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Chromium Removal from Industrial Effluent by Electrocoagulation: Operating Cost and Kinetics Study

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Abstract

This article reports the removal of chromium by electrocoagulation (EC) from metal complex dye (MCD) industrial effluent in a batch reactor using iron electrodes. The paper evaluates the application of EC process in assessing the impact of process parameters such as current density (CD), electrode distance (ED), pH, and supporting electrolyte (NaCl) concentration (Cs) for efficient removal of chromium from MCD effluent. The EC process showed, removal percentage 99.64 %, at the initial pH of 5.83, CD of 89.45 A/m^2, ED of 0.7 cm, and operating time of 50 minutes, are the optimal operating parameters for the treatment of MCD effluent. It was also noticed that the removal of chromium is appreciably enhanced for acidic pH values. It was observed that the removal of chromium follows the 1st order reaction by kinetic analysis of EC method. As CD raised from 17.89 to 89.45 A/m^2, the rate constant (k) was enhanced from 0.013 to 0.109 min^{-1}, as a result of the presence of more quantity of iron flocs throughout EC. Analysis of sludge was carried out using FESEM- EDX, which confirmed the presence of chromium, and iron hydroxide in sludge. The present work confirms that EC is an efficient process for chromium removal from MCD effluent with a calculated operating cost of 0.207 US$/m^3 and energy consumption of 2.499 kWh/m^3.

Keywords: Electrocoagulation, Iron electrode, Metal complex dye (MCD) effluent, Chromium removal, Kinetic study

1 Introduction

Industrial wastewaters such as Leather Company, metallurgical industry, electroplating, pigments, mining, paints etc. contain chromium ions [1]. Chromium (Cr) is present in two forms trivalent Cr(III) and hexavalent Cr(VI) in the effluent. Cr(VI) is present in the effluent in various forms like, ($HCrO^{4-}$),(CrO_4^{2-}) and ($Cr_2O_7^{2-}$) under typical pH [2]. In general, Cr(VI) has higher solubility as compared to Cr(III). Cr(VI) is very harmful to human life and the environment [3]. A release limit value of 0.05 mg/L is set by national environment protection act for chromium [4]. So before discharge into any water body, it should be treated to secure the environment [1]. The removal of chromium from industrial effluent is a big issue for the control of pollution [5]. For the treatment of chromium, many techniques are available and each technique has individual benefits and drawbacks, for example, the biological process is eco friendly [6] but it takes a long time for treatment, a large area required, and having less efficient [7], adsorption [8], an advantage of this process is less treatment time required and high efficient however high adsorbent required and efficiency depends on the type of adsorbent [9], ion exchange [10], membrane separation [11], photocatalysis [12] these methods are efficient but high cost required [13]. Precipitation and coagulation technique are mostly used due to their easy operation and less costly equipment needed. But these techniques require a large quantity of chemicals and more amount of generated sludge is its major problem [14]. Electrocoagulation (EC) is a superior technique because of its various favorable conditions, for example, no extra chemical necessity, simpler operation and system, economical, high efficiency, less sludge generation, color, and smell reduction, and lesser time required [15]. It was found that the various investigators have worked on artificial wastewater and few works of literature are available for real wastewater treatment. The chromium removal from different types of wastewater by electrocoagulation is given in Table 1. It was found that very few researchers have reported the cost estimation and kinetic analysis of the EC process. Both parameters need to be calculated before the industrial application of EC.

The present work aims to study the removal of chromium present in MCD effluent through EC techniques and optimization of variables such as pH of the effluent, current density (CD), the concentration of NaCl (Cs), the distance between electrodes (ED), and energy consumption (kWh m^{-3}). A kinetic study has been carried out to check the effect of different process variables such as current density and electrode distance on chromium removal.

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Table 1: EC applied for removal of chromium from industrial effluent

Industrial effluent	Current or Voltage, Current density	Types of electrode	pH	Operating time	Percentage of removal	Ref.
Leachate	2.5 V	SS-SS	7	60 min	88.35%	[16]
Groundwater samples	7.94 mA/cm^2	Fe-Fe	8	30 min	100%	[17]
Electroplating wastewater	20 mA/cm^2	Fe-Fe	2	30 min	99.85%	[18]
Leather finishing industrial	68 mA/cm^2	Fe-Fe	6.5	60 min	98.6%	[19]
Metal finishing industry	6.9 A	S.S.	4	90 min	83.5%	[20]
Industrial wastewater	200 A/m^2	Fe and Al	3	60 min	100 %	[21]
Tannery wastewater	0.81 A	Al-Fe	8	60 min	90%	[22]

In addition, energy consumption and operating cost are calculated with respect to operating conditions. Elementals analysis of sludge produced by EC has been carried out.

2 Theory

Electrocoagulation (EC) is a process that applies a current across electrodes through a liquid, this result in the dissolution of the anode. These ions then form hydroxides which complex with absorb contaminants and precipitate from wastewater. For chromium removal, solution reactions at anode and cathode with Fe electrode is given as follows [5,23,24]:

Mechanism 1

Anode: $4Fe_{(s)} \rightarrow 4Fe_{(aq)}^{2+} + 8e-$ (1)

Bulk of solution: $4Fe_{(aq)}^{2+} + 10H_2O_{(l)} + O_{2(aq)} \rightarrow 4Fe(OH)_{3(s)} + 8H^+_{(aq)}$ (2)

Cathode: $8H^+_{(aq)} + 8e- \rightarrow 4H_{2(g)}$ (3)

Overall: $4Fe_{(s)} + 10H_2O_{(l)} + O_{2(aq)} \rightarrow 4Fe(OH)_{3(s)} + 4H_{2(g)}$ (4)

Mechanism 2

Anode: $Fe_{(s)} \rightarrow Fe^{2+}_{(aq)} + 2e-$ (5)

Bulk of solution: $Fe_{(aq)}^{2+} + OH^-_{(aq)} \rightarrow Fe(OH)_{2(s)}$ (6)

Cathode: $2H_2O_{(l)} + 2e- \rightarrow H_{2(g)} + 2OH^-_{(aq)}$ (7)

Overall: $Fe_{(s)} + 2H_2O_{(l)} \rightarrow Fe(OH)_{2(s)} + H_{2(g)}$ (8)

The $Fe(OH)_{n(s)}$ produced in the solution as a gelatinous suspension, can remove the impurities from effluent by coagulation. The Fe^{2+} and OH^- react, and formed various monomeric and polymeric hydroxides. These hydroxides are acted as coagulants [23]. The removal of chromium can be given as a 1st order kinetic model by Eq. (9) [25,26].

$$-\ln\left(\frac{C_0}{C}\right) = kt \quad (9)$$

where, k denotes rate constant (min.$^{-1}$), C denotes chromium concentration in effluent, at any time 't' C_0 denotes initial chromium concentration.

3 Materials and methods

3.1 Effluent and its characterization

The metal complex dye industry effluent containing chromium used in this study was collected from GIDC Ankleshwar, Gujarat India. The wastewater was characterized for Cr, pH, COD, BOD, TDS, TS, and color. The real and treated wastewater resulted in the current work is shown in Table 2.

Table 2: Characteristic composition of effluent without and with treatment by EC at optimum condition (CD = 89.45 A/m^2, electrode distance = 0.7 cm.).

Name of parameters	**Effluent without treated**	**Effluent with treated at optimum condition**	**% of removal**
Chromium mg/L	82.7	0.291	99.64
COD mg/L	3642.86	618.18	83.03
BOD mg/L	1638.17	304.97	81.38
pH	5.83	7.71	-
TDS mg/L	14984	4491.6	70.02
TS mg/L	20252	7052	65.17
Color	Dark Brown	Light Brown (transparent)	

3.2 Electrocoagulation (EC) setup and procedure

All experiments were carried out in a 1-liter beaker filled with 500 ml of effluent. Iron plate (15 cm × 4.3 cm × 0.1 cm) was used as an electrode with an active surface area of 55.9 cm^2 (0.00559 m^2). The electrodes were attached with a DC power supply (Aplab, India, Model: L-1285) and worked on either steady voltage or constant current mode. The schematic design of the experimental setup is shown in Figure 1. The effluent was continuously stirred throughout the experiment. pH was varied by using 1N H_2SO_4 and NaOH. The conductivity of MCD solution was varied by using NaCl. Details of various process parameters studied for the removal of chromium are presented in Table 3.

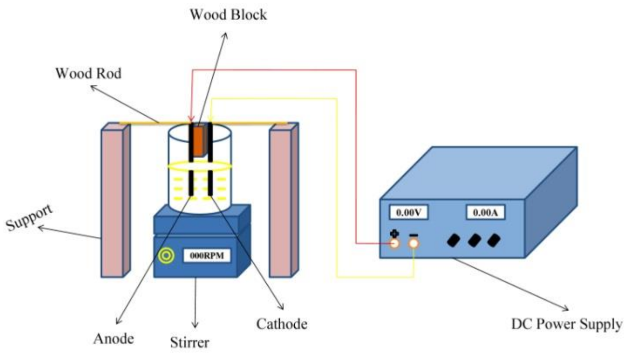


Figure 1: Electrocoagulation setup

Table 3: Various process parameters studied for EC

Variable studied	Variation in variable	Fix variable
pH	3, 5, 7, 9, 11	Cr concentration= 82.7 mg/L, Current= 0.5 A, ED= 0.7 cm,
Current (A)	0.1, 0.2, 0.3, 0.4, 0.5	Cr concentration= 82.7 mg/L, A, ED= 0.7cm. pH= 5.83
Electrolyte (NaCl) (g/l)	0.25, 0.50, 0.75, 1	Cr concentration= 82.7 mg/L, Current= 0.5A, pH= 5.83, ED= 0.7 cm.
Electrode distance (ED) (cm)	0.7, 1.4, 2.1, 2.8, 3.5	Cr concentration= 82.7 mg/L, Current= 0.5 A, pH= 5.83
Time of EC (minute)	50	

All the experiments were carried out at room temperature (~25 ^{O}C) for 50 minutes. The samples were taken in 10 minutes of EC interval and filtered by filter paper. All experiments were repeated two times and the average experimental error was approximately 4%. The chromium concentration was measured using Atomic Absorption Spectrometer (Chemito Instruments Pvt. Ltd. - AA201), and the chromium removal percentage was determined by the following equation:

$$\text{Chromium removal \%} = \frac{C_0 - C_t}{C_0} \times 100 \tag{10}$$

where, C_0 denotes initial chromium concentration (mg/L), C_t denotes chromium concentration at any time *t*. The specific electrical energy consumption (SEEC) in kWh/ (kg Fe) was calculated by Eqs (11-13) [27].

$$\varphi = \frac{\Delta M_{experimental}}{\Delta M_{theoritical}} \times 100 \tag{11}$$

$$\Delta M_{theoritical} = \frac{M.I.t_{EC}}{n.F} \tag{12}$$

$$SEEC = \frac{n \times F \times U}{3600 \times M \times \varphi} \tag{13}$$

where F is Faraday constant (F = 96487 C mol^{-1}), I is current (A), U is voltage (volt), M is molecular weight of iron (g mol^{-1}), φ is current efficiency, n is moles of electron, and tEC is the time of EC (minute). Energy consumption per unit volume (kWh m^{-3}) for various process variables was calculated by following Eq (14) [28]:

$$Energy\ consumption\ (E) = \frac{U\ I\ t_{EC}}{V} \tag{14}$$

where E denotes energy consumption (kWh/m^3), U indicates voltage (V), I denotes current (A), V indicates of effluent (m^3), and t_{EC} indicates the total time of EC (h).

4 Results and Discussion

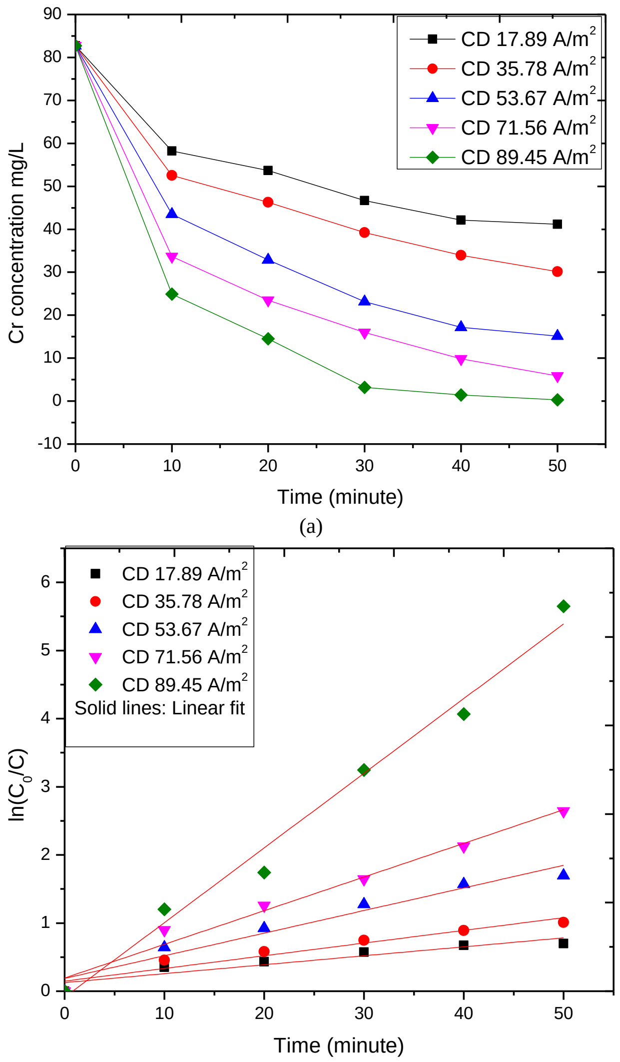
4.1 Effect of current density on chromium removal

The removal percentage depends on electrolysis time and current. These are key parameters that influence the removal percentage and the reaction rate of EC process. Inline to study the outcome of current density on chromium removal efficiency, experiments were performed at current density range from 17.89 A/m^2 to 89.45 A/m^2 and initial chromium concentrations 82.7 mg/l. Figure 2a demonstrates that the removal percentage enhanced with an increase in current density. The current density increased from 17.89 to 89.45 A/m^2 the removal percentage increased from 50.24 to 99.64 %. Current density 17.89, 35.78, 53.67, 71.56 and 89.45 A/m^2, the chromium removal efficiency of 50.24%, 63.55%, 81.69%, 92.90% and 99.64% respectively were achieved in 50 minute. As the current density is enhanced, the removal percentage of chromium increases. This is as per Faraday's law (Eq. (15)), it provides a relationship between CD and electrode (anode) that dissolves during the EC process [29].

$$m = \frac{M\ (CD)t}{n\ F} \tag{15}$$

where m is theoretical weight loss of electrode per unit area, CD is current density, F is Faraday constant (F = 96487 C mol^{-1}), M is molecular weight of iron (g mol^{-1}), n is moles of electron. As the CD increases, the rate of Fe^{2+} ions production rises. It resulted to the formation of iron hydroxides. These hydroxides helps into the higher chromium removal [30]. A kinetic study was performed for the first-order reaction model available in the literature [26,31]. It was observed that the best fitting for 1st order model was presented in Eq 9. Figure 2b demonstrates the plot of ln (C_0/C) vs time for 1st order kinetic. The rate constant (k) computed for different current densities are 0.013, 0.018, 0.033, 0.049, and 0.109 min.$^{-1}$ for different current densities of 17.89, 35.78, 53.67, 71.56 and 89.45 A/m^2 respectively. Form figure 2b, it was concluded that the value of k increase with increasing CD. It was due to the availability of more Fe^{2+} ions. The rate equation (16) with various CD is given as:

$$k = 12.5 \times\ 10^{-4} \times CD\ \left(\frac{A}{m^2}\right) \qquad R^2 = 0.910 \tag{16}$$



(a)

(b)

Figure 2: (a) Variation of chromium concentration with different current densities. (b) 1st order equation plot for various current densities

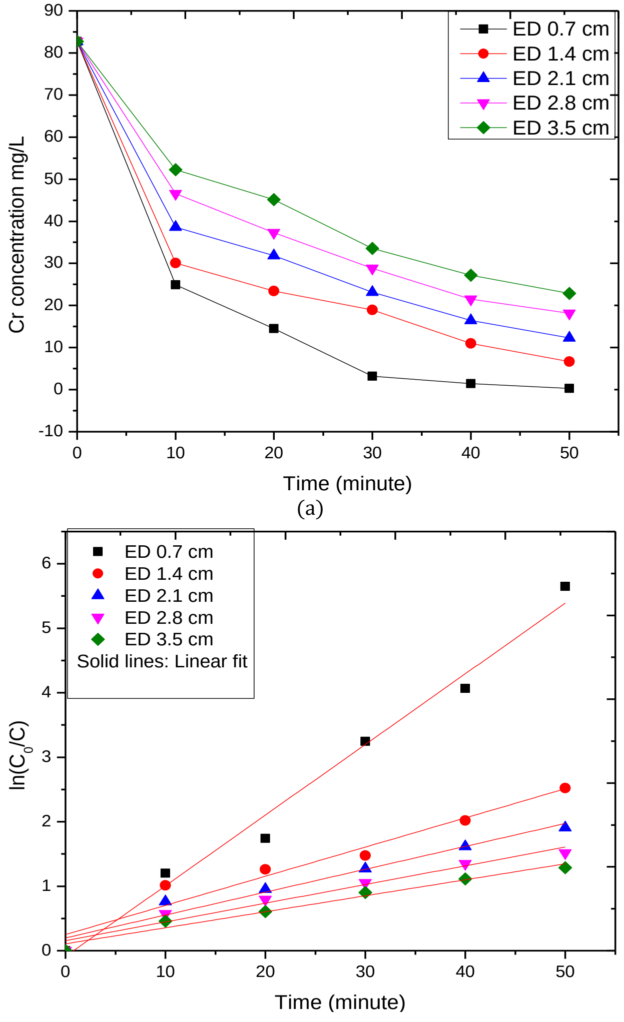
4.2 Effect of electrode distance on chromium removal and SEEC

Figure 3a shows the variation in chromium concentration at different electrode distance (ED). It was observed from figure 3a that the chromium removal percentage decreases with increasing ED. The chromium removal percentage reduced from 99.64 % to 72.37 %, as the ED increased from 0.7 to 3.5 cm. The corresponding residual concentration of chromium was raised from 0.291 to 22.84 mg/L due to a delay in the interactions between Fe^{2+} and OH- ions generated from the electrodes. Thus, as the rate of iron hydroxides production reduces, there is a decrease in removal percentages of chromium from the effluent [32]. Furthermore, with changes in ED from 0.7 to 3.5 cm, SEEC rises from 2.499 to 4.998 kWh/m^3 at constant current density i.e. 89.45 A/m^2. This is because increasing the ED from 0.7 to 3.5 cm causes voltage rise from 3.0 to 6.0 volt respectively with a corresponding increase in energy consumption [33]. Table 4 demonstrates the deviation of SEEC with different ED. The rate kinetics was performed for various ED and the plot of ln (C_0/C) versus time shown in Figure 3b. The rate constant (k) decreased from 0.109 to 0.024 min^{-1} as ED increased from 0.7 to 3.5cm. It was due to the lower rate of OH^- generation during the EC at higher ED. The rate constant (k) with ED is given as:

$$k = -26.5 \times 10^{-3} \times ED\ (cm) + 0.104 \qquad R^2 = 0.858 \qquad (17)$$

Table 4: Effect of ED on SEEC and chromium removal efficiency (CD 89.45 A/m^2, Cr concentration= 82.7 mg/L, pH 5.83, EC time 50 min)

ED in cm.	**SEEC**		**Removal efficiency %**
	kWh/ (kg Fe)	kWh/m^3	
0.7	3.57	2.499	99.64
1.4	4.29	2.9155	91.96
2.1	5.63	3.7485	85.17
2.8	6.41	4.165	78.03
3.5	7.81	4.998	72.37



(a)

(b)

Figure 3: (a) Variation of chromium concentration with different ED (b) 1st order equation plot for various ED

Figure 4a and figure 4b show the impact of ED on the SEEC and chromium removal percentage for various current densities of 53.67, 71.56, and 89.45 A/m^2. It can be seen from figure 4b that chromium removal decreases with increases in ED from 0.7 cm to 3.5 cm. For the ED value of 3.5 cm and the current density value of 53.67 A/m^2 minimum chromium removals of 63.87 % were observed. For the ED value of 0.7 cm and the current density value of 89.45 A/m^2 highest chromium removals of 99.64% were observed. The reason for this is discussed in previous sections. The same results have been reported in the literature [26,34]. It was seen that the SEEC increased significantly with an enhanced current density and ED. At a fix CD of 89.45 A/m^2, SEEC enhance from 3.57 to 7.81 kWh/ (kg Fe) when ED was increased from 0.7 cm to 3.5 cm. The reason was that increased ED, increases the resistance between the electrode plates, and hence the voltage drop increases. It resulted in rises in SEEC at higher ED for constant CD.

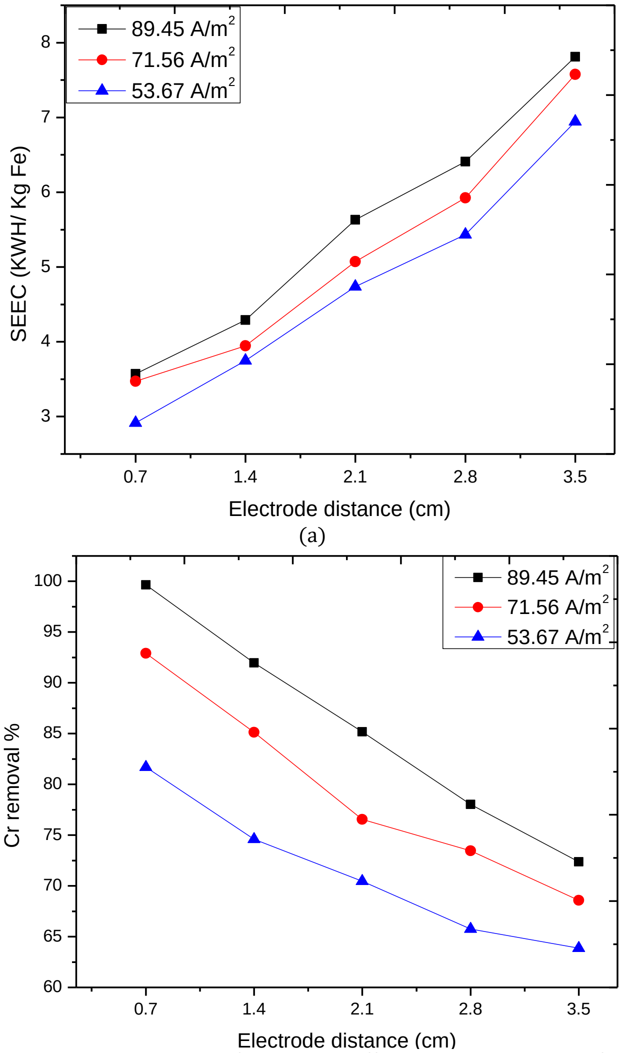


Figure 4: (a). Variation of SEEC with different ED. (b). Variation of chromium removal efficiency at different ED

4.3 Effect of NaCl concentration on chromium removal

In these experiments, NaCl was used as an electrolyte. The NaCl concentrations were varied from 0.25 g L^{-1} to 1.0 g L^{-1}. The impacts of NaCl concentration on chromium removal are shown in Figure 5. It can be seen that the removal percentage increases from 78.21 % to 86.99 % with increasing NaCl concentration from 0.25 g L^{-1} to 1.0 g L^{-1}. This result shows that at elevated NaCl concentrations, more chromium is removed. As the supporting electrolyte concentration increased, the voltage drop reduces at steady current and decreases the electrical energy in electrolytic cells. The overall conductivity of the solution increases with an increasing amount of NaCl and a reduction in residual concentration [23]. It was also concluded that the overall rate of removal percentage experienced a decrease. These results confirm that a rise in NaCl dose may not be helpful for chromium removal. It might be on the grounds that there were not adequate ions to carry out the current with a subsequent rise in power obstruction of the cell, thereby reducing removal percentage and the SEEC of the process. Literature also suggests that the addition of any supporting electrolyte leads to the addition of impurity in wastewater [3,23,34].

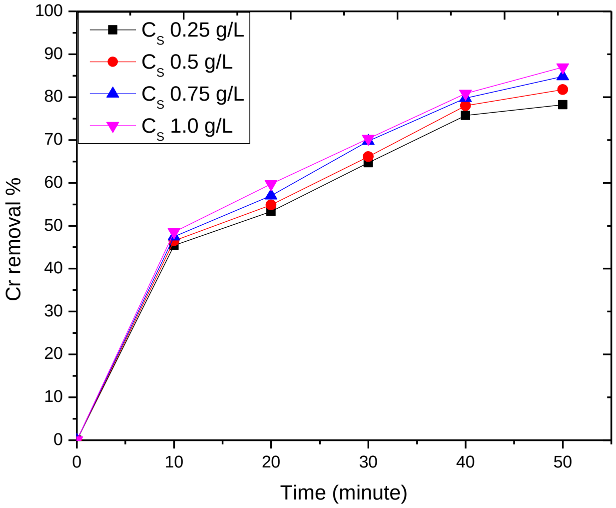


Figure 5: Effect of NaCl concentration on chromium removal

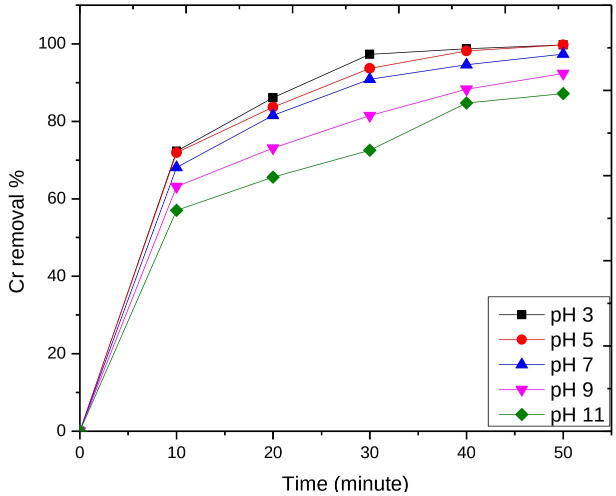


Figure 6: Effect of pH on chromium removal

4.4 Effect of pH concentration on chromium removal

For the electrocoagulation process, the initial pH is a significant operating variable to influence the performance of the process [35]. Figure 6 demonstrates the result of pH on chromium removal percentage. It was observed that the maximum chromium removal percentage 99.76% at pH 3, whereas the minimum chromium removal efficiency 87.16% at pH 11. From Figure 6, it is clear that the lower pH has more impact on chromium removal percentage. When the pH decreases, the chromium removal percentage increases with acidic conditions being appropriate for an increase in chromium removal rates while the alkali medium enhanced the rate of decrease in chromium removal. The same results have been reported in the literature [3,5,23,36]. From figure 6 it can be seen that the pH increased from 3 to 11 the chromium removal percentage decreased from 99.76 to 87.16 %. This is due to the high rate of Fe^{2+} production under acidic condition and this is confirmed by the simultaneous chemical dissolution of anode jointly with the electrochemical dissolution [21].

4.5 Assessment of operating cost at different current densities

The estimation of operating cost of EC technique for the removal of chromium after 50 minutes is computed as:

$$\text{Operating cost of EC} = A.\ C_{electrode} + B.\ C_{energy} \tag{18}$$

where A= 0.65 US$/kg Fe, cost of iron per kg and B= 0.083 US$/kWh, cost of electricity per unit. It is considered as per Indian market price [37]. Energy consumption per unit volume (kWh m^{-3}) calculated as per Eq. (14) and electrode consumption per unit volume was calculated as per Eq. (19):

$$C_{electrode} = \frac{I \times T \times M}{n \times F \times V} \tag{19}$$

where I is current, T is time of EC in hours, M is molecular weight of Fe (56 g/mol), n is mole of electron (n=2), F is faradays constant (F= 96487 C/mol) and V is treated volume in m^3. Figure 7 shows the deviation of electrical energy and operating cost at different current densities in 50 minutes of the EC process. It can be observed from the figure that as current density increased from 35.78 to 89.45 A/m^2, the operating cost and energy consumption increases from 0.0277 to 0.207 US$/$m^3$ and 0.333 to 2.499 kWh/m^3 respectively. The reason is explained by Faraday's law (Eq. 15) which provides a relationship between CD and the quantity of electrode (anode) that dissolves in the EC cell. At higher CD, the production of Fe^{2+} ions increases. It results in increases in the consumption of electrode [23].

4.6 Mass balance for Chromium

The residue after the EC process was collected. The residue was separated by filtration and dried. This dried residue was dissolved in aquarezia with slowly heating (at temperature 80 ^{0}C) until the residue mixed completely. The chromium concentration in residue and filtrate were measured by AAS. The mass balance of chromium is presented in Table 5. The overall mass balance shows that the chromium is the same with a maximum error of 4.85 %. Chromium can be recovered by various physicochemical treatment methods [15,18].

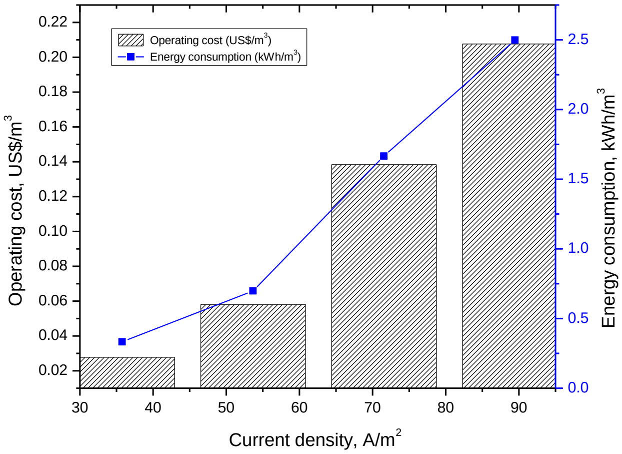


Figure 7: Operating cost and energy consumption at different current density

Table 5: Mass balance of chromium at various pH

pH	Initial Cr in effluent (mg/L)	Cr in residue (mg/L)	Cr in filtrate (mg/L)	Total Cr (mg/L) (Residue + Filtrate)	Error %
3	82.7	80.21	0.91	81.12	1.91
5	82.7	79.42	0.21	79.63	3.71
7	82.7	76.51	2.18	78.69	4.85
9	82.7	73.11	6.3	79.41	3.98
11	82.7	70.48	10.61	81.09	1.95

4.7 Characterization of sludge produced by electrocoagulation

FESEM- EDX was used for elemental analysis of sludge constituents of chromium, iron hydroxide, and other elements that are present in effluent shown in Figure 8b. Figure 8a shows the FESEM picture of the flocs. It can be observed that flocs are porous in the structure which explains the effective coagulation of $Fe(OH)_2$ flocs during the EC process. Figure 8b shows the EDX spectrum taken from FESEM-EDX analysis. It confirms that chromium, carbon, oxygen, iron, copper, and aurum are the elements present in the sludge. The existence of chromium and iron provides direct evidence that chromium is adsorbed on iron hydroxide. Disposal of sludge is a big problem however it can be used as industrial fuel or manufacturing of several products such as bricks, cementitious mixtures, fertilizer, etc.

Figure 8: (a) FESEM image of EC produces sludge

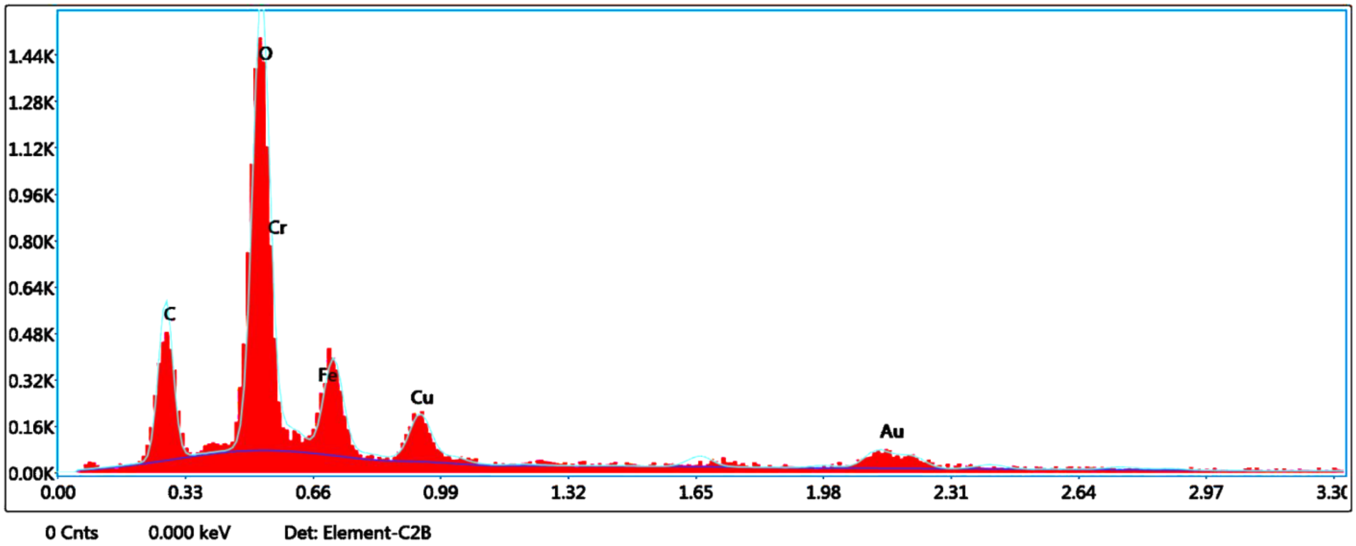


Figure 8: (b) EDX spectra of EC produced sludge

Table 6: Comparison of present work with other studies

Type of sample	Electrode	Current density A/m^2	pH	Initial concentration in mg/L	Treated volume in ml	Energy consumption in kWh/m^3	Operation cost in US$/$m^3$	EC time in min	% Removal	Ref.
Metal plating effluent	Fe-St	90	9	Cu=12.6, Cr=13.6, Ni=165	610	10.15	-	60	Cu=Cr=Ni=100	[38]
Electroplating effluent	Fe	73.5	3.5	Cr=55.3, Pb= 3.5	1500	17.14	1.86	90	Cr=91.7, Pb= 91.3	[29]
Industrial effluent	Fe-Al	200	6.32	Cr=75	3000	59.34	-	60	100	[21]
Tannery wastewater	Al-Fa	79.41	8	22.3	800	-	1.73	60	90	[22]
Aqueous solution	Al	9.14 volt	4.23	200	-	3.536	-	30	91	[39]
Metal complex dye effluent	Fe	89.45	5.83	82.7	500	2.499	0.207	50	99.64	Present work

4.8 Comparison of present work with other studies

Comparison of the present work with other studies for the treatment of chromium is presented in Table 6.

5 Conclusions

In the present study, the EC process for the treatment of metal complex dye industrial effluent is confirmed as an effective treatment method to remove chromium metal ions and other impurities. The results showed that the highest chromium removal i.e. 99.64 % was achieved at CD value of 89.45 A/m^2, pH value of 5.83 with ED of 0.7 cm, and EC time of 50 minutes. The energy consumption and operating cost of the EC process for the treatment of effluent was 2.499 kWh/m^3 and 0.207 US$/$m^3$ respectively. It was also found that other impurities like; total dissolved solids 70.02 %, COD 83.03 %, BOD 81.38 %, total solid 65.17 %, and color are removed from wastewater. Kinetic studies carried out on the EC process conclude that chromium ion removal follows a 1st order kinetic model concerning various process variables. The rate constant (k) was increased from 0.013 min^{-1} to 0.109 min^{-1} as CD increased from 17.89 A/m^2 to 89.45 A/m^2. The capture of chromium ions from effluent by iron hydroxides was confirmed with EDX analysis of sludge.

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