

Application of Electrocoagulation Process in Removal of Zinc and Copper From Aqueous Solutions by Aluminum Electrodes

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ABSTRACT: In this study, the performance of electro-coagulation in combination with aluminum sacrificial anode, in removal of zinc and copper has been investigated. Several parameters, such as pollutant concentration, pH, electrical potential, COD, turbidity and contact time were studied to achieve a higher removal capacity. Variable concentrations (5-50-500 ppm) of zinc and copper solutions were prepared. In order to follow the progress of the treatment, samples of 25mL were taken at 15 min intervals up to 60 min and then filtered with 0.45 μ diameter to eliminate sludge formed during electrolysis. The varying pH of the initial solution was also studied to measure their effects on the zinc and copper removal efficiency. Results obtained with synthetic wastewater revealed that the most effective removal capacities of studied metals could be achieved at 40 V electrical potential. In addition, the increase of electrical potential, in the range of 20-40 V, enhanced the treatment rate without affecting the charge loading required to reduce metal ion concentrations under the admissible legal levels. The process was successfully applied to the treatment of an electroplating wastewater where an effective reduction of zinc and copper concentration under legal limits was obtained, just after 15-60 min. Moreover, it can be concluded that the electro-coagulation process has the potential to be utilized for the cost-effective removal of heavy metals from water and wastewater.

Key words: Electro-coagulation, Zinc and copper removal, Aluminum electrodes

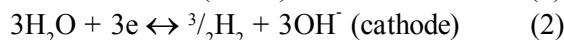
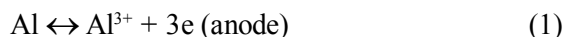
INTRODUCTION

In recent years, there has been an increasing public awareness of the long-term effect of water containing dissolved heavy metal ions (Son et al, 2004). Heavy metals, such as chromium, cadmium, copper, zinc and nickel, are contained in industrial waste water, which pollutes the environment and strongly affects human health; In this regard, the prevention of environmental pollution has actively been studied (Choi and Kim, 2003; Nouri *et al.*, 2006; Mahvi, 2008); Samarghandi, *et al.*, 2007; Nouri *et al.*, 2008). Toxic heavy metals are released into the environment from a number of industries such as mining, plating, dyeing, automo-

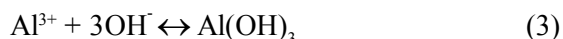
bile manufacturing and metal processing. The presence of heavy metals in the environment has led to a number of environmental problems (Vinodhini and Narayanan, 2008). In order to meet the water quality standards for most countries, the concentration of heavy metals in wastewater must be controlled (Kim et al, 2005; Mahvi *et al.*, 2007; Rahmani *et al.*, 2009; Asgari, 2008). Separation techniques of heavy metals, such as chromium, cadmium, copper, zinc and nickel, from industrial wastewater include precipitation, ion exchange, adsorption, electro-dialysis and filtration (Abdel-Ghani *et al.*, 2009; Malakootian *et al.*, 2009), but these techniques have limitations in

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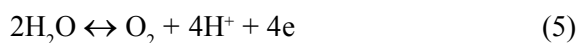
selective separation and the problems of high investment cost and equipment operation (Choi and Kim, 2005). During the past years, Electro-coagulation method has been proposed as an effective method to treat various wastewaters such as landfill leachate, restaurant wastewater, saline wastewater, tar sand and oil shale wastewater, urban wastewater, laundry wastewater, nitrate and arsenic bearing wastewater and chemical mechanical polishing wastewater (Niam *et al.*, 2007; Schulz *et al.*, 2009; Karbassi, *et al.*, 2008a). Electro-coagulation is a simple and efficient method to remove the flocculating agent generated by electro-oxidation of a sacrificial anode and generally made of iron or aluminum. In this process, the treatment is performed without adding any chemical coagulant or flocculants. Thus, reducing the amount of sludge which must be disposed (Cenkin and Belevstev, 1985). On the other hand, electrocoagulation is based on the in situ formation of the coagulant as the sacrificial anode corrodes due to an applied current, while the simultaneous evolution of hydrogen at the cathode allows for pollutant removal by flotation. This technique combines three main interdependent processes, operating synergistically to remove pollutants: electrochemistry, coagulation and hydrodynamics. An examination of the chemical reactions occurring in the electrocoagulation process shows that the main reactions occurring at the electrodes (aluminum electrodes) are:



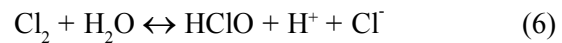
In addition, Al^{3+} and OH^- ions generated at electrode surfaces react in the bulk wastewater to form aluminum hydroxide:



If the anode potential is sufficiently high, secondary reactions may occur at the anode, such as direct oxidation of organic compounds and of H_2O or Cl^- present in wastewater:



The produced chlorine undergoes a dis-mutation reaction at pH higher than 3–4:



The aluminum hydroxide flocs normally act as adsorbents and/or traps for metal ions. Therefore, they would eliminate them from the solution. Simultaneously, the hydroxyl ions produced at the cathode increase the pH in the electrolyte and may induce co-precipitation of Cu^{2+} , Zn^{2+} in the form of their corresponding hydroxides. This acts synergistically to remove pollutants from water (Adhoum *et al.*, 2004). The objective of this study was to evaluate zinc and copper removal from synthetic solutions by electro-coagulation process method and to determine the effects of voltage, pH, COD, turbidity, and electrical conductivity on the removal efficiency. The study has been performed in 2009- 2010 in Tehran University of Medical Sciences, by financial support of Zahedan University of Medical Sciences.

MATERIALS & METHODS

All chemicals including copper sulphate (CuSO_4), zinc sulphate (ZnSO_4), sodium hydroxide pellets, concentrated sulfuric acid and potassium chloride were used as analytical grade. Desired concentrations of copper and zinc solutions were prepared by mixing proper amount of copper sulphate and zinc sulphate with de-ionized water. In order to increase the conductivity of the solution to 1.6 mS/cm, potassium chloride (1 N) was added to the solution before injecting it into the apparatus. The chloride salt added to the solution can also prevent the formation of the oxide layer on the anode and therefore reduce the passivation problem of the electrodes. The pH of influent solution was adjusted using sulfuric acid solution and sodium hydroxide (0.1 M).

The experiments were performed in a bipolar batch reactor (Fig.1). with four aluminum electrode connected in parallel (bipolar mode). Only the outer electrodes were connected to the power source, and anodic and cathodic reactions occurred on each surface of the inner electrode when the current passed through the electrodes. The internal size of the cell was 10 cm × 13cm ×

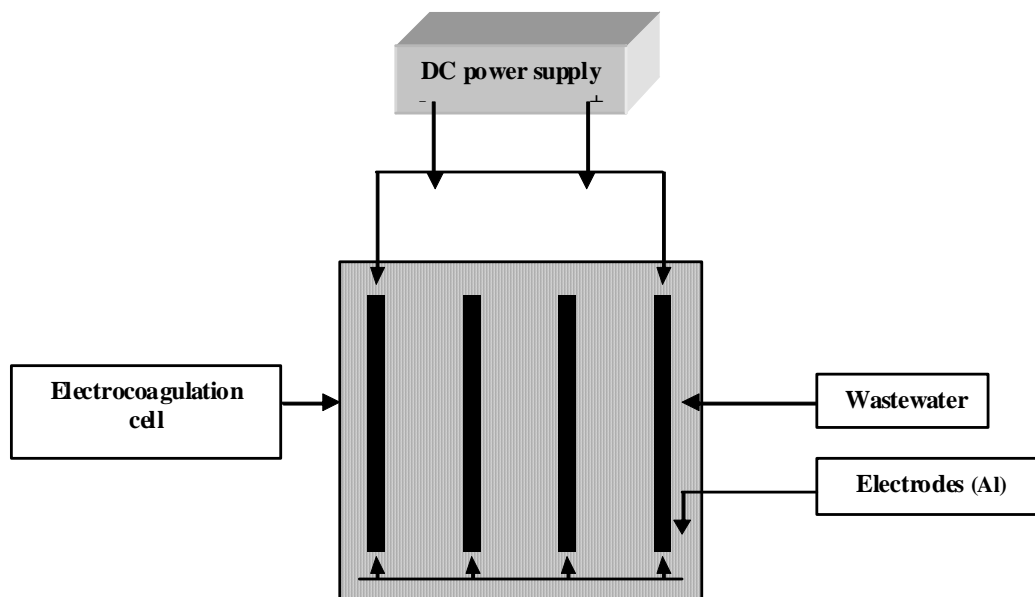


Fig.1. A schematic diagram of the experimental set up

12cm (width \times length \times depth) with an effective volume of 1000 cm³. The volume (V) of the solution of each batch was 1 l. The active area of each electrode was 10 \times 10 cm. The distance between electrodes was 1.5 cm. A power supply pack equipped to an input of 220V and variable output of 0–40V with maximum current of 5 ampere was used as direct current source.

The temperature of each system was maintained at 25 \pm 1 %C. The pH values in influent and reactor unit were measured using a pH meter model E520 (Metrohm Herisau, Switzerland). A Jenway Conductivity Meter (Model 4200) was employed to determine the conductivity of the solution. Samples were extracted every 15 minute for up to 60 min and then immediately filtered through a 0.45- μ m mixed cellulose acetate membrane. The residual copper and zinc concentration was determined using atomic absorption spectrophotometer method according to the standard method (APHA, 1992). The amount of copper and zinc ion removal was measured at pH 3, 7 and 10 and in electrical potential of 20, 30 and 40 volts. Also at the end of each stage of experiment, the volume of produced sludge was measured.

RESULTS & DISCUSSION

The electro-coagulation process is quite complex and may be affected by several operating parameters, such as pollutants concentrations, ini-

tial pH electrical potential (voltage), COD and turbidity. In the present study, electro-coagulation process has been evaluated as a treatment technology for zinc and copper removal from synthetic solutions. Zinc and copper removal efficiency at different condition (pH, electrical potential) in various times was evaluated. It has been established in previous studies (Chen *et al.*, 2000) that pH has a considerable effect on the efficiency of the electro-coagulation process. Also, as observed by other investigators the pH of the medium changed during the process. This change depends on the type of electrode material and initial pH and alkalinity.

In this study, the pH was varied in the range of 3–10 to investigate the influence of this parameter on the removal of zinc and copper. Removal efficiencies of zinc and copper as a function of initial pH with aluminum electrodes as presented in Tables 1-6. According to other investigators (Vik *et al.*, 1984; Yao Hu *et al.*, 2007; Bazrafshan *et al.*, 2007), pH would increase at low (< 7) initial pH. Vik *et al.* (1984) ascribed this increase to hydrogen evolution at cathodes. However, this was contested by Chen *et al.* (2000), who explained this increase by the release of CO₂ from wastewater owing to H₂ bubble disturbance. Indeed, at low pH, CO₂ is over saturated in wastewater and can release during H₂ evolution, causing a pH increase. In addition, if the initial pH is acidic, reactions would shift towards a pH increase. In alka-

line medium ($\text{pH} > 8$), the final pH does not vary very much and a slight drop was recorded. This result is in accord with previously published works and suggests that electro-coagulation can act as pH buffer. In this research, the influent pH did not affect the removal efficiencies significantly over a wide range. Therefore, pH adjustment before treatment is not required in practical applications. The pH variation of solution after electro-coagulation process in various voltages showed that the final pH for pH 3 and 7 of experiments is higher than initial pH, which is in agreement with results obtained later, but for initial pH ($\text{pH}=10$), the final pH was lower than 10, which is in agreement with results obtained later (Kobya *et al.*, 2003).

Preliminary laboratory testing of the electrolysis cell involved determining the effect of applied voltage on the efficiency of Zn^{2+} and Cu^{2+} removal. It is well-known that electrical current not only determines the coagulant dosage rate but also the bubble production rate and size and the floc growth (Letterman *et al.*, 1999; Holt *et al.*, 2002; Karbassi, *et al.*, 2007; 2008b), which can influence the treatment efficiency of the electro-coagulation. Therefore, the effect of current density or electrical potential (voltage) on the pollutants removal was investigated. As expected, it appears that for a given time, the removal efficiency increased significantly with the increase in current density. The highest electrical potential (40V) produced the quickest treatment with $> 90\%$ reduction occurring after only 15 min (for all concentrations) and the lowest Zn^{2+} and Cu^{2+} removal efficiency occurred in the lowest electrical potential (20V) and initial concentrations of 5.0 mg/L. This is ascribed to the fact that at higher voltage the amount of aluminum oxidized increased, resulting in a greater amount of precipitate for the removal of pollutants. In addition, it was demonstrated that bubbles density increases and their size decreases with increasing current density (Khosla *et al.*, 1991; Bazrafshan *et al.*, 2008), resulting in a greater upwards flux and a faster removal of pollutants and sludge flotation. As the current decreased, the time needed to achieve similar efficiencies increased and the results of this research confirm this fact. This expected behavior is explained by the fact that the treatment efficiency was mainly affected by charge loading ($Q = It$), as reported by Chen (Chen *et al.*, 2000). However, the cost of the process is deter-

mined by the consumption of the sacrificial electrode and the electrical energy which economically are the advantages of this method. These results suggest 40 V as an optimal electrical potential for the treatment of effluents containing zinc and copper, since it ensures the quickest removal rate with the lowest cost.

A set of experiments was performed with different initial concentrations of zinc and copper to determine the time required for removal under various conditions of electro-coagulation process. The results obtained at different electrical potential showed that initial concentration of Zn^{2+} and Cu^{2+} can not effect significantly on efficiency removal and for higher concentration of Zn^{2+} and Cu^{2+} , lower electrical potential is needed. On the other hand, there is not a direct correlation between pollutant concentration and removal efficiency. It is clear from Tables (1- 6) that in the higher concentrations, longer time is needed for removal of zinc and copper, but higher initial concentrations of zinc and copper were reduced significantly in relatively less time than lower concentrations. Thus, increases with the increase in concentration. This can be explained by the theory of dilute solution. In dilute solution, formation of the diffusion layer at the vicinity of the electrode causes a slower reaction rate, but in concentrated solution the diffusion layer has no effect on the rate of diffusion or migration of metal ions to the electrode surface (Chaudhary *et al.*, 2003).

The time dependence of zinc and copper removal by electro-coagulation process at different pHs is shown in Tables 1- 6. It can be seen from the Tables that up to 50-94 % of the initial concentration of zinc and copper decreased within 15-30 min of processing for all concentrations and the residual Zn^{2+} and Cu^{2+} concentrations in effluent at the end of reaction time (60 min) reached to < 2.0 mg/L. Thus, the discharge treated effluents to the environment can be performed safely. Comparison of Zn^{2+} and Cu^{2+} removal efficiency (Tables 1-6) showed that removal efficiency is similar.

A set of experiments was performed with different initial concentrations of Zn^{2+} and Cu^{2+} (5, 50 and 500 mg/L) to examine the effect of the presence of COD with concentration 100, 500 and 1000 mg/L and various levels of turbidity (10, 50 and 200 NTU) in wastewater on the removal effi-

Table 1. Percent of zinc removal during electro-coagulation process using aluminum electrodes (Initial concentration = 5 mg/L)

T= 60 min	T= 45 min	T= 30 min	T= 15 min	Voltage	pH
97.80	97.20	94.20	52.60	20	3
98.20	97.80	96.60	75.2	30	
99.20	98.80	97.20	85.60	40	
98.20	97.00	93.40	61.80	20	7
98.80	98.20	94.60	90.20	30	
>99.80	99.40	97.60	86.80	40	
98.00	96.60	92.60	62.80	20	10
98.80	97.40	93.80	89.40	30	
99.60	99.00	97.40	86.00	40	

Table 2. Percent of zinc removal during electro-coagulation process using aluminum electrodes (Initial concentration = 50 mg/L)

T= 60 min	T= 45 min	T= 30 min	T= 15 min	Voltage	pH
96.44	93.82	84.76	79.14	20	3
97.72	96.42	92.06	91.68	30	
98.32	97.46	95.14	93.76	40	
97.66	95.14	91.78	85.54	20	7
98.52	97.16	95.38	93.86	30	
98.74	97.78	96.12	94.74	40	
97.14	94.46	91.62	84.32	20	10
98.36	96.54	95.16	93.26	30	
98.58	97.46	95.74	95.02	40	

Table 3. Percent of zinc removal during electro-coagulation process using aluminum electrodes (Initial concentration = 500 mg/L)

T= 60 min	T= 45 min	T= 30 min	T= 15 min	Voltage	pH
96.562	95.034	93.576	91.818	20	3
97.196	96.314	95.608	94.492	30	
99.416	98.778	97.514	96.066	40	
96.962	95.858	94.354	93.822	20	7
99.578	97.342	96.074	95.656	30	
99.686	99.044	97.736	96.548	40	
96.766	95.652	94.616	93.506	20	10
99.454	97.412	96.392	95.238	30	
99.674	98.966	97.366	95.914	40	

Table 4. Percent of copper removal during electro-coagulation process using aluminum electrodes (Initial concentration = 5 mg/L)

T= 60 min	T= 45 min	T= 30 min	T= 15 min	Voltage	pH
98.18	97.44	95.26	75.72	20	3
99.72	99.66	99.38	97.66	30	
99.80	99.72	99.62	99.42	40	
98.52	97.58	96.60	78.60	20	7
99.78	99.76	99.44	97.76	30	
99.98	99.80	99.68	99.52	40	
98.26	97.38	95.92	76.60	20	10
99.72	99.68	99.40	97.70	30	
99.92	99.78	99.66	99.46	40	

Table 5. Percent of copper removal during electro-coagulation process using aluminum electrodes (Initial concentration = 50 mg/L)

T= 60 min	T= 45 min	T= 30 min	T= 15 min	Voltage	pH
96.72	95.78	92.92	84.74	20	3
96.96	96.12	93.62	89.46	30	
98.16	96.86	95.66	92.78	40	
97.86	95.92	93.48	88.52	20	7
98.06	96.48	93.84	91.68	30	
99.56	97.78	96.34	94.86	40	
97.66	95.80	92.58	87.16	20	10
97.94	96.20	93.64	91.06	30	
99.06	97.14	96.16	94.22	40	

Table 6. Percent of copper removal during electro-coagulation process using aluminum electrodes (Initial concentration = 500 mg/L)

T= 60 min	T= 45 min	T= 30 min	T= 15 min	Voltage	pH
96.762	95.514	93.848	93.178	20	3
97.366	96.172	94.522	94.172	30	
98.474	96.966	96.308	95.612	40	
97.386	96.114	94.976	94.536	20	7
97.794	96.706	95.874	95.45	30	
99.572	97.858	97.502	96.858	40	
96.976	95.988	94.906	94.076	20	10
97.458	96.562	95.892	95.172	30	
99.536	97.794	97.496	96.786	40	

ciency of Zn²⁺ and Cu²⁺. The results obtained at selected condition (pH=7, reaction time =60 min and voltage = 40V) showed that the removal efficiency for various concentrations of zinc and copper was constant and hence electro-coagulation process can be applied efficiently for Zn²⁺ and Cu²⁺ removal in the presence of COD and turbidity. With regard to a series of tests conducted with different concentration of zinc and copper in the solution, the weight of the electrode consumed with respect to different voltage of the pilot under study is given in Tables 7 and 8. As shown in the Tables 7 and 8, it can be concluded that the higher voltage of the system applied, the more electrode needs to be consumed in the process. Also, the higher the concentrations of the Zn²⁺ and Cu²⁺ in the solution, the higher consumption of the electrode would be. As the Tables represent, the electrode consumed with 40 volts in the process is much more than the process conducted with 20 volts. As the zinc and copper concentrations in the solution increased to 500 mg/L, the consumption of the electrode did not increase as much. However, the zinc and copper removal efficiency would occur, because much floc formation helped to sweep away zinc and copper and there was no need for as much electrode consumption as before. For example, aluminum elec-

trode consumption for initial concentration of zinc and for 5.0 mg/L and voltage 40V was 6.5 gram while for initial concentration of 500 mg/L, it was 2.8 g.

Table 7. Weight of aluminum electrode consumption during electro-coagulation process (g/L)

Zinc Concentration (mg/L)			Voltage (V)
500.0	50.0	5.0	
1.1	1.7	2.2	20
2.1	3.9	3.3	30
2.8	6.8	6.5	40

Table 8. Weight of aluminum electrode consumption during electro-coagulation process (g/L)

Copper Concentration (mg/L)			Voltage (V)
500.0	50.0	5.0	
1.3	1.7	4.1	20
1.4	5.3	7.9	30
13.1	7.7	10.2	40

Results showed that consumed energy for removal of one gram zinc and copper at electrical potential of 40V (Tables 9 and 10), initial concentration of Zn²⁺ and Cu²⁺ (5 mg/L) and pH 3, 7 and 10 was 20.74, 19.98 and 26.16 kWh and 31.15, 35.06 and 34.94 kWh, respectively. Also, consumed en-

ergy for removal of one gram zinc and copper at electrical potential 40V, initial concentration of 50 mg/L and pH 3, 7 and 10 was 1.67, 2.32 and 1.82 kWh and 2.24, 2.28 and 2.24 kWh, respectively. In addition, with initial concentration increase to 500 mg/L, pH of 3, 7 and 10 at electrical potential of 40V, the energy consumed for removal of one gram of zinc and copper was 0.07, 0.095 and 0.16 kWh and 0.07, 0.29 and 0.22 kWh, respectively. It can be concluded that the consumed energy decrease with increase in Zn²⁺ and Cu²⁺ concentration, because the enhanced floc formation help sweep pollutant away from the solution.

Table 9. Electrical energy consumption during Electro-coagulation process (kWh/g), using aluminum electrodes, at voltage = 40 V

Zinc Concentration, mg/L			pH
500.0	50.0	5.0	
0.07	1.67	20.74	3
0.095	2.32	19.98	7
0.16	1.82	26.16	10

Table 10. Electrical energy consumption during electro-coagulation process (kWh/g), using aluminum electrodes, at voltage = 40 V

Copper Concentration, mg/L			pH
500.0	50.0	5.0	
0.07	2.24	31.15	3
0.29	2.28	35.06	7
0.22	2.24	34.94	10

Finally, it can be concluded that electro-coagulation method is a reliable, safe, efficient and cost-effective method for removal of zinc and copper from industrial effluents, especially designed for voltage = 40V. On the other hand, in this study it was shown that electro-coagulation process achieves a fast and effective reduction of Zn²⁺ and Cu²⁺ (more than 98%) present in industrial effluents (such as plating baths wastewater). Indeed, the reported results show that electro-coagulation is an effective process for Zn²⁺ and Cu²⁺ removal from aqueous environments. Nevertheless, further studies should be carried out to confirm the practical feasibility of this method for treating various wastewaters and with different condition.

CONCLUSION

Using electro-coagulation process in removal of heavy metals by various electrode such as aluminum electrode has been extensively studied. The results obtained with synthetic wastewater revealed that the most effective removal capacities of proposed metals achieved at 40 V electrical potential. In addition, the increase of electrical potential, in the range of 20-40 V, enhanced the treatment rate without affecting the charge loading required to reduce metal ion concentrations under the admissible legal levels. The process was successfully applied to the treatment of an electroplating wastewater where an effective reduction of zinc and copper concentration under legal limits was obtained. Finally, it can be concluded that the consumed energy decrease with increase in Zn²⁺ and Cu²⁺ concentration, because the enhanced floc formation help sweep pollutant away from the solution.

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