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Simultaneous removal of Co, Cu, and Cr from water by electrocoagulation

Subramanyan Vasudevan*, Jothinathan Lakshmi and Ganapathy Sozhan

CSIR – Central Electrochemical Research Institute, Karaikudi 630 006, Tamil Nadu, India

This study provides an electrocoagulation process for the removal of metals such as cobalt, copper, and chromium from water using magnesium as anode and galvanized iron as cathode. The various parameters like pH, current density, temperature, and inter electrode distance on the removal efficiency of metals were studied. The results showed that maximum removal efficiency was achieved for cobalt, copper, and chromium with magnesium as anode and galvanized iron as cathode at a current density of $0.025\,A\,dm^{-2}$ at pH 7.0. First- and second-order rate equations were applied to study adsorption kinetics. The adsorption process follows second-order kinetics model with good correlation. The Langmuir and Freundlich adsorption isotherm models were studied using the experimental data. The Langmuir adsorption isotherm favors monolayer coverage of adsorbed molecules for the adsorption of cobalt, copper, and chromium. Temperature studies showed that adsorption was endothermic and spontaneous in nature.

Keywords: electrocoagulation; removal; metals; adsorption; isotherms; kinetics

Introduction

Cobalt, copper, and chromium are known to be the most toxic metals for living organisms and are most widespread contaminants of the environment (Vinikour, Goldstein, and Anderson 1980; James, Sampath, and Selvamani 2006). The potential sources of these contaminants in industrial effluents include metal cleaning and plating baths, pulp, paper board mills, petrochemical industries, paints and pigments, municipal and storm water run-off (Marshal 1979; Kimbrough et al. 1999; Rengaraj and Moon 2002; Boujelben, Bouzid, and Elouear 2009; Naeem et al. 2009). Exposure to these metals may cause health problems such as epigastria, nausea, vomiting, severe diarrhea, internal hemorrhage, cancer, dermatitis, liver and kidney damage (Bailey et al. 1999; Yu et al. 2000; Mohan, Singh, and Singh 2005; Ozcan et al. 2005). As recommended by the World Health Organization (WHO 2004), the drinking water guideline values are 0.02, 2.0, and $0.05\,mg\,L^{-1}$ for cobalt, copper, and chromium, respectively.

Ion exchange, reverse osmosis, co-precipitation, coagulation, electrodialysis, and adsorption technologies have been utilized for the removal of metals; but due to economic and energy considerations, these methods have not been adopted for practical purposes (Shukla and Sakhardane 1992; Ozcan et al. 2005; Saeed, Iqbal, and Akhtar 2005; Fiol et al. 2006). Recent studies have demonstrated that the electrocoagulation method offers an attractive alternative to the traditional methods for treating water. The advantages of electrocoagulation are high particulate removal efficiency, compact treatment facility, ease

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*Corresponding author. Email: vasudevan65@gmail.com; svasudevan65@gmail.com

of operation, reduced sludge production, and the possibility of complete automation (Chen 2004; Carlos, Huitle, and Ferro 2006; Onder, Koparal, and Ogutveren 2007). Besides, the main disadvantage in the case of aluminum electrode is the residual aluminum present in the treated water due to cathodic dissolution. This will lead to health problems like cancer. There is no such health problem in the case of magnesium electrode. Although there are numerous reports dealing with electrocoagulation as a means of the removal of many pollutants from water and wastewater, there are few studies on the simultaneous removal of metals by the electrocoagulation method using magnesium as anode.

This article presents the results of the studies undertaken on the electrochemical removal of cobalt, copper, and chromium using magnesium as anode and galvanized iron as cathode. To optimize the maximum removal efficiency of cobalt, copper, and chromium different parameters like the effect of temperature, concentration, pH, and current density were studied. The equilibrium adsorption behavior is analyzed by fitting models of the Langmuir and Freundlich isotherm. Adsorption kinetics of electrocoagulants is analyzed using first- and second-order kinetic models. Activation energy was evaluated to study the nature of adsorption.

Experimental

Cell construction and electrolysis

The electrolytic cell consisted of a 1 L plexiglas vessel that was fitted with a poly-(vinyl chloride) cell cover with slots to introduce the electrodes, pH sensor, a thermometer, and the electrolytes. The anode and cathode with a surface area of $0.2\,dm^2$ were made of magnesium (commercial grade) and galvanized iron (commercial grade) placed at an inter-electrode distance of 5 mm. The temperature of the electrolyte was controlled to the desired value with a variation of $\pm 2\,K$ by adjusting the rate of flow of thermostatically controlled water through an external glass-cooling spiral. A regulated direct current (DC) was supplied from a rectifier (10 A, 0–25 V; Aplab model, Chennai).

Cobalt nitrate, copper nitrate, and potassium dichromate (Merck, Darmstadt, Germany) were dissolved in deionized water for the required concentrations. The pH of the electrolyte was adjusted, if required, with HCl or NaOH (AnalaR Reagent, Merck, Darmstadt, Germany) solutions. Temperature studies were carried at varying temperature (313–343 K) to determine the type of reaction.

Analysis

The analysis of cobalt, copper, and chromium were characterized by UV-Visible spectrophotometer (MERCK, Pharo 300) using standard MERCK Kit. The SEM and EDAX of magnesium hydroxide were analyzed with a Scanning Electron Microscope (SEM) made by Hitachi (model s-3000 h).

Results and discussion

It has been established that the initial pH of the electrolyte is one of the important factors affecting the performance of electrocoagulation processes. To evaluate its effect, a series of experiments were performed, using $10\,mg\,L^{-1}$ cobalt, copper, and chromium containing solutions, with an initial pH varying in the range of 4–12. From Figure 1, it can be seen that the removal efficiency of cobalt, copper, and chromium was increased by increasing
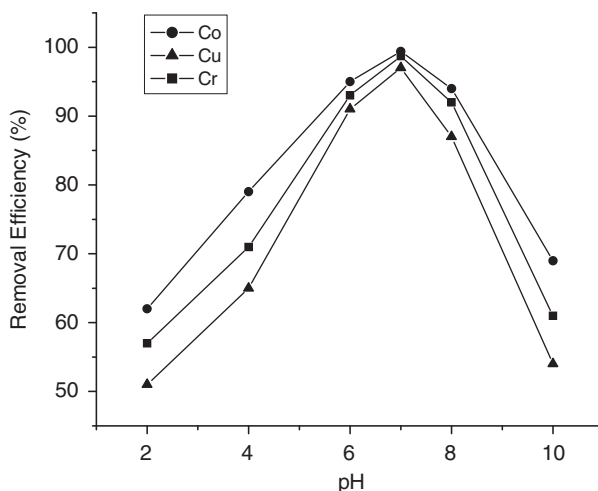
Figure 1. Effect of initial pH of the electrolyte on the removal of cobalt, copper, and chromium. Notes: Concentration of pollutants: $10 \, mg \, L^{-1}$, solution temperature: $305 \, K$, anode: magnesium, cathode: galvanized iron, current density: $0.025 \, A \, dm^{-2}$, duration: $30 \, min$.

the pH and the maximum removal efficiency was obtained at pH 7.0. The percentage of removal increased by increasing pH to 7, a decreasing trend in adsorption was observed below and above pH 7. At acidic and alkaline pHs, the oxide surfaces exhibit net positive and negative charges, respectively, and would tend to repulse the adsorption of cobalt, copper, and chromium, resulting the maximum adsorption at pH 7.0.

To determine the effect various inter-electrode distances between anode and cathode, they were kept at different distances of 3, 5, 7, 9, and 11 mm using $10 \, mg \, L^{-1}$ of cobalt, copper, and chromium each containing solutions at a current density of $0.025 \, A \, dm^{-2}$. The results of the inter-electrode distance on removal efficiency and energy consumption are presented in Figure 2. Decreasing the inter-electrode distance shows decrease in energy consumption and increase in removal efficiency. Short distance between each electrode requires lesser electrical energy for motion of ions due to shorter travel path that reduce the resistance of motion and the situation is reverse for the case of large distance between each electrode. Maintaining the inter-electrode distance of 3 mm was practically difficult, so further experiments were carried out at an inter-electrode distance of 5 mm. Inter-electrode spacing of 5 mm had low energy consumption and higher removal efficiency.

Operating current density is critical in electrocoagulation, as it is the only operational parameter that can be controlled directly. Current density directly determines both coagulant dosage and bubble generation rates, as well as strongly influencing both solution mixing and mass transfer at the electrodes. Thus, a set of experiments were carried out to quantify the impact of operating current on reactor performance. A series of experiments were carried out using $10.0 \, mg \, L^{-1}$ of cobalt, copper, and chromium containing electrolyte, at pH 7.0, with the current density being varied from 0.01 to $0.1 \, A \, dm^{-2}$. The plot shows that the uptake of cobalt, copper, and chromium ($mg \, g^{-1}$) increased with increase in the current density and remained nearly constant after the equilibrium time (figure not shown). The equilibrium time was found to be 45 min for all the studied concentration. After 45 min, the amount of cobalt, copper, and chromium adsorption (q_e) increases as the current density increases from 0.01 to $0.1 \, mg \, L^{-1}$. The plots
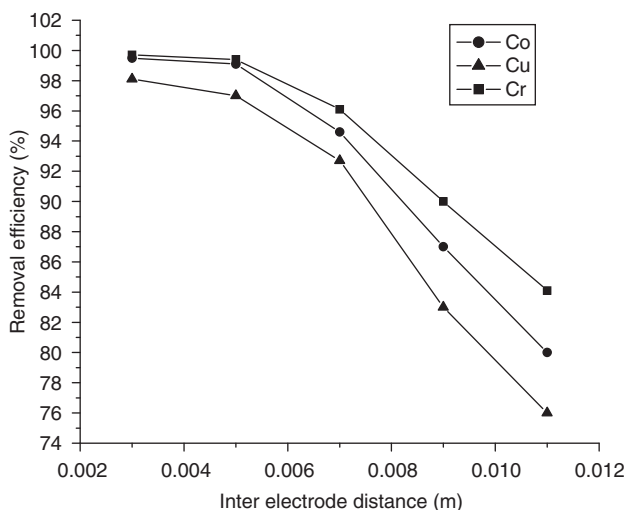
Figure 2. Effect of inter-electrode distance for the removal of cobalt, copper, and chromium.
Notes: Solution pH: 7.0, solution temperature: 305 K, anode: magnesium, cathode: galvanized iron, current density: $0.025\,A\,dm^{-2}$, duration: 30 min.

are single, smooth, and continuous curves leading to saturation, suggesting the possible monolayer coverage to cobalt, copper, and chromium on the surface of the adsorbent. Further, the amount of heavy metal in water removal depends upon the quantity of adsorbent (Magnesium hydroxide) generated, which is related to the time and current density.

Kinetic modeling

Two kinetic models, namely, Lagergren's first- (Wan Ngah and Hanafiah 2008) and second-order (Ho and McKay 1998; Wu, Joo, and Lee 2005; Benaissa and Elouchd 2007) kinetic models were tested with the cobalt, copper, and chromium concentration of $10\,mg\,L^{-1}$ at various current densities from 0.01 to $0.1\,A\,dm^{-2}$.

The experimental data were analyzed initially with the first-order Lagergren model. The plot of $\log(q_e - q_t)$ *versus* t should give the linear relationship from which k_1 and q_e can be determined by the slope and intercept, respectively. The computed results are presented in Table 1. The results show that the theoretical q_e(cal) values do not agree with the experimental q_e(exp) values at all the studied concentrations with poor correlation co-efficient. So, further the experimental data were fitted with the second-order Lagergren model.

The kinetic data were fitted to the second-order Lagergren model. The equilibrium adsorption capacity, q_e(cal), and k_2 were determined from the slope and intercept of the plot of t/q_t *versus* t (figure not shown) and are compiled in Table 1. The plots were found to be linear with good correlation coefficients. The theoretical q_e(cal) values agree well with the experimental q_e(exp) values at all the studied current density. This implies that the second-order model is in good agreement with the experimental data and can be used to favorably explain the cobalt, copper, and chromium adsorption on $Mg(OH)_2$.

Table 1. Comparison of experimental and calculated q_e values at different current densities for first- and second-order adsorption kinetics of cobalt, copper, and chromium with the concentration of $10\,\mathrm{mg\,L^{-1}}$ each at $303\,\mathrm{K}$.

Contaminant	Current density $(\mathrm{A\,dm^{-2}})$	q_e (exp)	First-order adsorption			Second-order adsorption		
			q_e (cal)	$k_1 \times 10^4$ $(\mathrm{min\,mg^{-1}})$	R^2	q_e (cal)	$k_2 \times 10^4$ $(\mathrm{min\,mg^{-1}})$	R^2
Cobalt	0.010	5.2650	11.88	−0.0049	0.7869	5.0798	0.0776	0.9976
	0.025	6.3375	12.76	−0.0056	0.7007	6.1933	0.0781	0.9944
	0.050	6.8781	13.45	−0.0059	0.7156	6.8126	0.0794	0.9988
	0.075	7.6331	14.01	−0.0065	0.7465	7.6821	0.0881	0.9964
	0.100	7.8647	14.36	−0.0071	0.7764	7.8135	0.0894	0.9997
Copper	0.010	5.1654	11.89	−0.0054	0.7755	4.9981	0.0781	0.9999
	0.025	6.2645	12.65	−0.0058	0.7677	6.1864	0.0789	0.9997
	0.050	6.7764	13.79	−0.0061	0.78640	6.7641	0.0804	0.9989
	0.075	7.6645	14.46	−0.0066	.8116	7.6027	0.0933	0.9993
	0.100	8.0021	14.84	−0.0074	0.7986	7.9645	0.0965	0.9987
Chromium	0.010	5.0119	11.32	−0.0056	0.7688	4.9931	0.0781	0.9999
	0.025	6.1279	12.33	−0.0061	0.7878	6.1155	0.0799	0.9987
	0.050	6.9956	13.64	−0.0063	0.7964	6.7645	0.0894	0.9999
	0.075	7.7583	14.25	−0.0074	0.8011	7.5646	0.0932	0.9989
	0.100	7.9864	14.66	−0.0079	0.7964	7.7789	0.0964	0.9987

Table 1 depicts the computed results obtained from first- and second-order models. From the tables, it is found that the second-order model shows good correlation coefficient than the first-order model. Further, the calculated q_e values agree well with the experimental q_e values for the second-order kinetics model, concluding that the second-order kinetics equation is the best fitting kinetic model.

Isotherm modeling

In testing the isotherm, the cobalt, copper, and chromium concentration used was $5–25\,\mathrm{mg\,L^{-1}}$ with various current densities, namely $0.01–0.1\,\mathrm{A\,dm^{-2}}$, and at an initial pH 7. The data obtained were analyzed using the well-known Freundlich (Leea, Yanga, and Hsiehb 2004; Prasanna Kumar, King, and Prasad 2006) and Langmuir (Sarioglu, May, and Cebeci 2005; Bouzid et al. 2008) isotherm models. The adsorption data is plotted as $\log q_e$ *versus* $\log C_e$ (figure not shown) should result in a straight line with slope "n" and intercept k_f. The intercept and the slope are indicators of adsorption capacity and adsorption intensity, respectively. The value of "n" falling in the range of 1–10 indicates favorable sorption. The k_f and "n" values are listed in Table 2 for each concentration and current density. It has been reported that the values of "n" lying between 0 and 10 indicate favorable adsorption. From the analysis of the results, it is found that the Freundlich plots fit satisfactorily with the experimental data obtained in this study.

The Langmuir isotherm was tested and the plots ($1/q_e$ *vs.* $1/C_e$) were found linear with good correlation coefficients (>0.99), indicating the applicability of the Langmuir model in this study. The values of monolayer capacity (q_m) and the Langmuir constant (b) are given in Table 2. The values of q_m calculated by the Langmuir isotherm were all close to

Table 2. Constant parameters and correlation coefficients for different adsorption isotherm models for cobalt, copper, and chromium at $5-25\,mg\,L^{-1}$.
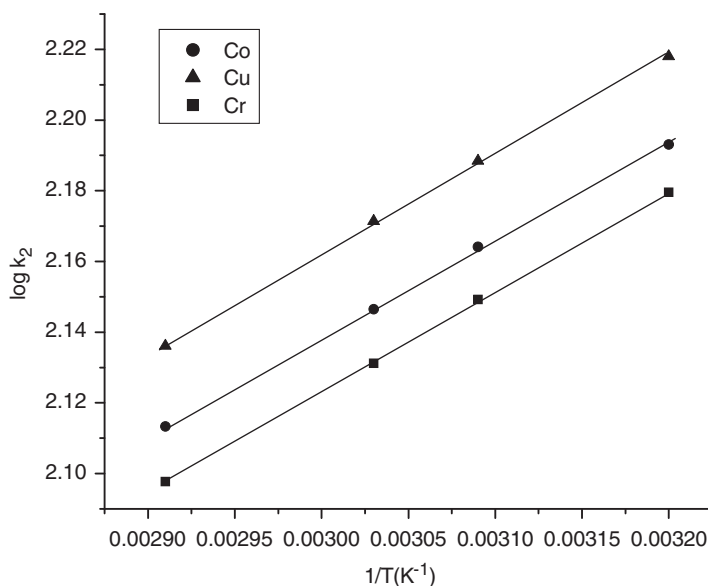
| Isotherm | Parameters | Contaminants | | |
		Cobalt	Copper	Chromium
Langmuir	q_m (mg g^{-1})	2.0261	1.9964	2.1165
	b (L mg^{-1})	4.8864	4.8277	4.9651
	R^2	0.9999	0.9994	0.9996
	R_L	0.5461	0.5546	0.5561
Freundlich	k_f (mg g^{-1})	1.6175	1.6465	1.5564
	n (L mg^1)	1.0412	1.0563	1.0126
	R^2	0.9866	0.9874	0.9864

the experimental values at given experimental conditions. These facts suggest that chromium, cobalt, and copper are adsorbed in the form of monolayer coverage on the surface of the adsorbent. The sorption isotherms of chromium, cobalt, and copper on magnesium hydroxide typically follow the Langmuirian behavior described by previous researchers.

The dimensionless constant R_L were calculated. The R_L values were found to be between 0 and 1 for all the studied concentration of cobalt, copper, and chromium. The correlation coefficient values of the Langmuir and Freundlich isotherm models are presented in Table 2. The Langmuir isotherm model has higher regression coefficient ($R^2 = 0.999$) compared to the other models, indicating that the Langmuir model provides a better description of the process.

Effect of temperature

Well-known equations (Golder, Samantha, and Ray 2006) were employed to calculate the change in enthalpy (ΔH), the change in entropy (ΔS), and the change in Gibb's free energy (ΔG) for the adsorption of contaminants by electrocoagulant. Figure 3 shows that the rate constants vary with temperature for cobalt, copper, and chromium. The activation energy is calculated from the slope of the fitted equation. The K_c and ΔG values are presented in Table 3. From the table it is found that the negative value of ΔG indicates the spontaneous nature of adsorption. The enthalpy change and entropy change were obtained from the slope and the intercept of the van't Hoff linear plots of $\ln K_c$ *versus* $1/T$ (Figure not shown) for cobalt, copper, and chromium. Positive value of enthalpy change (ΔH) indicates that the adsorption process is endothermic in nature, and the negative value of change in internal energy (ΔG) shows the spontaneous adsorption of cobalt, copper, and chromium on the adsorbent. Positive values of entropy change show the increased randomness of the solution interface during the adsorption of cobalt, copper, and chromium on the adsorbent (Table 3). Enhancement of adsorption capacity of electrocoagulant (magnesium hydroxide) at higher temperatures may be attributed to the enlargement of the pore size and/or activation of the adsorbent surface. Using the Lagergren rate equation, second-order rate constants and correlation coefficient were calculated for different temperatures (323–343 K). The calculated "q_e" values obtained from the second-order kinetics model agrees with the experimental "q_e" values better than the first-order kinetics model,

Figure 3. Plot of $\log k_2$ and $1/T$.
Notes: pH of the electrolyte: 7.0, current density: $0.025\,A\,dm^{-2}$, and concentration of pollutants: $10\,mg\,L^{-1}$.

Table 3. Thermodynamics parameters for adsorption of cobalt, copper, and chromium.

Contaminant	Temp (K)	K_c	ΔG^o ($J\,mol^{-1}$)	ΔH^o ($kJ\,mol^{-1}$)	ΔS^o ($J\,mol^{-1}\,K^{-1}$)
Cobalt	323	16.32	−214.32	2.864	0.8465
	333	18.65	−199.81		
	343	21.35	−176.39		
Copper	323	18.55	−245.31	2.764	0.8893
	333	20.31	−212.66		
	343	21.64	−178.22		
Chromium	323	19.22	−266.84	2.665	0.9961
	333	20.91	−204.35		
	343	21.88	−180.34		

indicating adsorption following second-order kinetics. Table 4 depicts the computed results obtained from first- and second-order kinetic models at different temperatures.

Surface morphology

To investigate the possibility of direct deposition of cobalt, copper, and chromium on the cathode surface, the EDAX analysis was carried out on the cathode surface. The results showed that the amount of deposited cobalt, copper, and chromium on the cathode surface is very low. The reason for this phenomenon may be due to the competition between hydrogen evolution from aqueous solutions and metal deposition on the

Table 4. Comparison of experimental and calculated q_e values for the cobalt, copper, and chromium at $0.025\,A\,dm^{-2}$ with the concentration of $10\,mg\,L^{-1}$ in first- and second-order adsorption kinetics.

Contaminant	Temp (K)	q_e (exp)	First-order adsorption			Second-order adsorption		
			q_e (cal)	$k_1 \times 10^4$ (min mg^{-1})	R^2	q_e (cal)	$k_2 \times 10^4$ (min mg^{-1})	R^2
Cobalt	323	6.3815	11.25	−0.0061	0.8125	6.3814	0.0961	0.9997
	333	6.4531	11.47	−0.0065	0.7947	6.5002	0.0992	0.9994
	343	6.5615	11.67	−0.0067	0.8022	6.5894	0.0998	0.9985
Copper	323	6.3556	11.87	−0.0051	0.8021	6.3124	0.0966	0.9999
	333	6.4345	12.45	−0.0054	0.7996	6.3964	0.0989	0.9989
	343	6.5567	12.57	−0.0059	0.7946	6.5891	0.0981	0.9993
Chromium	323	6.3314	12.46	−0.0065	0.8115	6.2127	0.0964	0.9999
	333	6.4412	12.66	−0.0066	0.8021	6.4123	0.0971	0.9991
	343	6.5012	12.83	−0.0068	0.8364	6.4987	0.0976	0.9989

cathode surface. Hence, the direct cathodic deposition has insignificant effect on the removal of cobalt, copper, and chromium ions from the aqueous solution.

The SEM images of magnesium anode, before and after electrocoagulation of cobalt, copper, and chromium electrolyte were obtained to compare the surface texture. The surface of the electrode is uniform prior to its use in electrocoagulation experiments. After several cycles of use in electrocoagulation experiments, the electrodes show rough and a number of dents. These dents are formed around the nucleus of the active sites where the electrode dissolution results in the production of magnesium hydroxides. The formation of a large number of dents may be attributed to the anode material consumption at active sites due to the generation of oxygen at its surface.

Energy-dispersive analysis of X-rays was used to analyze the elemental constituents of cobalt, copper, and chromium adsorbed by magnesium hydroxide is shown in Figure 4. It shows that the presence of Co, Cu, Cr, Mg, and O appears in the spectrum. EDAX analysis provides direct evidence that metals are adsorbed on magnesium hydroxide. Other elements detected in the adsorbed magnesium hydroxide come from the adsorption of the conducting electrolyte, chemicals used in the experiments, and the scrap impurities of the anode and cathode.

Conclusions

The results showed that the maximum removal efficiencies were achieved for cobalt, copper, and chromium at a current density of $0.025\,A\,dm^{-2}$ and a pH of 7.0 using magnesium as anode and galvanized iron as cathode. The magnesium hydroxide generated in the cell removed the cobalt, copper, and chromium present in the water and made it suitable for drinking. Adsorption of cobalt, copper, and chromium were preferably fitting the Langmuir adsorption isotherm. The adsorption process follows second-order kinetics. Temperature studies showed that adsorption was endothermic and spontaneous in nature. From the surface characterization studies, it is confirmed that the magnesium hydroxide
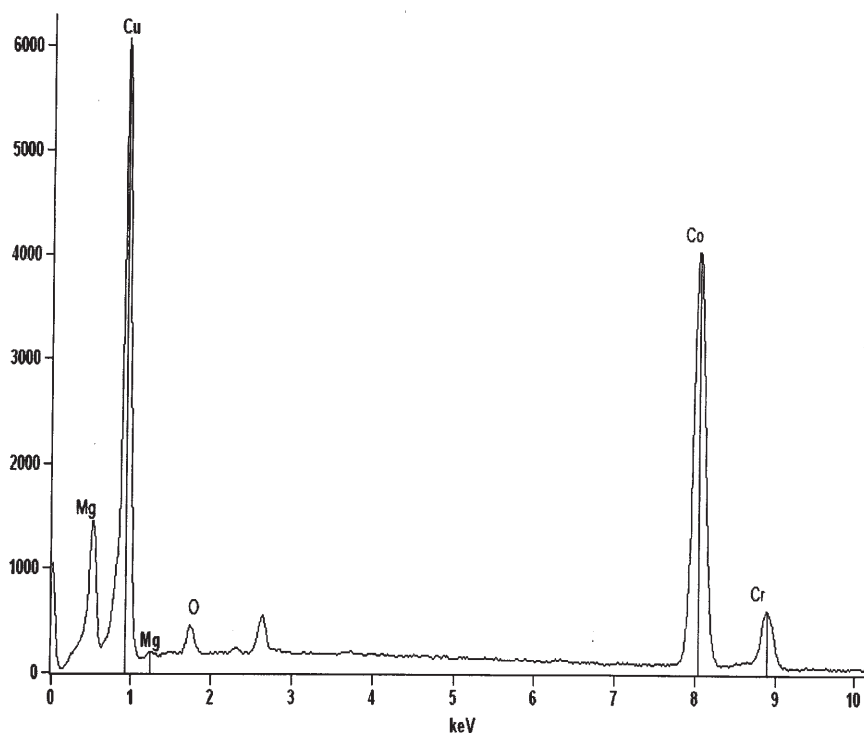
Figure 4. EDAX spectrum of cobalt, copper, and chromium adsorbed on magnesium hydroxide.

generated in the cell adsorbed cobalt, copper, and chromium present in the water than direct cathodic deposition.

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