Treatment of zinc plating wastewater by combination of electrocoagulation and ultrafiltration process

Merve Dönmez Öztel, Ayşe Kuleyin and Feryal Akbal

ABSTRACT

In this study treatment of zinc plating wastewater by combination of electrocoagulation (EC) and ultrafiltration (UF) processes was investigated. The effect of operating parameters such as wastewater pH, flow rate and membrane pore size on zinc removal was investigated to optimize the EC-UF process. The results showed that zinc removal was pH dependent and optimum pH was 9.0. The zinc removal over 99% was achieved by the combined EC-UF process with both 50 and 100 kDa membranes at pH 9.0 and current density of 5 mA cm^{-2}. Suspended solids and oil–grease were also removed completely by the combined EC-UF process. The EC-UF process proved to be a promising technology for the treatment and recycling of zinc plating wastewater.

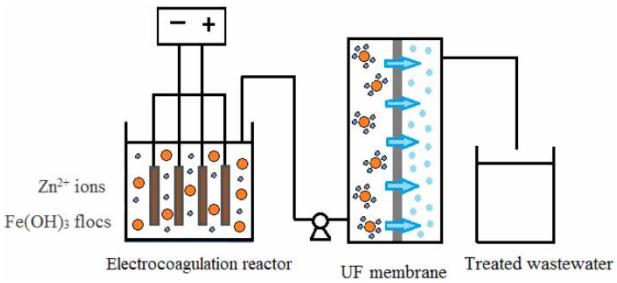
Key words | electrocoagulation, metal industry, ultrafiltration, wastewater, zinc

Merve Dönmez Öztel
Ayşe Kuleyin
Feryal Akbal (corresponding author)
Department of Environmental Engineering,
Ondokuz Mayıs University,
55139 Samsun,
Turkey
E-mail: *feryal.akbal@gmail.com*

HIGHLIGHTS

- The electrocoagulation–ultrafiltration (EC-UF) process was used for the treatment and recovery of metal plating wastewater.
- The effects of pH, flow rate, operating time and membrane pore size on the performance of EC-UF process were investigated.
- EC-UF process showed higher removal efficiencies than EC process.
- EC-UF process removed the zinc, suspended solids and oil–grease efficiently and produced high quality water.

GRAPHICAL ABSTRACT



INTRODUCTION

Toxic heavy metals discharged to the environment by industrial activities are one of the most significant contaminants in water. Heavy metals are non-degradable, persistent and accumulative in nature and most heavy metals cause important environmental problems (Gupta & Nayak 2012; Mahmoud & Hoadley 2012). Heavy metals in industrial wastewaters usually include zinc, cadmium, chromium, copper, nickel and lead. Zinc is widely used in galvanization of steel and battery production. Zinc is also used in plastics, printing inks, photocopier paper and wallpaper as a pigment

doi: 10.2166/wst.2020.357

and in rubber production as a catalyst (Gakwisiri *et al.* 2012). Zinc is an essential metal for biological reactions but it can cause common health problems, such as skin irritations, anemia, nausea and stomach cramps, at high concentrations (Fu & Wang 2011). Therefore, removal of zinc from wastewater prior to discharge, is of great importance. The US Environmental Protection Agency (USEPA) has set the maximum and average limit of zinc for metal finishing wastewater as 2.16 and 1.48 mg/L, respectively (USEPA 2020). According to Turkish Water Pollution Control Regulation (WPCR) the maximum allowable limit of zinc is 5 mg/L for metal plating wastewater (WCPR 2008).

The methods commonly used to remove heavy metals include adsorption (Sen & Gomez 2011; Bilal *et al.* 2013), ion-exchange (Mier *et al.* 2001; Dąbrowski *et al.* 2004), membrane filtration (Sheikholeslami & Bright 2002; Qdais & Moussa 2004) and electrochemical treatment (Mouedhen *et al.* 2009; Lakshmanan *et al.* 2010). Among many separation techniques, the membrane process is an efficient and widely applied separation process (Ennigrou *et al.* 2009). Membrane technology offers an important solution for the treatment of industrial wastewaters (Chen & Deng 2012). The membrane technologies used for the removal of metals from metal-containing wastewaters are nanofiltration, reverse osmosis and electrodialysis. However, high energy requirement of these processes increases their operational costs. Ultrafiltration is a low-pressure and cost-effective process for the removal of dissolved and colloidal pollutants (Sardari *et al.* 2018). However, since the pore size of ultrafiltration membranes is larger than dissolved metal ions in the hydrated form, ultrafiltration membranes are not capable for the separation of heavy metal ions. Ultrafiltration can be combined with a chemical or electrochemical process to meet discharge standards for several pollutants as hybrid membrane processes (Yahiaoui *et al.* 2011). Several techniques such as micellar enhanced ultrafiltration (Channarong *et al.* 2010; Zeftawy & Mulligan 2011), polymer enhanced ultrafiltration (Korus & Loska 2009; Jiao *et al.* 2013; Kalaiselvi *et al.* 2013) and hybrid systems (Zeng *et al.* 2009; Du *et al.* 2014) have been developed in order to improve removal efficiency of heavy metal ions by ultrafiltration membranes (Magnenet *et al.* 2013).

In membrane filtration applications, fouling is the major problem. Electrocoagulation can be used as a pre-treatment for membrane processes, where the particles can be coagulated resulting in the reduction of fouling (Kherti *et al.* 2013). Integration of electrocoagulation with membrane processes has been reported previously. Changmai *et al.* (2019) employed an electrocoagulation–microfiltration process for the treatment of oily wastewater containing oil, grease and metals. Tavangar *et al.* (2019) applied a hybrid electrocoagulation–nanofiltration (EC-NF) process for treatment of real textile industry wastewater in order to recycle the treated wastewater. They reported that EC-NF possessed excellent potential for real textile wastewater treatment and efficient fractionation of dyes and inorganic salts. Gonder *et al.* (2020) applied an integrated EC–NF process for carwash wastewater treatment and they showed that the water quality for reuse in carwashing process could be achieved with an integrated EC-NF process.

Electrocoagulation is based on the production of a coagulant in wastewater through electrolysis by applying a current to a pair of cathode and anode (Kumarasinghe *et al.* 2009). Electrocoagulation removes colloidal particles efficiently compared to conventional coagulation, because the electric field enhances the stabilization and coagulation of charged particles (Gandhimathi *et al.* 2014). Simultaneously, gas bubbles which are produced by electrolysis enhance flotation. Flocs formed electrochemically have much higher sorption capability than those obtained chemically, thanks to the finely dispersed structure and well developed specific surface (Mavrov *et al.* 2006). Also, electrocoagulation flocs contain less bound water and are much larger and more stable and they can be separated faster by filtration (Chen & Deng 2012). Electrocoagulation process was successfully used for the treatment of metal-containing wastewaters.

The objective of this study is to investigate the feasibility and performance of the combined electrocoagulation–ultrafiltration (EC-UF) process for the treatment and recycling of zinc plating wastewater. The effect of wastewater pH, flow rate and membrane pore size on EC-UF process performance was investigated. The treatment efficiencies and the effluent quality of EC and EC-UF processes were also compared.

EXPERIMENTAL

Wastewater characterization and analytical methods

The wastewater used in experiments was obtained from a spent zinc-plating bath of a metal finishing factory in Samsun, Turkey. The wastewater sample was transferred to the laboratory in plastic containers and stored at 4 °C before experiments. The zinc, chemical oxygen demand (COD), oil–grease (OG) and suspended solids (SS) were

analyzed in raw wastewater and effluents of EC and EC-UF processes by standard methods (APHA/AWWA/WEF 2017).

The wastewater pH was measured by a Sartorius PB pH meter, Sartorius Instruments, Germany. The zinc concentrations were measured using an atomic absorption spectrophotometer (UNICAM 929). COD analyses were performed by a NOVA 60 thermoreactor according to SM 5220D. OG analyses were performed by gravimetric method according to SM 5520B and SS analyses were performed according to SM 2540D. The chemical composition of the wastewater is listed in Table 1.

Electrocoagulation experiments

An EC reactor combined with UF system was used in the experimental studies. The experimental set-up is presented in Figure 1. Electrocoagulation experiments were conducted in an electrocoagulation reactor with steel electrodes in monopolar mode. The total submerged surface area of electrodes was 130 cm^2. The electrodes were placed 1.0 cm apart in the electrocoagulation reactor. The wastewater pH was adjusted by adding NaOH. The EC reactor was filled with 1,500 mL of the wastewater before starting the electrocoagulation experiments. The wastewater was constantly stirred using a magnetic stirrer (2MLH). The direct current was supplied by a power source for electrocoagulation experiments (GW GPC-3060D model). Before each experiment, the electrodes were cleaned with successive rinses of 0.1 N H_2SO_4 and deionized water. After the experiments, the electrodes were cleaned by scrubbing with sand paper to remove the rust and the oxy/hydroxides.

Table 1 | Characterization of industrial wastewater used for experiments

Parameter	Value
Chemical oxygen demand (mg/L)	600
Suspended solids (mg/L)	142
Oil–grease (mg/L)	44
Zinc (mg/L)	200
pH	2.57

Ultrafiltration experiments

After the electrocoagulation reactor, the wastewater was transferred to a quartz sand filter and then the ultrafiltration system with a peristaltic pump (Watson Marlow). The quartz sand filter material in the column was 12 cm in height and the filter material was supported with glass wool. A Sartocon Slice 200 filter holder was used for ultrafiltration experiments. Polyethersulfone membranes with average molecular weight 50 and 100 kDa were used in the ultrafiltration system. The surface area of membranes was 200 cm^2. Ultrafiltration membrane properties are listed in Table 2. Ultrafiltration experiments were conducted in cross-flow mode. The filtrate (permeate) was collected in a separate tank and the concentrate (retentate) was transferred back to the raw wastewater tank.

The separation capability of the membrane is defined as rejection, and given by Equation (1):

$$\%R = \left(1 - \frac{C_p}{C_0}\right) * 100 \quad (1)$$

where C_0 is the initial contaminant concentration in mg/L and C_p is contaminant concentration in the permeate in mg/L.

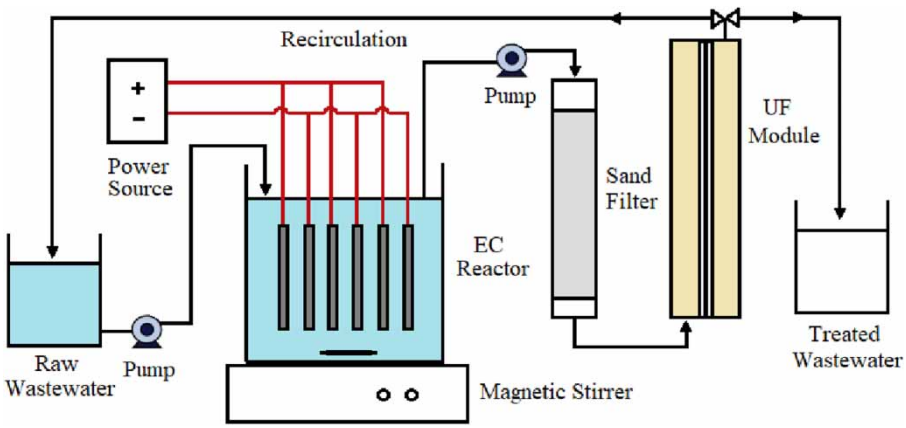


Figure 1 | The electrocoagulation–ultrafiltration experimental set-up.

Table 2 | Ultrafiltration membrane properties

Properties	Value
Filter surface area (m^2)	0.02
pH stability	1–14
Maximum pressure at 20 °C (bar)	4
Maximum temperature for the process conditions [°C]	50
Air diffusion rate at feed pressure of 14.5 psi (mL/min)	<1.5

The permeate flux (J) is defined as the permeate volume flowing through the membrane per unit membrane area per unit time in L/m^2.h (Desai & Murthy 2012). The J values were calculated by Equation (2):

$$J = \frac{Q}{A} \tag{2}$$

where Q is the permeate flow in L/h and A is total membrane surface area in m^2.

RESULTS AND DISCUSSION

Effect of wastewater pH on EC–UF process

The electrocoagulation process involves the application of direct electric current across sacrificial electrodes for the generation of coagulants (Gatsios *et al.* 2015). The pH is a significant operating factor as it influences the performance of electrochemical processes. The effect of wastewater pH on zinc removal was investigated by using a 100 kDa ultrafiltration membrane at pH 3, 5, 8, 9 and 11 and the results are presented in Figure 2. It can be seen from Figure 2 that the zinc removal increased from 92.4 to 99.9% when the pH increased from 3 to 9 and slightly decreased with the further increase in pH. It can be concluded that the wastewater pH influenced the performance of EC-UF process.

The results showed that removal of Zn decreased in acidic and basic conditions. In alkaline medium, decrease in removal efficiency can be attributed to the oxidation of hydroxyl ions at the anode and the formation of $Fe(OH)^{4-}$ and $Fe(OH)_6^{3-}$ anions. In acidic medium, the protons in the solution are reduced to hydrogen gas at the cathode and hydroxyl ions cannot be produced at the same proportion (Al-Shannag *et al.* 2015).

The pH of wastewater also affects the formation and solubility of zinc complexes. Zinc hydroxide is slightly soluble in water and solubility increases at acidic and basic pH values. Zinc can be removed from wastewater by the addition of base to form the insoluble $Zn(OH)_2$. However, in the case of excess base addition, zinc will form soluble complexes with OH^- and will transfer to solution according to the following equations:

Precipitation reaction

$$Zn(OH)_2(s) \leftrightarrow Zn^{2+} + 2OH^- \qquad K_{sp} = 8 \times 10^{-18} \tag{3}$$

Complex formation reactions

$$Zn^{2+} + OH^- \leftrightarrow ZnOH^+ \qquad K_1 = 1.4 \times 10^4 \tag{4}$$

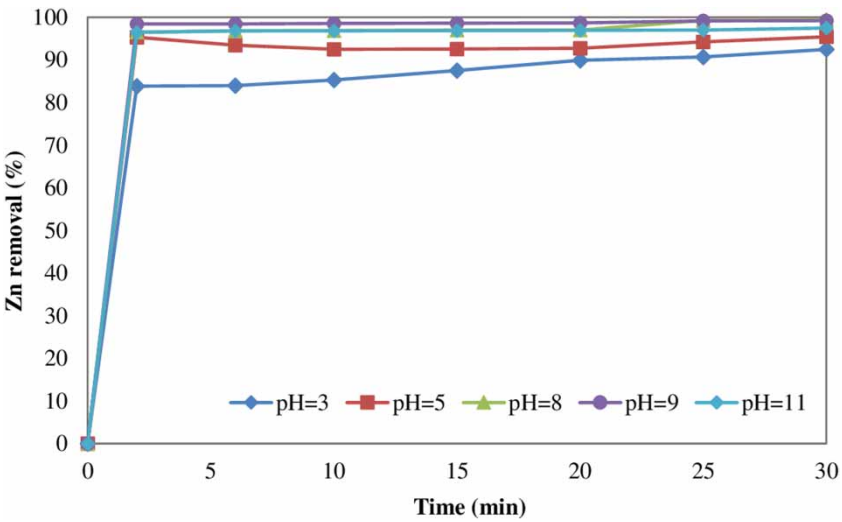


Figure 2 | Effect of pH on the Zn removal by EC-UF process; current density: 5 mA/cm^2, flow rate: 60 mL/min, molecular weight cut-off: 100 kDa.

$$ZnOH^{+} + OH^{-} \leftrightarrow Zn(OH)_2(aq) \quad K_2 = 1 \times 10^6 \tag{5}$$

$$Zn(OH)_2(aq) + OH^{-} \leftrightarrow Zn(OH)^{3-} \quad K_3 = 1.3 \times 10^4 \tag{6}$$

$$Zn(OH)^{3-} + OH^{-} \leftrightarrow Zn(OH)_4^{2-} \quad K_4 = 1.8 \times 10^1 \tag{7}$$

The increase in the pH from 3 to 9 led to increase in percentage of Zn removal efficiency. Zn removal was higher in the initial period when pH was between 5 and 9. Zn removal efficiencies were found as 83.8, 95.3, 96.5, 98.4 and 96.5% at operating time of 2 min and 92.4, 95.4, 99.4, 99.9 and 97.5% at operating time of 30 min for pH 3, 5, 8, 9 and 11, respectively. The maximum efficiency of Zn removal was observed at pH 9.0. Therefore, the subsequent experimental studies were performed at pH 9.

Effect of flow rate on EC–UF process

The effect of flow rate on the EC–UF process was examined by using 50 and 100 kDa UF membranes. The effect of flow rate on Zn and COD removal for 50 and 100 kDa UF membranes is shown in Figure 3(a) and 3(b) respectively. It can be seen that Zn removal decreased when the flow rate increased. Zn removal was found as 99.2 and 92.9% for 100 kDa UF membrane and 99.6 and 94.4% for 50 kDa UF membrane at flow rate of 60 and 100 mL/min and operating time of 30 min. COD removal was also decreased from 84% to 66.5% for 100 kDa UF membrane and from 85.9 to 74.9% for 50 kDa UF membrane at operating time of 30 min, when the flow rate increased from 60 to 100 mL/min.

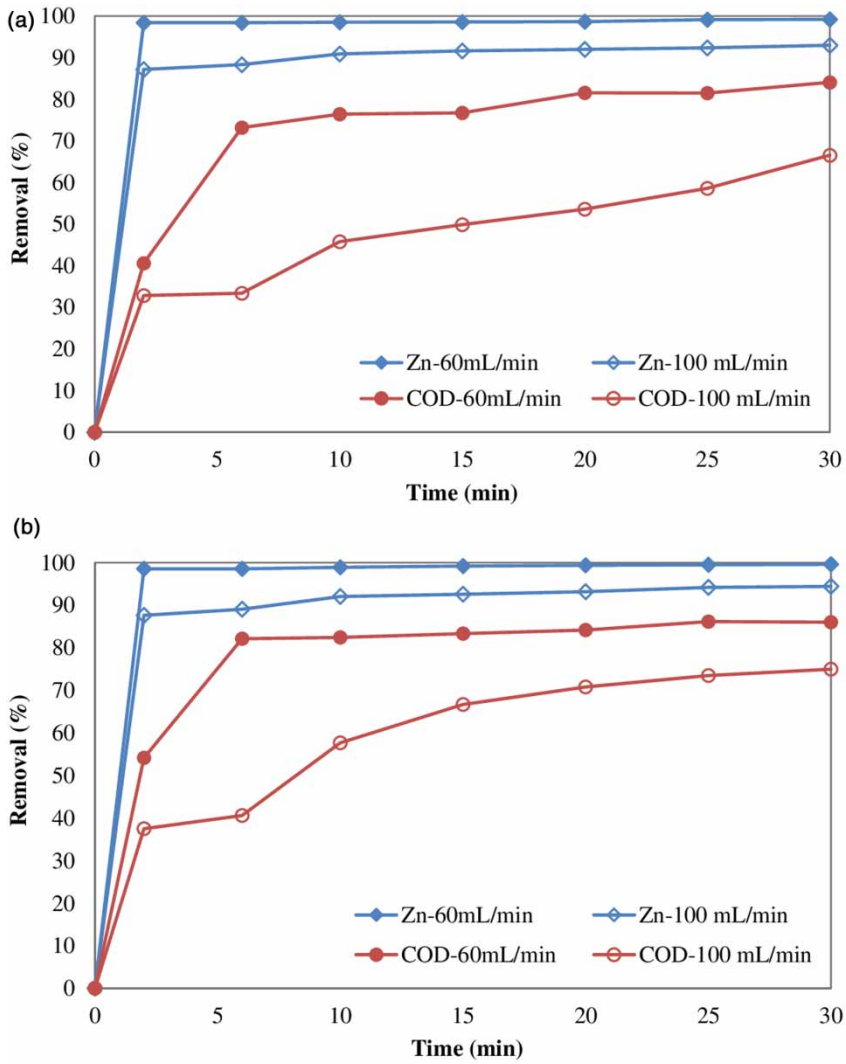


Figure 3 | Effect of flow rate on the Zn and COD removal by EC-UF process for (a) 50 kDa and (b) 100 kDa UF membrane; current density: 5 mA/cm^2, pH: 9.

The removal efficiency decreased with increasing flow rate, since the retention time decreased by the increasing flow rate. Higher retention time provided more time for the reactions in the EC reactor. The reduction in the retention time with increasing flow rate has led to a decrease in the Zn and COD removals from the wastewater. The optimal influent flow rate could therefore be considered to be 60 mL/min. Also, when the flow rates increased, the Zn and COD removal efficiency decreased since the quantity of dissolved iron per unit of time and volume was lower.

Zn removal of EC-UF process increased up to 10 min and then remained almost constant at flow rate of 100 mL/min, while it was very high in the initial period at flow rate of 60 mL/min for both 50 and 100 kDa membranes. The residual Zn concentration in the EC-UF reactor at 30 min was obtained as 1.58 and 14.07 mg/L for 100 kDa UF membrane and 0.81 and 11.2 mg/L for 50 kDa UF membrane at flow rate of 60 and 100 mL/min, respectively. Zn removal efficiency of EC-UF reactor at 30 min was 99.2 and 99.6% for 100 and 50 kDa membranes at flow rate of 60 mL/min.

It can be seen that COD removal efficiency increased with time. On the other hand, the increase of the flow rate reduced COD removal efficiency. For flow rates of 60 and 100 mL/min, the COD removal at operating time of 30 min was 84% and 66.5% for 100 kDa UF membrane and 85.9 and 74.9% for 50 kDa UF membrane, respectively. The residual COD concentration in the EC-UF reactor at 30 min was 95 and 200 mg/L for 100 kDa UF membrane and 84 and 150 mg/L for 50 kDa UF membrane at flow rate of 60 and 100 mL/min, respectively.

Effect of membrane pore size on EC–UF process

The effect of membrane pore size on EC-UF process was examined by using 50 and 100 kDa UF membranes at current density of 5 mA/cm^2, flow rate of 60 mL/min and pH 9.0. It was found that zinc and COD removal was slightly higher with the 50 kDa ultrafiltration membrane and removal efficiency increased when the operating time increased.

It can be seen from Figure 4(a) that Zn removal was high in the initial period and increased slightly with time. Zn removal efficiency achieved for the EC-UF system at 30 min was 99.2% for 100 kDa UF membrane and 99.6% for 50 kDa UF membrane. As can be seen from Figure 4(b)

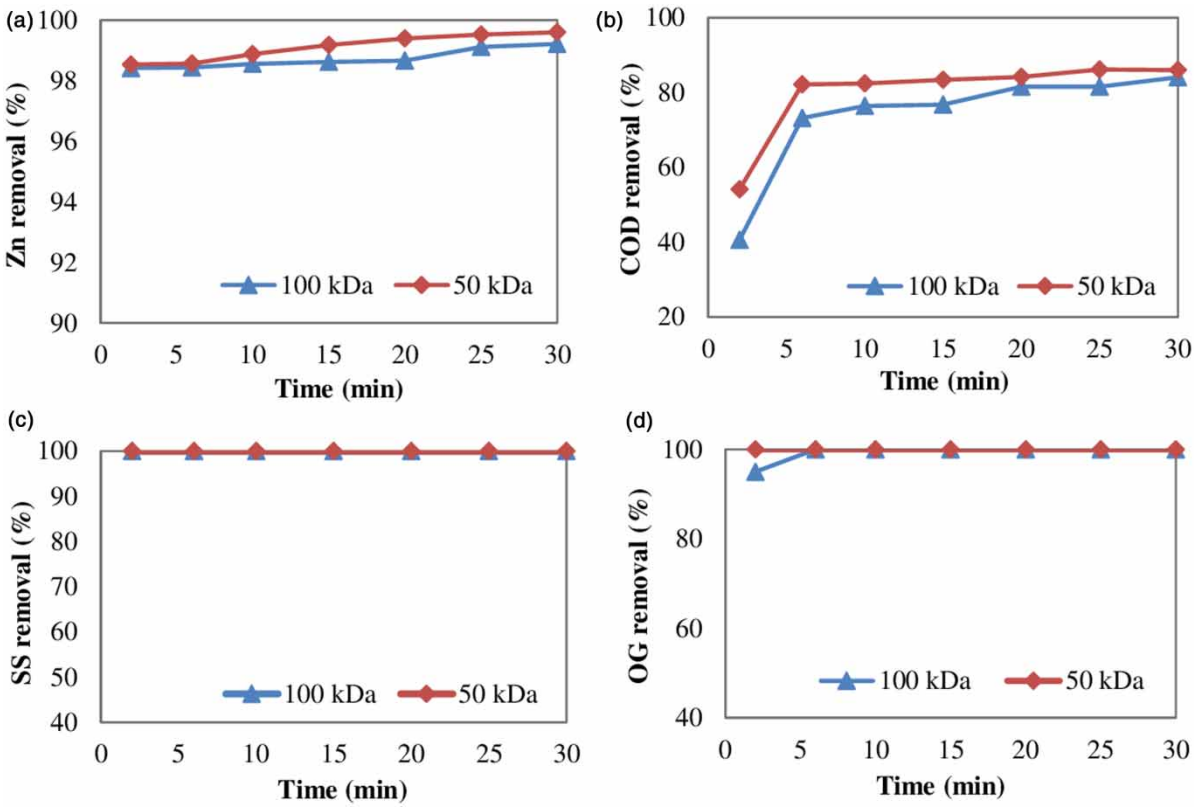


Figure 4 | Effect of membrane pore size on (a) zinc, (b) COD, (c) SS and (d) OG removal by EC-UF process; current density: 5 mA/cm^2, flow rate: 60 mL/min, pH: 9.

that COD removal increased rapidly up to 6 min, then slightly up to 30 min. The COD removal efficiency for the EC-UF system at 30 min was 84 and 86% for 100 and 50 kDa UF membranes, respectively.

Figure 4(c) indicates that SS removal of EC-UF system reached steady state in approximately 2 min for both 50 and 100 kDa UF membranes. OG removal of EC-UF system reached steady state in approximately 2 min for 50 kDa UF membrane, while the steady state in OG removal was achieved in approximately 6 min for 100 kDa UF membrane (Figure 4(d)). The removal efficiencies of SS and OG by the EC-UF system were found as 100%.

Effect of membrane pore size on permeate flux

Ultrafiltration was carried out under constant transmembrane pressure of 1 bar and the permeate flux was measured with filtration time. Figure 5 shows the permeate flux decline for 50 and 100 kDa UF membranes. The initial permeate flux at the first 2 minutes was found to be 37.2 and 21.9 L/m^2·h for 100 and 50 kDa membranes, respectively. Permeate flux decline for 50 kDa membrane was higher than that of 100 kDa membrane. The flux decline during membrane filtration can be attributed to the concentration polarization, the formation a gel layer, the pore blockage and the adsorption of contaminants on the membrane structure. Pollutant removal efficiencies of 50 kDa membrane and 100 kDa membrane were very similar, but permeate flux of 100 kDa membrane was higher than that of 50 kDa membrane. Hence, it was concluded that the 100 kDa UF membrane was more suitable for EC-UF process.

Comparison of EC and EC-UF processes

The removal efficiencies of Zn, COD, SS and OG by EC and EC-UF processes are shown in Figure 6(a) and 6(b) respectively. It can be seen that removal efficiency of EC-UF process was remarkably higher than that of the EC process. Zn and COD removal efficiencies were found as 80.3 and 83.3% for EC and 99.6 and 86% for EC-UF process. SS and OG removal efficiencies were 82.4 and 100% for EC process and 100% for EC-UF process. These results showed the superior performance of the EC–UF process over that of the EC process, which was likely due to the formation of $Fe(OH)_3$ flocs during the electrocoagulation process, and effective rejection of the pollutants adsorbed on $Fe(OH)_3$ flocs by the UF membrane. It can be concluded that EC and UF processes can complement each other for the treatment of zinc plating wastewater.

Table 3 shows the raw and EC and EC-UF treated wastewater characteristics. The combination of electrocoagulation and ultrafiltration processes allowed significant reduction in wastewater pollutants. The EC-UF process could reduce zinc concentration from 200 to 1.58 mg/L, COD from 600 mg/L to 95 mg/L, SS from 144 to 0 mg/L and OG from 44 to 0 mg/L. In Table 3, discharge standards of USEPA and Turkish WPCR for metal plating wastewater are also shown. It can be seen from Table 3 that EC effluent met the discharge limits for COD, OG and SS, but did not meet the discharge limit for Zn. EC-UF effluent met the

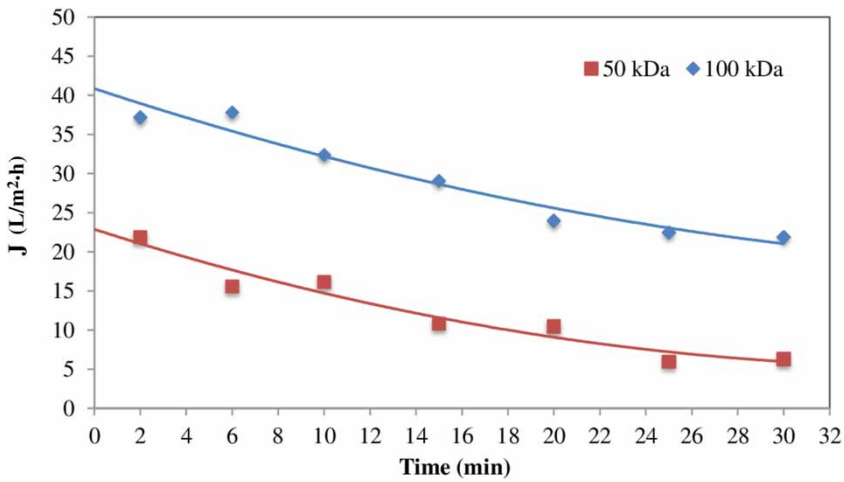


Figure 5 | Permeate flux decline during EC-UF process for 50 and 100 kDa UF membranes at pH 9.

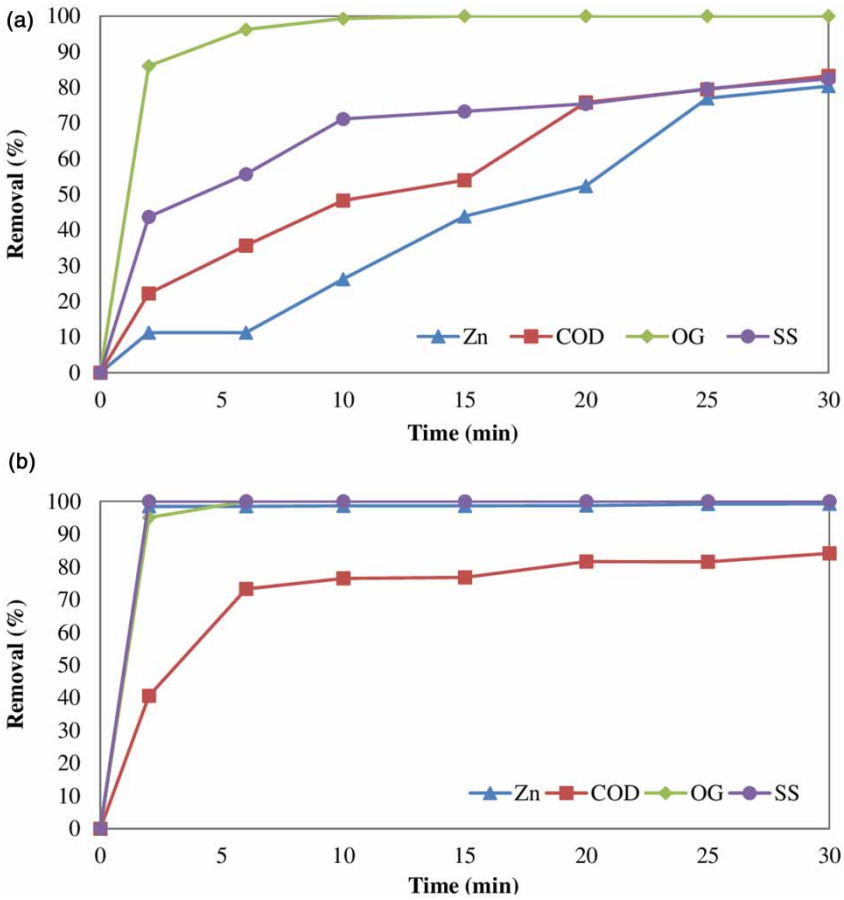


Figure 6 | Zn, COD, SS and OG removal by (a) EC and (b) EC-UF process; current density: 5 mA/cm^2, flow rate: 60 mL/min, pH: 9.

Table 3 | Wastewater characteristics after EC and EC-UF treatment

					USEPA limit[b]	
Parameter	**Wastewater**	**EC effluent**	**EC-UF effluent**	**WPCR limit[a]**	**Max.**	**Monthly avg.**
Chemical oxygen demand (mg/L)	600	100	95	200	–	–
Suspended solids (mg/L)	142	25	0	125	60	31
Oil–grease (mg/L)	44	0	0	20	52	26
Zinc (mg/L)	200	39	1.58	5	2.61	1.48

[a]Discharge standard for metal industry in Turkish Water Pollution Control Regulation (WPCR 2008).
[b]USEPA effluent guidelines and standards for metal finishing (40 CFR Part 433 (USEPA 2020)).

discharge limits for all the parameters and produced high quality water.

CONCLUSIONS

In this study, EC-UF process was used for the treatment and recovery of zinc plating wastewater. The effect of wastewater pH, flow rate and molecular weight cut-off of membrane on the performance of EC-UF process was investigated. The results showed that zinc removal was pH dependent and higher at high pH values. The optimum pH for zinc removal was obtained as 9.0. Two different membranes (50 and 100 kDa) were used in ultrafiltration experiments. It was found that zinc, COD, OG and SS removal was slightly higher for 50 kDa UF membrane than

that of 100 kDa UF membrane. The zinc removal with the combined EC-UF process was over 99% for both 50 and 100 kDa membranes at 1 A applied electric current (current density 5 mA/cm^2) and pH 9.0. It was also found that COD was reduced significantly and SS and OG were completely removed with EC-UF process. As a result, the combined EC-UF process was found to be a promising method in the treatment and recovery of zinc plating wastewater.

ACKNOWLEDGEMENT

This study was financially supported by Ondokuz Mayıs University Scientific Research Projects Commission (Project numbers PYO.MUH.1901.16.005).

DATA AVAILABILITY STATEMENT

All relevant data are included in the paper or its Supplementary Information.

REFERENCES

Al-Shannag, M., Al-Qodah, Z., Bani-Melhem, K., Qtaishat, M. R. & Alkasrawi, M. 2015 Heavy metal ions removal from metal plating wastewater using electrocoagulation: kinetic study and process performance. *Chemical Engineering Journal* **260**, 749–756.

APHA/AWWA/WEF 2017 *Standard Methods for the Examination of Water and Wastewater*, 23rd edn. American Public Health Association/American Water Works Association/Water Environment Federation, Washington, DC, USA.

Bilal, M., Shah, J. A., Ashfaq, T., Gardazi, S. M. H., Tahir, A. A., Pervez, A., Haroon, H. & Mahmood, Q. 2013 Waste biomass adsorbents for copper removal from industrial wastewater–A review. *Journal of Hazardous Materials* **263**, 323–333.

Changmai, M., Pasawan, M. & Purkait, M. K. 2019 Treatment of oily wastewater from drilling site using electrocoagulation followed by microfiltration. *Separation and Purification Technology* **210**, 463–472.

Channarong, B., Lee, S. H., Bade, R. & Shipin, O. V. 2010 Simultaneous removal of nickel and zinc from aqueous solution by micellar-enhanced ultrafiltration and activated carbon fiber hybrid process. *Desalination* **262**, 221–227.

Chen, X. & Deng, H. 2012 Removal of humic acids from water by hybrid titanium-based electrocoagulation with ultrafiltration membrane processes. *Desalination* **300**, 51–57.

Dąbrowski, A., Hubicki, Z., Podkościelny, P. & Robens, E. 2004 Selective removal of the heavy metal ions from waters and industrial wastewaters by ion-exchange method. *Chemosphere* **56** (2), 91–106.

Desai, K. R. & Murthy, Z. V. P. 2012 Removal of silver from aqueous solutions by complexation–ultrafiltration using anionic polyacrylamide. *Chemical Engineering Journal* **185–186**, 187–192.

Du, X., Qu, F., Liang, H., Li, K., Yu, H., Bai, L. & Li, G. 2014 Removal of antimony (III) from polluted surface water using a hybrid coagulation–flocculation–ultrafiltration (CF–UF) process. *Chemical Engineering Journal* **254**, 293–301.

Ennigrou, D. J., Gzara, L., Romdhane, M. R. B. & Dhahbi, M. 2009 Cadmium removal from aqueous solutions by polyelectrolyte enhanced ultrafiltration. *Desalination* **246**, 363–369.

Fu, F. & Wang, Q. 2011 Removal of heavy metal ions from wastewaters: a review. *Journal of Environmental Management* **92**, 407–418.

Gakwisiri, C., Raut, N., Al-Saadi, A., Al-Aisri, S. & Al-Ajmi, A. 2012 A critical review of removal of zinc from wastewater. *Proceedings of the World Congress on Engineering*, Vol. I. London, UK, pp. 627–630.

Gandhimathi, R., Babu, A., Nidheesh, P. V., Ramesh, S. T. & Anantha Singh, T. S. 2014 Laboratory study on leachate treatment by electrocoagulation using fly ash and bottom ash as supporting electrolytes. *Journal of Hazardous, Toxic, and Radioactive Waste (ASCE)* **19** (3), 04014033.

Gatsios, E., Hahladakis, J. N. & Gidarakos, E. 2015 Optimization of electrocoagulation (EC) process for the purification of a real industrial wastewater from toxic metals. *Journal of Environmental Management* **154**, 117–127.

Gonder, Z. B., Balcıoglu, G., Vergili, I. & Kaya, Y. 2020 An integrated electrocoagulation nanofiltration process for carwash wastewater reuse. *Chemosphere* **253**, 126713.

Gupta, V. K. & Nayak, A. 2012 Cadmium removal and recovery from aqueous solutions by novel adsorbents prepared from orange peel and Fe_2O_3 nanoparticles. *Chemical Engineering Journal* **180**, 81–90.

Jiao, A. Y., Li, Z. S. & Bao, L. C. 2013 Poly(acrylic acid-co-maleic acid) for the enhanced treatment of Cu (II) loaded aqueous solution and its reuse by ultrafiltration–electrolytic process. *Desalination* **322**, 29–36.

Kalaiselvi, G., Maheswari, P., Balasubramanian, S. & Mohan, D. 2013 Synthesis, characterization of polyelectrolyte and performance evaluation of polyelectrolyte incorporated polysulfone ultrafiltration membrane for metal ion removal. *Desalination* **325**, 65–75.

Kherti, Vinduja, V. & Balasubramanian, N. 2013 Removal of heavy metals by hybrid electrocoagulation and microfiltration processes. *Environmental Technology* **34**, 2897–2902.

Korus, I. & Loska, K. 2009 Removal of Cr(III) and Cr(VI) ions from aqueous solutions by means of polyelectrolyte-enhanced ultrafiltration. *Desalination* **247**, 390–395.

Kumarasinghe, D., Pettigrew, L. & Nghiem, L. D. 2009 Removal of heavy metals from mining impacted water by an electrocoagulation-ultrafiltration hybrid process. *Desalination and Water Treatment* **11**, 66–72.

Lakshmanan, D., Clifford, D. A. & Samanta, G. 2010 Comparative study of arsenic removal by iron using electrocoagulation and chemical coagulation. *Water Research* **44** (19), 5641–5652.

Magnenet, C., Jurin, F. E., Lakard, S., Buron, C. C. & Lakard, B. 2013 Polyelectrolyte modification of ultrafiltration membrane for removal of copper ions. *Colloids and Surfaces A: Physicochemical and Engineering Aspects* **435**, 170–177.

Mahmoud, A. & Hoadley, A. F. A. 2012 An evaluation of a hybrid ion exchange electrodialysis process in the recovery of heavy metals from simulated dilute industrial wastewater. *Water Research* **46** (10), 3364–3376.

Mavrov, V., Stamenov, S., Todorova, E., Chmiel, H. & Erwe, T. 2006 New hybrid electrocoagulation membrane process for removing selenium from industrial wastewater. *Desalination* **201**, 290–296.

Mier, M. V., Callejas, R. L., Gehr, R., Cisneros, B. E. J. & Alvare, P. J. J. 2001 Heavy metal removal with Mexican clinoptilolite: multi-component ionic exchange. *Water Research* **35** (2), 373–378.

Mouedhen, G., Feki, M., Petris-Wery, M. D. & Ayedi, H. F. 2009 Electrochemical removal of Cr(VI) from aqueous media using iron and aluminium as electrode materials: towards a better understanding of the involved phenomena. *Journal of Hazardous Materials* **168**, 983–991.

Qdais, H. A. & Moussa, H. 2004 Removal of heavy metals from wastewater by membrane processes: a comparative study. *Desalination* **164** (2), 105–110.

Sardari, K., Askegaard, J., Chiao, Y.-H., Darvishmanesh, S., Kamaz, M. & Wickramasinghe, S. R. 2018 Electrocoagulation followed by ultrafiltration for treating poultry processing wastewater. *Journal of Environmental Chemical Engineering* **6**, 4937–4944.

Sen, T. K. & Gomez, D. 2011 Adsorption of zinc (Zn^{2+}) from aqueous solution on natural bentonite. *Desalination* **267**, 286–294.

Sheikholeslami, R. & Bright, J. 2002 Silica and metals removal by pretreatment to prevent fouling of reverse osmosis membranes. *Desalination* **143** (3), 255–267.

Tavangar, T., Jalali, K., Shahmirzadi, M. A. A. & Karimi, M. 2019 Toward real textile wastewater treatment: membrane fouling control and effective fractionation of dyes/inorganic salts using a hybrid electrocoagulation–nanofiltration process. *Separation and Purification Technology* **216**, 115–125.

USEPA 2020 Electronic Code of Federal Regulations, e-CFR, 40 CFR, Chapter I, Part 433. https://www.epa.gov/eg/metal-finishing-effluent-guidelines.

WPCR (Water Pollution Control Regulation) 2008 Turkish Environment and Forestry Ministry.

Yahiaoui, O., Lounici, H., Abdi, N., Drouiche, N., Ghaffour, N., Pauss, A. & Mameri, N. 2011 Treatment of olive mill wastewater by the combination of ultrafiltration and bipolar electrochemical reactor processes. *Chemical Engineering and Processing* **50**, 37–41.

Zeftawy, M. A. M. E. & Mulligan, C. N. 2011 Use of rhamnolipid to remove heavy metals from wastewater by micellar-enhanced ultrafiltration (MEUF). *Separation and Purification Technology* **77**, 120–127.

Zeng, J., Ye, H. & Hu, Z. 2009 Application of the hybrid complexation–ultrafiltration process for metal ion removal from aqueous solutions. *Journal of Hazardous Materials* **161**, 1491–1498.

First received 23 February 2020; accepted in revised form 20 July 2020. Available online 3 August 2020