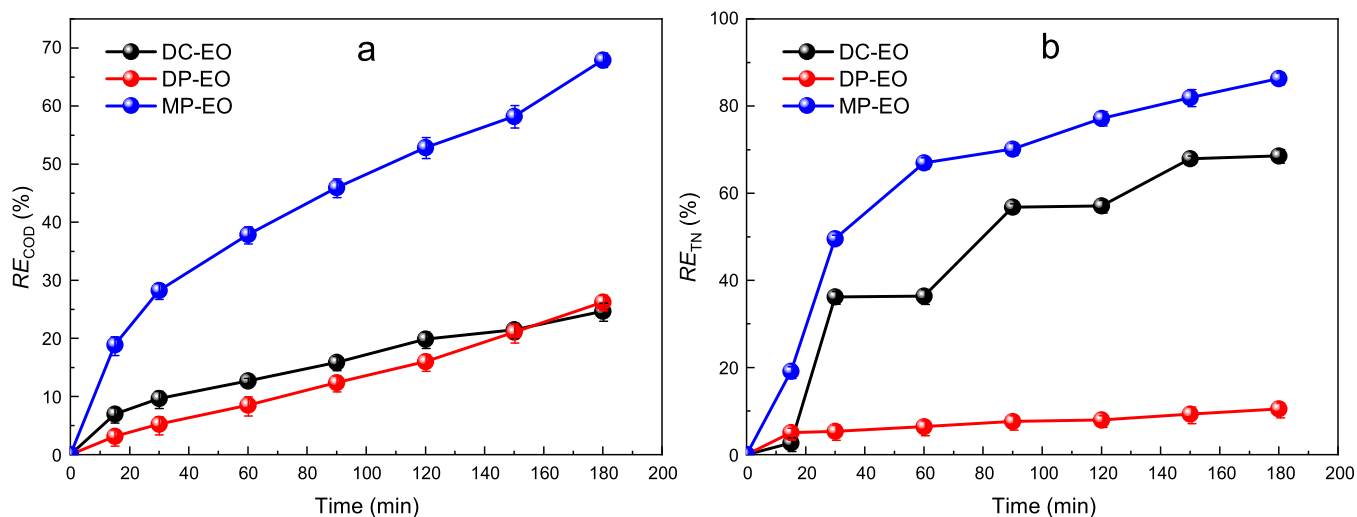


Figure 2. Effect of power supply on COD removal (a) and TN removal (b).

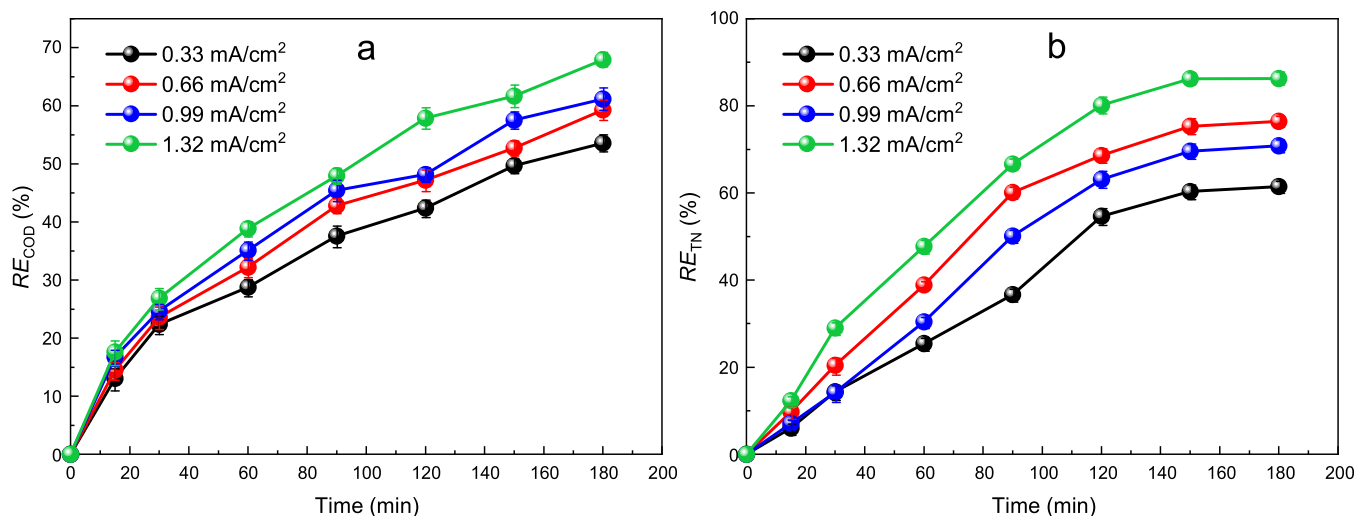
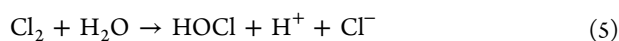


Figure 3. Effect of I_A on COD removal (a) and TN removal (b).

shown in Figure 2. As can be seen from Figure 2 that the RE_{COD} and RE_{TN} increase with the extension of reaction time. The RE_{COD} and RE_{TN} of MP-EO are the best, followed by DC-EO, and DP-EO is the worst. In the initial stage, the degradation rate of COD is fast, and the RE_{COD} of MP-EO reached 28.25% in the first 30 min. At this time, the content of oxidizing substances produced by electrolysis was high, and the COD degradation rate was fast in BTL.⁴⁵ Then, with the decrease in the concentration of refractory organic matter, the degradation rate of COD slows down gradually. The RE_{TN} showed a similar pattern to that of RE_{COD} . The removal of pollutants was mainly by indirect oxidation, and the possible reactions that happened in the BTL are shown in eqs 4–6^{46,47}



These results may be because the instantaneous current of MP-EO is several times or even dozens of times higher than that of the traditional DC-EO process, and more active chlorine (Cl_2/OCl^-) could be produced, leading to a much

higher degradation rate and efficiency of COD and TN.^{36,37,48} Second, the oxidants and other intermediate products, produced on the electrodes by MP-EO, could be better to be mass transferred into other parts of the BTL when the pulse transmitted off during the t_{off} times.³⁸ At the same time, raw material Cl^- of the EO process could also be transferred into the surface of the electrodes, resulting in the elimination of concentration polarization and benefits to the next pulse electrolysis cycles.³⁸ However, the continuous and periodic alternating of the positive and negative electrodes of the DP-EO process adversely affected the formation of Cl_2/OCl^- because the original cathode plates of 304 stainless steel are not designed for the chlorine evolution reaction of eqs 4–6.⁴⁹ As expected, inevitably corrosion happened on the electrode plates of 304 stainless steel because of the reactions shown in eqs 7 and 8^{50,51}



Experimental conditions: $V = 2$ L, I_A was 1.32 mA/cm², U_L was 0.46 cm/s, f_P was 5000 Hz, R_D was 40%, and C_{Cl^-} was 4129 mg/L.

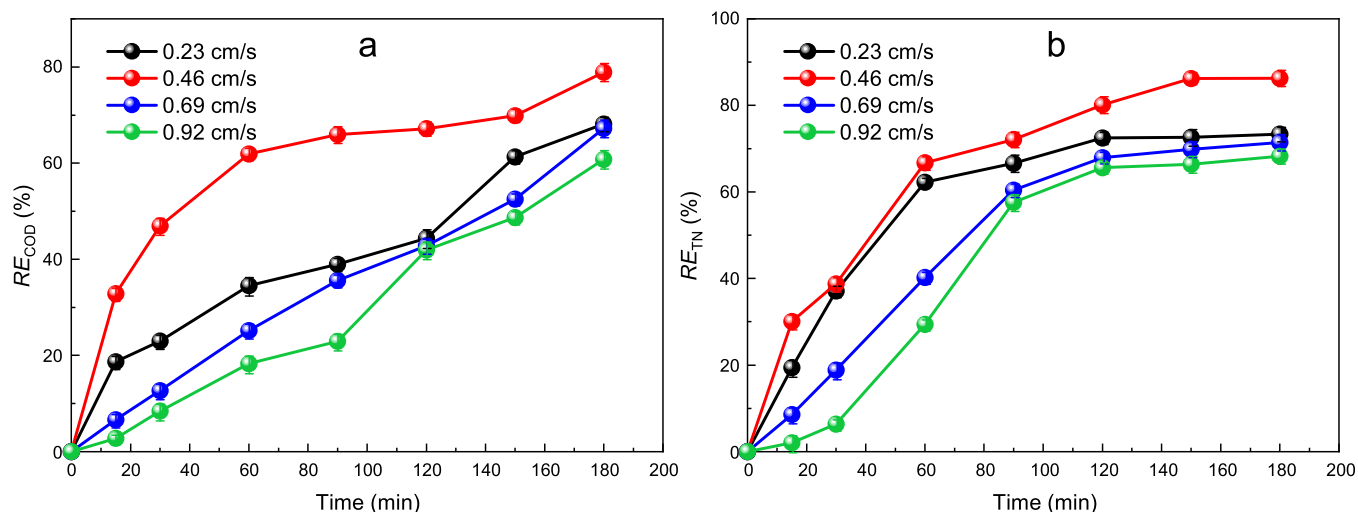


Figure 4. Effect of U_L on COD removal (a) and TN removal (b).

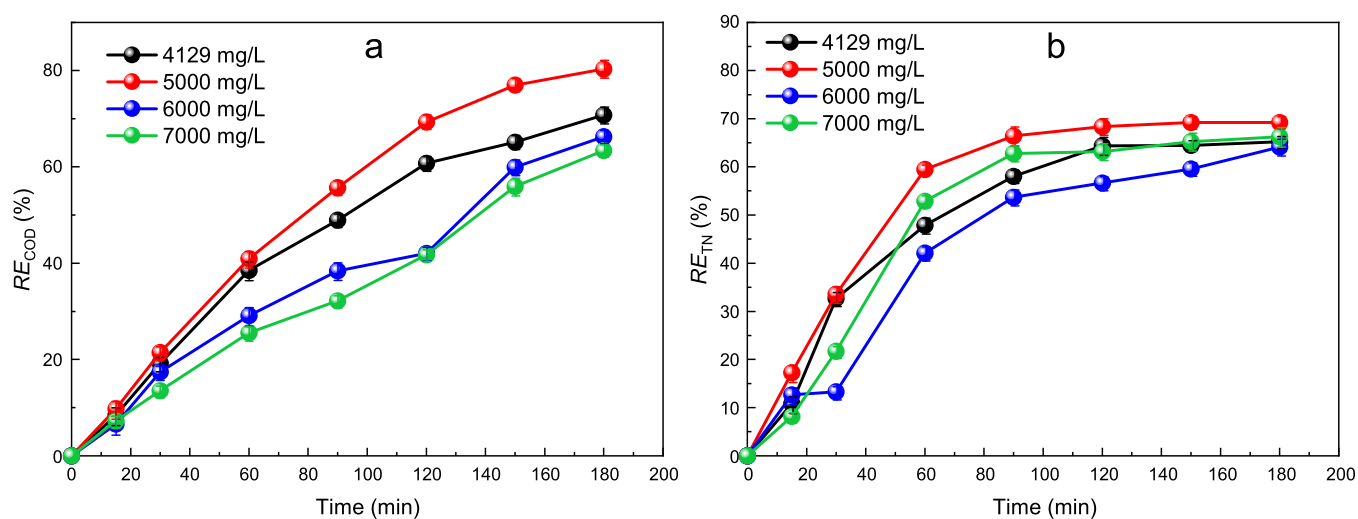


Figure 5. Effect of C_{Cl^-} on COD removal (a) and TN removal (b).

2.2. Effects of Process Parameters of MP–EO on COD and TN Removal.

2.2.1. Effect of I_A . As shown in Figure 3, the RE_{COD} and RE_{TN} of the BTL gradually increase with the increase in I_A , resulting in a higher I_A for a better RE_{COD} and RE_{TN} . When I_A increased to 1.32 mA/cm², the RE_{COD} and RE_{TN} reached 67.9 and 86.29%, respectively. When the low I_A is applied, the potential required for the reaction of eq 4 is higher than that can be provided by the anodes, which cause adverse effects to the reaction, resulting in less production of Cl_2/OCl^- .⁵² Contrarily, with the increase in I_A , more electrons pass through the cathodes and anodes, leading to the enhancement of EO, increasing more production of Cl_2/OCl^- , thus accelerating the degradation of COD and TN.⁵³ However, when the I_A increase to a certain extent, the RE_{COD} and RE_{TN} are mainly controlled by mass transfer factors, and the CE is also significantly reduced.⁵³

Experimental conditions: $V = 2$ L, MP power supply, U_L was 0.46 cm/s, f_p was 5000 Hz, R_D was 40%, and C_{Cl^-} was 4129 mg/L.

2.2.2. Effect of U_L . As shown in Figure 4, the RE_{COD} and RE_{TN} first increased but then decreased with the increase in U_L , there existed an optimum value of U_L . This is because the mixing and mass transfer of organic pollutants and oxidizer of

active chlorine in BTL were strengthened by an increasing U_L , which favored the removal rate of COD and TN when U_L increased from 0.23 to 0.46 cm/s.⁵³ However, with the further increase in U_L from 0.46 to 0.92 cm/s, the drastically mixing effect of the fluid between the electrode plates also promotes the mass transfer of Cl_2/OCl^- to the cathodes to be consumed, resulting in a decrease in the oxidizer and bringing about undesirable effects on RE_{COD} and RE_{TN} , which was also reported by other researchers.^{54,55}

Experimental conditions: $V = 2$ L, MP power supply, I_A was 1.32 mA/cm², f_p was 5000 Hz, R_D was 40%, and C_{Cl^-} was 4129 mg/L.

2.2.3. Effect of C_{Cl^-} . As observed in Figure 5, the RE_{COD} and RE_{TN} showed a trend of first increasing and then decreasing with the increase in C_{Cl^-} . When C_{Cl^-} increased from the initial concentration of BTL of 4129 to 5000 mg/L, the RE_{COD} and RE_{TN} were the highest of 78.90 and 68.66%, respectively. However, when C_{Cl^-} continued to increase to 7000 mg/L, the removal rate of organics and TN slowed down and RE_{COD} and RE_{TN} also descended. The RE_{COD} and RE_{TN} in the reaction system are mainly based on the indirect oxidation, appropriately increasing the C_{Cl^-} is beneficial to the generation of more Cl_2/OCl^- in the BTL and accelerates the removal of COD and

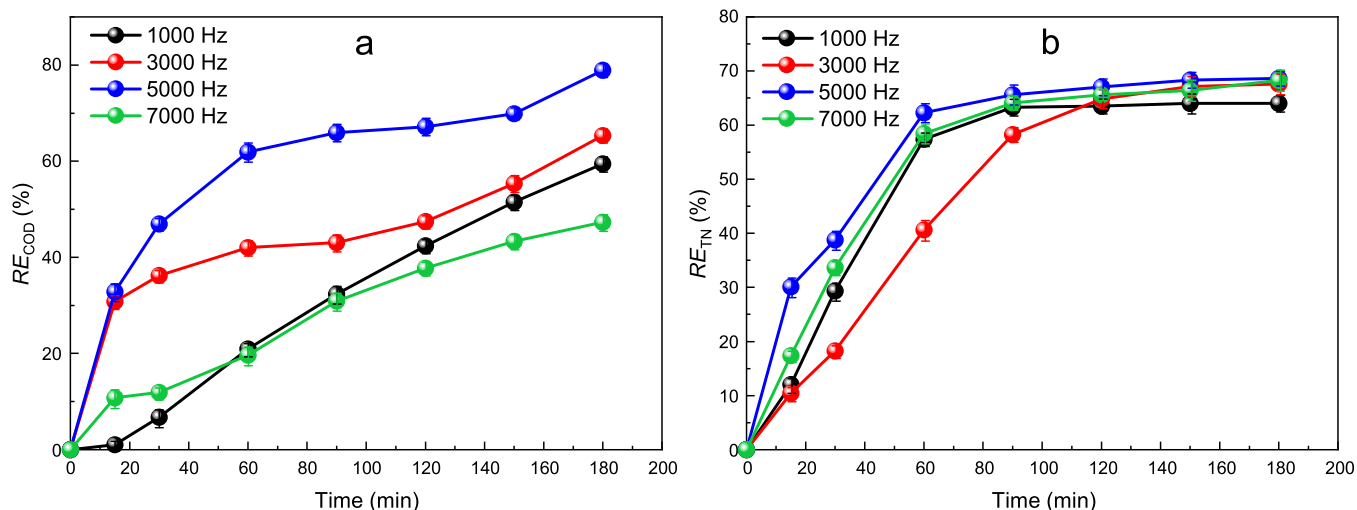


Figure 6. Effect of f_p on COD removal (a) and TN removal (b).

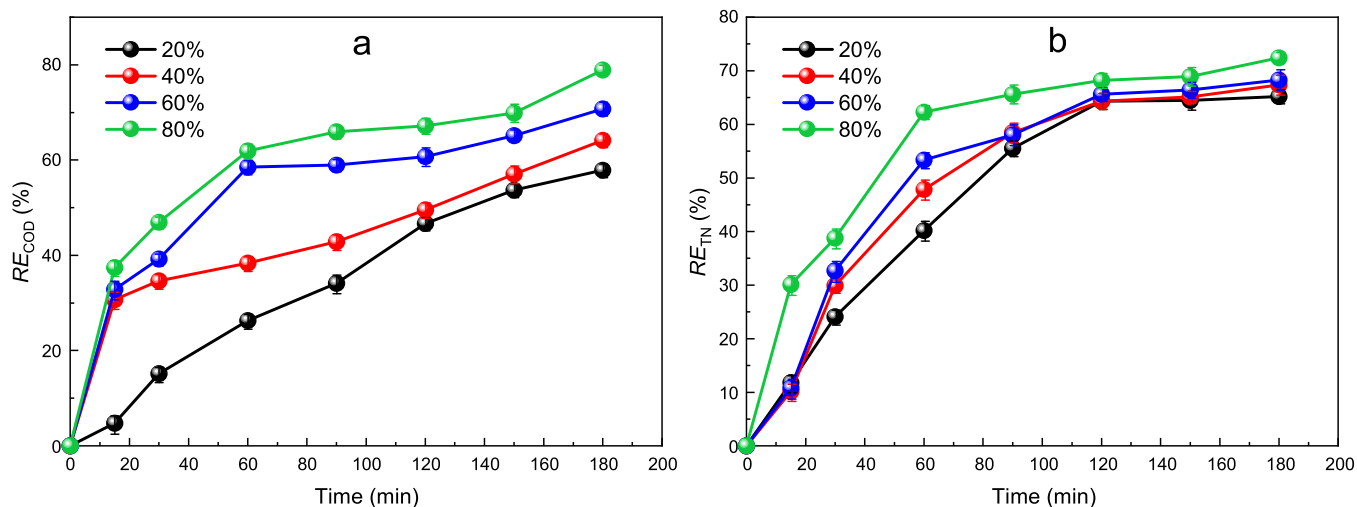


Figure 7. Effect of R_D on COD removal (a) and TN removal (b).

TN.^{47,56} Nevertheless, from the kinetic perspective, the kinetics of removal of COD and TN by Cl_2/OCl^- can be regarded as controlled by two K values: the available Cl_2/OCl^- (K_1) generated by electrolytic oxidation of Cl^- and the available active chlorine (K_2) participated in the removal of COD and TN.⁵² The increase in C_{Cl^-} only affects K_1 but has little effect on K_2 . Therefore, the COD and TN removal rates do not increase linearly with the increase in C_{Cl^-} . However, excessive addition of Cl^- could produce more toxic by-products.⁵⁷ Therefore, the appropriate C_{Cl^-} should be chosen as 5000 mg/L.

Experimental conditions: $V = 2$ L, I_A was 1.32 mA/cm², U_L was 0.46 cm/s, f_p was 5000 Hz, R_D was 40%, and MP power supply.

2.2.4. Effect of f_p . Figure 6 shows the trend of RE_{COD} and RE_{TN} with the increase f_p from 1000 to 7000 Hz. When the f_p increased from 1000 to 5000 Hz, the removal pattern of COD and TN was different, the rate of RE_{COD} increased gradually while RE_{TN} kept almost constant, and RE_{COD} and RE_{TN} reached 78.90 and 68.66% after 180 min treatment of BTL, respectively. Further increasing the f_p to 7000 Hz decreased the RE_{COD} and RE_{TN} . When the f_p was as low as 1000 Hz, the pulse time of T was long, and the frequency of charge–

discharge circles of the pulse was correspondingly low. As a result, the less transient current was generated by discharge,^{37,48} and the mass-transfer effect of MP–EO was also not obvious.³⁸ However, with the increase in f_p to 5000 Hz, the number of the transient current increased at the moment of charge–discharge in a short T , leading to an increased generation of active chlorine and mass-transfer effect. However, t_{on} and T are continually decreased with the further increase in f_p of 7000 Hz, which is closer to the DC–EO, which exhibited an adverse effect on the production of active chlorine and the mass-transfer efficiency.⁵⁸

Experimental conditions: $V = 2$ L, I_A was 1.32 mA/cm², U_L was 0.46 cm/s, C_{Cl^-} was 4129 mg/L, R_D was 40%, and MP power supply.

2.2.5. Effects of R_D . As can be seen in Figure 7, the RE_{COD} and RE_{TN} of BTL increase with the increase in R_D . When the R_D is 80%, the treatment performance is the best, and the RE_{COD} and RE_{TN} reach 78.90 and 72.43% after 180 min of BTL treatment, respectively. Because the intermittent reaction of pulse electrolysis improves the diffusion rate of different chemicals of active chlorine, organic pollutants, and intermediate product, and so forth, it benefits the EO reaction to reduce the overpotential and energy consumption.^{59,60}

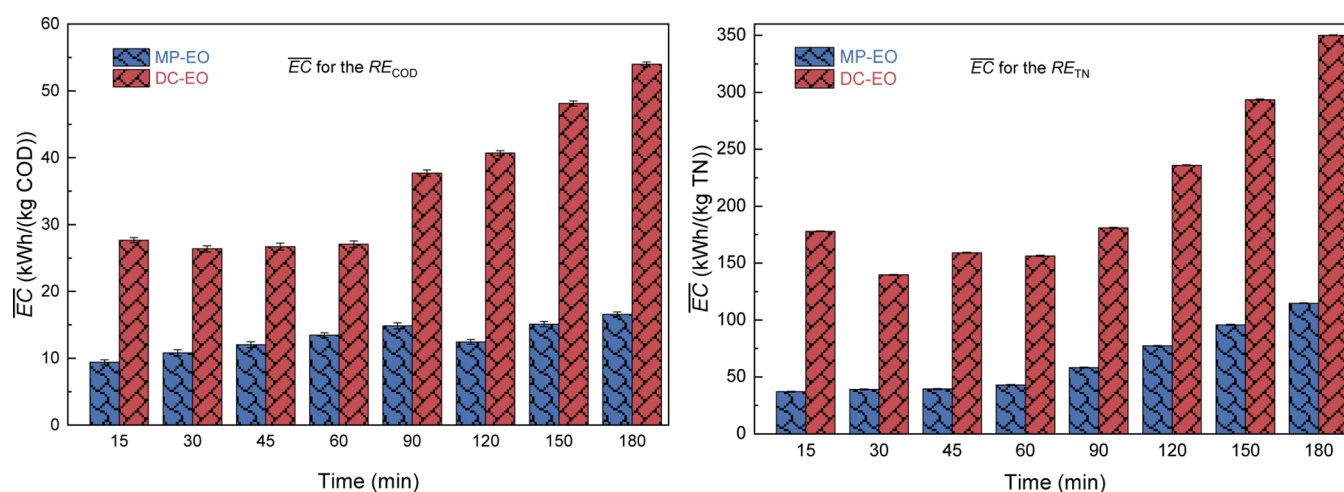


Figure 8. Comparison of \overline{EC} of MP-EO and DC-EO supply at different times.

However, the ineffective intermittent electrolytic time can lead to the prolongation of electrolytic working time, which is attributed to the fact that increasing R_D favors better removal rate and efficiency of COD and TN.⁶⁰

Experimental conditions: $V = 2$ L, I_A was 1.32 mA/cm², U_L was 0.46 cm/s, f_p was 5000 Hz, MP power supply, and C_{Cl^-} was 4129 mg/L.

2.3. Comparison of Energy Consumption of MP-EO and DC-EO. As optimized above, better treatment conditions for organics and TN removal of MP-EO are as follows: I_A of 1.32 mA/cm², U_L of 0.46 cm/s, C_{Cl^-} of 5000 mg/L, R_D of 80%, and f_p of 5000 Hz. For a better comparison, the conditions for the DC-EO process are strictly the same. Under optimal conditions, 2 L of BTL was treated with MP-EO for 180 min, and the RE_{COD} , RE_{TOC} , and RE_{TN} could reach as high as 80, 30, and 80%, respectively. The effect of process parameters on RE_{TOC} are shown in Figure S1. Besides, the treatment data of BTL under the optimal conditions of MP-EO and DC-EO were used to calculate the \overline{EC} by eq 17, and the results are illustrated in Figure 8 and Table S1.

As can be seen in Figure 8, not only \overline{EC} of COD but also of TN, MP-EO exhibits much more energy saving than that of DC-EO, and the percentage of \overline{EC} for COD of DC-EO was 201.5–326.6% of that of MP-EO and 304.9–479.0% for TN. The overall \overline{EC} of MP-EO for 3 h treatment of BTL is 16.58 kW h/(kg COD) and 114.83 kW h/(kg TN), while that of DC-EO is 53.97 kW h/(kg COD) and 350.12 kW h/(kg TN). Moreover, Figure 8 also shows that \overline{EC} for the first 60 min treatment of BTL of both MP-EO and DC-EO is lower than that of the other longer time, especially for the DC-EO. At the first 60 min, the concentration of COD and TN was higher in BTL, so that CE was higher, leading to a lower \overline{EC} of the first 60 min. However, with the extension of the treatment time, the concentration of COD and TN decreased, reducing of the EO treatment efficiency and increasing the energy consumption. Because of the existence of on-off-on circles of the PS-EO process, the working time of MP-EO is much shorter than that of the DC-EO under the same conditions. Besides, the higher mass transfer and instantaneous current effect of MP than that of DC-EO, as discussed in the Section 2.1,³⁸ also improves EO treatment efficiency. As a result, compared with DC-EO, the MP-EO could obtain higher CE but lower energy consumption.

The \overline{EC} of COD of various EO processes for the treatment of the MSW leachate with different reactors was compared and is summarized in Table 1. As can be noted in the table, commonly the \overline{EC} is rather high for the treatment of leachate by EO without a specially designed system, which is from dozens to more than a hundred kW h/(kg COD), because of the high concentration of refractory organics.^{61–64} However, this work, with the help of MP-EO and a multi-channel flow reactor with an electrode gap of 5 mm, exhibits higher treatment efficiency but lower energy consumption with \overline{EC} of only 16.58 kW h/(kg COD), which may be suitable for industrial applications of advanced treatment of the BTL.

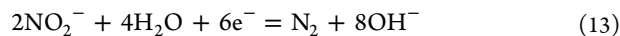
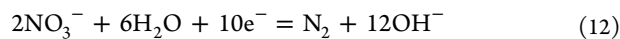
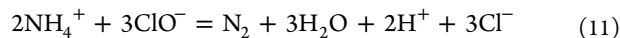
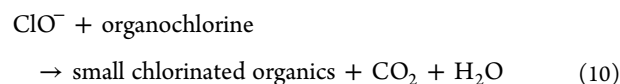
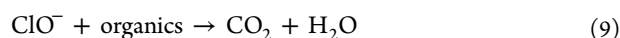
Experimental conditions of MP-EO: $V = 2$ L, the I_A of 1.32 mA/cm², the U_L of 0.46 cm/s, C_{Cl^-} of 5000 mg/L, the R_D of 80%, and f_p of 5000 Hz; experimental conditions of DC-EO: $V = 2$, the I_A of 1.32 mA/cm², the U_L of 0.46 cm/s, and C_{Cl^-} of 5000 mg/L.

2.4. Comparison of the Chemical Composition of the Treated Effluent with MP-EO and DC-EO. Agilent 7890A-5975C gas chromatography–mass spectrometry (GC–MS) was used to analyze the organic content of the original BTL and treated the effluent with MP-EO and DC-EO for 3 h, and the detected organic matters with the credibility more than 60% are listed and shown in Figures 9 and 10 and Table S2. As observed in Figures 9 and 10, the organic matter in the BTL reduced significantly after the EO treatment from 53 kinds in BTL to the treated effluent with MP-EO of 11 kinds and with DC-EO of 29 kinds. Besides, the species of 24 organic compounds, including seven hydrocarbons, seven esters, three ketones, one alcohol, and six others, were significantly eliminated by the DC-EO. Correspondingly, the species of 45 organic compounds of 21 hydrocarbons, 8 esters, 4 ketones, 3 alcohols, and 9 others, were also significantly eliminated, while three organochlorines were increased by the MP-EO. These results were attributed to the EO degradation of macromolecular organics into small molecules, or even completely oxidized into carbon dioxide and water by Cl_2/OCl^- ,⁶⁶ and higher production of Cl_2/OCl^- of MP-EO than that of DC-EO (as discussed in the section comparison of different EO processes). The emerging of new organochlorines may be caused by the side effect of addition and substitution reactions of the higher concentration of active chlorine produced by the MP-EO process.⁶⁷

Table 1. Comparison of \overline{EC} under the Different Systems for the Treatment of the Leachate

| reactors | electrodes/power types | electrolysis channels | wastewater/treatment capacity | other experimental condition | RE and \overline{EC} | references |
|---|---|-----------------------|---|---|---|------------|
| electrolytic cell of thermostated glass reservoir | anode: Ti/PbO ₂ ; cathode: stainless steel (304); electrolyte: 1800 mg/L Cl ⁻ ; DC power supply | 2 | landfill leachate/0.35 L | initial COD: 780 mg/L; flow rate: 420 L/h; reaction time: 3 h; I_A : 18.52 mA/cm ² | COD: <160 mg/L 145.16 kW h/(kg COD) | 61 |
| electrolytic cell of cylindrical vessel | anode: Ti/Pt; cathode: perforated stainless steel cylinder; electrolyte: 4% NaCl; 4 g/L FeSO ₄ ·7H ₂ O; 3120 mg/L Cl ⁻ ; DC power supply | 1 | landfill leachate/6 L | initial COD: 60,000 mg/L; flow rate: 6 mL/min; reaction time: 5 h; applied voltage: 20 V | RE _{cod} : 41.6% 4.29 kW h/(kg COD) | 65 |
| electrochemical system of cylindrical vessel | anode: boron-doped diamond on silicon; cathode: stainless steel; electrolyte: 1630 mg/L Cl ⁻ ; DC power supply | 1 | BTL/250 L | initial COD: 860 mg/L; flow rate: 300 L/min; reaction time: 7 h; I_A : 30 mA/cm ² | COD: <160 mg/L 53.00 kW h/(kg COD) | 62 |
| electrolytic reactor of glass pot | anode: Ti/IrO ₂ -RuO ₂ ; cathode: zirconium; electrolyte: 1 mol/L HClO ₄ ; 6150 mg/L Cl ⁻ ; DC power supply | 2 | BTL/120 L | initial COD: 2960 mg/L; reaction time: 4 h; I_A : 32 mA/cm ² | RE _{cod} : 90% 35.00 kW h/(kg COD) | 63 |
| electrolytic cell of undivided cylindrical glass containing a MCFER | anode: Ti ₄ O ₇ ; cathode: FeIFeII-LDH-modified carbon felt; electrolyte: 0.2 mM Fe ²⁺ ; DC power supply | 1 | landfill leachate (NF concentrate)/0.22 L | initial COD: 860 mg/L; reactiontime: 4 h; I_A : 4.2 mA/cm ² | RE _{cod} : 45% 110.00 kW h/(kg COD) | 64 |
| | anode: Ti/RuO ₂ -IrO ₂ ; cathode: stainless steel (304); electrolyte: 5000 mg/L Cl ⁻ ; MP power supply | 4 | BTL/2 L | initial COD: 844.31 mg/L; I_A : 1.32 mA/cm ² ; UL: 0.46 cm/s; R _D : 80%; f_P : 5000 Hz; treatment time: 3 h | RE _{cod} : 80% 16.58 kW h/(kg COD) | this work |

2.5. Mechanism Analysis of MP–EO. The EO and reduction exist simultaneously in the treatment process of the MSW leachate by electrolysis (Figure 11).⁶⁶ The formation of active indirect oxidation species (such as chlorine, persulfate, percarbonate, etc.) is based on the anodic oxidation of ions in the leachate.⁶⁶ However, in this study of the electrolysis treatment of BTL, it was found that the main indirect oxidation species was of Cl₂/OCl⁻ produced by Cl⁻ oxidation because of the high concentration of Cl⁻ in the MSW leachate (as shown in eqs 4–6). The indirect oxidation of Cl₂/OCl⁻ plays a leading role in removing COD and ammonia nitrogen (NH₃-N) of the leachate,^{19,68} and the process can be described in eqs 9–11.^{18,69,70} The main form of TN of BTL is NH₃-N,⁶⁸ which accounts for 98.7% of TN, as shown in Table 2, which may be removed by eq 11. In addition, the electrochemical reduction of the cathode determines the removal of nitrate or nitrite (eqs 12 and 13).^{22,71}



3. CONCLUSIONS

In this study, a self-made four-channel flow electrochemical reactor was used for the post treatment of the BTL for further removal of COD, TOC, and TN by the EO process. The effects of different power sources and process parameters of I_A , U_L , C_{Cl^-} , R_D , and f_P were investigated. The results showed that MP–EO exhibits better removal performance of refractory organics and ammonia nitrogen than that of the DC–EO. The optimal conditions for MP–EO is that of I_A 1.32 mA/cm², U_L 0.46 cm/s, C_{Cl^-} 5000 mg/L, R_D 80%, and f_P 5000 Hz. Under these conditions, the RE_{COD}, RE_{TOC}, and RE_{TN} of MP–EO could reach as high as 80, 30, and 80%, respectively, after the treatment of 2 L BWL for 180 min. The MP–EO process is highly energy saving, and the overall average energy consumption of it is only 16.58 kW h/(kg COD) and 114.83 kW h/(kg TN), while that of DC–EO is 53.97 kW h/(kg COD) and 350.12 kW h/(kg TN), which is only 30.72 and 32.80% of that of the latter ones. Moreover, the treated effluent of MP–EO is safer to discharge into the ecosystem than that of DC–EO, and the organic kinds of which is reduced from 53 in BTL to 11 which is also much less than 29 kinds treated with the DC–EO. The MP–EO demonstrates an excellent performance and economic prospects in the continuous and industrial treatment of refractory wastewater.

4. MATERIALS AND METHODS

4.1. Water Quality of BTL. The BTL was sampled from a local MSW incineration power plant in Chongqing, China. Its physicochemical characteristics are shown in Table 2. As shown in the table, the COD, NH₃-N, and TN of BTL are as high as 844.31, 125.62, and 127.26 mg/L, respectively, which are far beyond the allowable limits of COD of 100 mg/L, NH₃-N of 25 mg/L, and TN of 40 mg/L that must be further

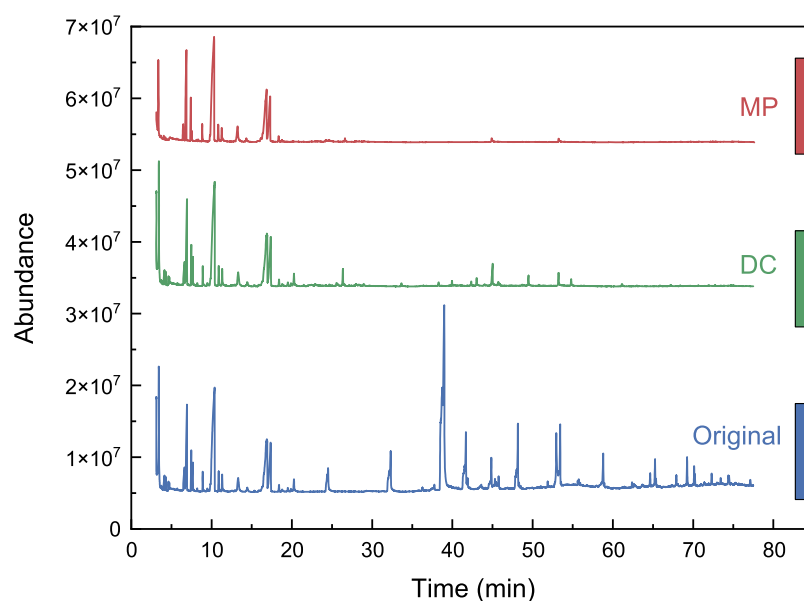


Figure 9. GC-MS analysis of the organic content of BTL after treatment under DC-EO and MP-EO.

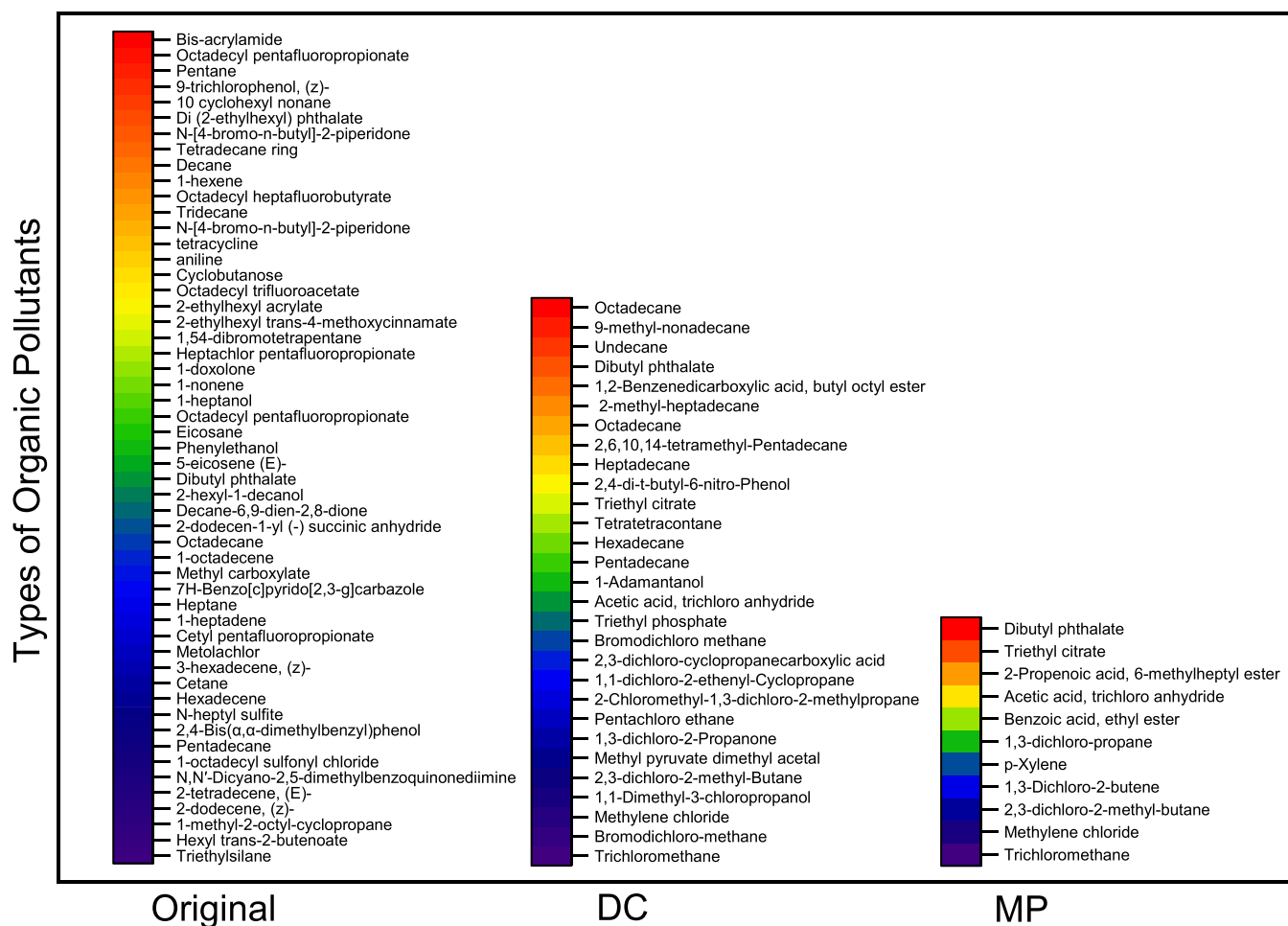


Figure 10. Variations of organic pollutants in BTL before and after DC-EO and MP-EO treatment.

well treated. The high concentration of Cl^- of 4128.91 mg/L indicates that the BTL is suitable for the EO treatment.

4.2. Design of the Electrochemical Reactor and Experimental Setup. The MCFER (190 mm \times 32 mm \times 314 mm, $L \times W \times H$) was made of polymethyl methacrylate

with four electrolytic channels of 5 mm—electrode—gap. Two titanium plates (190 \times 200 mm) coated with a mixed oxide layer of RuO_2 and IrO_2 ($\text{Ti/RuO}_2\text{-IrO}_2$) were used as the anodes, and three plates of 304 stainless steel with the same size (190 \times 200 mm) were used as the cathodes. The details of

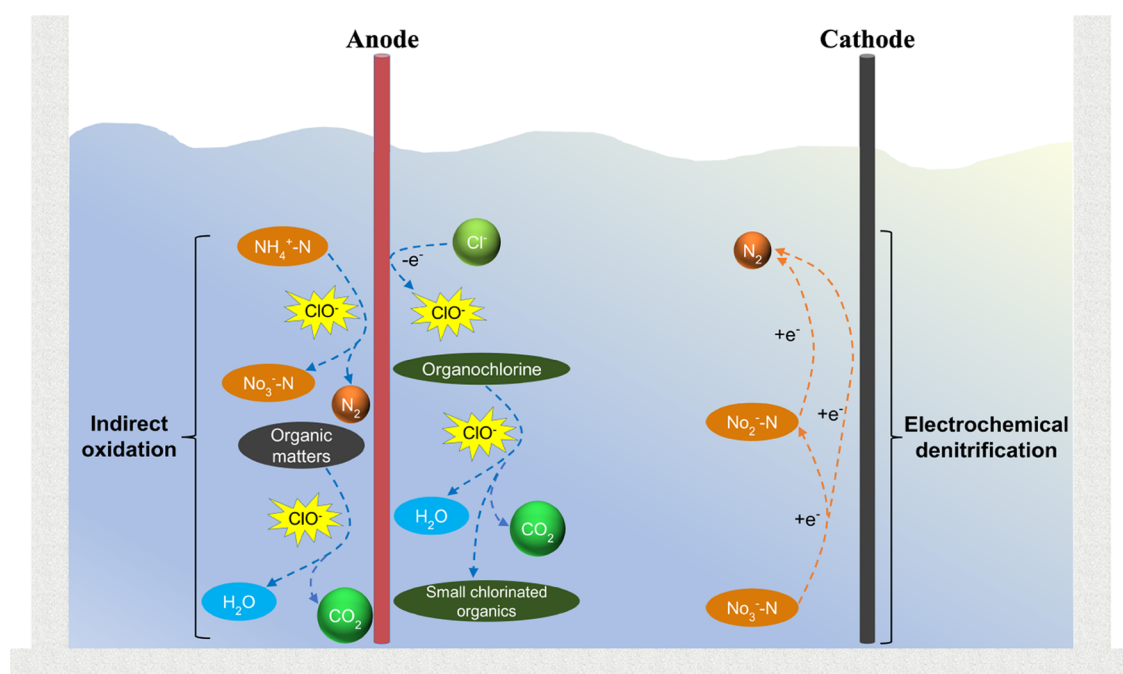


Figure 11. Schematic diagram of the EO process for BTL treatment.

Table 2. Characteristics of BTL

| parameters | BTL |
|---------------------------|---------------|
| color (times) | 350–400 |
| Cl ⁻ (mg/L) | 4128.91 ± 100 |
| COD (mg/L) | 844.31 ± 40 |
| TOC (mg/L) | 539.17 ± 20 |
| NH ₃ -N (mg/L) | 125.62 ± 2 |
| TN (mg/L) | 127.26 ± 2 |
| pH (mg/L) | 8.381 ± 0.02 |

the MCFER and the experimental setup of EO treatment of BTL are shown in Figure 12. A pulse power supply (KXN-

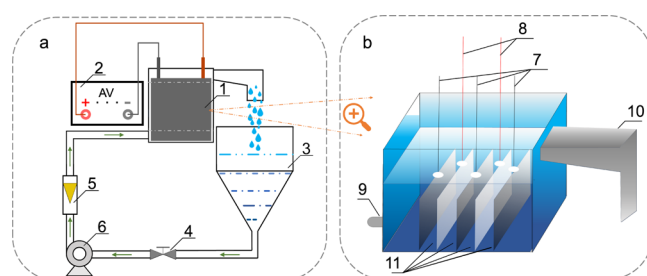


Figure 12. Diagram of the MCFER (a) and experimental setup of MP-EO (b) 1-MCFER reactor; 2-pulse power supply; 3-conical bottom water tank; 4-water valve; 5-rotameter; 6-circulating pump; (b) 7-cathodes; 8-anodes; 9-water inlet; 10-water outlet; 11-electrolytic channels.

6050D) with DC, MP, and DP models was used as the power source of DC-EO, MP-EO, and DP-EO processes in the experiments, respectively. A conical container with a diameter of 350 mm and a height of 300 mm with a capacity of 10 L was used as a water tank to store the BTL. The wastewater was circulated between the MCFER and water tank by a circulating pump (HQB-2500). The fluid flow was recorded by a

rotameter (LZS-15), which could be controlled by a valve set ahead of the pump.

4.3. EO Treatment Experiments. During each experiment, 2 L of BTL was added to the water tank. After being circulated for 10 min with the circulating pump, a 10 mL of the sample named the zero-time-sample was collected to determine the original water quality of the BTL. After that, the power supplier was turned on to start the EO treatment, and its parameters were set to the preset values of the experimental conditions. The EO treatment experiments were carried out at a constant ambient temperature (25.0 ± 1.0 °C), different power supply sources (DC, MP, and DP), I_A (0.33, 0.66, 0.99, and 1.32 mA/cm²), U_L (0.23, 0.46, 0.69, and 0.92 cm/s), C_{Cl^-} (4129, 5000, 6000, and 7000 mg/L), f_p (1000, 3000, 5000, and 7000 Hz), and R_D (20, 40, 60, and 80%). During each EO treatment process, 10 mL of samples was taken out from the water tank to detect the water quality of the treated effluent at certain time intervals. Each experiment was conducted in duplicate, and the average and the deviation of the results between the same experiments were calculated, correspondingly.

4.4. Analytical Methods. The COD of BTL was analyzed according to the Chinese standard water quality determination method of the fast digestion spectrophotometry. The absorbance was recorded using a UV-vis spectrophotometer (TU-1901, Beijing Puxi analyzing devices Ld. Co., China). The RE_{COD} was calculated in terms of the initial COD and COD after EO treatment of t time. The TOC and TN were analyzed using a TOC/TN analyzer (TOC-LCPH/CPN, Shimadzu, Japan). The RE_{COD} , RE_{TOC} , and RE_{TN} of the EO process were calculated by eqs 14–16 accordingly

$$RE_{COD} = \frac{C_0 - C_t}{C_0} \times 100\% \quad (14)$$

$$RE_{TOC} = \frac{C_0 - C_t}{C_0} \times 100\% \quad (15)$$

$$RE_{TN} = \frac{C_0 - C_t}{C_0} \times 100\% \quad (16)$$

where C_0 and C_t (mg/L) are the concentrations of COD, TOC, and TN at the initial and t time of EO treatment, respectively.

The organic compounds of the BTL before and after the EO treatment were identified by GC–MS (Shimadzu GCMS-QP2010 Plus, Japan), and the analytical procedures and conditions are described in our another work in detail.⁶⁷ The Cl^- was measured using a silver nitrate titration method. The color and pH were determined using a dilution multiple method and pH meter (pHS-3E, Leici, China), respectively.

The \overline{EC} for COD/TN of EO treatment can be calculated by the following eq 17³⁹

$$\overline{EC} = \frac{1000 UIR}{V(c_0 - c_t)} \quad (17)$$

where \overline{EC} is the average energy consumption, kW h/(kg COD) or kW h/(kg TN); U is the voltage, V; I is the current, A; T is the treatment time, h; R is the R_D ; V is the wastewater volume, L; C_0 is the initial COD/TN of BTL; and C_t is the COD/TN after EO treatment at t time, mg/L.

■ ASSOCIATED CONTENT

SI Supporting Information

The Supporting Information is available free of charge at <https://pubs.acs.org/doi/10.1021/acsomega.1c03567>.

Comparison of organic compounds in BTL after EO of MP and DC; effect of power supply, I_A , U_L , f_p , R_D , and C_{Cl^-} on TOC removal; and comparison of average energy consumption by COD and TN of MP–EO and DC–EO (PDF)

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Notes

The authors declare no competing financial interest.

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■ ABBREVIATIONS

| | |
|---------------------------------------|--|
| AOPs | advanced oxidation processes |
| BDD | boron-doped diamond |
| BOD | biochemical oxygen demand |
| BTL | biologically treated leachate |
| C_{Cl^-} | initial chloride ion concentration |
| CE | current efficiency |
| COD | chemical oxygen demand |
| DC | direct-current |
| DC–EO | electrochemical oxidation with DC |
| DP | double pulse |
| DP–EO | electrochemical oxidation with DP |
| \overline{EC} | average energy consumption |
| EO | electrochemical oxidation |
| f_p | pulse frequency |
| GC–MS | gas–mass spectrometry |
| HB | proton donor |
| I_A | current density |
| MCFER | multi-channel flow electrochemical reactor |
| MO_x | metal oxide anode |
| MP | monopulse |
| MP–EO | electrochemical oxidation with monopulse |
| MSW | municipal solid waste |
| nitrate-N | ammonia nitrogen |
| nitrite-N | nitrogen oxides |
| PS–EO | pulse power supply applied to the EO process |
| R_D | duty ratio |
| RE_{COD} | the removal efficiency of COD |
| RE_{TN} | the removal efficiency of TN |
| RE_{TOC} | the removal efficiency of TOC |
| T | pulse period |
| Ti/RuO ₂ –IrO ₂ | mixed oxide layer of RuO ₂ and IrO ₂ |
| TN | total nitrogen |
| t_{off} | electrolytic interval time |
| t_{on} | pulse width |
| U_L | superficial liquid velocity |

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