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Removal of heavy metals (Cr, Cu, and Zn) from electroplating wastewater by electrocoagulation and adsorption processes

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ABSTRACT

Heavy metal intrusion into the water systems is a global environmental problem. Rivers, lakes, and ponds are usually contaminated by direct discharge from industries and these contaminants may leach into groundwater systems via the transport mechanisms. While some of the heavy metals are essential for the metabolic activities of living organisms but their presence in high concentrations in water may be very hazardous. The present study deals with the removal of trace metals including chromium (Cr), copper (Cu), and zinc (Zn) using electrocoagulation and adsorption techniques. The removal of Cr, Cu, and Zn was found to increase with the increase in electrocoagulation time, sodium chloride concentration and applied an electric current. The optimum conditions evaluated were pH around 4, applied electric current 2 A, and 60 min of electrolysis time. This experimental study showed that under the optimal conditions, 87.6% Cr, 100% Cu and 99.2% Zn were successfully removed. The adsorption percentages of these ions by TiO_2:AC increased sharply by increasing adsorbent dose. The results show that an optimum dose of 5 and 4 g/L of TiO_2:AC can remove about 97% Cr(VI), 97.45% Cu, and 96% Zn from the wastewater sample containing initially 50 mg/L concentration of each heavy metal. Electrocoagulation and TiO_2:AC exhibited a high degree of Cr, Cu, and Zn removal and therefore they can be utilized for the treatment of industrial effluents.

Keywords: Electrocoagulation; Adsorption; Electroplating wastewater; Trace metals

1. Introduction

Population explosion, urbanization, and industrialization have resulted in the generation of many types of wastewater [1,2]. Discharge of the wastewater can pollute surface, groundwater, and soil due to the presence of many organic and inorganic contaminants [3–5]. Heavy metals contamination is one of the major issues universally [4,6–8]. Some of the industrial wastewater generally contains metal ion concentrations much more than the permissible limits. The electroplating industry is one such industry that generates large amounts of metal-rich wastewater. These metals include Cd, lead, Hg, Cr, Ni, Cu, Zn, Co, etc. Heavy metals can damage the environment and consequently may result

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in different human diseases due to their toxicity even at very low concentrations [9]. They can enter the human body via food, air, and water, and bio-accumulate over time [10]. Since the solubility of heavy metals in wastewater is low, they cannot be degraded; therefore, they tend to accumulate. The removal of poisonous heavy metals from wastewaters is considered one of the most important fields of water treatment since many industries generate a lot of hazardous contamination issues to the environment [11]. Therefore, the use of proper methods for the treatment of wastewater containing heavy metals is essential [12,13]. Electrocoagulation (EC) is a new method in the field of water and wastewater purification, as it combines the advantages of coagulation, flotation, and electrochemistry. It is an efficient electrochemical method for the purification of various types of contaminated water that has received much attention recently because of its high efficiency in the removal of numerous pollutants. This method has been efficient in the removal of organic and inorganic pollutants with low or almost no generation of by-product wastes [14,15]. Many studies have been focused on the utilization of electrocoagulation for the purification of various types of wastewater such as polluted groundwater to highly contaminated refinery effluents [16–21]. They have demonstrated the evaluation of the efficiency of the electrocoagulation process in removing toxic metals from industrial wastewater. Adsorption process is another method that has also been used in many studies due to operationally straightforward, highly efficient, and relatively inexpensive properties for the removal of heavy metals [22,23]. Many types of adsorbents have been used for metal ions adsorption [24–26]. Bazrafshan et al. [27] quantitatively compared chemical precipitation and electrocoagulation in removing heavy metals such as Fe, Al, Ca, Mg, Mn, Zn, Si, Sr, B, Pb, Cr, and As from coal mine drainage wastewater at a laboratory scale [27,28]. Kobya et al. [26] applied electrocoagulation technique to remove arsenic from drinking water. Akbal and Camcı [29] investigated the removal of copper (Cu), chromium (Cr) and nickel (Ni) from metal electroplating wastewater by electrocoagulation with iron and aluminum electrodes with monopolar configuration. Nouri et al. [30] investigated the efficiency of electro-coagulation in combination with aluminum sacrificial anode, in the uptake of Zn and Cu. Arroyo et al. [31] investigated the effect of pH and chloride ions concentration on the removal of Cr(VI) from wastewater by batch electrocoagulation by the use of iron plate electrodes. Adhoum et al. [32] studied the efficiency of electrocoagulation, with aluminum sacrificial anode in the purification of wastewater containing some metal ions such as Cu(II), Zn(II) and Cr(VI). Ince and Kaplan [33] reviewed the adsorption process for heavy metal removal from water/wastewater. Kırbıyık et al. [34] compared the performance of Fe(III) and Cr(III) metal adsorption processes with three adsorbents including sesame stalk without pre-treatment, biochar derived from thermal decomposition of biomass, and activated carbon obtained from chemical activation of biomass in batch experiments. In another work, the suitability of prepared acid activated carbons derived from oil palm and coconut shells were investigated in removing heavy metal ions such as Ni(II), Pb(II) and Cr(VI) from aqueous solution [35]. In the study, the adsorption of Cd(II), Ni(II)) and Zn(II) was studied by activated carbon derived from coconut-based activated carbon of commercial-grade (ACC) [36]. Gin et al. [37] studied the kinetics and isotherms of the adsorption process of Zn, Cu, Pb and Fe ions from electroplating wastewater using activated carbon derived from Cassava peel (CPAC) developed as an adsorbent. Thajeel [38] and studied the adsorption isotherms, kinetics, and thermodynamics of uptake of Fe^{3+}, Cu^{2+}, Pb^{2+} and Zn^{2+} ions by activated carbon derived from rice husk [39]. Momčilović et al. [40] studied the influence of important parameters including pH, initial concentration of Pb ions, contact time, and adsorbent dosage on the removal efficiency in a batch process mode. Kobya et al. [26] investigated the ability of the activated carbon to remove Ni(II), Co(II), Cd(II), Cu(II), Pb(II), Cr(III) and Cr(VI) ions from aqueous solutions by adsorption. Monser and Adhoum [41] investigated the adsorption of toxic ions including Cu, Zn, Cr, and CN^- from wastewater on the adsorbents modified with tetra butyl ammonium (TBAI) and sodium diethyl dithiocarbamate. Therefore, this study aimed to explore the feasibility of using electrocoagulation and adsorption for the removal of trace metals from electroplating wastewater. The results of this study provide good information for the removal of heavy metals for heavy metal-rich effluents.

2. Materials and methods

The electroplating wastewater was obtained from "Shamshad electroplaters, Anoopshahar road, Aligarh", India, and analyzed for various parameters such as pH, alkalinity, total solids, and heavy metal (chromium, copper and zinc) contents. TiO_2 and activated carbon (AC) was purchased form Merck Co., Germany. Synthetic wastewater was prepared from stock solutions of 1,000 mg/L Cr, Cu, and Zn prepared using $K_2Cr_2O_7$, electrolytic copper wire, and zinc metal respectively.

2.1. Experimental set up

The electrocoagulation cell was constructed from a thick plastic container having a dimension of 16 cm × 16 cm × 18 cm. The experimental equipment is shown schematically in Fig. 1. The total volume of wastewater taken in each experiment was 2 L. Aluminum (Al) plates 8 cm high, 14 cm wide and 1 mm in thickness were used for the sacrificial electrodes, arranged in monopolar configurations. Six plates were constructed in the electrochemical reactor and the distance between plates was fixed at 15 mm. A constant current was maintained using a DC power supply (Sigma PS 30 V/5 A) characterized in the current range of 0–5 and 0–30 V.

2.2. Experiments

Effect of pH (4, 6, and 8), applied current (1, 1.5, and 2 A), and contact time (0–60 min) in the electrocoagulation process were studied. For adsorption, the effect of parameters including adsorbent dose (100, 200, 300, 400, and 500 mg/100 ml), pH (2, 4, 6 and 8) and contact time (0–180 min) were was studied. 0.1 N H_2SO_4, 0.1 N NaOH and NaCl were used to adjust the pH and electrical conductivity of solutions. The pH values were measured by a digital pH meter and metal concentrations were determined using an atomic adsorption

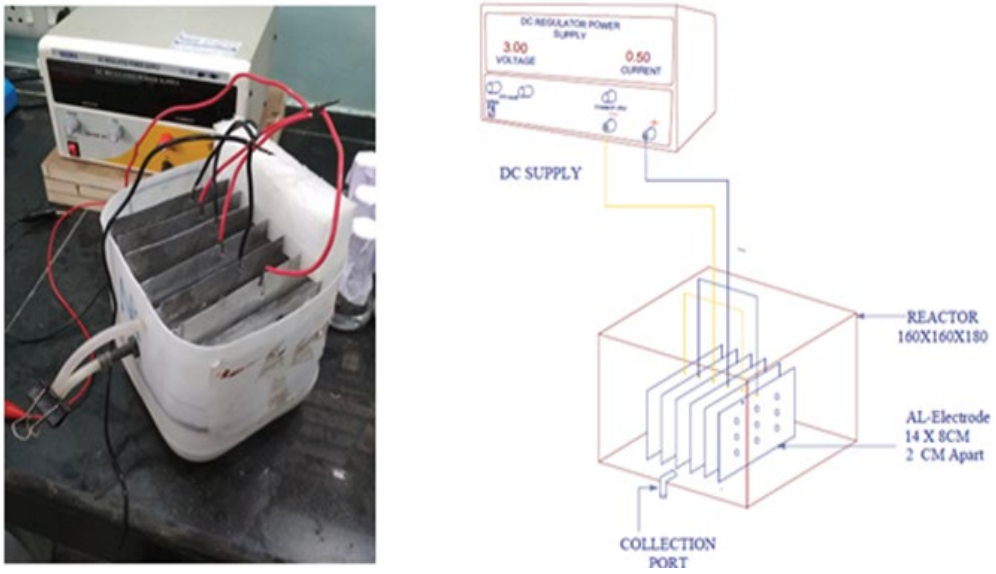


Fig. 1. Schematic diagram of the electrocoagulation setup used in this study.

spectrophotometer (Model: GBC, 932 Plus, GBC Scientific Instruments, Australia). TiO_2 and activated carbon (AC) were used as adsorbents in this study.

3. Results and discussion

3.1. Results of electrocoagulation

3.1.1. Effect of pH on chromium removal

Fig. 2 shows Cr removal at different pH values. The maximum removal efficiency of 87.64% was observed at pH 4. The efficiency of removal for Cr decreased with pH increase. The current increase resulted in better removal efficiency with maximum efficiency at a current value of 2 A. The minimum removal of 20.5% was observed at pH 6 at 1 A current and electrocoagulation time of 60 min.

3.1.2. Effect of pH on copper removal

Fig. 3 demonstrates the effect of pH, time and applied current on copper concentration. It shows that copper was removed effectively at the initial stage of the process and about 99% removal was achieved at 30 min of electrocoagulation time in the acidic pH range of 3–5.

3.1.3. Effect of pH on Zn removal

Fig. 4 demonstrates the removal of zinc from wastewater at different pH values. The removal of zinc was achieved effectively at an early stage of electrocoagulation. About 90% removal was achieved after 20 min of electrocoagulation time.

3.1.4. Effect of applied current on metal removal

Electric current is an important variable that controls the reaction rate in electrochemical systems, and it determines the magnitude of Al ions produced by the anode. Furthermore, it is very important in EC because it is the only operational parameter that can be controlled directly. In EC systems, electrode spacing is fixed and the current is continuously supplied. Current density directly affects both the coagulant dose and the amount of bubble production and considerably influences both solution mixing and mass transfer at the electrodes. The amount of metal dissolved or deposited is dependent on the amount of electricity which passess through the electrolytic solution. At higher current densities, the magnitude of anodic dissolution rises, and in turn, the magnitude of hydroxocationic complexes rises too, which leads to a rise in the removal efficiency of the pollutants. The supply of current affects the magnitude of aluminum ions (Al^{+3}) being produced from the electrodes. At high current densities, the amount of anodic dissolution of aluminum increases, leading to a greater amount of precipitate for the removal of pollutants. Moreover, bubble generation rate increases and the bubble size decreases with increasing current.

3.2. Results of adsorption experiments

Different parameters of adsorption including adsorbent dose, pH and contact time were studied for the effectiveness of TiO_2:AC as an adsorbent.

3.2.1. Effect of adsorbent dose on Cr(VI) removal

The effect of various adsorbent doses (TiO_2:AC) on Cr removal from wastewater at 24 h contact time, room temperature and fixed pH is presented in Fig. 5. At a pH value of 2, there was a sharp increase in Cr(VI) removal with an increase in adsorbent dose in the beginning and then it became more or less constant. The maximum removal of 97.44% of Cr(VI) was observed at a dose of 500 mg/100 ml and a minimum removal of 66.16% at a dose of 100 mg/100 ml.

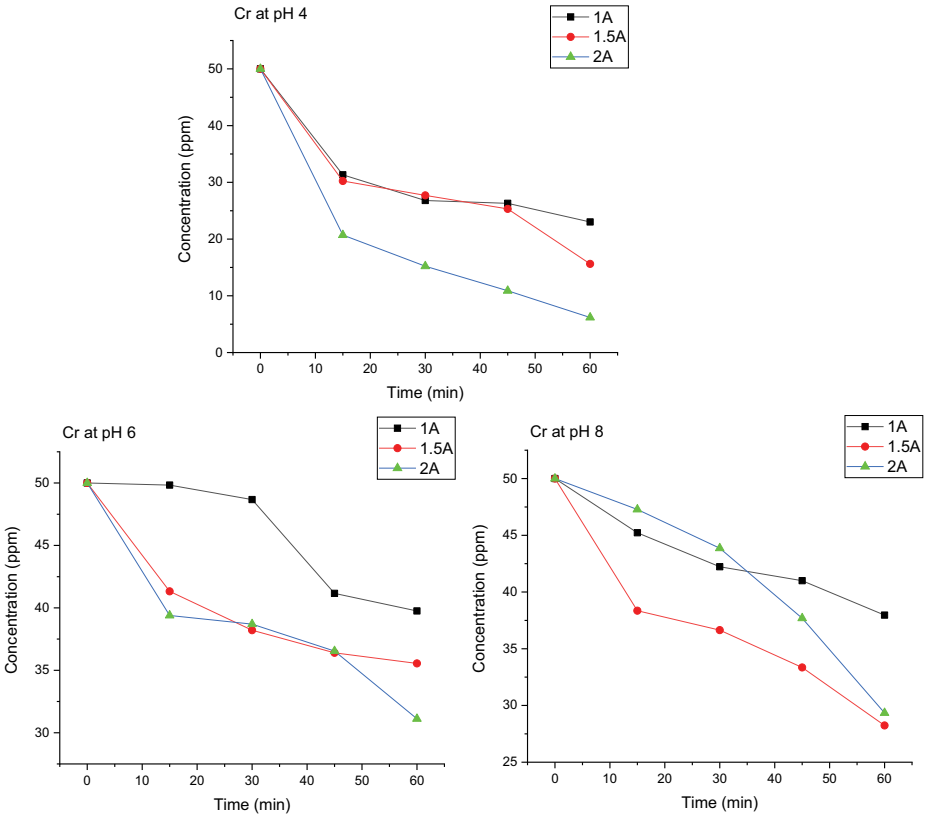


Fig. 2. Chromium concentration at different pH and current values.

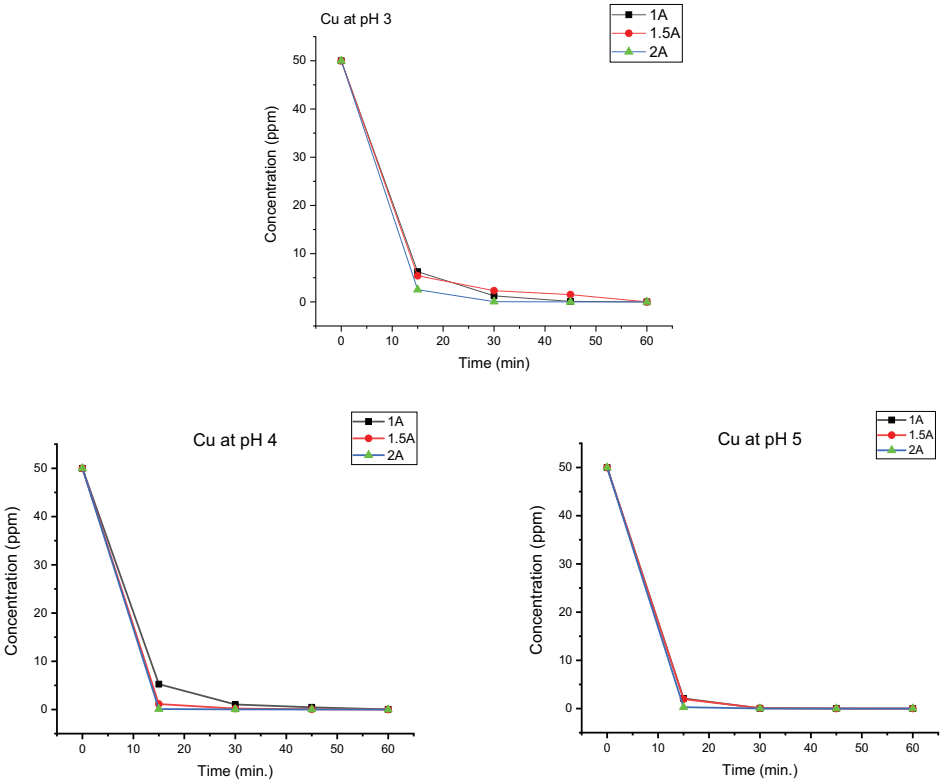


Fig. 3. Copper concentration at different pH and current values.

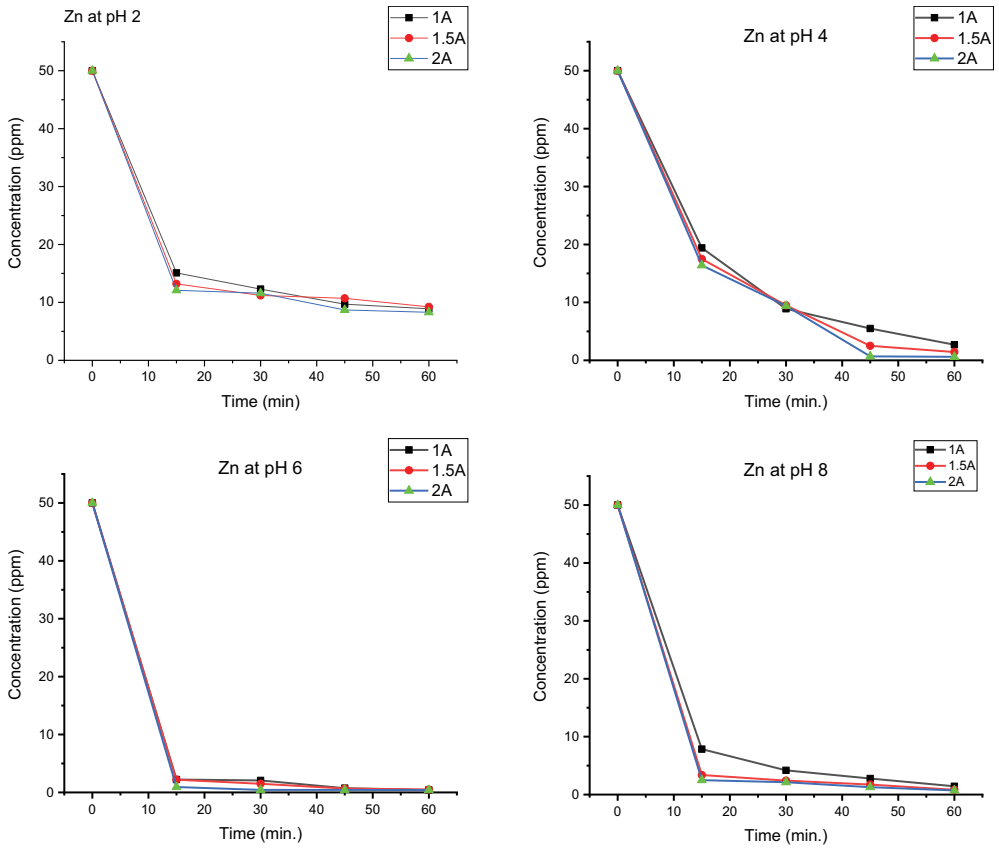


Fig. 4. Time vs. concentration of Zn at pH 2, 4, 6, and 8 at the current value of 1A, 1.5A, and 2A.

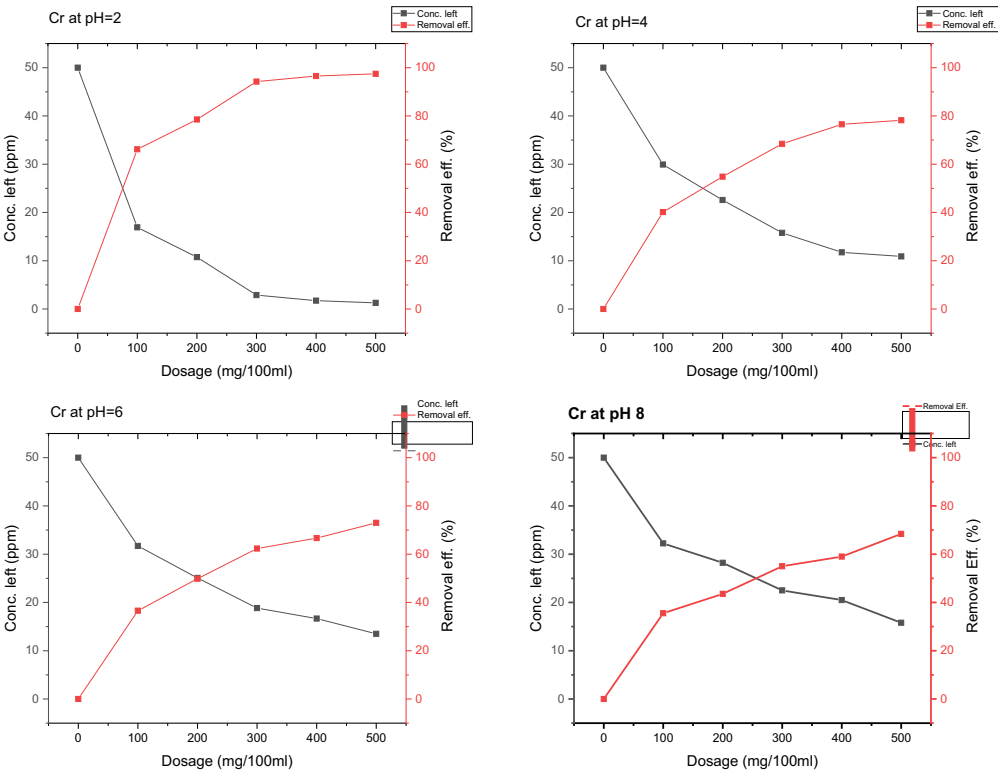


Fig. 5. Concentration and removal efficiency of Cr(VI) at different dosages and pH values.

A similar trend was observed at pH values of 4, 6 and 8 with decreasing removal efficiencies.

3.2.2. Effect of adsorbent dose on Cu removal

The effect of various adsorbent doses on Cu removal from wastewater is presented in Fig. 6 at a contact time of 24 h, and room temperature (27°C ± 1°C). The results indicated an increase in Cu removal with increasing adsorbent dosage up to a certain limit and beyond that, a less constant removal was observed. The maximum limit (97.45%) of Cu has been observed at a dose of 500 mg/100 ml and the minimum (60.16%) at a dose of 100 mg/100 ml.

3.2.3. Effect of adsorbent dose on Zn removal

Fig. 7 shows the variation of zinc removal at different pH values and adsorbent dosages at 24 h contact time. The results indicated an increase in Zn removal with an increasing adsorbent dose and then it became constant. The maximum removal of 74.18% was observed at a dose of 500 mg/100 ml and the minimum removal of 28.94% at a dose of 100 mg/100 ml.

3.2.4. Effect of pH on Cr(VI), Cu and Zn removal

Solution pH is an important parameter in the adsorption process [42]. The effect of varying pH on Cr(VI) removal at 24 h contact time, room temperature (27°C ± 1°C) and fixed doses are shown in Fig. 8. The maximum adsorption of 97.4% Cr(VI) was obtained at pH 2 and it decreased with an increase in pH.

Fig. 9 shows the variation of Cu removal for different pH values at 24 h contact time and a variable dose of 100 mg/100 ml to 500 mg/100 ml. The maximum adsorption of 97.4% was obtained at pH 3 and it decreased with an increase in pH.

Fig. 10 shows the variation of Zn removal for different pH values at 24 h contact time and a dose of 100 mg/100 ml. The maximum adsorption (96.26%) of Zn was obtained at pH 6 and the adsorption decreased with an increase or decrease in pH. The maximum adsorption of 76.66%, 95.42%, 95.42%, and 96.26% for Zn was observed at the adsorbent doses of 200, 300, 400, and 500 mg/100 ml, respectively.

3.2.5. Effect of contact time on Cr(VI), Cu and Zn removal

The results of contact time on Cr(VI) removal is presented in Fig. 11. The observations reveal that the Cr(VI) removal percentage increases with the increase in contact time. 81.43% Cr(VI) removal was observed after a contact time of 105 min. The removal efficiency increased to 97.42% on increasing contact time to 160 min. On further increasing the contact time to 24 h, removal efficiency remained the same. It is interesting to note that even at a low contact time of 75 min, the adsorbent demonstrates a chromium removal

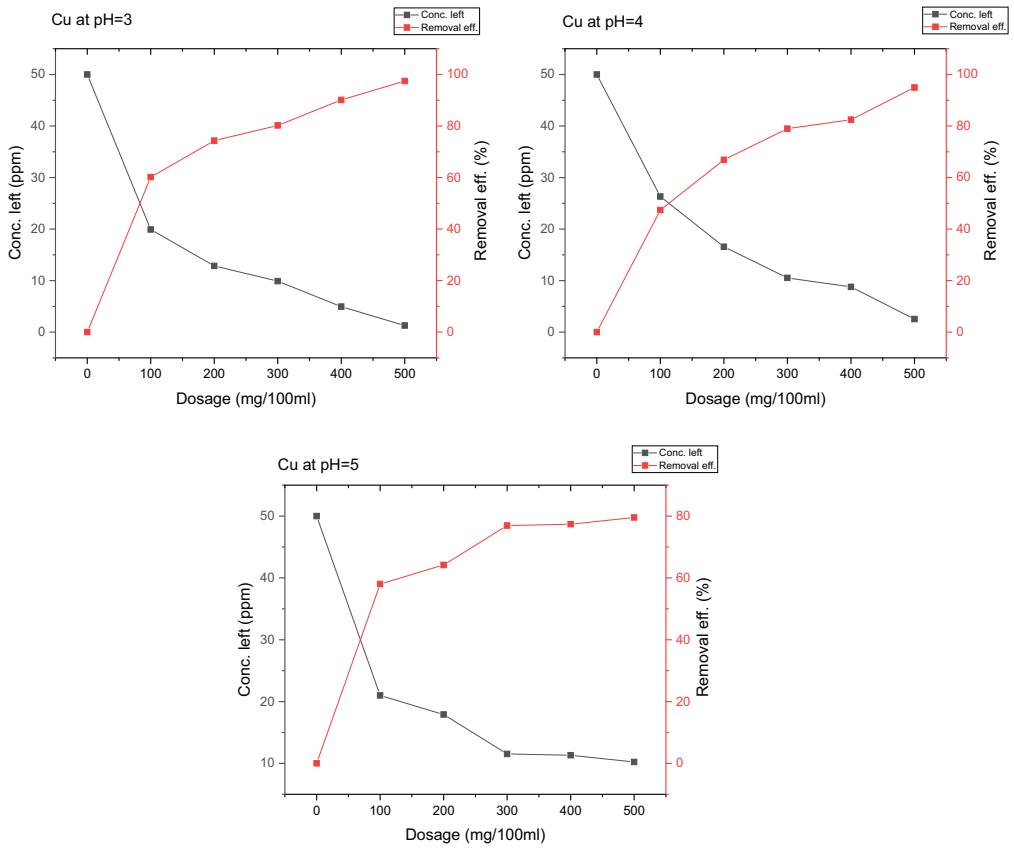


Fig. 6. Concentration and removal efficiency of Cu at different dosages and pH.

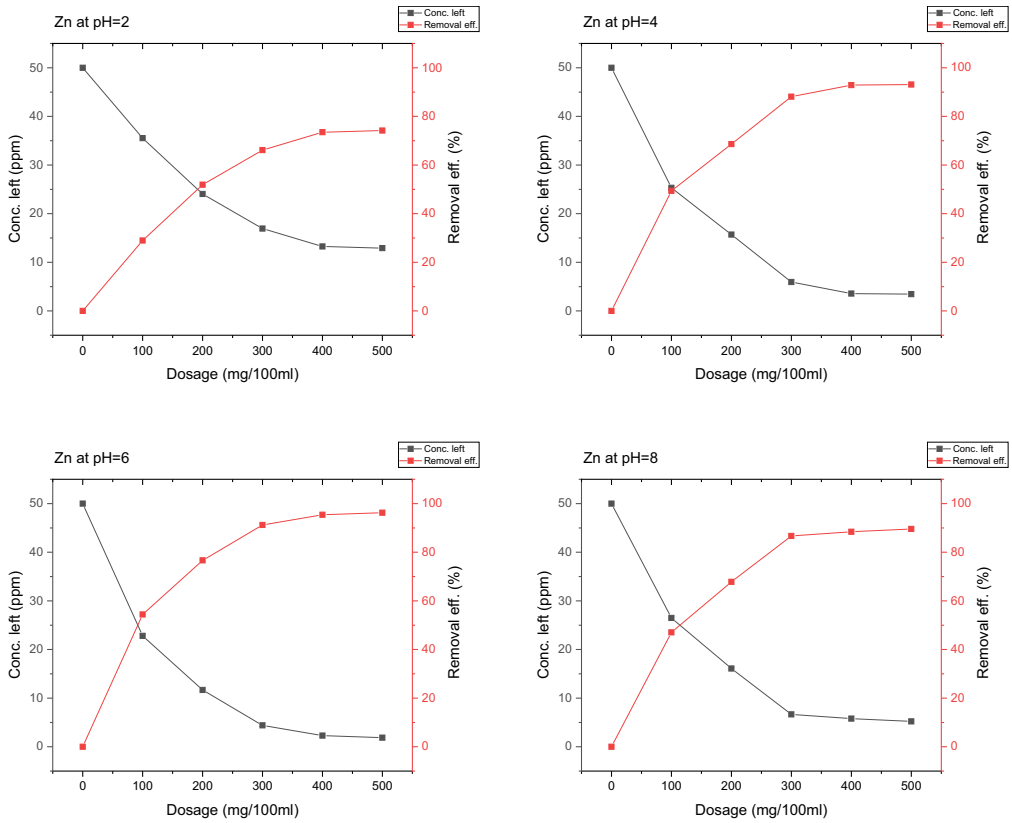


Fig. 7. Concentration and removal efficiency of Zn at different dosages and pH values.

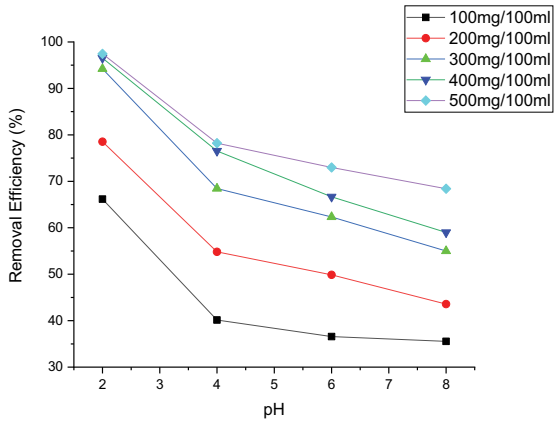


Fig. 8. Effect of pH and adsorbent dose on Cr removal.

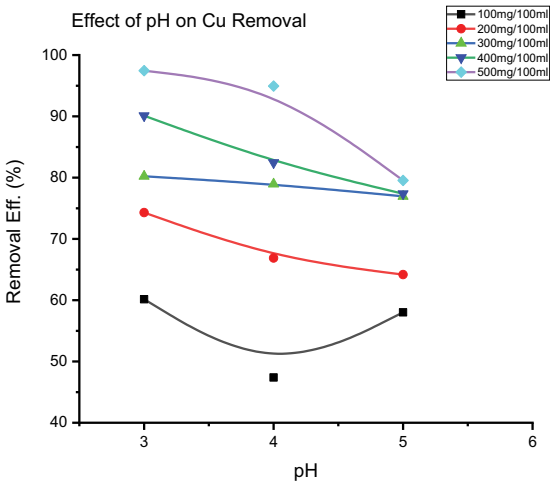


Fig. 9. Effect of pH and adsorbent dose on Cu removal.

of about 85% at a pH value of 2. It is interesting to note that even at a contact time of 105 min adsorption was efficient for removing about 80% Cr, 90% Cu, and 89.04% Zn from the wastewater sample containing initially 50 mg/L of these ions at pH 2, 3 and 6, respectively.

A similar response of contact time on copper and zinc removal is shown in Fig. 12. The observations reveal that the removal of copper increased with the increase in the contact time. About 90% of removal was observed at a contact time of 100 min. The removal efficiency increased to about 95% on increasing contact time to 160 min.

4. Conclusions

EC was found to be an effective method for the treatment of electroplating wastewater characterized by high heavy metal concentrations. The treatment of wastewater using aluminum electrodes was affected by the initial pH, current density, electrolysis time, and NaCl concentration. The removal percentage of heavy metals (Cr, Cu, and Zn) were found to increase with the increase in electrocoagulation

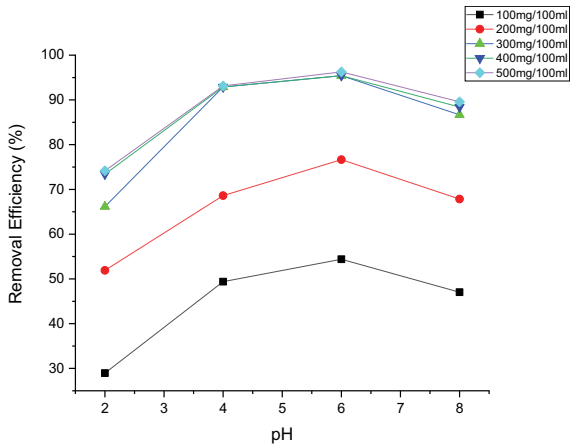


Fig. 10. Effect of pH and adsorbent dose on Zn removal.

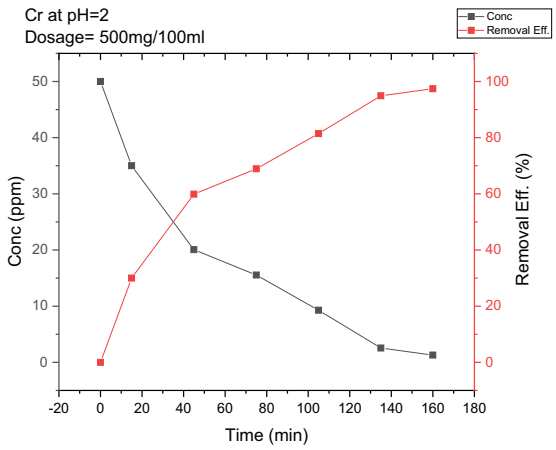
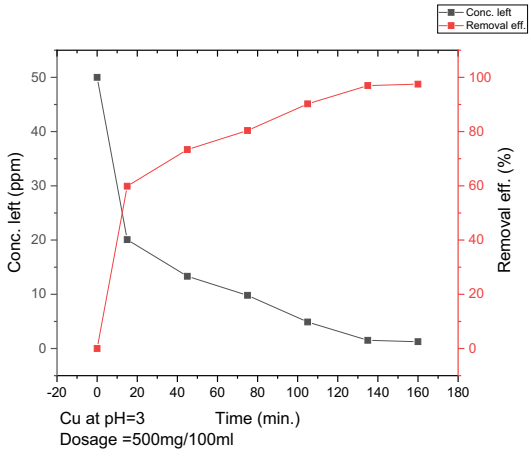


Fig. 11. Effect of contact time on chromium removal.



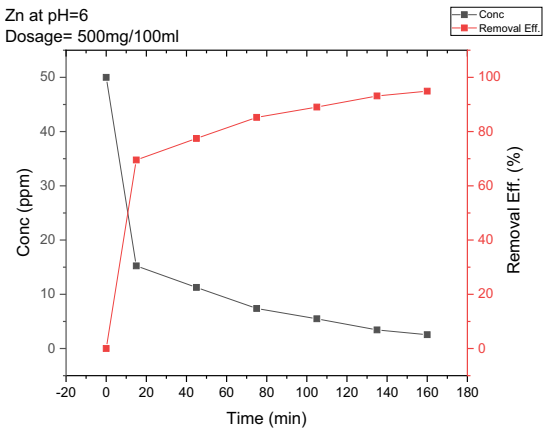


Fig. 12. Effect of contact time on copper and zinc removal.

time, sodium chloride concentration and applied an electric current. The optimum conditions evaluated were pH around 4, applied electric current 2 A, and 60 min of electrolysis time. This experimental study showed that under the optimal conditions, 87.6% Cr, 100% Cu, and 99.2% Zn were successfully removed. At the optimal condition, the pH of effluent was in the range of 6–7 which means effluent can be discharged without neutralization. The minimum power consumption for getting maximum removal efficiency was 0.2625 kWh/m^3 of wastewater treated in the process after 15 min. The power consumption for maximum zinc removal was 6.9 kWh/m^3 removal.

TiO_2:AC exhibits a high degree of adsorption and it can be utilized for the treatment of industrial wastes effluents containing Cr(VI), Cu and Zn. The adsorption percentages of these ions increased sharply by increasing adsorbent dose. The results showed that an optimum dose of 5 and 4 g/L of TiO_2:AC can remove about 97% Cr(VI), 97.45% Cu, and 96% Zn from the wastewater sample containing initially 50 mg/L concentration of each heavy metal. Data obtained during the presented study may be quite helpful in designing and studying the performance of full-scale adsorbents for the treatment of industrial wastewater containing chromium, copper, and zinc.

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