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A Comparison Between monopolar and bipolar Electrode configurations on Removal of chromium from synthetic wastewater by Electrocoagulation Process

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□ ABSTRACT □

In this work, the performance of batch electrocoagulation (EC) treatment using iron electrodes with monopolar and bipolar electrode configurations for trivalent chromium (Cr (III)) removal from a synthetic wastewater was investigated. The influences of current density (from 2 to 25 mA/cm^2) and initial metal concentration (from 100 to 250 mg/L) on the removal efficiency were explored in a batch stirred cell for monopolar and bipolar configurations.

Removal of Cr (III) by EC process from aqueous solution with both monopolar and bipolar electrode configurations was a feasible process. For the initial Cr (III) concentration of 250 mg/L, almost complete removal (99.88%) of Cr (III) was noted after 20 min of EC in case of bipolar electrode arrangement at 25 mA/cm^2 with 4.5 mmol/L of supporting electrolyte (Na_2SO_4) against 89.58% of Cr (III) removal for monopole electrode configuration.

At the same electrolysis time, the power consumption was 47 and 15.3 kWh/m^3 in case of monopolar and bipolar configuration respectively for the previous removal conditions.

Keywords: electrocoagulation; chromium removal; monopolar and bipolar electrode configurations; iron electrodes; synthetic wastewater treatment.

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مقارنة فعالية إزالة الكروم من مياه الصرف التركيبية باستخدام الوصل الفردي والوصل المزدوج لأقطاب المختر الكهربائي

الدكتور بسام العجي [*]

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ملخّص

تم في هذا البحث اختبار أداء عملية التخثير الكهربائي الدفقي باستخدام أقطاب الحديد الكهربائية ذات الوصل الفردي والمزدوج، بهدف إزالة الكروم ثلاثي التكافؤ **Cr (III)** من مياه الصرف التركيبية. كان استخدام كل من الوصل الفردي والمزدوج لعملية التخثير الكهربائي من أجل إزالة الكروم الثلاثي عمليةً ملائمةً.

جرت دراسة تأثير كل من كثافة التيار الكهربائي (ضمن المجال من **2** وحتى **25** ميلي أمبير/سم^2) وتركيز الكروم الابتدائي (ضمن المجال من **100** وحتى **250** مغ/ل) على كفاءة إزالة الكروم في خلية دفقية مضطربة من أجل كل من الوصلين الفردي والمزدوج. لوحظت إزالة شبه كاملة للكروم الثلاثي % 99.88 من مياه الصرف الحاوية على تركيز أولي للمعدن قدره **250** مغ/ل بعد 20 دقيقة من عمل المختر الكهربائي في حالة الوصل المزدوج لدى استخدام كثافة تيار كهربائي قدرها **25** ميلي أمبير/ سم^2 عند إضافة 4.5 ميلي مول /ل من داعم الالكتروليت مقابل إزالة قدرها % 89.58 لدى استخدام الوصل المفرد.

وكان مصروف الطاقة خلال هذه العملية لنفس الشروط السابقة حوالي 47 كيلو واط ساعي/م^3 للوصل المزدوج وحوالي 15.3 كيلو واط ساعي/م^3 للوصل المفرد من أجل الحصول على نفس الفعالية السابقة.

الكلمات المفتاحية: التخثير الكهربائي، إزالة الكروم، الوصل الفردي والمزدوج للأقطاب الكهربائية، الأقطاب الحديدية، معالجة مياه الصرف التركيبية.

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1. Introduction

Chromium can exist under six oxidation states like most transition metals but the only common ones in industrial effluent are trivalent Cr (III) and hexavalent Cr (VI) [1].

The use of (Cr (III)) and (Cr (VI)) forms in several industrial processes (leather tanning, mining of chrome ore, production of steel and alloys, dyes and pigment manufacturing, glass industry, wood preservation, textile industry, film and photography, metal cleaning, plating and electroplating, etc.) leads to contamination of the environment and threaten natural life and public health for their non-degradable and carcinogenic properties [2].

Exposure to Cr (VI) may cause health problems of epigastria, nausea, vomiting, severe diarrhea, internal hemorrhage, dermatitis, liver and kidney damage. Long time contamination with Cr (VI) can result in digestive tract and lung cancers [3,4].

Trivalent Cr (III) is about 300 times less toxic than Cr (VI) and due to the fact that has limited hydroxide solubility, it is less mobile. Cr (III) has effective function for reducing blood glucose levels, and is often offered to diabetes sufferers. However, long time exposure to excessive doses of Cr (III) may also cause some adverse health effects [5].

Thus environmental regulations usually define limitation values for cromium concentrations in waters. Based on the environmental regulations, almost all the wastewater of plants that use chromium during industrial process needs to be treated before being discharged into the environment [6].

Currently, using trivalent chromium in stead of hexavalent is one of the most common and efficient processes [7].

This pollutant sparkes concern to societies and regulation authorities around the world, due to its toxicity [8].

Various techniques employ for recovering or removing Cr from wastewater such as coagulation [9], adsorption by several types of adsorbents such as activated carbon [10]; lignin [11]; diatomite [12]; natural hydrothermal zeolites [13]; synthetic zeolites [14]; bentonite clay [15], biosolid sorbent from waste-activated sludge [16], biosorption by aquatic weeds [17], ion-exchange [18] and membrane technologies [19-21].

Most of these methods suffer from some drawbacks such as high capital and operational costs, large input of chemicals, incomplete removal and their operational problems.

Among the various techniques mentioned above, Electrocoagulation- EC is found to be quite interesting both for economical and environmental reasons. EC is successfully applied to remove soluble ionic species from solutions, particularly heavy metals.

EC has the potential to be the distinct economical and environmental choice for treatment of industrial wastewater and other related water management issues. It is an efficient technique since adsorption of hydroxide on mineral surfaces are a 100 times greater on in situ than on precipitated hydroxides when metal hydroxides are used as coagulant. EC needs simple equipments, designable for virtually any size. It is cost-effective, and easily operable. The start-up and operating costs are relatively low. It requires low maintenance cost with no moving parts. Since practically no chemical addition is required in this process, it brings minimum chance of secondary pollution. It needs low current, and it can be run even by green processes, such as, solar, wind-mills and fuel cells. EC process can effectively destabilize small colloidal particles. It produces minimal sludge. Gas bubbles produced during electrolysis can enhance flotation. It removes the smallest colloidal particles efficiently compared to conventional chemical and biological techniques, because the smallest charged particles have greater probability of being coagulated by the electric field that sets them in motion [22]. In addition to these, the following advantages of EC process can be added: electrochemical-based systems allow controlled and rapid reactions, smaller systems become viable and, instead of using chemicals and microorganisms, the systems employ only electrons to facilitate water treatment.

The advantages of EC have encouraged many studies on the use of this technology for treatment of several industrial wastewater such as laundry [23]; dairy [24]; biodiesel [25]; paper [26], oil [27], electroplating [28] and textile dyes [29].

In recent years, there is much interest in using EC for treatment of water and wastewater containing phosphate [30], fluorine [31], nitrate [32], phenol[33], suspended particles [34], humic acids [35], sulfide, sulphate and sulfite ions [36], and heavy metals including Cr [37-40].

The EC process is generally accepted that the EC process involves three successive stages:formation of coagulants by electrolytic oxidation of the electrode,including metal anode dissolution and water electrolysis; destabilization of the contaminants, particulate suspension, and breaking of emulsions; aggregation of the destabilized phases to form flocs [41].

Many systems and reactors (batch or continuous mode) with monopolar or bipolar EC connection with Fe or Al electrodes were investigated.

One of the most widely used electrode materials in EC process is iron. When iron electrodes are used, the generated Fe^{3+} ions will immediately undergo further spontaneous reactions to produce corresponding hydroxides and/or polyhydroxides, and the hydrolysis products may form many monomeric ions and/or polymeric hydroxy complexes. [42].

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.

In the presence of dissolved iron, trivalent chromium is coprecipitated with ferric iron to form the solid solution of $[Cr,Fe](OH)_3$ and $Cr(OH)_3(s)$ [43].

Monopolar and bipolar EC connections were used for chromium remove. Both of them have own advantages and disadvantages. Monopolar electrodes need an external electrical contact to the power supply and their two faces are active with the same polarity. The current in parallel connections at monopolar electrodes is divided between all the electrodes in relation to the resistance of the individual cells. Hence, a lower potential difference is required in parallel connection when compared with serial connections [41, 44].

Monopolar electrodes in serial connections for each pair of electrodes is internally connected with each other, because the cell voltages sum up, a higher potential difference is required for a given current [43] .

There is no electrical connection between inner electrodes for bipolar electrodes in serial connections. The outer electrodes are only connected to the power supply. Outer electrodes are monopolar and inner ones are bipolar. The voltage applied between the latter electrodes by the power supply causes the polarization of the intermediate bipolar electrodes, which then present different polarities in the opposite faces. This connection mode has a simple setup and requires less maintenance cost during operation. The costs of higher voltage/lower current transformer rectifiers are lower for the same total power output than the lower voltage/higher current required for monopolar electrode configurations. However, the large terminal voltage between a pair of feeder electrodes also causes a proportion of the current to flow in the solution, by passing the bipolar electrodes [44].

The target of the present study was to compare the treatability of synthetic wastewater containing Cr (III) by batch EC using iron monopolar in parallel connection and bipolar electrodes in serial connection, and to compare the effects of varying operating parameters such as the applied current density, initial concentration and supporting electrolyte on Cr (III) removal.

2. Materials and methods

To demonstrate the effect of varying operating parameters, a stock solution of 1 L containing 1000 mg for Cr (III) was prepared with basic cromiuom sulfat (Merck). A set of experiments were conducted with three different solutions containing Cr (III) concentrations of 100, 150 and 250 mg/L by diluting the stock solution.

The effect of initial metal concentrations with different current densities on the system performance was investigated. Experiments were made by using a cylindrical glass cell of 500 mL on a magnetic stirrer (Falc Instruments F60 model, Italy). Six plates of iron electrodes were installed vertically with a spacer to ensure fixed distance and immersed to a 4 cm depth with an effective area of 100 cm^2 each, were used as electrodes in monopolar mode and bipolar with serial connections mode in the experiments. The inter-electrode distance was 0.3 cm. The Bench-scale EC reactor with monopolar in parallel connection and bipolar electrodes in serial connection is shown in Fig. 1 and 2 respectively.

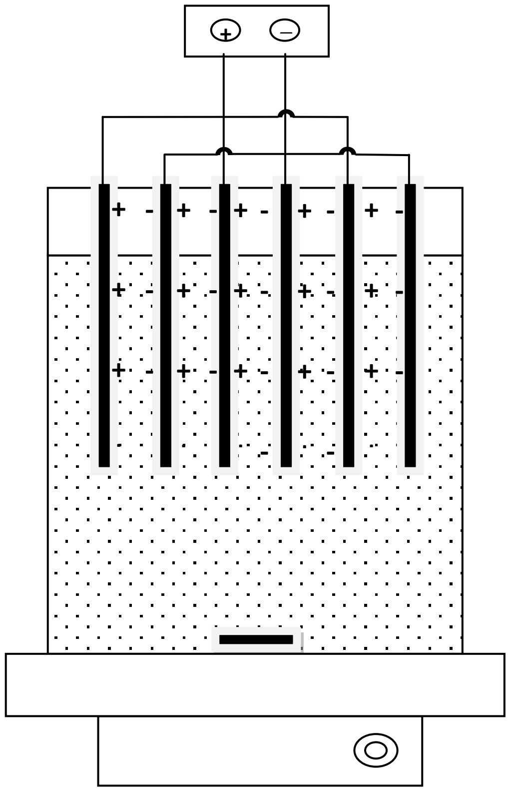

Figure 1. Bench-scale EC reactor with monopolar electrodes in parallel connection.

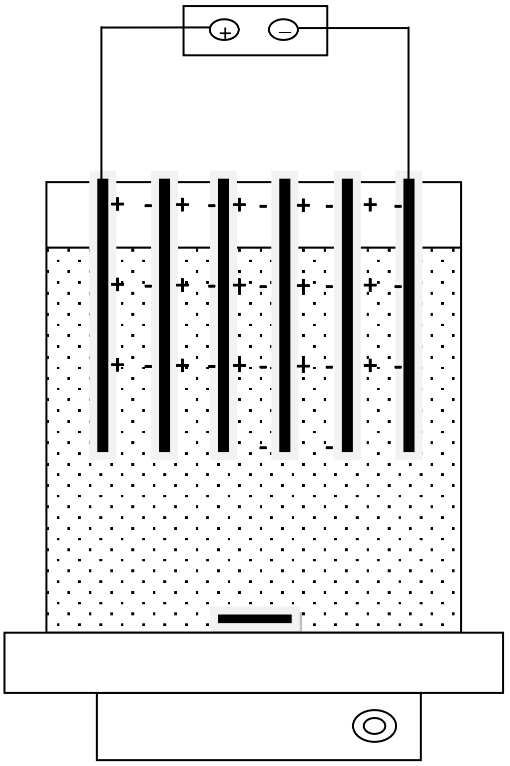

Figure 2. Bench-scale EC reactor with bipolar electrodes in serial connection.

The electrode surfaces were rinsed with diluted HNO_3 and distilled water and then dried to eliminate the oxides and passivation layers between the experiments. A DC-power supply (Statron type 3262, Germany) characterized by the ranges 0–5 A for current and 0–80 V for voltage was used to apply different current densities i.e. 2, 8, 15 and 25 mA/cm^2. The polarity of the cell was reversed after each experiment to limit the formation of the passivation layers on the electrodes. To determine the system performance, samples of 5 mL were taken periodically from the reactor at predetermined time intervals (5 or 10 min), and then filtered using Whatman filter paper (Grade 40) to eliminate sludge formed during electrolysis. The residual Cr (III) concentrations were determined by ICP-OES (Varian 720 ES model, Australia). The pH and conductivity of solutions during the EC were measured by a pH meter (Thermo-Orion 420 A model, USA).

In electrochemical processes which consist of heterogeneous ion transfer reactions, the conductivity of the solution to be treated is a significant parameter. Solution conductivity affects the applied current efficiency, voltage, and consumption of electrical energy in electrolytic systems, because the current passing through the circuit is a function of the conductivity under a certain applied voltage. More energy is required for overcoming a high ohmic resistance between the anode and the cathode in bipolar connection when electrical conductivity of the solution is low. The most common method used to overcome this problem is to add a small amount of electrolyte, which increases the electric conductivity of the solution and thus reduces the energy consumption during electrocoagulation. Conductivity of the solution increases as the supporting electrolyte concentration increases, so current passing through the circuit increases in potentiostatic mode. The different pollutants which exist in the real indstrial wastewater serve as supporting electrolyte.

Series of experiments were carried out at differnet supporting electrolyte concentrations of 0, 1.5 and 4.5 mmol/L Na_2SO_4, which equal (0, 213 and 639 mg/L) for EC process with bipolar connection. All the runs were performed at laboratory temperature and at the stirring speed of 300 rpm.

4. Results and discussions

Various parameters of EC were investigated for both electrode arrangements and the results are presented in this section.

4.1. Effect of current density

Current density combines the effect of cell current and effective surface area of electrodes. Current density during EC experiments was varied by changing cell current. The effect of the current density on removal of Cr (III) was studied at 2,8,15 and 25 mA/cm^2. These experiments were conducted with same initial concentration of 100,150 and 250 mg/L Cr(III). Figures 3, 4 and 5 show a comparison of current density effect on EC treatment for both monopolar and bipolar connections.

For the initial Cr (III) concentration of 100 mg/L, the removal efficiency values for monopolar electrode arrangement were 45.66% and 74.35% at the current densities of 2 and 8 mA/cm^2, respectively after 20 min EC treatment. The removal efficiency values for bipolar electrode arrangement were 60.35, 96.90% at the current densities of 2, 8 mA/cm^2 respectively with the supporting electrolyte of 0.0, and 1.5 mmol/L respectively as shown in Fig. 3.

For the initial Cr (III) concentration of 150 mg/L, the removal efficiency values for monopolar electrode arrangement were 70.80 and 77.50 % at the current densities of 8 and 15 mA/cm^2, respectively after 20 min EC treatment. The removal efficiency values for bipolar electrode arrangement were 96.36 and 99.70 % at the current densities of 8 and 15 mA/cm^2, respectively with the supporting electrolyte of 1.5 and 4.5 mmol/L respectively as shown in Fig. 4.

For the initial Cr (III) concentration of 250 mg/L, the removal efficiency values for monopolar electrode arrangement were 52.00, 79.60 and 89.58 % at the current densities of 8, 15 and 25 mA/cm^2, respectively after 20 min EC treatment. The removal efficiency values for bipolar electrode arrangement were 95.48, 98.74 and 99.88 % at the current densities of 8, 15 and 25 mA/cm^2 respectively with the same supporting electrolyte 4.5 mmol/L as shown in Fig. 5.

The iron hydroxides generation is lowest for monopolar electrode arrangement, therefore, effective removal of Cr (III) by electrocoagulation is observed at higher current densities for bipolar electrode arrangement, because of higher dissolution of electrode material (Faraday's law), and more generated bubbles.

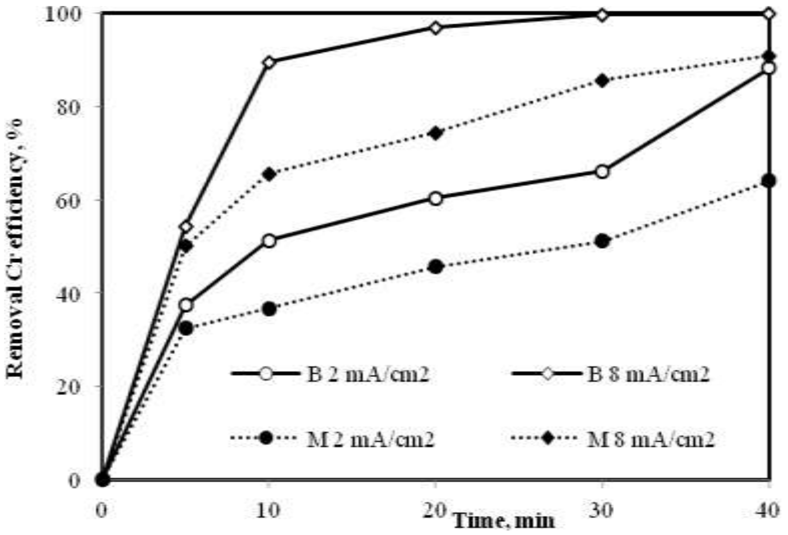

Figure 3. Comparison of current density effect on EC treatment for both monopolar and bipolar connections (C_o= 100 mg/L)

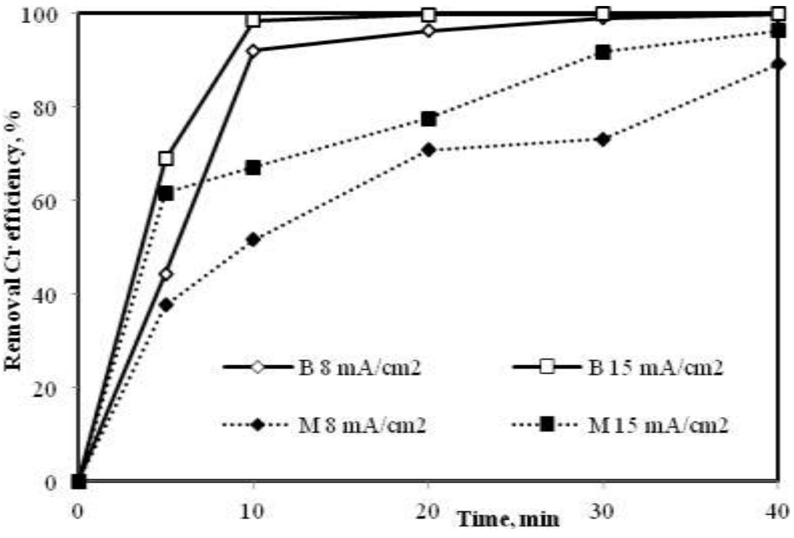

Figure 4. Comparison of current density effect on EC treatment for both monopolar and bipolar connections (C_o= 150 mg/L)

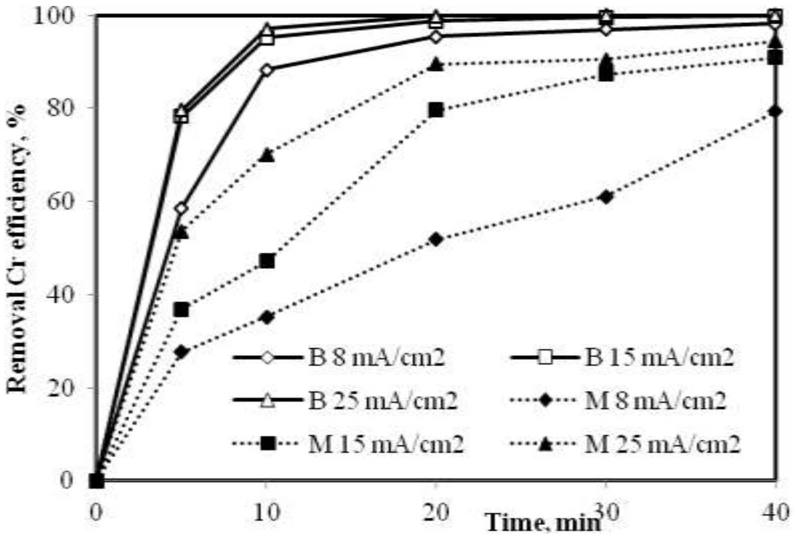

Figure 5. Comparison of current density effect on EC treatment for both monopolar and bipolar connections (C_o= 250 mg/L)

4.2. Effect of initial concentration

EC treatment for initial concentrations of 100, 150 and 250 mg/L for both electrode arrangements were tested. Initial concentrations did not influence the removal rates, whereas higher initial concentrations caused higher removal rates for both monopolar and bipolar connections . Fig. 6, 7 and 8 show the variations of initial Cr (III) concentrations as a function of time at different current densities of 8, 15 and 25 mA/cm^2. All these graphics exhibited a logical trend related with time depending on initial concentrations whithout direct correlations between Cr (III) concentration and removal rates. For the current density of 8 mA/cm^2 the Cr (III) residual concentrations for monopolar electrode arrangement were 25.6 and 43.8 mg/L for the initial Cr (III) concentrations of 100 and 150 mg/L respectively after 20 min EC treatment. The Cr (III) residual concentrations for bipolar electrode arrangement were 3.1 and 5.5 mg/L for the initial Cr (III) concentrations of 100 and 150 mg/L respectively with the same supporting electrolyte 1.5 mmol/L as shown in Fig. 6.

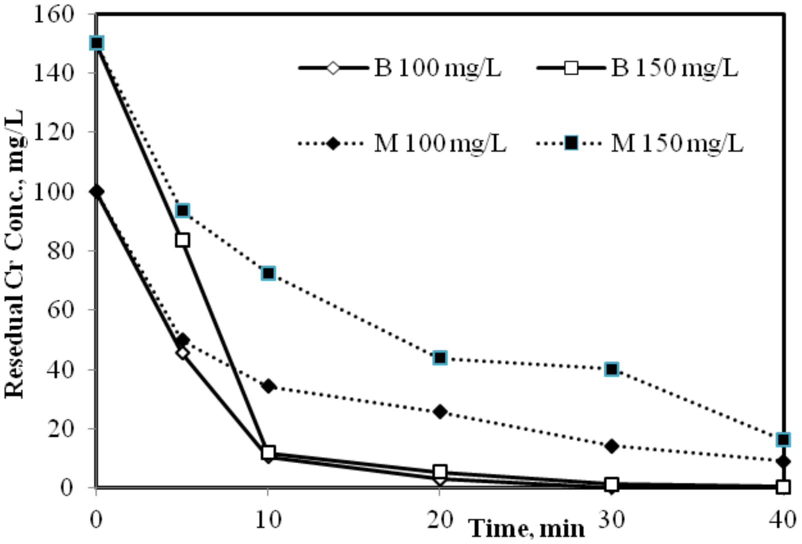

Figure 6. Comparison of initial concentration on EC treatment for both monopolar and bipolar connections (i = 8 mA/cm2)

For the current density of 15 mA /cm^2 the Cr (III) residual concentrations for monopolar electrode arrangement were 33.8 and 51.0 mg/L for the initial Cr (III) concentrations of 150 and 250 mg/L respectively after 20 min EC treatment. The Cr (III) residual concentrations for bipolar electrode arrangement were 0.4 and 3.15 mg/L for the initial Cr (III) concentrations of 150 and 250 mg/L respectively with the same supporting electrolyte 4.5 mmol/L as shown in Fig. 7.

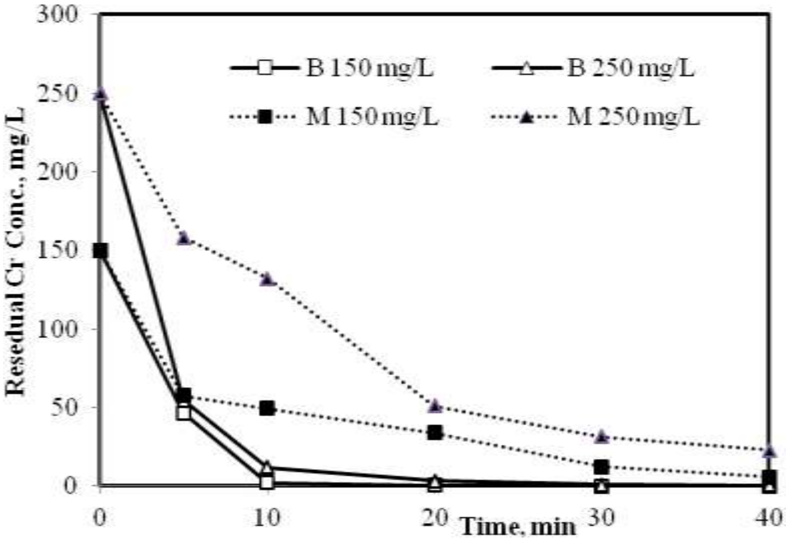

Figure 7. Comparison of initial concentration on EC treatment for both monopolar and bipolar connections (i = 15 mA/cm2)

For the current density of 25 mA/cm^2 the Cr (III) residual concentrations were 26.0 and 0.3 mg/L for monopolar and bipolar electrode arrangements for the initial Cr (III) concentrations of 250 mg/L after 20 min EC treatment. The supporting electrolyte of 4.5 mmol/L was used for bipolar connection as shown in Fig. 8.

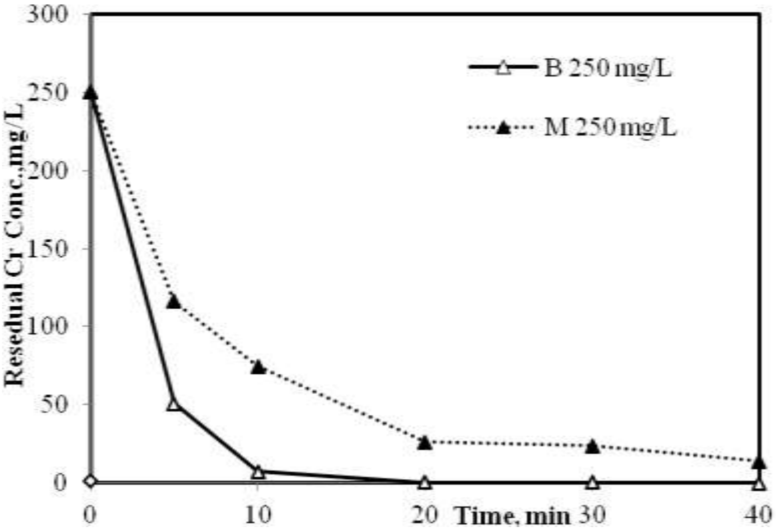

Figure 8. Comparison of initial concentration on EC treatment for both monopolar and bipolar connections (i = 25 mA/cm2)

4.3. Evaluation of energy consumption

Variation of total energy consumption values with time for Cr (III) removal as a function of current density for monopolar and bipolar connection is given in Fig. 9.

For the same energy consumption - 23 kWh/m^3 the electrolysis time was 10 min to gain more than 97% removal value for initial Cr (III) concentration of 250 mg/L at the current density of 25 mA/cm^2 with 4.5 mmol/L of supporting electrolyte for bipolar electrode arrangement, against 30 min to gain less than 91% Cr (III) removal value for monopolar electrode configuration.

Other cost items such as consumed steel electrode, maintenance and solid/liquid separation costs were not taken into account of energy consumption.

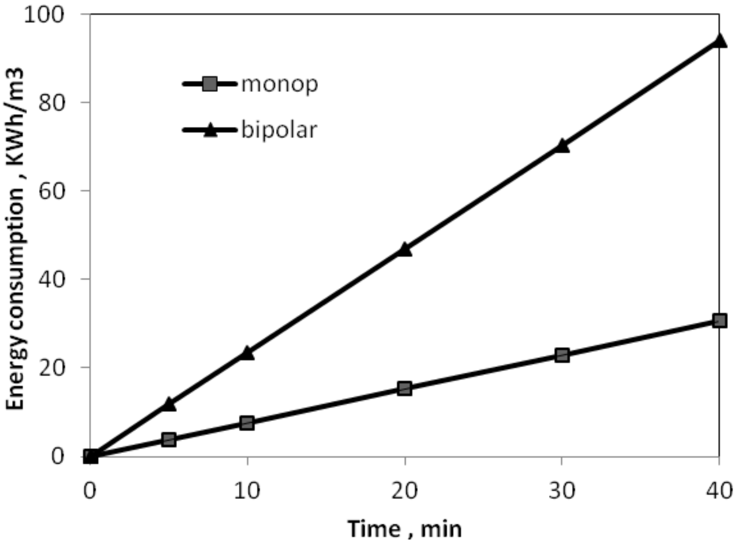

Figure 9.Variation of total energy consumption values with time for Cr (III) removal for monopolar and bipolar connection. C_0=250 mg/L, (i = 25 mA/cm2)

5. Conclusions

According to all results, EC with iron electrodes is a convenient route for effective removal of Cr (III) from a synthetic wastewater by monopolar and bipolar configurations. Bipolar configuration EC was more effective than monopolar configuration EC for Cr (III) removal from a synthetic wastewater. The effects of current density, supporting electrolyte and initial metal concentration on the removal efficiency were examined in a parallel plate electrochemical reactor. Other parameters such as pH and effect of other water pollutants on the Cr (III) removal efficiency were not taken in this study.

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