Optimization of a continuous flow electrocoagulation as pretreatment for membrane distillation of the waste stream in vinyl ester resin production

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Abstract

Vinyl ester resin production wastewater (VERW) contains high concentrations of organics particularly, methacrylic acid and bisphenol A, which are hazardous chemicals and harmful to the aquatic environment. Therefore, there is an urgent need to properly treat the effluent before discharge into the aquatic system. In this work, direct contact membrane distillation (DCMD) was explored as an advanced treatment of the VERW pre-treated by a continuous flow electrocoagulation (EC) and peroxi-electrocoagulation (PEC) processes. Optimization of EC and PEC processes were investigated and the DCMD performance was evaluated. Results showed that the optimal value of current density and polyacrylamide (PAM) dosage was 15 mA/cm^2 and 1 mg/L, respectively in the EC process. For the PEC process, the optimal addition of hydrogen peroxide (H_2O_2) dosage was four times of the chemical oxygen demand (COD) concentration of EC effluent. The COD of VERW was effectively removed via EC followed by PEC (EC-PEC), resulting in the significant alleviation of membrane fouling during DCMD filtration of VERW. The initial flux of DCMD filtration of VERW pre-treated via EC-PEC improved by 35%, compared that only pre-treated by EC. Moreover, the concentration factor (CF) of the DCMD system reached up to 8.1 and the conductivity of distillate was less than 33.2 μS/cm. Hence, the EC and membrane distillation hybrid process paves a new way for the effective treatment of waste steam from resin production.

Keywords: VERW; EC; Peroxi-electrocoagulation; DCMD; Saline high organic wastewater

1. Introduction

Vinyl ester resin (VER) is a thermosetting-modified epoxy resin obtained by the reaction of bisphenol-type or phenolic-type epoxy resin with methacrylic acid[1, 2]. It is a high-quality matrix for anti-corrosion composite materials such as glass fiber-reinforced plastics[1, 3, 4], and widely used in a wide range of industrial processes, such as automobile, petrochemical and, electronic information industry[5, 6]. However, during the VER manufacturing process, a large amount of alkaline production wastewater containing methacrylic acid and bisphenol A (BPA) is generated, which is difficult to treat because of the high salinity, high COD concentration, and low biodegradability[7, 8]. Therefore, the effective treatment of this type of wastewater is urgently needed.

Membrane distillation (MD) has received attention because of its effectiveness in treating high-salinity wastewater. Compared to conventional pressure-driven membrane processes (e.g., nanofiltration, reverse osmosis), MD can be operated under normal pressure and allows only volatile substances to pass through porous hydrophobic membranes, with almost 100% of non-volatile substances rejection and high-quality water recovery[9-12]. In addition, the operating temperature of MD is lower than that of traditional distillation processes with the possibility of using waste heat as energy input[13, 14]. Lokare et al. showed that treating shale gas produced water with DCMD using exhaust gas from a natural gas compression station as a waste heat source was sufficient to concentrate all produced water produced in Pennsylvania to 30wt %[15]. Recently, MD technology has been widely applied in seawater desalination, high-salinity wastewater treatment, and was also regarded as an effective way to achieve

near-zero discharge of wastewater[10, 11]. Sarallakhniknezhad et al. used DCMD to treat hypersaline drilling mud water, producing high quality distillates with water recovery of over 60%[16]. Yan et al. used DCMD process to treat reverse osmosis brine, and the water recovery rate was higher than 70%[17]. Ngo et al. used MD to purify reverse osmosis-concentrated wastewater and achieved 98% overall water recovery for potable water reuse[18]. Therefore, MD has become a promising application in the treatment of VERW to recover high quality water. However, the high concentration of refractory organics in VERW limits the application of MD[19]. First, the higher temperature at the feed side made the volatile organics pass through the membrane and destroyed the distillate quality[20]; second, once organics were adsorbed on the membrane surface, membrane fouling was accelerated, resulting in a rapid decline in membrane flux[21, 22]. Consequently, it is necessary to remove organics from VERW prior to the application of MD.

A variety of methods have been used to treat refractory organic wastewater with high concentration of COD, including condensation, adsorption, extraction, micro-electrolysis, Fenton oxidation, catalytic ozonation, coagulation, and sedimentation[23-28]. Coagulation is an established method to remove non-biodegradable organics[29, 30]. Particularly, compared with aluminum salt, ferric salt has better elimination effect on organic compounds[31]. Moreover, in-situ formation of ferric ions is more conducive to the coagulation of organic compounds than the direct addition of iron salts[31, 32]. Therefore, the EC process based on the in-situ formation of coagulants is expected to be an alternative treatment for the partial or total removal of organics from wastewater

to alleviate membrane fouling during MD process[8, 33, 34].

EC is an attractive approach with the advantages of environmental compatibility, selectivity, compatible of automation, and cost-effectiveness for treating various wastewater[8]. The method includes three successive stages: (a) sacrificial anode forming coagulant; (b) demulsification and destabilization of pollutants; (c) coagulant captures pollutants to form flocs[35, 36]. Moreover, sequential batch and continuous flow are the two common modes of EC process. During the EC process, the electrolytic dissolution of metal anodes produces a variety of hydroxyl metal ion complexes, which are hydrolyzed products and readily polymerized. The monomer and polyhydroxy complexes act as adsorbents to promote the adsorption and coprecipitation of compounds[36]. The electrostatic attraction between the particles causes the destabilized pollutants to aggregate and form readily settleable flocs[37, 38]. Ikhlaq et al. treated veterinary drug wastewater with an ozonic-catalyzed oxidation-coupled EC process. The removal efficiency of COD was 85.1% under optimal conditions[39]. Eryuruk et al. used an EC process with an iron electrode to treat poultry slaughter wastewater, and the COD removal efficiency was up to 95.5%, and the concentration decreased from 8800 to 425 mg/L[36]. Al-Qodah et al. reported coupling EC process with biological treatment for industrial wastewater treatment and found EC had high efficiency on the removal of color and residual COD in water[40]. Moreover, the removal of heavy metal ions and toxic substances in wastewater by EC improved the biodegradability for biological treatment. Meanwhile, the current in the EC process stimulated microorganisms to enhance the wastewater treatment effect[41]. These favorable factors promoted the

development of EC process. However, some dissolved refractory organics might still be in the effluent. Some hybrid processes such as ozone-EC, PEC, adsorption-EC, and ultrasound-EC were used to further improve the removal efficiency of dissolved organic matter in EC process[34, 42]. And the PEC process is suitable to be applied in acidic condition[43]. In the PEC process, externally-added H_2O_2 generates Fenton reactive system and synergistically works with EC[44, 45]. In this process, H_2O_2 is used to react with ferrous ions (Fe^{2+}) to generate hydroxyl radicals (·OH) in an acidic medium, which is conducive to the degradation of organic pollutants.

In previous studies, the integrated EC and MD process was used as an advanced treatment approach to effectively treat various types of waste streams[46]. Chen et al. used MD combined with EC and electro-oxidation to treat concentrated landfill leachate, and the removal efficiency of the organics reached 82.2%[47]. Sardari et al. used MD combined EC to treat the produced water with high salinity, and stable water flux with negligible fouling was reported over a 434 h experimental run[14]. Jebur et al. treated hydraulic fracturing produced water with a combined process of EC, microfiltration, and MD, reducing total dissolved solids (TDS) from 245,300 to 56 mg/L, and total organic carbon (TOC) from 120 to 1 mg/L[48]. However, the existing studies on EC-coupled MD mainly focus on the improvement of MD system performance, ignoring the maximum utilization of EC pretreatment process. In addition, most EC processes investigated are sequential batch mode, and the application of continuous flow EC and PEC coupling MD process to wastewater treatment has not been reported extensively.

In this work, the continuous flow EC and PEC processes were used as pretreatment approaches prior to DCMD filtration of VERW. Optimization of current density, flocculant (polyacrylamide, PAM) dosage and H_2O_2 dosage on the pretreatment performance was investigated. Mechanism of pollutant removal via the continuous flow EC and PEC processes was discussed. Additionally, system performance of DCMD filtration of VERW pre-treated by EC and PEC processes was evaluated.

2. Materials and methods

2.1 Characteristics of VERW

VERW was obtained from Shanghai Showa Polymer Co., LTD, China. The main characteristics of the VERW were summarized in Table S1. VERW was a milky solution with a conductivity of 19.3 mS/cm and an initial pH of 13.8. The VERW had high COD concentration and turbidity, up to 16,796.9 mg/L and 39,200 NTU, respectively. In addition, the concentration of NH_3-N, Na^+, and Ca^{2+} was 502 mg/L, 2511.9 mg/L, and 52.6 mg/L respectively.

2.2 Experimental setup

The VERW contained large amounts of methacrylic acid and BPA with a pungent odor. The white substances in VERW cannot be removed directly through filtration with filter papers or membranes. In the actual industrial treatment process, the pollutants were removed by adding a large amount of special organic acid coagulant and bio-treatment. As shown in Table S2 and Fig.S1, single addition of flocculant, coagulant or

acid had no effective removal effect on pollutants. However, under acidic conditions (pH < 1.8), the simultaneous addition of flocculant and coagulant achieved the demulsification effect and removed pollutants. Therefore, in this work, the wastewater was first acidified with H_2SO_4 to a pH of 1.8. Then, pollutants were removed through continuous flow EC&PEC (Fig.S2-3) and the voltage and quality of effluent were recorded continuously. The stability tests showed the effluent quality was generally stable and did not change with the volume of influent (Fig.S4)[36]. Hence, the representative average concentrations of pollutants in samples were used to evaluate the effluent quality. Finally, suspended particles were removed by microfiltration, before entering the DCMD (Fig.1).

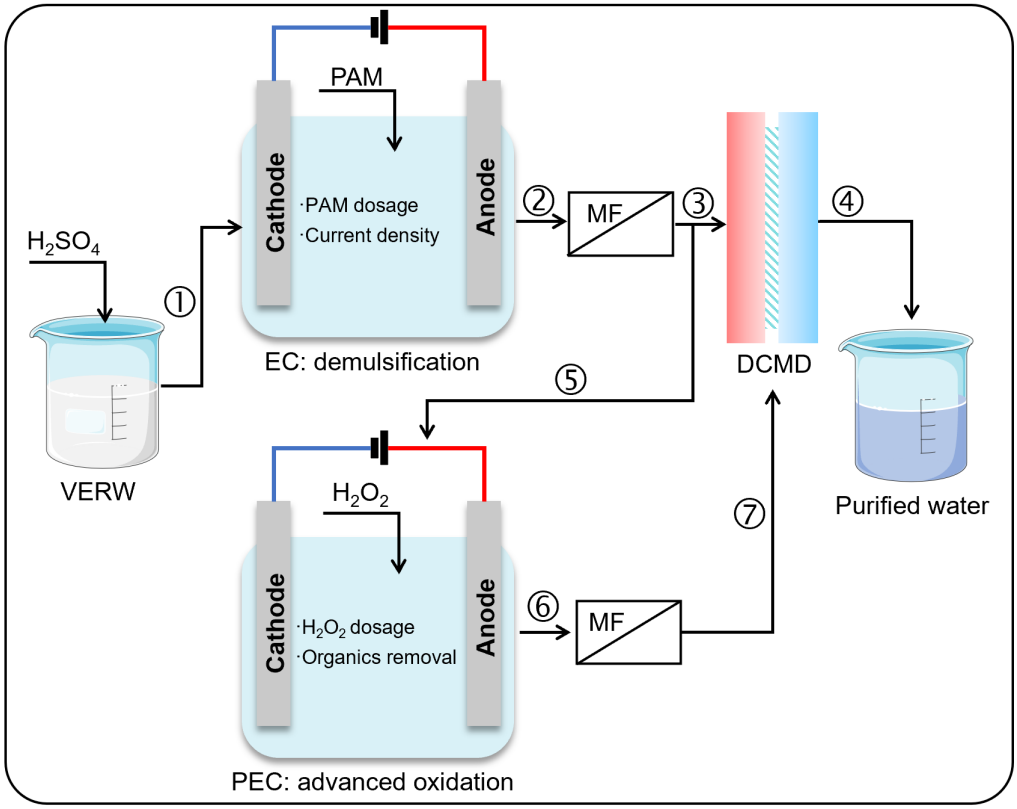

Fig.1 Diagram of the main experimental flowchart for advanced treatment of VERW pre-treated by the continuous EC and PEC processes.

2.2.1 DCMD

A laboratory-scale DCMD was employed in this work and a commercial flat sheet PVDF membrane (GVHP29325, nominal pore size of 0.22 μm) was used as the DCMD membrane (Table S3). More details of the DCMD setup can be found in our previous report[21, 46, 49]. 1200 mL of VERW and 1500 mL of deionized water were used as the feed and distillate solutions. The temperature of the feed and distillate was maintained at 60 ± 1℃ and 20 ± 1℃. Both circulatory flow rates were 300 mL/min. After operation, the PVDF membrane was observed by field emission scanning electron microscopy with energy dispersive X-ray fluorescence spectrometer (FESEM, Hitachi SU8010, Japan) for surface morphology and scale composition identification.

The distillate flux (J, kg/m^2·h) and concentration factor (CF) were calculated according to equations (9) and (10), respectively.

$$J=\frac{\Delta W}{\Delta t \times A} \quad (1)$$

where Δw (kg) is the change of distillate mass, Δt (h) is the distillate collection time and A (m^2) is the effective area of the membrane.

$$CF=\frac{V_i}{V} \quad (2)$$

where V_i (L) is the initial feed volume, V (L) is the concentrated volume of the feed.

2.3Analytical methods

2.3.1 EC process

During the EC process, the demulsification effect would be affected by a range of

operating parameters, including current density, electrode connection mode, flow rate, flocculant dosage[47, 50]. In this work, the fixed flow rate was 1000 mL/h and electrodes were monopolar connected. The influence of current density and PAM dosage on the EC demulsification process was investigated. The current density was calculated as follows:

$$CD = \frac{I}{S} \quad (3)$$

where CD (mA/cm^2) is the current density, I (mA) is the main current, and S (cm^2) is the effective area of the electrode.

Flocs produced by EC treatment of resin wastewater were small and dispersed (Fig.S1), which could be filtered and removed by adding flocculants (Table S2). PAM as a common flocculant was used in this work. However, DCMD performance and COD removal might be deteriorated due to the use of PAM[22, 51]. The COD contribution of PAM (COD_{PAM}) was calculated as follows:

$$(C_3H_5NO)_n + \frac{15n}{4}O_2 = 3nCO_2 + \frac{5n}{2}H_2O + \frac{n}{2}N_2 \quad (4)$$

$$COD_{PAM} = \frac{4M_{PAM}}{15nM_{O_2}}C_{PAM} \quad (5)$$

where n is the degree of polymerization of PAM, COD_{PAM} (mg/L) is the COD of wastewater increased by adding PAM, M_{PAM} (g/mol) is the molar mass of PAM, and M_{O2} (g/mol) is the molar mass of oxygen (32 g/mol). C_{PAM} (mg/L) is the concentration of PAM in wastewater. Although PAM presents different molar masses due to different degrees of polymerization, the theoretical consumption of 1 g of PAM is about 1.7 g O_2, i.e $\frac{4M_{PAM}}{15nM_{O_2}} \approx 1.7$.

The electrical energy consumed per kilogram of COD (q) was calculated as

follows[52]:

$$q=\left(\frac{I\times\int_{V_i}^{V_e}U\,dV}{1000\times(V_1-V_i)}\right)\times\left(u\times\frac{COD_i-COD_e}{10^6}\right)^{-1} \quad (6)$$

where q (kWh/kg COD) is the energy required for COD removal per unit mass, U (V) is trunk voltage, V_i (L) is the influent volume at the beginning of collecting EC effluent, V_e (L) is the influent volume at the end of collecting EC effluent, u (L/h) is the set flow rate. COD_i (mg/L) is the initial COD concentration, and COD_e (mg/L) is the effluent COD.

2.3.2 Determination of VERW composition

Concentrations of metal ions in VERW were determined by inductively coupled plasma atomic emission spectrometry (Prodigy-ICP, Leeman, USA). The anion concentrations in VERW were detected with ion chromatography (Dionex Aquion IC, Thermo Fisher, USA). The concentrations of COD and ammonia nitrogen were measured by dichromate method and Nessler's reagent spectrophotometry, respectively. Turbidity was determined by turbidity meter (WGZ-1A, QIWEIYIQI, China). The removal efficiency of pollutants was calculated using the following equation:

$$R=\frac{C_i-C_e}{C_i}\times100\% \quad (7)$$

where R (%) is removal efficiency, C_i (mg/L) and C_e (mg/L) are the concentration of pollutants in influent and effluent, respectively. The composition of organics in VERW was determined by a fluorescence spectrometer (F7000, Hitachi, Japan) and subtracted the spectrum of deionized water from the EEM of all samples to correct the inner filter effects[53, 54].

3. Results and discussion

3.1 Demulsification of VERW via EC

3.1.1 Effect of current density

Current density was the main factor affecting the removal of pollutants during EC process. In Fig.2(a), when the applied current density increased from 5 to 20 mA/cm^2, the pH of the effluent increased from 1.85 to 2.86, while the conductivity decreased from 17.9 to 11.9 mS/cm. These results were due to the constant consumption of hydrogen ions during the EC process. VERW in the reaction tank was disturbed by the hydrogen produced near the cathode (equation (8)). Moreover, the gas was beneficial to the coagulation process[50, 55]. Notably, the removal efficiency of COD decreased first and then increased with the increase of current density, reaching 94.9% at the current density of 20 mA/cm^2 (Fig.2(b)). As shown in equations (8-10), this phenomenon was related to the removal of organic matters by air flotation and flocculation precipitation in EC process[36, 56]. Previous study suggested that many small bubbles were conducive to the removal of pollutants during the flotation process and the bubble size increased with the increase of current density[57]. When the current density increased to 10 mA/cm^2, the COD removal efficiency decreased due to the increased of bubble volume. With the further increase of current density, the production of coagulants and bubbles increased, thereby increasing COD removal efficiency with the increase of current density. However, higher current density led to more energy being consumed, from an initial 0.039 to 0.53 kWh/kg COD at 20 mA/cm^2. Consequently, the current density of

15 mA/cm^2 was selected as the optimum value for the COD removal efficiency with low energy consumption.

At cathode:

$$2H_2O+2e^- \rightarrow 2OH^-+H_2 \uparrow \quad (8)$$

At anode:

$$Fe \rightarrow Fe^{2+}+2e^- \quad (9)$$

$$4Fe^{2+}+10H_2O+O_2 \rightarrow 4Fe(OH)_3 \downarrow +8H^+ \quad (10)$$

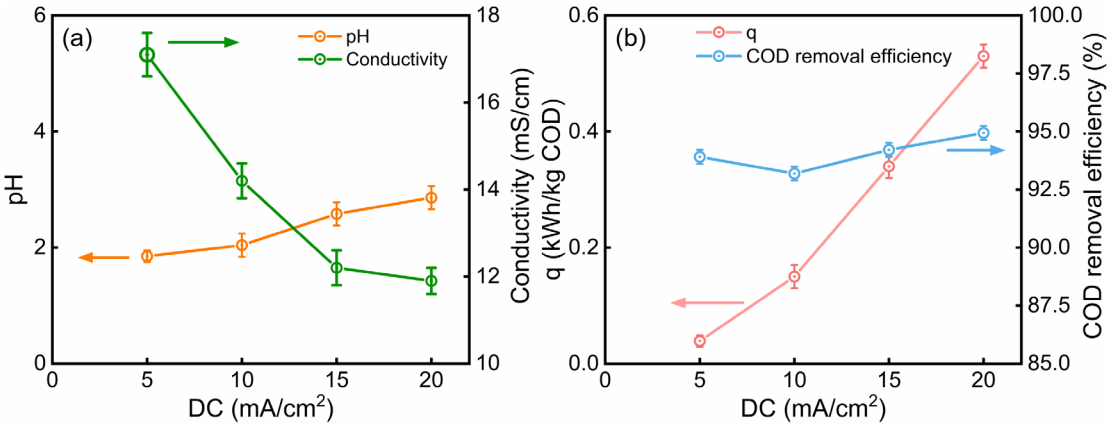

Fig.2 Performance of EC with different current densities. (a) pH and conductivity of the effluent, (b) COD removal efficiency and energy consumption. (PAM = 10 mg/L, effluent volume = 50 mL).

3.1.2 Effect of PAM dosage

A series of PAM experiments were carried out under the optimal current density to define the optimal dosage of PAM. As shown in Fig.3(a), when the dosage of PAM increased to 100 mg/L, the COD concentration in wastewater increased to 169.0 mg/L, while the COD removal efficiency decreased from 94.2 to 91.8%. Moreover, the energy consumption increased from 0.28 to 0.40 kWh/kg COD. These results indicated that the

overdosage of PAM led to a decrease in COD removal efficiency and an increase in energy consumption. On one hand, the addition of PAM increased the COD load, resulting in a decrease of COD removal efficiency. On the other hand, PAM overdosage destabilized and redissolved steady-state pollutants in VERW, increasing the concentration of COD[57]. When the dosage of PAM was 1 mg/L, the system energy consumption was low to 0.28 kWh/kg COD. When the dosage of PAM was 10 mg/L, the COD removal efficiency was the highest. Consequently, a lower dosage of PAM was beneficial to the EC process.

The impact of adding 10 mg/L PAM in the EC process on DCMD system performance was more significant than addition of 1 mg/L. In Fig.3(b-c), when the PAM dosage was 10 mg/L, the system flux decreased significantly after 8 hours, and the conductivity increased sharply after 12 hours, reaching 550 μS/cm until 21 hours. In contrast, when the PAM dosage was 1 mg/L, the flux remained stable for a longer time, gradually decreased at 20 h, and reached the same conductivity at 30.5 h. At the end of the experiment, the CF of DCMD system with the PAM dosage of 1 mg/L was 3.2, which was higher than that with the PAM dosage of 10 mg/L, indicating that the DCMD process after the EC pretreatment with addition of less flocculant could recover more distillate. This phenomenon can be attributed to the PVDF membrane fouling by PAM. The added PAM molecules might adhere to or deposit on the external surface of the membrane via establishing non-specific interactions with the functional groups of membrane materials, adding additional mass transfer resistance[58]. In addition, PAM molecules might trap inside the membrane pores, reducing the pore size and enhancing

the resistance to vapor transfer. Especially, PAM has high tendency to interact with each other at high concentrations[59]. The result that adverse effects of adding large amounts of PAM in feed on DCMD was also reported in previous studies[59]. In conclusion, 1 mg/L was selected as the optimal PAM dosage in the EC process.

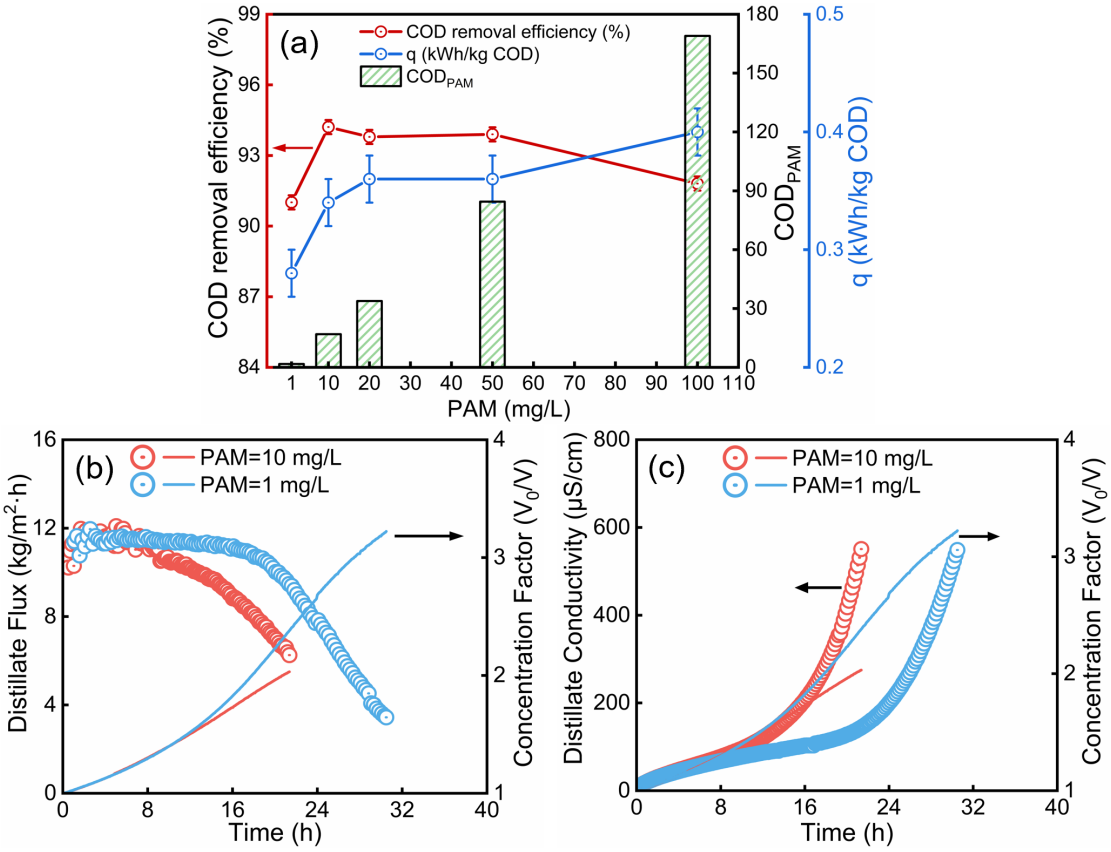

Fig. 3 Effect of PAM dosage on organic matter removal efficiency, COD contribution and energy consumption in the continuous EC process (a); DCMD system performance, CF with (b) distillate flux and (c) distillate conductivity as a function of time during treatment of VERW (I = 1.5 A, PAM dosage = 1 and 10 mg/L).

3.2 Organics removal via PEC

3.2.1 Effect of H_2O_2 dosage

Although the COD of the VERW was significantly reduced via the continuous

flow EC process, the effluent of VERW treated by EC still contains about 10% COD with a concentration of 2136.6 mg/L. Herein the PEC was used to further treat the effluent from EC process. As shown in Fig.4(a), the COD removal efficiency increased with the increase of H_2O_2 dosage. In addition, the energy consumption decreased with the increase of the H_2O_2 dosage, which was possibly due to the sharp increase of the COD removal efficiency. Specifically, when the addition dosage of H_2O_2 was equal to a quarter of the COD concentration, the COD concentration of the effluent from PEC was 1329.7 mg/L, and the energy consumption was 7.6 kWh/kg COD. When the dosage of H_2O_2 increased to four times of the COD concentration, the COD concentration decreased to 806.4 mg/L, the removal efficiency increased to 62.3%, and the energy consumption decreased to 3.72 kWh/kg COD. Interestingly, with the increase of H_2O_2 dosage, the color of precipitation gradually changed from light green to dark green, and finally to reddish brown (shown in Fig.4(a)). The reason for the phenomenon occurred was that more Fe^{2+} ions were oxidized to Fe^{3+} ions (equation 11). However, with the increase of H_2O_2 dosage, the Fenton reaction was inhibited (equation 12-14)[60, 61]. Therefore, it was unsuitable to further increase the addition dosage of H_2O_2, and the optimal H_2O_2 dosage was four times of the COD concentration of the EC effluent.

$$Fe^{2+}+H_2O_2 \rightarrow Fe^{3+}+ OH^-+ {}^{\cdot}OH \quad (11)$$

$$Fe^{3+}+H_2O_2 \leftrightarrow Fe\text{-}OOH^{2+}+H^+ \leftrightarrow Fe^{2+}+HO_2^{\cdot}+H^+ \quad (12)$$

$${}^{\cdot}OH+H_2O_2 \rightarrow HO_2^{\cdot}+H_2O \quad (13)$$

$$Fe^{2+}+{}^{\cdot}OH \rightarrow Fe^{3+}+ OH^- \quad (14)$$

To further characterize the organic pollutants removed via EC-PEC process, 3D-

EEM fluorescence spectra of the influent and effluent were illustrated in Fig.4(b-g). The fluorescence peaks of organics in the influent were mainly distributed in region V, IV, III, and I (Fig.4(b)). The peaks in region V and III were related to humic acid-like and fulvic acid-like substances, respectively. Peaks at 230.0/310.0 (Ex/Em(nm)) and 280.0/310.0 (Ex/Em(nm)) in the region I and IV might represent BPA[62, 63]. The fluorescence peak intensity of the organic in effluent weakened gradually with the increase of the H_2O_2 dosage. Result confirmed that the PEC was an effective process for organics removal. Notably, with the increase of H_2O_2 addition dosage, the characteristic peaks of BPA (Fig.4(e)) disappeared before the peaks in region III and V (Fig.4(g)) disappeared. The result indicated that PEC process was particularly effective in removing BPA. Similar results were found in previous study by Escalona et al. where a small amount of H_2O_2 could quickly remove most of BPA in wastewater[64]. There was no obvious fluorescence peak observed in Fig.4(g), which indicated that most of the fluorescent organics was removed.

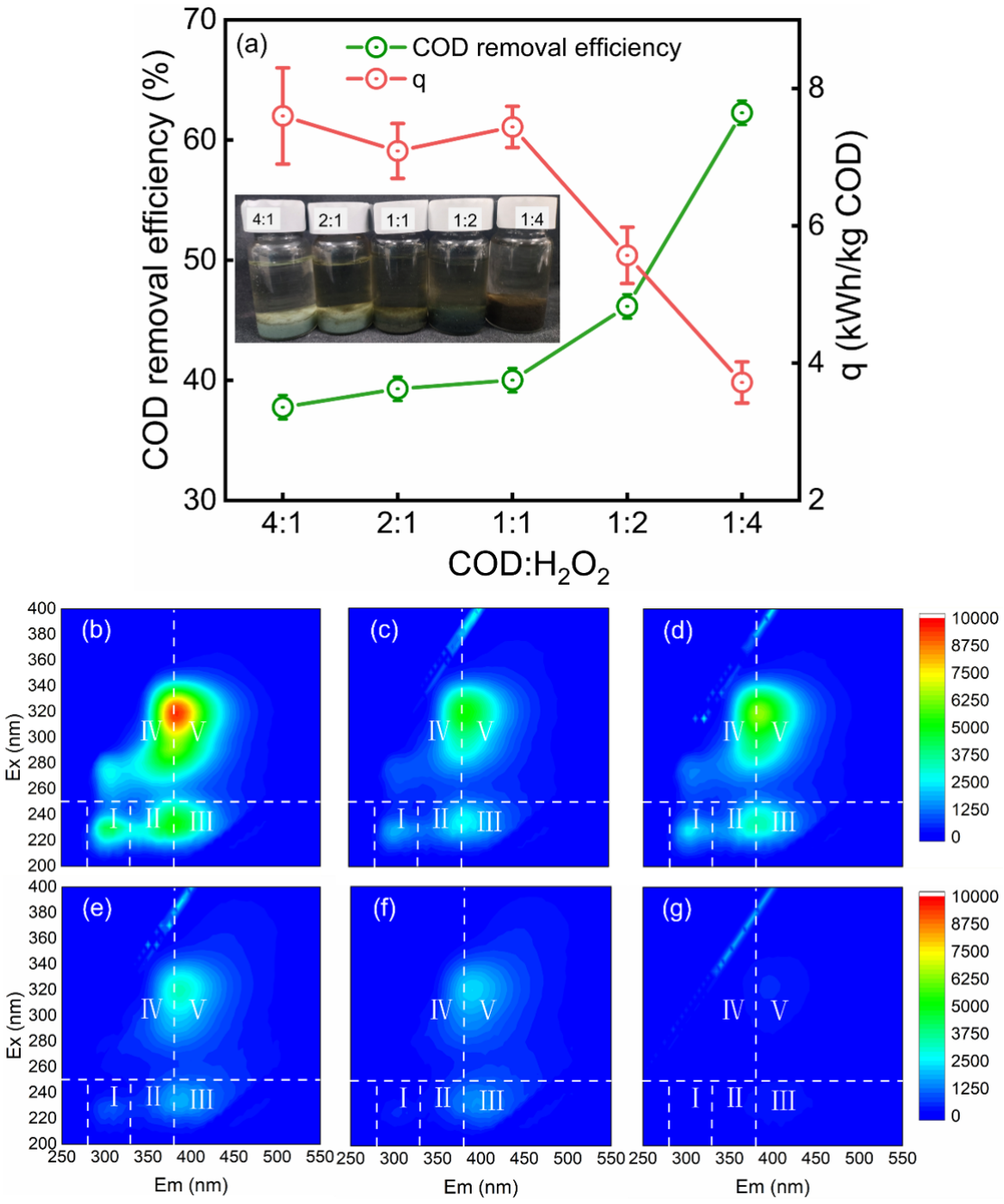

Fig.4 Effect of COD/H_2O_2 mass ratios on PEC process, (a) energy consumption and COD removal efficiency; 3D-EEM spectra of influent (b) and effluent, COD/H_2O_2 mass ratios were (c) 4:1, (d) 2:1, (e) 1:1, (f) 1:2 and (g) 1:4 respectively (25 times dilution).

3.2.2 Mechanism of organics removal via PEC

The generation and occurrence of hydroxyl radicals during the PEC process was determined by electron paramagnetic resonance (EPR) spectroscopy (Fig.5(d)).

Subsequently, the removal mechanism of organics via the PEC process was verified by the quenching experiments on the hydroxyl radicals. In this work, methanol was used as the quenching agent and the dosage was determined according to relevant reports[65, 66]. Meanwhile, the effect of coagulation on organics removal via the PEC process was determined by comparing the EEM spectra results of quenching experiments (Fig.5(b)), double-EC experiment (Fig.5(a)) and EC experiment (Fig.4(b)). The weak intensity of the fluorescent peak reported in Fig.5(a) meant that some contaminants were removed by coagulation during the PEC process. Moreover, similar EEM spectra of the quenching experiment (Fig.5(b)) and double-EC experiment (Fig.5(a)) indicated that coagulation did not contribute significantly to the removal of organics. On the contrary, the significant inhibitory effect of the methanol on the removal of organic (Fig.5(b) and (c)) indicated that the contribution of ·OH oxidation was more significant than coagulation. Removal mechanism of PEC process was shown in Fig.5(e), the combination of advanced oxidation and coagulation led to the removal of organic. Particularly, the ·OH was produced by the reaction of the Fe^{2+} generated on the anode with H_2O_2. PEC process realized the reutilization of residual Fe^{2+} during the EC demulsification process and enhance the organic removal by advanced oxidation.

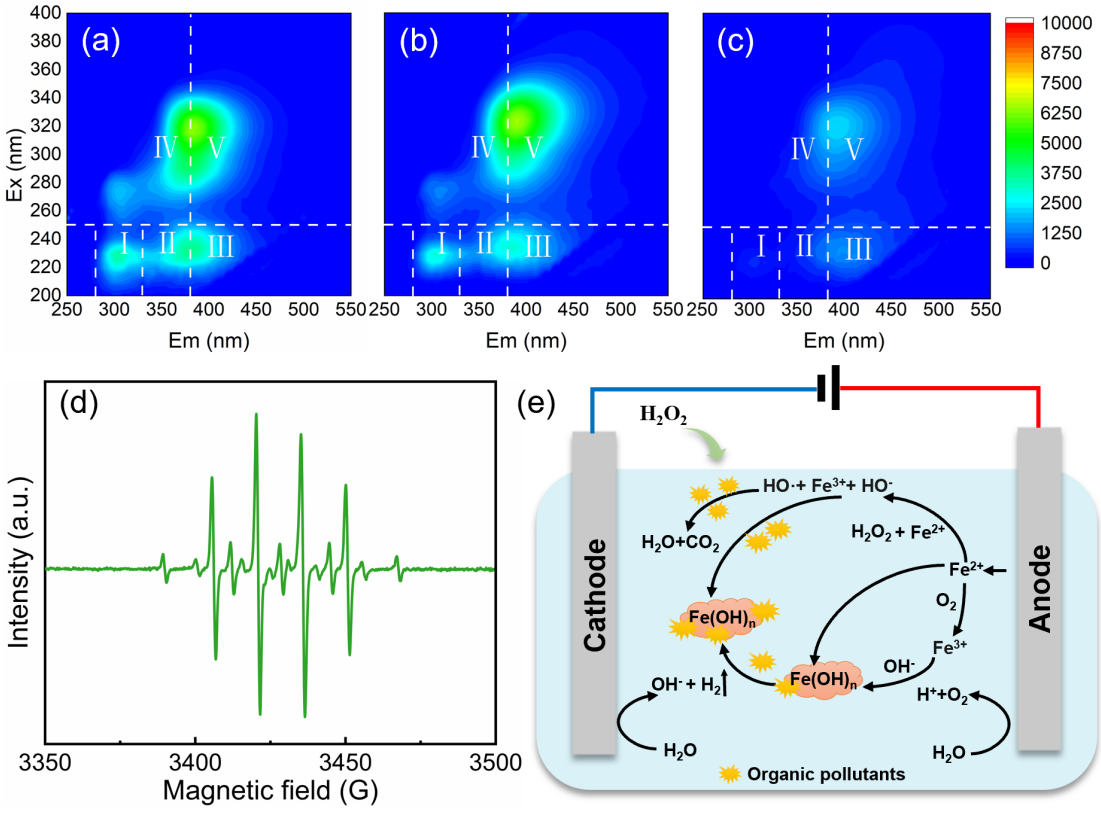

Fig.5 Organics removal in PEC process, 3D-EEM spectra of effluent treated with (a) double EC experiment, (b) with quenching agent, (c) without quenching agent (25 times dilution); (d) EPR spectra of hydroxyl radicals; (e) mechanism of organics removal.

3.3 DCMD performance

3.3.1 Effect of pretreatment on DCMD performance

The initial distillate flux of DCMD filtration of VERW pre-treated by EC was 11.4 kg/m^2·h, while the flux of DCMD filtration of VERW pre-treated via EC-PEC increased to 15.3 kg/m^2·h (Fig.6(a)). The DCMD process maintained the stability of high distillation flux for a long operating time. The conductivity of the distillate increased sharply to779 μS/cm at the end of DCMD filtration of VERW pre-treated by EC. The corresponding CF was 3.4. In contrast, the DCMD filtration of VERW pre-treated via

EC-PEC kept running until the hot side water was insufficient for operation and the conductivity remained below 33.3 μS/cm (Fig.6(b)) with the CF finally reached up to 8.1. Results showed that after EC demulsification treatment, there were still pollutants in the VERW that could result in membrane fouling. After further PEC treatment, the degree of membrane fouling was mitigated due to the efficient removal of organics. Therefore, the removal of organics via EC-PEC was considered as an effective pretreatment process prior to DCMD filtration of VERW to reduce the volume of the waste steam and recover a large amount of high-quality clean water.

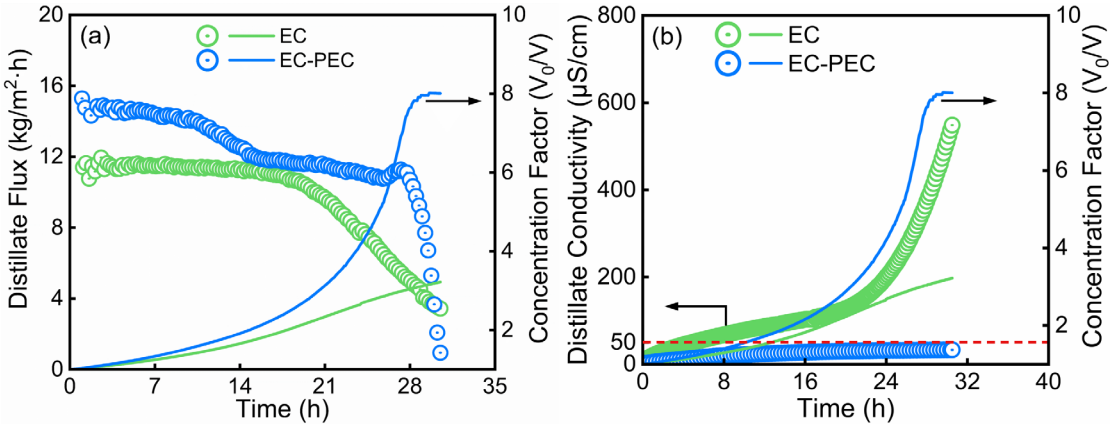

Fig.6 Performance of DCMD filtration of VERW pre-treated by the continuous flow EC and PEC process: CF with (a) distillate flux and (b) distillate conductivity as a function of time during treatment of VERW (I=1.5 A, PAM dosage =1 mg/L, COD: H_2O_2 = 1:4).

3.3.2 Membrane autopsy

SEM-EDS was used to characterize the PVDF membrane to analyze the influence of different pretreatment on membrane fouling. After DCMD filtration of VERW pre-treated via EC, it was found that the brown deposits distributed on the membrane

surface (Fig.7(a)). Compared to the clean MD membranes (Fig.S5), there was a thick fouling layer on the membrane surface composed of C, O, N, Fe, Na, and S (Fig.7(b-c)). Based on the results, it can be reasonably speculated that the foulants were mixture of organics and salt containing sodium and iron. On the contrary, the membrane obtained from DCMD filtration of VERW pre-treated via EC-PEC was almost clean without obvious fouling layer (Fig.7(d)). According to SEM-EDS analysis, only a small amount of calcium carbonate crystals was randomly deposited on the membrane surface (Fig.7(e-f)). As the removal of dissolved organics in VERW via EC-PEC, membrane fouling was significantly mitigated. Thus, organics in VERW were the main reason for the deterioration of DCMD performance and acceleration of membrane fouling.

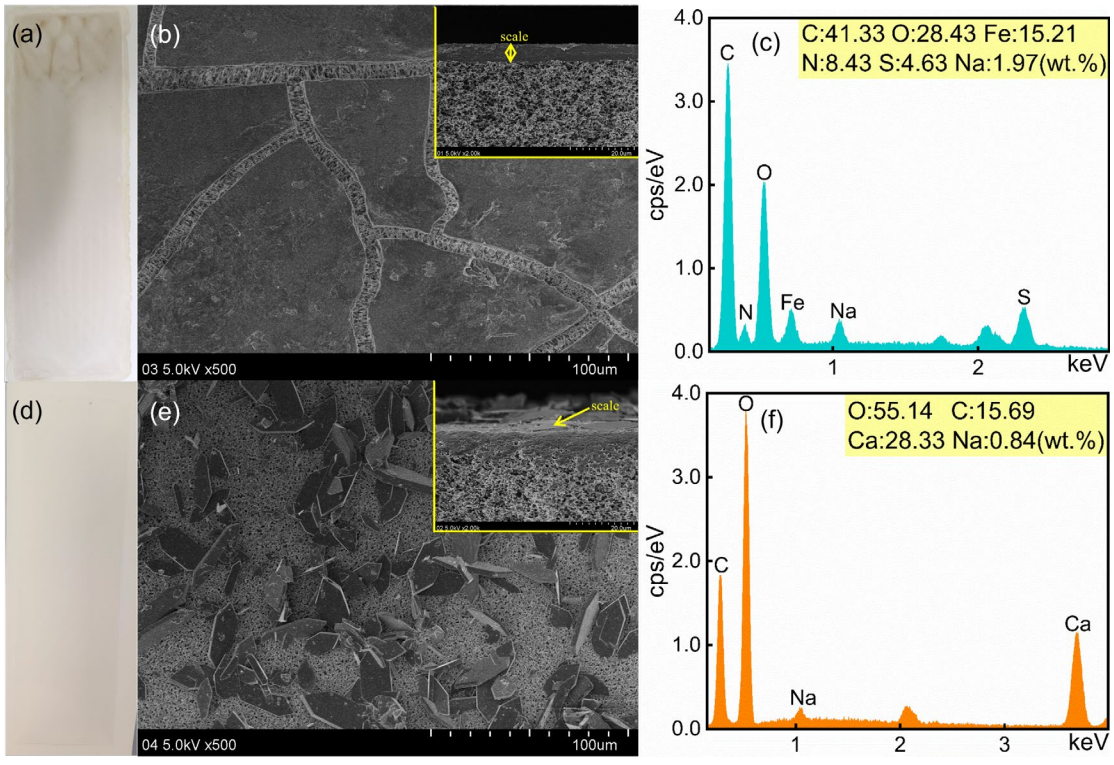

Fig.7 The photos and SEM images of the surface and cross-section of the membranes after DCMD filtration of VERW pre-treated via EC (a & b) and PEC (d & e), (c), and the corresponding results of EDS analysis (f).

410

411 **3.3.2 Comparison of distillate quality**

412 The pH of the distillate after DCMD filtration of the VERW pre-treated by EC was
413 3, with many metal ions and high COD, which indicated that the pollutants in the
414 VERW were not rejected effectively via the DCMD process (Table S4). Moreover, the
415 dissolved ions from the EC process partially permeates into the distillate during the
416 DCMD filtration of VERW pre-treated via EC process. However, at the conclusion of
417 DCMD filtration of VERW pre-treated via EC-PEC, the distillate was alkaline (pH =
418 9), and only ammonia nitrogen was detected. Volatilization and ionization of ammonia
419 were the main reasons for the changes of the pH and conductivity of the distillate. In
420 addition, the lower COD concentration of the distillate may be due to the
421 transmembrane transport of volatile organic compounds. EEM spectra showed that the
422 organics in the distillate after DCMD filtration of VERW pre-treated via EC-PEC were
423 distributed in region IV, and the two strong fluorescence peaks at 270.0/300.0 (Ex/Em
424 (nm)) and 290.0/355.0 (Ex/Em (nm)) represented BPA[62]. The distillate of the DCMD
425 filtration of the VERW pre-treated by EC-PEC detected no obvious fluorescence peak
426 (Fig.8). In summary, the DCMD process as an advanced treatment approach provided
427 a potential alternative for treatment of the waste steam pre-treated by EC-PEC from
428 resin production.

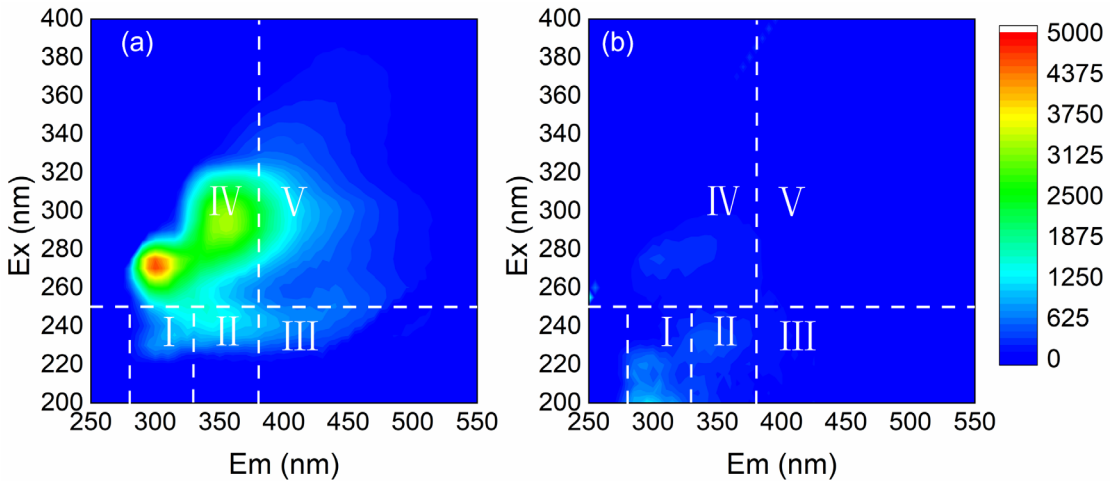

Fig.8 3D-EEM spectra of organic pollutants in distillate after (a) EC pretreatment (b) PEC pretreatment.

4. Conclusion

In this work, the continuous flow EC and PEC processes were used as the pretreatment approaches prior to advanced treatment of VERW via DCMD. The optimization of the two pretreatment processes was conducted. Besides, system performance and membrane fouling of DCMD filtration of the VERW pre-treated by the two different pretreatment processes were evaluated. The main conclusions were summarized as follows:

- Continuous flow EC and PEC processes were effective to remove the organics in VERW. The optimal current density and PAM dosage was 15 mA/cm^2 and 1 mg/L, respectively in EC process. For the PEC process, the optimal H_2O_2 dosage was four times of EC effluent COD concentration.
- The energy consumption of the continuous flow EC and PEC process under the optimal conditions was 0.34 kWh/kg COD and 3.72 kWh/kg COD, respectively.

- The initial flux of DCMD filtration of VERW pre-treated via EC-PEC increased by 35%, and membrane fouling was significantly mitigated, compared with that pre-treated by EC.
- The volume of VERW was reduced via DCMD with evidence that the CF reached up to 8.1. Additionally, the high-quality distillate with conductivity less than 33.2 μS/cm was obtained.

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