



Short communication

Use of adsorption using granular activated carbon (GAC) for the enhancement of removal of chromium from synthetic wastewater by electrocoagulation

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ABSTRACT

The present work deals with removal of hexavalent chromium from synthetic effluents in a batch stirred electrocoagulation cell with iron–aluminium electrode pair coupled with adsorption using granular activated carbon (GAC). Several working parameters such as pH, current density, adsorbent concentration and operating time were studied in an attempt to achieve higher removal capacity. Results obtained with synthetic wastewater revealed that most effective removal capacities of chromium (VI) could be achieved when the initial pH was near 8. The removal of chromium (VI) during electrocoagulation, is due to the combined effect of chemical precipitation, coprecipitation, sweep coagulation and adsorption. In addition, increasing current density in a range of 6.7–26.7 mA/cm² and operating time from 20 to 100 min enhanced the treatment rate to reduce metal ion concentration below admissible legal levels. The addition of GAC as adsorbent resulted in remarkable increase in the removal rate of chromium at lower current densities and operating time, than the conventional electrocoagulation process. The method was found to be highly efficient and relatively fast compared to existing conventional techniques.

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1. Introduction

Industrial growth and environmental pollution being two sides of the same coin run parallel to each other. Industrial growth is of utmost importance to mankind but the environmental pollution due to it is never desired. Despite several measures taken to prevent it, environmental pollution, especially due to tannery industries is growing uncontrollably. Excessive release of heavy metals into the environment is a major concern worldwide. Heavy metal contamination exists in aqueous wastes of many industries such as metal plating, mining operations, tanneries, chloro alkali, radiator manufacturing, smelting, alloy industries, storage batteries industries, etc. [1] and these usually contain metal-ion concentrations much higher than the permissible levels and do not degrade easily into harmless end products [2]. Due to their high toxicity, industrial wastewaters containing heavy metals are strictly regulated and must be treated before being discharged in the environment. Various techniques have been employed for the treatment of heavy metals, including precipitation, adsorption, ion exchange and reverse osmosis. Precipitation is most applicable among these techniques and considered to be the most economical [3]. It is based

on chemical coagulation by adding lime to raise the pH and aluminium salt to remove colloidal matter as gelatinous hydroxides. Activated silica or polyelectrolytes are added to stimulate coagulation. The former treatment may be followed by adsorption onto activated carbon to complete metals removal at the ppm level [4]. Although, it is shown to be quite effective in treating industrial effluents, chemical coagulation induces secondary pollution caused by added chemical substances.

This drawback, together with the need for low cost effective treatment, encouraged many studies on the use of electrocoagulation for the treatment of several industrial effluents. Electrocoagulation is an efficient method where the flocculating agent is generated by electro-oxidation of a sacrificial anode, generally made of iron or aluminium. In this process, treatment is done without adding any chemical coagulant or flocculants, thus reducing the amount of sludge, which must be disposed [5]. Electrocoagulation has been successfully used to treat oil wastes, with removal efficiencies as high as 99% [6]. A similar success was obtained when treating dye-containing solutions [7,8], potable water [9], urban and restaurant wastewater [10,11] and nitrate or fluoride containing waters [12,13]. In addition, a great deal of work performed in the last decades [14–16] has proved that electrocoagulation is an effective technology for the treatment of heavy metal containing solutions.

On the other hand, adsorption on activated carbon has been found to be a suitable alternative for wastewater treatment. The

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Nomenclature

EC	electrocoagulation
Cr	chromium
Fe	iron
Cr ⁶⁺	chromium (VI)
Cr ³⁺	chromium (III)
Fe ³⁺	iron (III)
GAC	granular activated carbon

contaminants are generally physically adsorbed or chemically adsorbed onto the surface of the adsorbent. There has been a tremendous increase in the use of adsorption for wastewater treatment over the last few decades in an attempt to search for a low cost adsorbent [18]. Several low cost adsorbents like bagasse fly ash [19], activated slag and biosorption using green algae have been used [20,21]. However, bagasse fly ash has an alternative use in the cement industry to modify the compactive effort and shear strength properties of cement [22]. Due to the above reasons and the abundance of jackfruit along the coastal regions of South India, jackfruit peel has been chosen as the alternative source of carbon for preparation of GAC.

Due to the operational difficulties and high cost of treatment of electrocoagulation for large-scale treatment of wastewater rich in Cr (VI) in developing countries like India, adsorption process using low cost adsorbents (GAC) has been coupled with electrocoagulation in this present study for the removal of Cr (VI). Unlike other attempts, we have chosen to study the effect of adsorption in enhancing the process of removal of chromium when coupled with electrocoagulation. The effect of the wastewater characteristics such as initial pH, current density, treatment time and adsorbent concentration on the removal efficiency is explored and discussed to determine the optimum operational conditions.

2. Theory

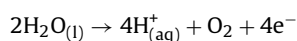
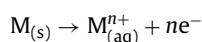
2.1. Electrocoagulation

In any EC process the coagulating ions are produced 'in situ' and it involves the following successive stages of coagulant formation and subsequent reduction of Cr⁶⁺ to Cr³⁺.

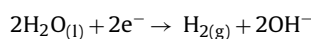
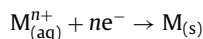
2.1.1. Formation of coagulants by electrolytic oxidation of the 'sacrificial electrode'

In this process, a potential is applied to the metal (M) anodes, typically fabricated from either iron or aluminium, which causes two separate reactions depicted as follows:

- At the anode:



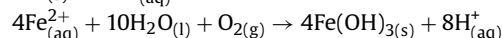
- At the cathode:



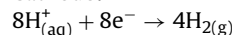
Fe/Al is dissolved from the anode, as depicted by Fig. 1, generating corresponding metal ions, which almost immediately hydrolyze to polymeric iron or aluminium hydroxides. The following mechanisms describe the formation of the iron hydroxides:

- Mechanism 1

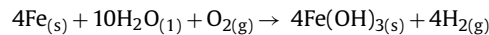
- Anode:



- Cathode:

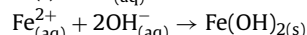
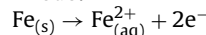


- Overall:

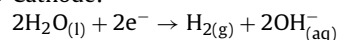


- Mechanism 2

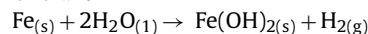
- Anode:



- Cathode:



- Overall:



The Fe(OH)_{n(s)} formed remains in the aqueous stream as a gelatinous suspension, which can remove the pollutants from wastewater either by complexation or by electrostatic attraction, followed by coagulation.

Thus, rather than adding coagulating chemicals as in conventional coagulation process, these coagulating agents are generated in situ [23]. These polymeric hydroxides are excellent coagulating agents. The consumable metal anodes are used to continuously produce polymeric hydroxides in the vicinity of the anode.

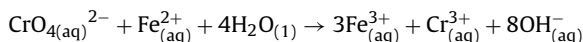
2.1.2. Destabilization of the contaminants, particulate suspension, breaking of emulsion and chromium reduction

This may be summarized as follows:

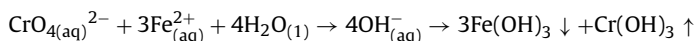
- Compression of the diffuse double layer around the charged species by the interactions of ions generated by oxidation of the sacrificial anode.
- Charge neutralization of the ionic species present in wastewater takes place due to the counter ion produced by the electrochemical dissolution of the sacrificial anode. These counter ions reduce the electrostatic inter particle repulsion to the extent that the van der Waals attraction predominates, thus causing coagulation. A zero net charge results in the process.
- The floc formed as a result of coagulation creates a sludge blanket that entraps and bridges colloidal particles still remaining in the aqueous medium.

The following reactions highlight how Cr (VI) gets reduced to Cr (III) due to the formation of Fe²⁺.

The ferrous ion (Fe²⁺) generated by electro oxidation of the iron anode can reduce Cr⁶⁺ to Cr³⁺ under alkaline conditions and is itself oxidized to ferric (Fe³⁺) ion according to



or



The Cr²⁺_(aq) ion is then precipitated as Cr(OH)_{3(s)} by raising the pH of the solution. The Fe²⁺_(aq) ions can also reduce Cr₂O₇²⁻ under acidic

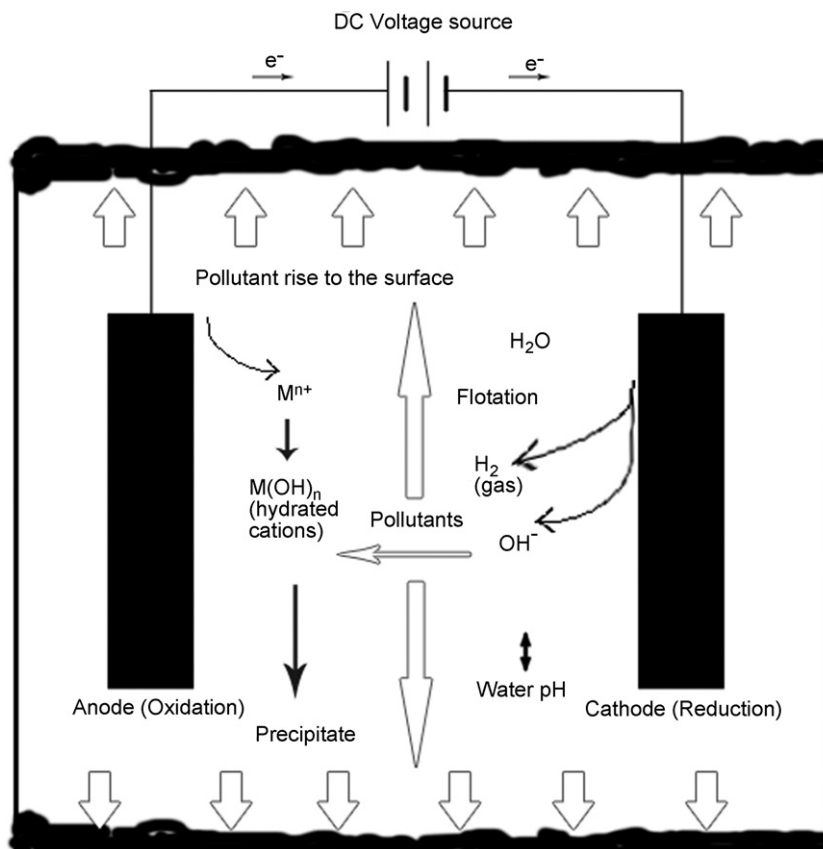
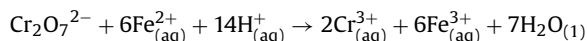


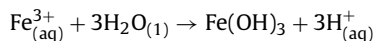
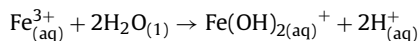
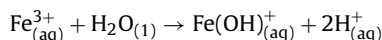
Fig. 1. Schematic diagram of bench-scale two-electrode electro coagulation.

conditions according to the following reaction:



However, the Fe^{3+} ions may undergo hydrolysis depending on the pH of the solution $\text{Fe}(\text{OH})_2^+$, $\text{Fe}(\text{OH})_2^+$ and $\text{Fe}(\text{OH})_3$ species may be present under acidic conditions.

The reactions involved are



Under alkaline conditions, $\text{Fe}(\text{OH})_6^-$ and $\text{Fe}(\text{OH})_4^-$ ions may also be present. It is, therefore, quite apparent that EC of both anionic and cationic species is possible by using a metal plate/rod as a sacrificial electrode. Simultaneously the adsorption of chromium on GAC takes place. The solid oxides, hydroxides and oxy-hydroxides provide active surfaces for the adsorption of the polluting species. Bubbles of oxygen and hydrogen generated due to the electrolysis of water floats the flocculated pollutants to the surface due to natural buoyancy [36–38].

2.2. Theory of adsorption

Subsequently, studies on the treatment of heavy metal containing effluents have revealed adsorption to be an equally effective technique for the removal of heavy metals from waste streams,

with wide use of activated carbon as an adsorbent [24]. Additionally, adsorption has been found to be superior to other techniques for water re-use in terms of the initial cost, simplicity of design, ease of operation and insensitivity to toxic substances [17]. Contaminants can be physically (physisorption) or chemically adsorbed (chemisorption) onto the GAC which acts as an adsorbent while the adsorbate chromium (VI) will accumulate on the GAC surface. In South India, jackfruit peels are available as the sources of low-cost adsorbent or activated carbon, which can be effectively used in the industrial wastewater treatment for Cr (VI). This is because these materials are considered as waste products from the country's economy and re-using them creates a large-scale recycling mechanism reducing the impact of pollution on the environment and helping to rationally consume natural resources. Traditional theories like Langmuir and Freundlich equations cannot be used separately to study the extent of adsorption on percentage removal of chromium. These equations have to be coupled with the kinetics of electrocoagulation to determine the effective percentage removal and, to the best of our knowledge no such studies have been carried out, till date.

3. Materials and method

3.1. Materials

Potassium dichromate (Ranbaxy (P) Chemicals, New Delhi, India) dissolved in de-ionized water was used as our source of chromium. 0.7071 g of the dichromate was dissolved in water and made up 250 ml in a standard flask thus giving a chromium solution of 1000 ppm concentration. From this bulk, quantitative measures were retrieved to obtain solutions of 50 ppm concentrations. 0.25%

Diphenyl carbazide solution prepared using acetone (Reachem Laboratory Chemicals (P) Ltd, Chennai, India) for spectrophotometric analysis was added to the chromium solution as a coloring agent. GAC was obtained from finely powdered jackfruit peels treated with H_2SO_4 (Ranbaxy (P) Chemicals, New Delhi, India) and dried in sunlight for 3 days, until no significant change in weight was observed. It was then crushed using a ball mill and then sieved in sieve tray system of size 800 and 1000 micrometers (μm). The GAC retained between these two sieves was used as the adsorbent. This was washed with 1.0 M HCl to remove oil and impurities and rinsed with de-ionized water to remove leachable impurities until the washed water showed the same UV absorbance as the de-ionized water [17]. After this, the GAC was dried to constant weight at $100^\circ C$ in an oven [25].

3.2. Experimental setup

The experiment was conducted in an electrocoagulation cell of capacity 1000 ml fabricated out of borosilicate glass. Mild steel containing 96% iron was used as the anode and grade 6063 aluminium containing 99% aluminium was used as the cathode. Plates of dimension 150 mm length, 100 mm breadth and 2 mm thickness provided an effective electrode area of 150 cm^2 . The electrode assembly was seated on the non-conducting wedges fixed to the bottom plate of the cell. In order to avoid short-circuiting, spacers were provided between anode and cathode plates. The gap between anode and cathode was maintained at 4 mm to minimize ohmic losses [26]. The electrodes were sanded and washed with dilute H_2SO_4 before each run. The stirrer (Spectralab Whirlmatic SP-1) was maintained at 200 rpm. The schematic diagram of the complete system and the electrode assembly is shown in Fig. 2. The apparatus used was immersed in nitric acid (1.0N) for a day to remove any metal contaminants and then immersed in de-ionized water to remove leachable impurities so that they do not affect the performance of the adsorbent [27]. The adsorbent was admitted to the system at varying dosages. A series of standard (2–10 μg) Cr (VI) was prepared and 0.25% of diphenyl carbazide in acetone was added and diluted to 25 ml maintaining an overall acidity equal to $0.1\text{ mol dm}^{-3} H_2SO_4$. The chromium–diphenyl

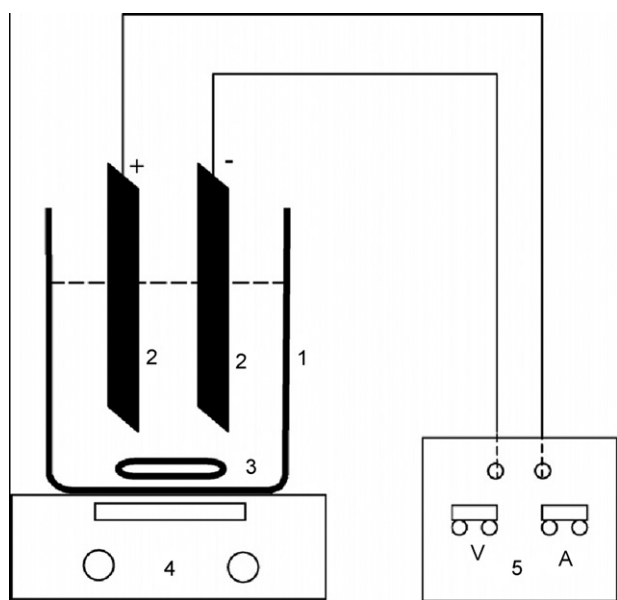


Fig. 2. Schematic process arrangement. (1) Borosil cell; (2) iron and aluminium electrode pair; (3) magnetic pellet; (4) magnetic stirrer; (5) dc power supply, voltage regulator (V) and current regulator (A).

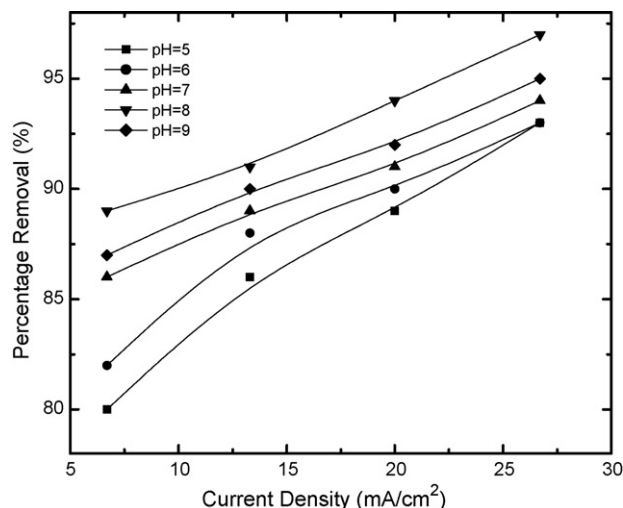


Fig. 3. Effect of current density on percentage removal (operating time = 100 min, adsorbent concentration = 100 ppm).

carbazine complex was analyzed spectrophotometrically at a wavelength of 540 nm and the calibration was found to be linear in the range 2–10 $\mu g/25\text{ ml}$ aqueous volume. Upon completion of the process, the test samples were centrifuged (Weiber ACM 67891-C) at 5000 rpm for 15 min and the supernatant was collected and its absorbance was analyzed using a UV–visible spectrophotometer (Elico Double beam UV–Vis spectrophotometer SL164). The feed was kept at a concentration of 50 ppm of chromium (VI) for all the test cases. A regulated power source (Kpas KE01) was used to provide desired cell current.

4. Result and discussions

4.1. Effect of current density and initial chromium concentration

The current density is the amount of current per area of the electrode. Current density not only determines the amount of iron coagulant produced in the process but also affects the reducing reaction of hexavalent chromium to trivalent chromium. In the reaction unit, the iron electrodes lose electrons and dissolve into the solution as iron (II) ions. Chromium (VI) ions can react with Fe (II) ions to produce chromium (III) and Fe (III). The current density applied is 6.7, 13.3, 20 and 26.7 mA/cm^2 . The current densities were maintained for different times as 20, 40, 60, 80 and 100 min. The results showed that as current density increases, reduction of chromium (VI) and removal of chromium also increases. Fig. 3 depicts the effect of current density on the percentage removal of chromium (VI) at various current densities and pH after an operating time of 100 min. It was found that, with an increase in the current density, there has been a substantial increase in the chromium removal. This can be attributed to the increase in the amount of Fe ions being generated in situ thereby resulting in rapid reduction of chromium (VI). A detailed discussion about the effect of higher current density can be obtained from refs. [28–30]. At higher current densities, the temperature of the solution may increase and hence affect the solubility of chromium in the solution. However, in the present system, no such effects were observed or studied.

4.2. Effect of pH

Effect of pH on percentage removal is shown in Fig. 4 for various current densities. Solubility of metal hydroxide species (both

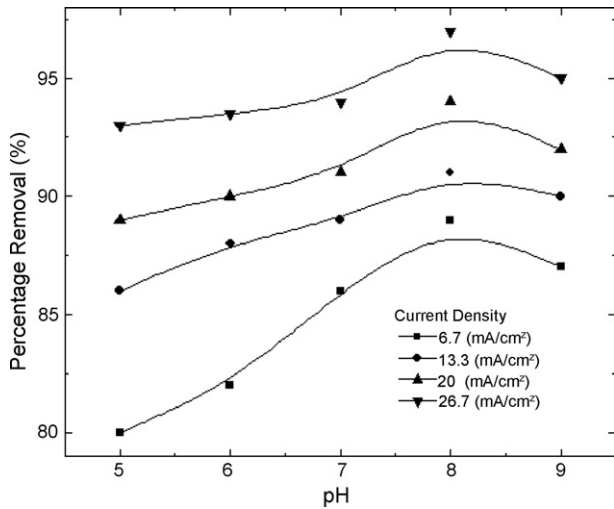


Fig. 4. Effect of pH on percentage removal (operating time = 100 min, adsorbent concentration = 100 ppm).

chromic and iron hydroxides) strongly depends on the chemistry of the continuous media. Both initial pH and the elevation of pH during EC affect Cr^{3+} solubility and hence its removal. Hydrated $\text{Cr}(\text{OH})_3$ at 298 K has minimum solubility (1.66 mg/l) at pH 8.5 in the solutions containing chlorides ions whereas $\text{Fe}(\text{OH})_3$ is least soluble at pH 8.0 [31].

It has been established that the initial pH is of vital importance in the performance of many electrochemical processes and chemical coagulation operations. It was found that as the initial pH was increased from 5 to 8, coagulation is enhanced. The final pH of the solution always increased after the electrocoagulation process [34]. It was found that for an initial pH beyond 8, percentage removal of chromium decreased, thus determining 8 to be the optimum pH value for efficient percentage removal. Electrocoagulation also exhibits some pH buffering capacity in the alkaline medium [32].

4.3. Effect of operating time

Reaction time influences the treatment efficiency of the electrocoagulation process. It determines the rate of production of Fe^{2+} ions from iron electrode [29]. Fig. 5 shows the relationship between

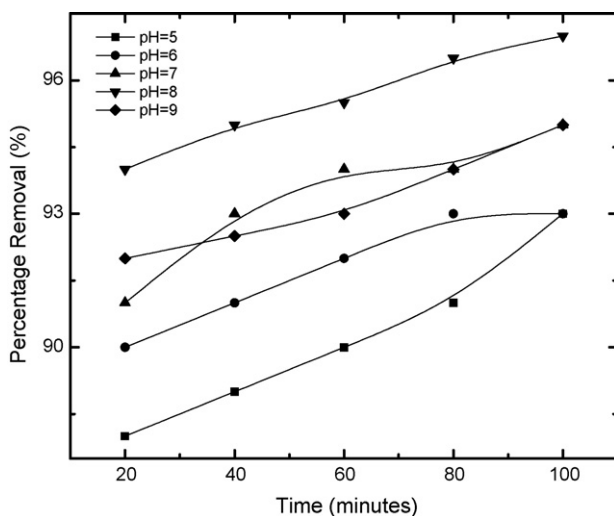


Fig. 5. Effect of operating time on percentage removal (adsorbent dosage = 100 ppm, current density = 26.7 mA/cm²).

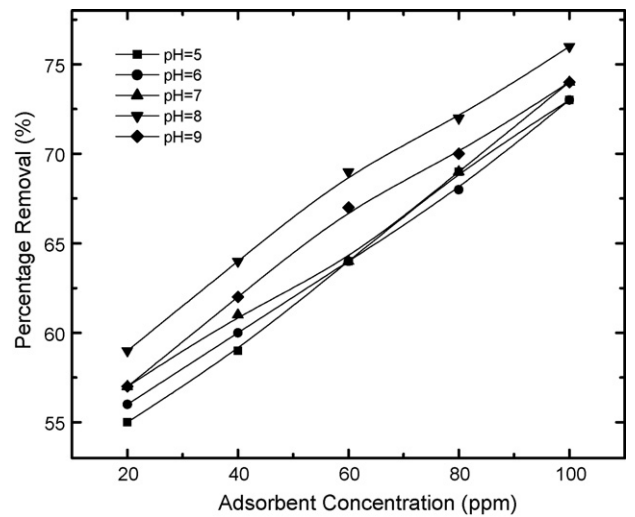


Fig. 6. Effect of adsorbent concentration on percentage removal (operating time = 20 min, current density = 6.7 mA/cm²).

chromium removal efficiency and operating time. The efficiency depends directly on the concentration of the hydroxyl and metal ions produced on the electrodes. Therefore, as expected, higher chromium removal has been observed for higher operating time.

4.4. Effect of adsorbent dosage

The following graphs show the variation of chromium removal from the effluent with the addition of GAC to the electrocoagulation setup. The adsorbent concentration was varied from 20 to 100 ppm in steps of 20 ppm.

- (i) As the adsorbent dosage is increased, keeping the other parameters constant, the percentage removal of Cr is also increased as shown in the Figs. 6 and 7.
- (ii) The highest percentage removal of 97% (current density = 26.7 mA/cm², pH 8, chromium concentration = 50 ppm, operating time = 100 min) is at an adsorbent dosage of 100 ppm.
- (iii) It is interesting to note that a percentage removal of 94% (current density = 26.7 mA/cm², pH 8, chromium concentra-

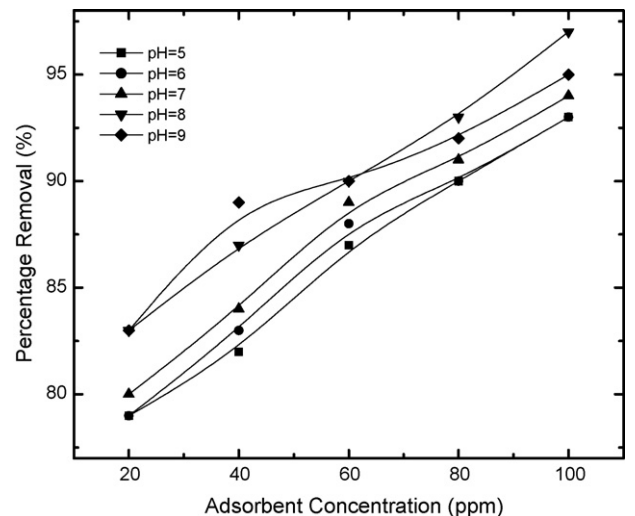


Fig. 7. Effect of adsorbent concentration on percentage removal (operating time = 100 min, current density = 26.7 mA/cm²).

tion = 50 ppm, operating time = 20 min) is achieved at an adsorbent dosage of 100 ppm whereas there has been only 75% chromium removal (current density = 26.7 mA/cm², pH 8, chromium concentration = 50 ppm, operating time = 20 min) with an adsorbent dosage of 20 ppm. Therefore, addition of adsorbent increases the extent of chromium removal.

(iv) It is also observed that a percentage removal of 94% (current density = 26.7 mA/cm², pH 8, chromium concentration = 50 ppm, operating time = 20 min) is achieved at an adsorbent dosage of 100 ppm whereas at an adsorbent dosage of 20 ppm, 83% removal has been observed (current density = 26.7 mA/cm², pH 8, chromium concentration = 50 ppm, operating time = 100 min). Therefore, addition of adsorbent enhances the process of electrocoagulation and reduces the operating time.

(v) Since chromium removal is due to the combined effect of EC and adsorption, we cannot obtain the adsorption isotherms using the existing models like Langmuir and Freundlich.

5. Conclusion

From our present study, it was found that the removal of chromium (VI) using iron and aluminium electrodes by electrocoagulation and adsorption using GAC is a feasible process. The addition of GAC as adsorbent resulted in remarkable increase in the removal rate of chromium at lower current densities and operating time than the conventional electrocoagulation process. At elevated pH, the removal of chromium is higher due to simultaneous co-precipitation and chemical precipitation. This effect is observed till the pH is increased to 8 and the percentage removal starts decreasing beyond that point. The treatment rate was found to increase upon increasing the current density from a range of 6.7–26.7 mA/cm². Also, increasing the operating time from 20 to 100 min further enhanced the removal of chromium (VI) and combined with optimum pH and current density, the percentage removal is highest. Increasing the adsorbent concentration from 20 to 100 ppm improved the percentage removal significantly; however excess addition of GAC might defeat the purpose and increase the sludge volume. In comparison to conventional techniques, the present method achieves faster removal of pollutants [11,33]. Hence, coupling of different mechanism to develop hybrid treatment process, like carried out by Chen et al. [35], may pave way for a new dimension in the field of water treatment.

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