

## Treatment of industrial electroplating wastewater for metals removal via electrocoagulation continuous flow reactors

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### ABSTRACT

A real industrial electroplating rinsing wastewater was collected and subjected to physical and chemical examination. The study showed that it can be categorized as high strength wastewater, at pH- 2, COD 1430 mg/l, and high level of metals above permissible limits, namely: 150, 30, 25, and 2.9 for Ni, Cu, Zn, and Fe mg/l respectively. Therefore, metals must be adequately removed before discharging to avoid any hazardous impact on the environment. Similar synthetic wastewater was prepared to study the effect of chemical coagulation for the precipitation of metals. The optimum removal rate was achieved by using a combination of lime and ferric chloride at 100 and 30 mg/l respectively. The chemically treated electroplating wastewater was subjected to an electrocoagulation study. A comparison between iron and stainless-steel electrodes for the removal of metals was investigated. Furthermore, the effect of different electric voltages, and contact time on metals removal efficiency were also examined. It was found that the optimum removal capacity was achieved when a stainless steel electrode was employed in the presence of ferric chloride as coagulant, at 10 volts, 30 min. contact time, and pH 9 for synthetic solution. In a batch treatment system, the real industrial wastewater was treated at the predetermined optimum operating conditions; the removal of metals was 92.1%, 87.8% and 82.9% for Ni, Zn, and Cu respectively. By employing a continuous flow reactor for the treatment of the same real wastewater and under the same operating conditions, metals removal rate increased to 98.9%, 97.4% and 96.6% for Ni, Zn, and Cu respectively. The level of metals in the final treated wastewater copes with Egyptian Environmental Regulation. The overall results confirmed that the electro-coagulation (EC) technology offers an effective alternative process in combination with the conventional chemical coagulation process for reaching high removal performance of toxic metals from the electroplating wastewater. The advantage of EC technique is achieving high treatment efficiency instead of expensive chemical reagents, high construction cost and/or other conventional processes. In addition, the final treated water can be reused for rinsing process in the electroplating industry and/or discharging without any environmental hazard effect. It is also recommended to employ solar energy instead of electricity to reduce cost of operation.

**Key words:** chemical coagulation, electrocoagulation, heavy metals, industrial electroplating wastewater, wastewater treatment

### HIGHLIGHT

- Treatment of industrial wastewater and removal of heavy metal by electrocoagulation process using stainless steel electrode.

### INTRODUCTION

Electroplating industrial activities are among the major pollutants to the environment and waterways (Abdel-Shafy 2015; Ghernaout *et al.* 2019). Such activities led to the formation of hazardous wastewater associated with toxic heavy metals that threaten the environment and aquatic lives (El-Bahy *et al.* 2005). Meanwhile, electroplating industries produce large amounts of polluted wastewater presenting hazard and impose a negative environmental impact (Rajemahadik *et al.* 2013). On the other hand, heavy metals are categorized among the most hazardous pollutants to the environment, including groundwater (Abdel-Shafy *et al.* 2016). It is well documented that discharging of heavy metals could cause adverse impact to the environment, Man's health and other living beings due to their bioaccumulation effects (Abdel-Shafy & Kamel 2016). Contamination of waterways by

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heavy metals could induce adverse impact to the aquatic lives including fish and algae (Abdel-Shafy & El-Saharty 2015). The electroplating wastewaters are characterized by high acidic values and ionic strength (Kabdaşlı *et al.* 2012). Besides, it usually contains several valuable metals such as gold, silver, copper, nickel, and zinc.

Furthermore, the conventional physical processes and chemical coagulation for treatment of the electroplating wastewater proved to be high capital investment as well as high running costs (Rajemahadik *et al.* 2013; Abdel-Shafy & Al-Sulaiman 2014). Several conventional processes including neutralization followed by precipitation, or physical treatment including ion-exchange, and membrane technology are also used for wastewater treatment (Malathi *et al.* 2014; Abdel-Shafy & Abdel-Shafy 2017). Other treatment techniques include adsorption, filtration, and chemical coagulation (Abdel-Sahfy *et al.* 1998). The application of these treatment techniques is problematic to the developing countries as a high technique. The most applicable process for the treatment of electroplating wastewater is neutralization followed by sedimentation (Abdel-Shafy *et al.* 2020). Nevertheless, this process results in producing large amounts of precipitated metallic sludge that requires additional handling and cost. Furthermore, the process is not feasible because the precipitated metals are lost through discharge (Abdel-Shafy *et al.* 1997; Muchlis & Ardeniswan 2018). It would be highly desirable from the economic point of view as well as environmentally beneficial to recover these metals.

Therefore, electrocoagulation process is an alternative promising solution using electricity instead of expensive chemical reagents (Hafez *et al.* 2018). This process attracted great attention for the treatment of industrial wastewater due to its versatility and environmental compatibility. (Rajemahadik *et al.* 2013). The advantages of this processes compared to the other conventional methods is that it uses simple equipment, is easy to operate, shorter time is allowed, and limited chemicals are needed (Banupriya *et al.* 2017). Moreover, this process provides rapid sedimentation of the electro-generated flocs besides producing less sludge (Abdel-Shafy 2015; Banupriya *et al.* 2017).

Furthermore, the electrocoagulation process is capable of removing the smallest colloidal particles in comparison to the convention flocculation/coagulation method (Beyazit 2014; Abdel-Shafy *et al.* 2019). The obtained charged particles pose a greater capability for coagulation and destabilization due to the electric field of these processes that sets them in motion (Behbahani *et al.* 2011).

Electrocoagulation technology (EC) is an emerging process for the treatment of water and wastewater that proves successful handling of both organic and inorganic pollutants (Ghernaout *et al.* 2019). The technique requires a limited amount of chemicals and involves the dissolution of a sacrificial anode that produces metal hydroxide as an active and strong coagulant (Abdel-Shafy *et al.* 2002). The removal of the contaminants from wastewater proceeds via destabilization/neutralization of the repulsive forces that keep all pollutants suspended in the wastewater. By neutralizing the repulsive forces, the suspended pollutants form large particles that can be settled for easier separation from the treated wastewater. The achievement of such technology depends on its ability to satisfy the commercial criteria including minimizing the cost, time of operation, and maintenance as well as recycling the metals instead of disposing of them as wasted components (Abdel-Shafy *et al.* 2002; Rajemahadik *et al.* 2013).

The most employed electrodes in the EC are soluble ones including mostly iron and aluminum. The insoluble electrodes include carbon or stainless steel in combination with a coagulant (such as ferric chloride, alum, or lime). Each electrode yields different results (Beyazit 2014; Abdel-Shafy *et al.* 2015; Santhosh *et al.* 2015).

Several studies were conducted in electro-coagulation using stainless steel as a cathode (Beyazit 2014; Santhosh *et al.* 2015). The study was concerned with the removal of Cu(II), Cr(VI), and Ni(II) from wastewater of metal plating in a electrocoagulation (EC). Beyazit (2014) employed aluminum (Al), iron (Fe), and stainless steel (St) electrodes. He also used an Fe-St pair electrode for subsequent experiments. This study reported that by using the Fe-St pair, 100% removal could be achieved for all metals at pH 9, and the current density was 90 A/m<sup>2</sup> (Beyazit 2014).

Continuous treatment reactors are open thermodynamic systems with a continuous wastewater feed (Santhosh *et al.* 2015). The Electrocoagulation reactor consists of cathode and anode that are designed in a proper way to let the reactions occur under well-controlled conditions to obtain the specific products (Packialakshmi & Bhawani 2018). It works at specific dimensions using electrodes supplied with adequate electric direct current via power supply (Kabdaşlı *et al.* 2012).

The metal plates, which are known as sacrificial electrodes, are usually connected in parallel with a specified inter-electrode distance (namely: 1.5–3.5 cm). Meanwhile, the supplied electric current is distributed on the

electrodes depending on the resistance of the individual electrodes. It is important to mention that the distance between the electrodes has a direct relationship with the electricity consumption (Verma & Khandegar 2013).

The aim of the present work is to evaluate the present electrocoagulation technology, which gives zero or minimum environmental contaminants with heavy metals. This study is concerned with real industrial electroplating wastewater effluent containing nickel, copper, and zinc. The study focuses on employing an electrocoagulation system with electrochemical corrosion using insoluble stainless steel electrodes in combination with lime and ferric chloride as coagulant and coagulant aid in a continuous flow system. The objective is to reach the permissible limits and the legal regulations of discharging treated industrial wastewater into the environment and/or reuse in the electroplating industry in the rinsing process.

## MATERIALS AND METHODS

### Real industrial electroplating wastewater

Industrial electroplating wastewater effluent was collected from Candilars metal electroplating plant, Cairo. This plant consumes about 100 m<sup>3</sup> of water daily in the electroplating process, 80% of wastewater is reused after treatment for rinsing the electroplated pieces. This plant produces more than 35,000 m<sup>3</sup> electroplating wastewater per year as rinsing water, containing variable amounts of heavy metals, namely Cu, Ni, and Zn. Samples of the raw rinsing electroplating wastewater 20 litre each were monthly collected and transferred to our laboratories for the determination of the physical and chemical characteristics including the level of heavy metals.

### Chemically synthetic electroplating wastewater

Synthetic electroplating wastewater was laboratory prepared resembling the real industrial wastewater in terms of heavy metals using NiCl<sub>2</sub>, NiSO<sub>4</sub>, CuSO<sub>4</sub> and ZnCl<sub>2</sub> (Table 2). The concentration of metals was 450, 90, and 75 mg/l for Ni, Cu, and Zn respectively. The pH of the synthetic wastewater was adjusted to pH 2 using HCl.

### Chemical coagulation

This examination was conducted by bench scale study using the jar-test apparatus (Abdel-Shafy 2015). The experiment was conducted using 6 jars each at 1,000 ml wastewater each. The synthetic and real electroplating wastewater were subjected to chemical coagulation except the control jar. The jar-test experiment consists of flash mixing for 1 min., flocculation for 20 min, and settling for 30 min. The determined parameters and the results are given in Table 1. Different coagulants were examined to select the most efficient one. The examined coagulants were NaOH, alum, alum in combination with NaOH, lime, and lime in combination with FeCl<sub>3</sub>. The most efficient coagulant was selected based on percentage of metals removal. Such highest efficiency coagulant was further employed for the treatment of the real industrial wastewater in a continuous system. All the used chemicals were selected from Analytical Grade reagents.

### Continuous flow electrocoagulation treatment system

#### Reactor design

The laboratory scale electrocoagulation reactor consists of stainless steel plate electrodes, DC power supply, voltmeter, electric pump, cables and electrolyte (Figures 1 and 2). The design of the reactor was as follows: a plastic rectangular tank reactor with external dimension at 35 cm length 18 cm width, 22 cm height, and wall thickness at 2 mm. (Figure 1). The maximum liquid height was 20 cm, thus batch electrocoagulation reactor has a maximum 10 L capacity. The reactor divided into six sets as follow: the first five sets consisted of two stainless steel electrodes assembled in two parallel ones. The last set was designed to collect the clear treated water through a hole connected with a hose to eject the clear treated water. All anodic and/or cathodic electrodes were connected together separately in an external way with a copper wire in mono-polar series.

#### The electro-coagulation device

The electrode design consists of twelve sheets of AISI 304 stainless steel (Figure 1). They were used as electrode material, where each two electrodes were fixed in each set of the batch reactor. Each electrode had a surface area of 30 cm<sup>2</sup> (10 cm × 3 cm) and 0.1 cm thickness. The purpose of such intermediate electrode coverage is to ensure sufficient bubble dispersion for metal removal by flotation as well as enabling the formation of currents that induced agitation and mixing. Electrode passivation is the accumulation of an inhibiting layer (usually a metal oxide) on the surface of the electrode. The electrodes are connected with each other into 2 opposite rows,

**Table 1** | Physical and chemical characteristics of the raw electroplating wastewater before and after treatment using chemical coagulation

Parameter	N	Raw EWW	EWW after treatment	% of removal
Temp °C	11	25	26	–
pH	11	2	8	increased
DO (mg/L)	11	4	5.71	increased
Turbidity (NTU)	9	256	2	95
EC (mS/cm)	9	1.695	1.69	zero
TDS (mg/L)	9	1180	130.3	88.96
TSS (mg/L)	7	100	50	50
TOC (mg/L)	7	370	198	46.5
COD (mg/L)	7	1430	937	65
BOD (mg/L)	7	1.56	1	52.98

N, number of samples.

Raw EWW = raw electro-plating wastewater.

EWW after treatment = electro-plating wastewater after Electrocoagulation treatment.

% of removal = percentage of removal.

**Figure 1** | Set up of the electrocoagulation (EC) cell unit consisted of stainless steel plate electrodes, DC power supply, voltmeter, electric pump, cables and electrolyte.

where one row is connected with anode and the other is connected with cathode. The distance between the electrodes was 2 cm each. The set of electrodes was submerged into the examined electroplating wastewater. This system was designed to determine the released coagulant and the bubble type, thereby influencing flotation, mixing, mass transfer and metal removal. It is worth mentioning that flotation is a major separation mode, therefore, electrolytic bubbles production is required.

Thus, electrochemically inert electrodes were needed. Stainless steel is electrochemically inert and does not readily decay in the presence of ferric chloride as coagulant. Such electrodes were selected for the required reaction. An electrode with a known flat surface area, as a plate electrode, and constant spacing from each other can ensure a constant current density. The electrodes were arranged in mono-polar mode with the rectifier. The combination of a potential difference and an electrode arrangement can determine the bubbles removal path rate and the mixing. As current density increased, so did the production of the bubbles increased. Thus greater upward momentum flux and an increase in the mixing rate could be maintained. The electrodes arrangement as well as their physical placement within the reactor can determine the bubble dispersion throughout the reactor. Larger electrode surface area resulted in a greater dispersion of bubbles throughout



**Figure 2** | set up (surface view) of electrocoagulation treatment cell for electroplating wastewater effluent.

**Table 2** | Characteristics of metal plating industrial wastewater

Characteristics	Units	Value
pH	–	2.0
Electric Conductivity	mS/cm	560
Zinc	mg/l	25
Copper	mg/l	30
Nickel	mg/l	150
Iron	mg/l	2.9

the reactor, whereas a smaller electrode surface area resulted in a concentrated source of bubbles within the reactor.

The Inductive Coupled Plasma (ICP) OES: A Unique Double Monochromator Optical System was employed for metals determination throughout all the present experimental study. All experiments were conducted at 25 °C room temperature, constant 10 voltage and 100 ml/min. flow rate.

## RESULTS AND DISCUSSION

### Raw wastewater

The physical and chemical characteristics of the raw electroplating wastewater are given in Table 1. The given results indicated that this wastewater can be classified as ‘highly strength’ wastewater where the E.C., turbidity, TDS are 1.695 mS/cm, 256 NTU, and 1,180 mg/l respectively. Meanwhile, the COD value is 1,430 mg/l. It is highly acidic wastewater at pH = 2, and non-biodegradable as indicated from the level of BOD and COD (Table 1). The high value of the turbidity may be explained by the continuous use of the commercial chemicals as well as the repeated use of the same water during the same operation process.

In addition, level of heavy metals in this industrial electroplating wastewater is given in Table 2. Level of Zn, Cu, Ni, and Fe was 25, 30, 150 and 2.9 mg/l respectively. This industrial wastewater should be adequately to prevent any hazard impact to the environment.

### Chemical coagulation

The synthetic solution was subjected to different coagulants, namely: NaOH, alum, alum in combination with NaOH, lime, and lime in combination with FeCl<sub>3</sub>. The results (Table 3) indicated narrow variations in the efficiency of the studied coagulants. However, the combination of lime and FeCl<sub>3</sub> exhibited the most efficient one

**Table 3** | Correlation between the efficiency of the studied chemical coagulants on the removal of Cu, Ni and Zn at the optimum pH from the synthetic solution

Chemical coagulant	Optimum pH	% of metal removal					
		Metal* Cu (mg/l)	% R	Metal* Ni (mg/l)	% R	Metal* Zn (mg/l)	% R
A.WW*		90	–	450	–	75 mg/l	–
NaOH	8.5	11.66	87.0	76.5	83.0	19.5	74.0
Alum	8.4	19.35	78.5	89.55	80.1	20.63	72.5
Alum + NaOH	8.5	18.27	79.7	78.75	82.5	19.65	73.8
Lime (CaO)	9.0	13.79	84.68	67.5	85.0	16.0	78.7
Lime (CaO) + FeCl <sub>3</sub>	8.5	3.51	96.1	60.0	86.66	15	80.0

(initial metal concentration was 90 mg/l for Cu, 450 mg/l for Ni, and 75 mg/l for Zn).

A.WW\* = initial metal concentration (as mg/l) in the artificial wastewater before chemical coagulation.

% R = percentage of metal removal.

Metal\* = metal concentration after chemical coagulation.

at which the removal of Cu, Ni, and Zn reached 96.1, 86.66%, and 80.0% respectively (Table 3). The corresponding residual concentration of metals was 3.51, 60, and 15 mg/l for Cu, Ni, and Zn successively.

### Electro-coagulation

Removal of Cu, Ni, and Zn from synthetic solution was studied under variable factors including different electrode types, variation in electric voltage and different contact times. For this purpose two types electrodes namely: Iron electrode and stainless steel electrode were examined. Each type of electrode was examined under different electric currents and contact times to determine the optimum metal removal rate using the synthetic electroplating wastewater. The obtained conditions with regards to the suitable electrode, electric voltage, and contact time would be used for the continuous treatment reaction of both the synthetic solution and real wastewater sample.

### Electro-Coagulation of the artificial synthetic solution after chemical treatment

#### Effect of different electric voltage

The effect of the different electrical Voltage on the removal of metals from the synthetic solution as artificial electroplating wastewater using iron electrode was investigated. This study was conducted at variable electric voltage namely: 5, 10, 15, and 20 mv. This study was conducted at 30 min. contact time. The obtained results are given in Table 4. These results indicated that the removal of the studied metals from the aqueous solution increased by increasing electric voltage from 5 to 10 V. As the voltage increase the removal efficiency increased to 90.6%, 89.2% and 89.8% for Ni, Zn, and Cu respectively at 10 volts. Further voltage increase showed slight decrease in the removal rate. At 15 volts metal removal was 90.2%, 88.4% and 88.5% for Ni, Zn, and Cu respectively. At 20 volts the removal of the same metals was 89.8%, 88.1% and 87.9%

**Table 4** | Effect of different voltage on the removal of metals from the chemically treated electroplating synthetic wastewater<sup>a</sup> via electrocoagulation using iron electrode at 30 min

Volt Mv	Ni		Zn		Cu	
	conc. (mg/l)	%R	conc. (mg/l)	%R	Conc. (mg/l)	%R
Initial metal concentration after chemical coagulation <sup>a</sup>	80	–	20	–	22.5	–
5 mv	9.6	88.0	2.82	85.9	2.88	87.2
10 mv	7.52	90.6	2.16	89.2	2.30	89.8
15 mv	7.84	90.2	2.32	88.4	2.59	88.5
20 mv	8.16	89.8	2.38	88.1	2.72	87.9

conc. (mg/l) = metal concentration as (mg/l).

%R = percentage of removal.

<sup>a</sup>Initial metal concentration after chemical coagulation using Lime (CaO) + FeCl<sub>3</sub>.

successively. This attributed to the fact that by increasing the value from (5 V) to (10 V) the removal rate increase, further increase in to (15 V) or (20 V) was not in the favor of metal removal.

In addition, it was demonstrated that bubbles density increased by increasing the voltage from 5 to 10 V, but their size decreases with increasing current density to 15 and 20 V. This can be explained that increasing the current density above the value (10 V) increases the dissolution of  $\text{Fe}^{+2}$ . This leads to excessive generation of oxygen which; consequently; cause heat generation.

It was demonstrated that the efficiency of metal removal from wastewater depends mostly on the applied current intensity and material of electrodes. These enhancing effects are attributed to the increase in the driving force of the electrode reaction and the increase in current voltage (Holt *et al.* 2002). It is well known that the electrical current not only determines the coagulant dosage rate but also the bubbles production rate and the size of the flocs which can influence the treatment efficiency of the electrocoagulation. Therefore, it was found essential to study the effect of current density or electrical potential (voltage) on the pollutant removal was investigated (Kabdaşlı *et al.* 2012).

*Effect of different electrical Voltage on the removal of metals from the synthetic wastewater using Stainless steel electrode.* The insoluble stainless steel electrode (304) was employed. In this study 100 mg/l of  $\text{FeCl}_3$  was added to the synthetic wastewater to enhance the process. This electrode was made of (iron and carbon) mixed with elements such as chromium to keep it resistant to rusting and with nickel to enhance corrosion resistance properties (Khosa *et al.* 2013). This study was conducted at 30 min. contact time.

The results are presented in Table 5. The Optimum achievement was at 10 volt at which the removal rate of Ni, Zn, and Cu was 93.5, 89.8, 90.1% respectively. By increasing the voltage to 15 V under the same contact time; metal removal rate decreased to 87.1%, 85.6% and 86.0% for Ni, Zn, and Cu respectively. Further increase in the voltage to 20 V induced further decrease in the removal rate to 85.5%, 83.9%, and 85.3% for Ni, Zn, and Cu respectively (Table 5).

**Table 5** | Effect of different voltage on the removal of metals from the synthetic wastewater<sup>a</sup> via electrocoagulation using stainless steel electrode at 30 min.

Volt mv	Ni		Zn		Cu	
	conc. (mg/l)	%R	conc. (mg/l)	%R	conc. (mg/l)	%R
Initial metal concentration after coagulation <sup>a</sup>	80	–	20	–	22.5	–
5 mv	9.04	88.7	2.78	86.1	2.75	87.8
10 mv	5.20	93.5	2.04	89.8	2.23	90.1
15 mv	10.32	87.1	2.88	85.6	3.15	86.0
20 mv	11.60	85.5	3.22	83.9	3.31	85.3

contact time and after chemical coagulation with Lime and ferric chloride.

conc. (mg/l) = metal concentration as (mg/l).

%R = percentage of removal.

<sup>a</sup>Initial metal concentration after chemical coagulation using Lime (CaO) +  $\text{FeCl}_3$ .

This achievement may be attributed to the fact that the presence of the dissolved iron traces resulted from addition of  $\text{FeCl}_3$  as coagulant aid performs. The later (i.e.  $\text{FeCl}_3$ ) acted in same manner as the iron electrodes in terms of charging the particulates, thereby allow them to agglomerate and settle at the bottom of the tank. It seems that increasing the applied potential enhanced the generation of  $\text{Fe}^{+3}$  and  $\text{OH}^{-1}$  in the electrochemical cell at 10 volt as optimum removal value. Above such optimum value (namely: 15 and 20 volt) dissolution of  $\text{Fe}^{+3}$  exceeded. This leads to excessive generation of oxygen with consequently caused heat generation. Therefore, no more  $\text{Fe}^{+3}$  ions remain in the solution, as a result the removal rate decreased (Shammas *et al.* 2010).

#### Effect of different contact time

In this study the effect of the variable contact times namely 15, 30, 45, and 60 minutes was investigated at the predetermined optimum current voltage (10 V) using either iron electrodes and/or stainless steel electrodes with  $\text{FeCl}_3$ .

*By using iron electrode at different contact times.* The effect of different operating contact time on the removal of Ni, Zn, and Cu by the electrocoagulation process is shown in Table 6. Results indicated that the optimum removal efficiency was observed at 30 minutes contact time where the removal rate reached 90.6, 89.2 and 89.8% for Ni, Zn, and Cu respectively. Further increase in the contact time to 45 min. induced a notable decrease in metal removal rates to 89.9, 88.4 and 88.8% for Ni, Zn, and Cu respectively, and to 89.1, 88.1 and 87.9% at 60 minutes contact time for the same corresponding metals.

**Table 6** | Electrocoagulation using iron electrode at 10 V and different contact times namely 15, 30, 45, and 60 minutes

Contact Time Min.	Ni		Zn		Cu	
	conc. mg/l	%R	conc. mg/l	%R	conc. mg/l	%R
Initial metal concentration after coagulation <sup>a</sup>	80	–	20	–	22.5	–
15 Min.	9.28	88.4	2.46	87.7	2.66	88.2
30 Min.	7.52	90.6	2.16	89.2	2.29	89.8
45 Min.	8.08	89.9	2.32	88.4	2.50	88.8
60 Min.	8.72	89.1	2.38	88.1	2.72	87.9

After chemical coagulation using Lime (CaO) + FeCl<sub>3</sub>.

Min. = minutes.

conc. (mg/l) = metal concentration as (mg/l).

%R = percentage of removal.

<sup>a</sup>Initial metal concentration after chemical coagulation using Lime (CaO) + FeCl<sub>3</sub>.

In this system the anodic and cathodic reduction occurs at positive and negative electrodes respectively. Active evolution of oxygen is responsible for the anode dissolution in the liquid which is related to the dissolving of certain amount of iron. The released ions neutralized the particles charges thus initiate the coagulation effectively. Initially, minor formation of oxygen at the sacrificial anode competes with iron dissolution in the given synthetic electroplating wastewater which in turn reduces the dissolved amount of iron at this sacrificial anode. This induces the co-precipitation and sweeping effect which attributed to higher removal rate of the concerned metals (Chaturvedi 2013).

Meanwhile, optimum removal efficiency that achieved at 30 minutes contact time is mainly due to formation of enough coagulant iron hydroxides concentration. On the contrary, the decrease in the removal efficiency observed at both 45 and 60-minutes contact time is due to the continuous electrocoagulation which leads to the formation of hydroxides ions that oxidized at anode. As a result, the generation of iron is reduced; and the efficiency of metals removal decreased (Chopra *et al.* 2011).

#### By using stainless steel electrode at different contact times

Results are given in Table 7. The optimum removal rate of the studied metals was reached at 30 minutes at which the removal efficiency was 93.5%, 89.8% and 90.0% for Ni, Zn, and Cu respectively. Increasing the contact time to 45 minutes decreased the removal rate to 91.1%, 88.1% and 86.0% to Ni, Zn, and Cu respectively. Further

**Table 7** | Electrocoagulation using stainless steel electrode at 10 V and different contact times namely 15, 30, 45, and 60 minutes

Contact Time Min.	Ni		Zn		Cu	
	conc.	%R	conc.	%R	conc.	%R
Initial metal concentration after coagulation <sup>a</sup>	80	–	20	–	22.5	–
15 Min.	11.6	85.5	13.6	83	3.65	83.8
30 Min.	5.2	93.5	2.04	89.8	2.24	90.0
45 Min.	7.12	91.1	2.38	88.1	3.15	86.0
60 Min.	9.68	87.9	2.88	85.6	3.71	85.3

conc. = metal concentration as (mg/l).

%R = percentage of removal.

<sup>a</sup>Initial metal concentration after chemical coagulation using Lime (CaO) + FeCl<sub>3</sub>.



increase in the contact time to 60 min. showed further decrease in the removal rate to 87.9%, 85.6% and 85.3% for the same metals successively.

Comparison between using iron electrodes (Table 6) and stainless-steel electrodes (Table 7) indicated that the latter is slightly more efficient than the former in term of metals removal in particularly with Ni. The dissolution of traces iron that resulted from using FeCl<sub>3</sub> in combination with lime could compensate the presence of such ion for the stainless steel electrode. Meanwhile, the use of stainless steel electrode proved to be more suitable than iron electrode because there is no consumption of the mass of this electrode. Meanwhile, the excess amount of flocs formation sweeps away the metals from the solution. By employing the optimum treatment conditions in a continuous system, the final treated effluent can be discharged safely to the water bodies without any pollution threat while meeting the required standard regulation (Saleem *et al.* 2011; Kashefiasl *et al.* 2014).

## CONTINUOUS TREATMENT PROCESS

### Synthetic wastewater

The synthetic wastewater sample was subjected to the optimum studied conditions namely: chemical coagulation using lime in combination with FeCl<sub>3</sub>, followed by a continuous treatment using electrocoagulation cells. This experiment was running according to the optimum operating conditions previously obtained from the batch experiments. These conditions are: 10 mv electric current, and 30 min. contact time, and stainless steel electrodes.

The electrocoagulation cells were divided into 6 parts, each part consisted of two parallel stainless-steel electrodes that are connected in mono-polar series. Each pair of electrodes is internally connected with each other and has no interconnections with the outer electrodes. It is worth to mention that such arrangement of the mono-polar electrodes with cells in series is electrically similar to a single cell with many interconnected electrodes. A higher potential difference in such mono-polar series cell arrangement is required to allow the flowing of the given current due to the fact that connecting the cells in series are characterized by higher resistance. In such arrangement, the electrical current flows equally through all the electrodes (Fathy *et al.* 2020).

The results are given in Table 8. The removal efficiency of Ni, Zn, and Cu reached 93.9%, 90.3% and 91.1% respectively. These results indicated that the continuous treatment process was more efficient than the batch treatment system in terms of metals removal.

**Table 8** | Comparison between the efficiency of batch process and continuous treatment process in the removal of metals from synthetic solution using electrocoagulation at 10 mv electric voltage and 30 min.

	Ni		Zn		Cu	
	conc.	%R	conc.	%R	conc.	%R
Initial metal concentration after chemical coagulation <sup>a</sup>	80	–	20	–	22.5	–
Electro-coagulation In Batch process <sup>b</sup>	5.2	93.5	2.04	89.8	2.24	90.0
Electro-coagulation by Continuous treatment process <sup>b</sup>	4.88	93.9	1.94	90.3	2.00	91.1

contact time = metal concentration as (mg/l).

%R = percentage of removal.

<sup>a</sup>Initial metal concentration after chemical coagulation using Lime (CaO) + FeCl<sub>3</sub>.

<sup>b</sup>After electro and chemical-coagulation.

It is worth to mention that increasing the number of electro-plates increases the removal efficiencies of heavy metals from solutions. This is mainly due to decreasing the space between the electrodes (Fathy *et al.* 2020). The given results showed that the removal efficiency of heavy metals increases when the distance between the plates decrease. This is mainly due to the fact that the resistance between plates at constant voltage decreases, thus the current increases. The later induces an increase in the coagulation efficiency (Kashefiasl *et al.* 2014). Therefore, the performance of the electrocoagulation process is enhanced.

### Real electroplating wastewater

Real electroplating industrial wastewater samples were collected and subjected to the studied chemical coagulation using the previously determined optimum doses of lime and FeCl<sub>3</sub>, followed by electrocoagulation in a continuous treatment system. The results are given in Table 9.

**Table 9** | Chemical coagulation and electrocoagulation of real industrial wastewater in continuous process system

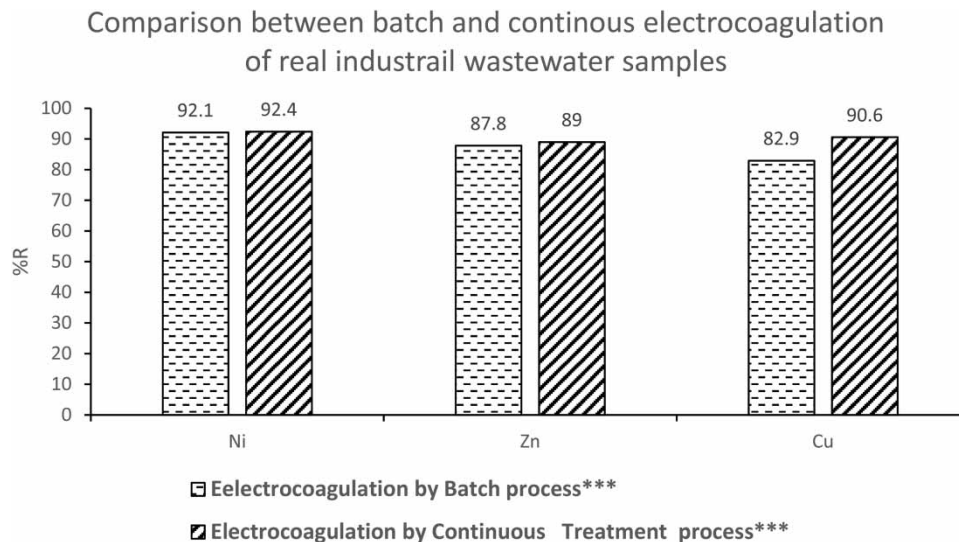
Wastewater	Ni		Zn		Cu	
	conc.	%R	conc.	%R	conc.	%R
Raw <sup>a</sup>	150	–	25	–	30	–
After chemical coagulation <sup>b</sup>	20.01	86.66	5.0	80.0	1.17	96.1
Eelectrocoagulation by Batch process <sup>c</sup>	1.58	92.1	0.61	87.8	0.20	82.9
Electrocoagulation by Continuous Treatment process <sup>c</sup>	0.22	98.9%	0.13	97.4%	0.04	96.6%

<sup>a</sup>Raw = real raw electroplating wastewater.

<sup>b</sup>Initial metal concentration after chemical coagulation using Lime (CaO) + FeCl<sub>3</sub> at pH 8.5.

<sup>c</sup>After electro and chemical-coagulation.

These results showed that the removal efficiency was 98.9%, 97.4% and 96.6% for Ni, Zn, and Cu respectively in the continuous treatment process. Correlation between the efficiency of the batch and the continuous treatment process is illustrated in Figure 3 indicating that the latter is more efficient than the former.



**Figure 3** | Comparison between batch and continuous electrocoagulation process for the removal of heavy metals from real electro-plating wastewater sample using stainless-steel electrodes at the pre-determined optimum conditions.

## CONCLUSIONS AND RECOMMENDATIONS

The application of the electro-coagulation process proved to be an efficient treatment technology for the removal of metals from electroplating wastewater. Combination of lime and ferric chloride was more efficient for the precipitation of heavy metals from industrial electroplating wastewater than other chemical coagulants. The use of stainless steel electrode in the presence of ferric chloride is more efficient than using iron electrode in the electro-coagulation process, due to the fact that the later electrode decays much faster than the former one. The treated electroplating wastewater can be reused again in the rinsing process of industrial electroplating industry instead of disposal. It is recommended to consider the power of solar energy instead of electricity in this electro-coagulation to reduce the cost of operation.

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## DATA AVAILABILITY STATEMENT

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

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