Environ Sci Pollut Res (2014) 21:14166–14173
DOI 10.1007/s11356-014-3331-8

RESEARCH ARTICLE

Effect of operational parameters on heavy metal removal by electrocoagulation

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Received: 8 April 2014 / Accepted: 11 July 2014 / Published online: 25 July 2014

Abstract In the present paper, the performance of electrocoagulation (EC) for the treatability of mixed metals (chromium (Cr), copper (Cu), lead (Pb), nickel (Ni), and zinc (Zn)) from metal plating industrial wastewater (EPW) has been investigated. The study mainly focused on the affecting parameters of EC process, such as electrode material, initial pH, distance between electrodes, electrode size, and applied voltage. The pH 8 is observed to be the best for metal removal. Fe–Fe electrode pair with 1-cm inter-electrode distance and electrode surface area of 40 cm^2 at an applied voltage of 8 V is observed to more efficient in the metal removal. Experiments have shown that the maximum removal percentage of the metals like Cr, Ni, Zn, Cu, and Pb are reported to be 96.2, 96.4, 99.9, 98, and 99.5 %, respectively, at a reaction time of 30 min. Under optimum conditions, the energy consumption is observed to be 51.40 kWh/m^3. The method is observed to be very effective in the removal of metals from electroplating effluent.

Keywords Electroplating · Heavy metal · Electrocoagulation · Metal hydroxide

Introduction

Electroplating industrial wastewaters (EPW) contain various kinds of toxic substances such as alkaline cleaning agents, degreasing solvents, and metals. Among these, heavy metals are very toxic and generally appear in electroplating industrial effluents. Most of the metals such as copper, nickel, chromium, lead, and zinc are harmful when discharged into sewages or rivers, which is a major environmental issue when disposed of without a proper treatment (Ashok and Sharma 2013 and Bhagawan et al. 2014). Hence, these metals are of considerable concern, because they are non-biodegradable, highly toxic, and potentially carcinogenic. To meet environmental regulations, this metal containing wastewater must be treated before its discharge. Various conventional metal removal techniques include precipitation, adsorption, solvent extraction, bio-sorption, membrane separation, ion exchange, and reverse osmosis (Akbal 2013; Nafaa Adhoum et al. 2004; Golder et al. 2007a, b; Arroyo et al. 2009); precipitation is considered to be the most economical and applicable among all the techniques. It is based on chemical coagulation where lime is added to raise the pH, and metal salt is used to remove colloidal matter as gelatinous hydroxides. Activated silica or polyelectrolytes may also be added to stimulate coagulation. The former treatment may also be followed by adsorption onto activated carbon to remove metals at a ppm level. Although precipitation has proved to be quite effective in treating industrial effluents, the chemical coagulation may induce secondary pollution caused by added chemical substances. This drawback, together with the need for low-cost effective treatment, encouraged many researchers to use electrocoagulation for the treatment of several industrial effluents (Akbal 2013; Nafaa Adhoum et al. 2004; Dermentzis et al. 2011a, b; Ramakrishnan et al. 2013) such as petroleum refinery wastewater (Ichrak et al. 2013), textile wastewater (Anissa et al. 2009), winery wastewater (Visnja et al. 2013), biodiesel wastewater (Saeb et al. 2013), laundry wastewater (Chih and Kuo 2009), dairy effluent (Yusuf et al. 2011), and dye containing effluent (Fatiha et al. 2008). Recently, some studies have been reported in the literature on the use of

Responsible editor: Bingcai Pan

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electrocoagulation (EC) for the treatment of metals from electroplating rinse water (Meyyappan et al. 2012), chrome tanning effluent (Golder et al. 2011), Ni from aqueous solutions (YaoXing et al. 2013), copper removal from synthetic sample (Subramanyan et al. 2013), cobalt from aqueous solution (Ashraf et al. 2011), Zn(II), Cu(II), Ni(II), Ag(I) Cr(VI) from aqueous solutions (Ilona and Wolfgang 2008), Cd from simulated wastewater (Abbas et al. 2014), and Cr from wastewaters (Toktam et al. 2014).

Electrocoagulation is a process consisting of creating metallic hydroxide floc within the solution by electrodissolution of soluble anodes, usually made of iron or aluminum. The generation of metallic cations takes place at the anode, due to the electrochemical oxidation of the electrode, whereas H_2 gas is typically produced at the cathode (Arroyo et al. 2009). The overall reactions are as follows:

At anode:

$$\mathrm{Fe} \rightarrow \mathrm{Fe}^{2+} + 2\mathrm{e}^- \tag{1}$$

$$\mathrm{A1} \rightarrow \mathrm{A1}^{2+} + 3\mathrm{e}^- \tag{2}$$

$$\mathrm{Zn} \rightarrow \mathrm{Zn}^{2+} + 2\mathrm{e}^- \tag{3}$$

$$\mathrm{Cr_2O_7}^{2-} + 6e^- + 7\mathrm{H_2O} \rightarrow 2\mathrm{Cr}^{3+} + 14\mathrm{OH}^- \tag{4}$$

$$\mathrm{Cr6}^+ + 3\,\mathrm{Fe}^{2+} \rightarrow \mathrm{Cr}^{3+} + 3\,\mathrm{Fe}^{3+} \tag{5}$$

$$\mathrm{Cu} \rightarrow \mathrm{Cu}^{2+} + 2\mathrm{e}^- \tag{6}$$

$$\mathrm{Ni} \rightarrow \mathrm{Ni}^{2+} + 2\mathrm{e}^- \tag{7}$$

These above metals finally form as metal hydroxides as follows:

$$\mathrm{Me}^{n+} + n\mathrm{OH}^- \rightarrow \mathrm{Me(OH)}_{n(\mathrm{s})} \tag{8}$$

At cathode:

$$2\mathrm{H_2O} + 2\mathrm{e}^- \rightarrow \mathrm{H_2(g)} + 2\mathrm{OH}^- \tag{9}$$

EC process involves many chemical and physical phenomena, such as discharge, anodic oxidation, cathodic reduction, coagulation, electrophoretic migration, and adsorption (Cheng 2006). Unlike chemical metal precipitation, in an electrocoagulation process, liquid is not enriched with anions and, thus, the salt content does not increase. This contributes to the production of more compact metallic sludges during electrocoagulation than those generated by chemical precipitation (Meunier et al. 2006). Other advantages include small footprint, shorter reaction time, low cost of equipment and operation, and easy operation (Arash et al. 2011).

The metal hydroxide floc formed either from Al or Fe electrode normally acts 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 Cr, Cu, Pb, Ni, and Zn in the form of their corresponding hydroxides (reaction 8).

The performance of an electrotreatment process using iron (Fe) and/or aluminum (Al) as the electrode material strongly depends on the pH of the solution, which varies continuously during the process. Minimum solubility of heavy metals such as copper, zinc, lead, nickel, and chromium, etc. occurs at a particular pH, and when the pH of the solution changes, metal hydroxide precipitate/sludge tends to be resolubilized (Golder et al. 2007a, b and 2009). Vasudevan and Dermentzis also described the decrease in metal removal efficiency at strong acidic and basic conditions. It ascribed to an amphoteric behavior of $\mathrm{Fe(OH)_3}$ which leads to soluble cations Fe^{3+}, $\mathrm{Fe(OH)}^{2+}$, $\mathrm{Fe(OH)_2}^{+}$, and Al^{3+} and to monomeric anions $\mathrm{Fe(OH)_4}^{-}$, $\mathrm{Fe(OH)_6}^{3-}$, and $\mathrm{Al(OH)_4}^{-}$ (Vasudevan et al. 2009; Dermentzis et al. 2011a, b; Akbal 2011a, b). Best removal capacities for all metals have been reported at a pH range of 4–8 (Dermentzis et al. 2011a, b).

In the present work, the efficiency of electrocoagulation in removing copper, nickel, chromium, lead, and zinc from EPW is reported. The effect of electrode material, initial pH, distance between electrodes, electrode size, and applied voltage on the removal efficiency is explored and discussed to determine the optimum operational conditions. The operating cost of treatment is also investigated, by considering the cost of consumption of electric power and electrode.

Material and methods

Electroplating wastewater

Electroplating wastewater was collected from an electroplating industry in Hyderabad, India. The initial characterization of the sample (one) has been given in Table 1.

Methodology

The reactor used in this study is given in Fig. 1. Batch mode experiments were carried in a 250-ml beaker with the working volume of 200 ml at room temperature (27 °C). The two electrode materials (Fe and Al) were tested as anode and cathode with dimensions of 100 mm×50 mm×2 mm. The pH of the EPW varied from 4–8 (4, 6, and 8), effective reactive surface area studied between 10–40 cm^2 (10, 20, and 40 cm^2), distance between electrodes studied between

Table 1 Initial characteristics of the waste water sample

S. no	Parameter	Unit	Concentration before treatment	Concentration after treatment	CPCB limits
1	pH	–	<1	9	6.0–9.0
2	Electrical conductivity	ms/cm	79.2	44.8	NA
3	Total solids	mg/L	30,070	24,200	NA
4	Total dissolved solids	mg/L	28,100	22,000	NA
5	Chromium	mg/L	39	1.5	2.0
6	Nickel	mg/L	13.5	0.5	3.0
7	Zinc	mg/L	2,356	0.08	5.0
8	Copper	mg/L	4	0.08	3.0
9	Lead	mg/L	5.5	0.03	0.1
11	Chlorides	mg/L	53,175	19,852	NA
12	Oil and grease	mg/L	4.5	0.79	10
13	Sulfates	mg/L	138	31	400
14	Phosphates	mg/L	1.6	0.2	5

NA not applicable

CPCB limits (2012)

1–4 cm (1, 2, and 4 cm), and effect of applied voltage studied at 4 and 8 V, respectively. The adjustment of pH is made with 0.1 N/1.0 N solution of HCl or 0.1 N/1.0 N NaOH. Working electrodes are connected to a DC power supply (APLAB regulated DC power supply L6403) unit with 0 to 84-V voltage supply capacity. The samples are collected at 10-minute (min) time intervals for analyzing residual metal content.

Metal content is determined after acidification with HCl to a pH of less than 2.0 to avoid further removal during the post-EC period. The concentrations of metals are measured using an atomic absorption spectrophotometer.

Operation cost evaluation (OC)

One of the most important parameter affecting the application of any method of water and treatment is the operating cost, which heavily determines cost of the treatment process. The OC of the EC is calculated by including the material cost (mainly electrodes), utility cost (mainly electrical energy), and chemicals' fixed costs (Abdurrahman et al. 2013; Kobya et al. 2010).

$$\mathrm{OC} = \mathrm{a}C\,\mathrm{energy} + \mathrm{b}C\,\mathrm{electrode} + \mathrm{c}C\,\mathrm{chemicals} \qquad \text{(i)}$$

where "*a*" is the electricity consumed (kWh/m^3, formulae ii), "*b*" is the electrode material consumed (kg/m^3), and "*c*" chemicals are consumption quantities of chemicals (kg/m^3, formulae iii) of the wastewater treated. The cost values ($) of *a*, *b*, and *c* are calculated according to the Indian market (Mar. 2014). It is the energy price as 0.1$/kWh (6.40 Rs/kWh); electrode price as Fe 1.61$/kg, Al 3.27$/kg (97 and 197 Rs/kg); and chemical cost (NaOH) as 8.17$/kg (492 Rs/kg).

The electrode and energy consumption were calculated using the following equations.

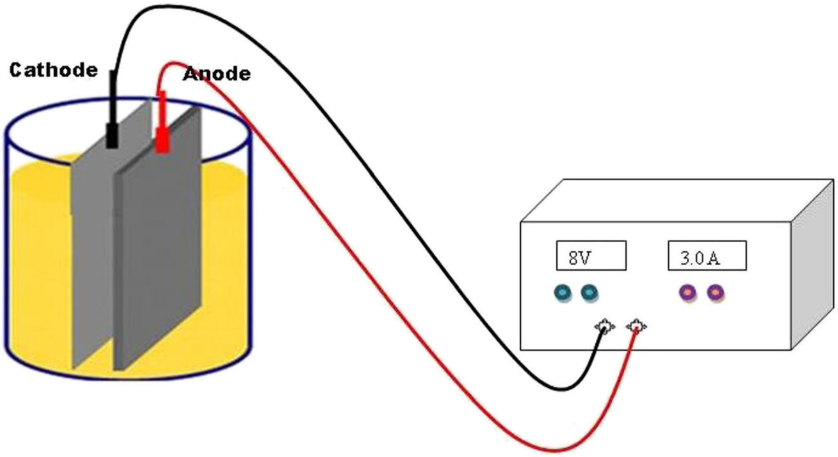


Fig. 1 Schematic view of the experiment

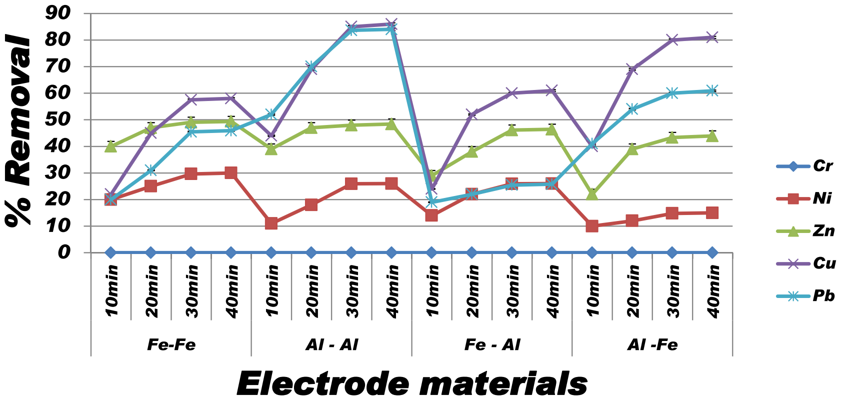


Fig. 2 Effect of reaction time on metal removing efficiency using different electrode materials. Conditions: electrode material Fe and Al, reaction time 10, 20, 30, and 40 min, volume of the sample 200 ml, pH <1, applied voltage 8 V, reaction surface area 40 cm^2, and inter-electrode distance 4 cm

$$C\,\text{energy} = \frac{\text{UIt}_{\text{EC}}}{\text{V}} \tag{ii}$$

$$C\,\text{electrode} = \frac{It_{\text{EC}}M}{ZFV} \tag{iii}$$

where U is cell voltage (V), I is the current (A), t_{EC} is the operating time (h), V volume of the sample (m^3), M is the molecular weight of electrode (for Fe 55.84 g/mol, Al 26.98 g/mol), Z is number of electrons transferred (Z=3 for Al and 2 for Fe), and F is the Faraday constant (96487 C/mol).

Results and discussion

Effect of electrode material on metal removal

The reaction time for EPW treatment is investigated with four electrode (anode–cathode) combinations (Fe–Fe, Al–Al, Fe–Al, and Al–Fe). The maximum metal removal was observed (Fig. 2) to be 30 and 49.4 % of Ni and Zn, respectively, with Fe–Fe electrode combination and 86 and 84 % of Cu and Pb with Al–Al electrode combination after 40 min. These results revealed that at the beginning of the process, i.e., within 30 min, metal removal rate was observed to be faster. After 30 min, it decreased gradually which might be due to inefficient dosage of the coagulant. Similar results were also observed by Bazafshan et al. (2006) and Sepideh et al. (2013). Therefore, to avoid excess operational cost, the reaction time was optimized to be 30 min.

Effect of initial pH on metal removal

The initial pH of the electrolyte (EPW) has a considerable effect on the efficiency of the electrocoagulation process. In the present study, it was observed (Fig. 3) that metal removal efficiency increased with the increase in pH and the maximum removal has been observed at pH 8. At this pH, the removal % of Ni, Zn, Cu, and Pb was 96.4, 98.9, 97.5, and 98.2 %, respectively, when using iron electrodes, whereas with aluminum electrodes, it was reported to be 93.9, 93.2, 95, and 92.7 %, respectively. The reason for this behavior might be due to the fact that adsorption capacity of ferric oxides is much higher than aluminum oxides (Akbal 2011a, b; Denial et al. 2007). The Cr removal % was similar (81 %) with both iron and aluminum electrodes. Hence, further experiments were conducted with iron electrodes.

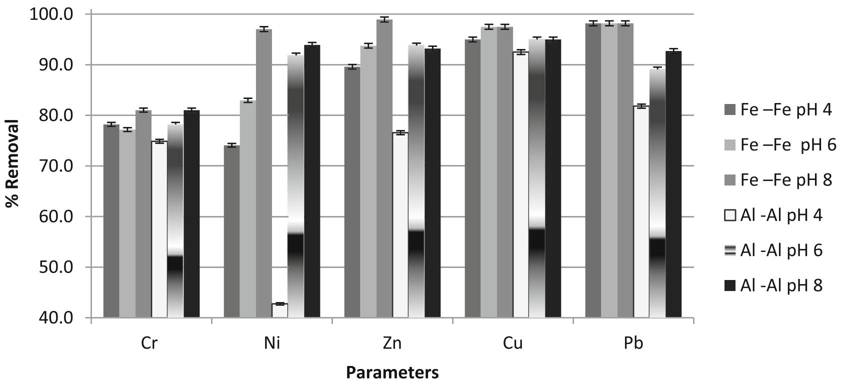


Fig. 3 Effect of pH on metal removal efficiency using various electrode materials. Conditions: electrode material Fe and Al, reaction time 30 min, volume of the sample 200 ml, pH 4, 6, and 8, applied voltage 8 V, reaction surface area 40 cm^2, and inter-electrode distance 4 cm

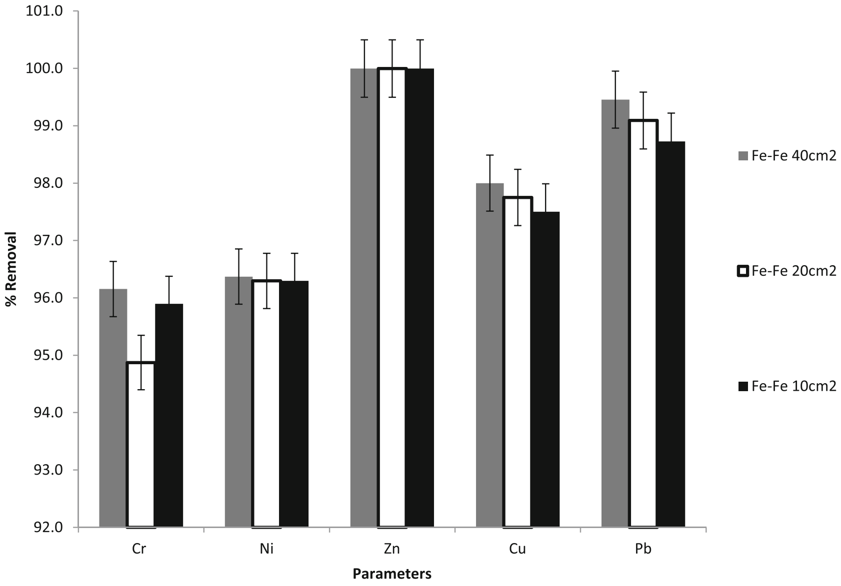


Fig. 4 Effect of reaction (electrode) area on metal removal efficiency. Conditions: electrode material Fe, reaction time 40 min, volume of the sample 200 ml, pH 8, applied voltage 8 V, reaction surface area 10, 20, and 40 cm^2, and inter-electrode distance 4 cm

Effect of reaction area on metal removal

This parameter has been varied as 10, 20, 30, and 40 cm^2 to minimize reactive surface area and electrode consumption for the EPW treatment. The metal removal efficiency increased with an increase in electrode surface area from 10 to 40 cm^2. The maximum % removal efficiency of Cr, Ni, Zn, Cu, and Pb was observed (Fig. 4) to be 96.2, 96.4, 99.9, 98, and 99.5 %, respectively. This might be attributed to a greater electrode area that produced larger amounts of anions and cations from the electrodes. The larger the electrode's surface, it increased the rate of floc's formation, which in turn influenced the removal efficiency (Ashok and Sharma 2013).

Effect of inter-electrode distance on metal removal

The inter-electrode distance was studied as a parameter to minimize electricity consumption for the treatment of EPW. Inter-electrode distance was varied from 1 to 4 cm. The removal percentage of metals was raised with a decrease in inter-electrode distance from 4 to 1 cm (Fig. 5), whereby it exhibited the maximum removal of Cr (96 %), Ni (96.4 %), Zn (99.9 %), Cu (98 %), and Pb (99.5 %) at the shortest distance (1 cm) between the electrodes with an electrode area of 40 cm^2. Similar observations have also been reported by Ashok and Sharma (2013) and Ghosh et al. (2008). That removal efficiency increase might be due to the faster anion discharge at the anode and improved oxidation. It also reduces

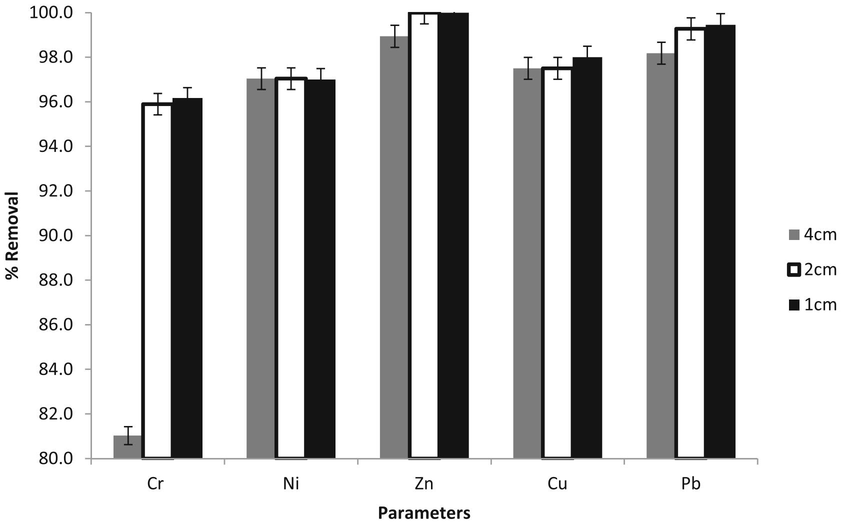
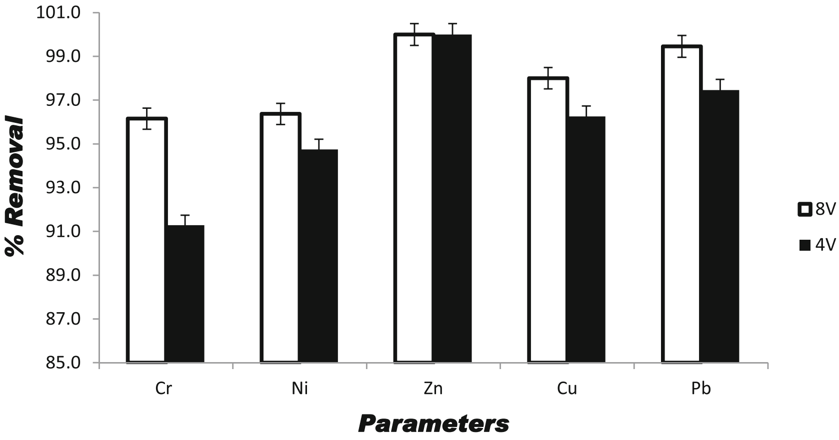


Fig. 5 Effect of inter-electrode distance on metal removal efficiency. Conditions: electrode material Fe, reaction time 40 min, volume of the sample 200 ml, pH 8, applied voltage 8 V, reaction surface area 40 cm^2, and inter-electrode distances 1, 2, and 4 cm

Fig. 6 Effect of applied voltage on metal removal efficiency. Conditions: electrode material Fe, reaction time 40 min, volume of the sample 200 ml, pH 8, applied voltages 4 and 8 V, reaction surface area 40 cm^2, and inter-electrode distance 1 cm



resistance, the electricity consumption, and, consequently, the cost of the wastewater treatment (Arash et al. 2011; Mohd and Wahid 2012).

Effect of applied voltage on metal removal

Applied voltage is one of the operating parameters directly affecting the performance and the operating cost. EC experiments were carried at 4 and 8 V (Fig. 6). At 8 V, the maximum removal of metals was found to be 96, 96.4, 99.9, 98, and 99.5 % for Cr, Ni, Zn, Cu, and Pb, respectively. The concentration of the metals reached the dischargeable limits of Central Pollution Control Board (CPCB) for wastewater. With reducing voltage, the metal removal decreased. This might be attributed to decrease in the voltage directly decreases both the coagulant dose and bubble generation rate as well as influences both mixing of solution and mass transfer at the electrodes (Golder et al. 2007a, b). Akbal (2011a, b) also observed the same order of removal while treating the metal plating wastewater (Cu, Cr, and Ni).

Table 2 Cost estimation of the treatment process

Process	kWh/m^3	Kg/m^3	NaOH (Kg/m^3)	Total ($/$m^3$)
pH optimization				
Fe–Fe pH4	44.60	1.61	8.5	76.5
Fe–Fe pH6	50.00	1.81	10	89.7
Fe–Fe pH8	54.40	1.97	12	106.7
Al–Al pH4	46.20	0.54	8.5	75.8
Al–Al pH6	49.00	0.57	10	88.5
Al–Al pH8	50.60	0.59	12	105.0
Surface area optimization				
Fe–Fe 40 cm^2	54.40	1.97	12	106.7
Fe–Fe 20 cm^2	52.00	1.88	12	106.3
Fe–Fe 10 cm^2	50.00	1.81	12	106.0
Electrode distance optimization				
Fe–Fe 4 cm	54.40	1.97	12	106.7
Fe–Fe 2 cm	52.60	1.90	12	106.4
Fe–Fe 1 cm	51.40	1.86	12	106.2
Voltage optimization				
Fe–Fe 8 V	51.40	1.86	12	106.2
Fe–Fe 4 V	14.00	1.01	12	101.1

Relationship between heavy metal removal % and changes in conductivity

Conductivity in water is affected by the presence of inorganic dissolved solids. In the present study, conductivity was observed to decrease proportionally with increase in the % removal of heavy metals, which is similar to Kim et al.'s (2013) observations. However, at a certain stage, the conductivity remained constant without an increase or decrease, which might be due to the presence of chloride ions.

Energy consumption and cost analysis of metal removal

The cost estimation for the treatment process is given in Table 2. From the table, it is observed that with an increase in initial pH of the solution, reaction area, inter-electrode distance, and applied voltage, the cost of the process increased gradually. At optimized conditions (electrode material Fe–Fe, pH 8, area of the electrode 40 cm^2, inter-electrode distance 1 cm, and applied voltage 8 V), the energy and electrode consumption is observed to be 51.40 kWh/m^3 and 1.86 kg/m^3, respectively.

Conclusions

- This study concluded that EC is efficient and effective for the removal of heavy metals from metal plating wastewaters.
- The maximum removal of metal is observed at a reaction time of 30 min, initial pH of 8, electrode material as Fe–

Fe, electrode reaction area of 40 cm^2, applied voltage of 8 V, and electrode spacing of 1 cm.

- The maximum dischargeable values of pollutants given by the official CPCB norms have been met in this study.
- Iron electrodes are found to be most ideal electrodes, compared with aluminum and hybrid Al/Fe electrodes for the treatment of metal plating wastewater.
- Metal removal rate increased with increasing the current applied voltage and electrode surface area.
- Metal removal rate decreased with increasing inter-electrode distance.
- Under optimum treatment conditions, metal removal % was found to be 96.2, 96.4, 99.9, 98, and 99.5 % for Cr, Ni, Zn, Cu, and Pb, respectively.

Acknowledgments The authors would like to thank Environmental Solutions Pvt. Ltd., Hyderabad, India, for providing experimental setup and electroplating industrial effluent.

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