

Investigation of the effect of different electrode connections on the removal efficiency of Tartrazine from aqueous solutions by electrocoagulation

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Abstract

This study investigates the influence of variables on the decolorization efficiency of a solution containing Tartrazine (a synthetic yellow azo dye) by D.C. electrocoagulation (EC). The efficiency of different electrode connections and materials (iron, aluminium) for color removal is compared. Current density, time of electrolysis, interelectrode distance, supporting electrolyte concentration and pH of the solution were the variables that mostly influenced the color removal. Initially, a simple electrochemical cell was prepared with an anode and a cathode, then the effect of each variable was studied separately using aqueous Tartrazine in a batch mode. For a solution of 40 mg l^{-1} Tartrazine + 400 mg l^{-1} NaCl with chemical oxygen demand (COD) of approximately 30 mg l^{-1} , almost 100% color and 90% COD were removed, when the pH was about 5.78, time of electrolysis was approximately 6 min, current density was approximately 120 A m^{-2} and interelectrode distance was 1.5 cm. In the second series of experiment, the efficiency of EC cells with monopolar electrodes in series and parallel connections and an EC cell with bipolar electrodes was compared with that of a simple electrochemical cell. The results revealed that EC cell with monopolar electrodes in series connection was more effective where aluminium electrodes were used as sacrificial and iron was used as anode and cathode. Electrocoagulation with Fe/Al (anode/cathode) was more effective for the treatment process than Fe/Fe electrode pair.

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Keywords: Electrocoagulation; Tartrazine; Color removal; Sacrificial electrodes; Iron; Aluminium electrode

1. Introduction

Dyestuff effluent is one of the industrial wastewaters which is difficult to treat because of the strong color, rich organic pollutants and high suspended solids. Its decolorization is one of the indispensable processes in wastewater treatment. Color is usually the first contaminant to be recognized in wastewater and a very small amount of dye in water ($10\text{--}20 \text{ mg l}^{-1}$) is highly visible and does affect water transparency and gas solubility of waterbodies. Electrocoagulation is a very effective technique in coagulating the colloidal, found in natural water so that it reduces the turbidity and color. It is also used in the removal

or destruction of algae or microorganisms. In addition, EC has been applied to treat water containing foodstuff wastes [1], oil wastes [1–4], dyes [5], suspended particles [6–10], organic matter from landfills leachate [10], chemical and mechanical polishing waste [11], defluorination of water [12], synthetic detergent effluents [13], mine wastes [14] and heavy metal-containing solution [15–19].

Electrocoagulation involves the generation of coagulants in situ by dissolving electrically either aluminium or iron ions from aluminium or iron electrodes, respectively. The metal ions generation takes place at the anode and hydrogen gas is released from the cathode. The hydrogen gas would also help to float the flocculated particles out of the water. The electrodes can be arranged in a monopolar or bipolar mode. The materials can be aluminium or iron in plate form or packed form of scraps such as steel turnings, millings, etc [20].

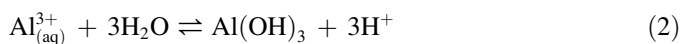
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In addition, the following physicochemical reactions may also take place in the EC cell [21]:

- Cathodic reduction of impurities present in wastewater.
- Discharge and coagulation of colloidal particles.
- Electrophoretic migration of the ions in solution.
- Electroflotation of the coagulated particles by O₂ and H₂ bubbles produced at the electrodes.
- Reduction of metal ions at the cathode.
- Other electrochemical and chemical processes.

The electrolytic dissolution of the aluminium anode produces the cationic monomeric species such as Al³⁺ and Al(OH)₂⁺ at low pH, which at appropriate pH values are transformed initially into Al(OH)₃ and finally polymerized to Al_n(OH)_{3n} according to the following reactions:

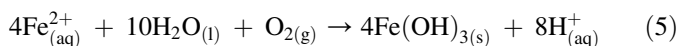


Iron upon oxidation in an electrolytic system produces iron hydroxide, Fe(OH)_n, where *n* is 2 or 3.

Two mechanisms have been proposed for the production of Fe(OH)_n [22–24].

• Mechanism 1

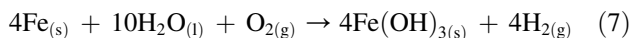
Anode:



Cathode:

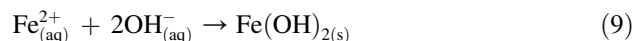


Overall:

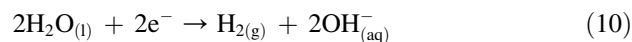


• Mechanism 2

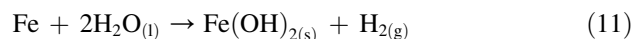
Anode:



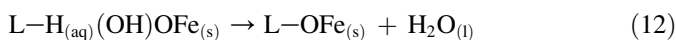
Cathode:



Overall:



The formed Fe(OH)_{n(s)} 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 [25]. In the surface complexation mode, the pollutant acts as a ligand (L) to chemically bind hydrous iron:



Furthermore, another strong oxidant of hypochlorite may be produced in many wastewaters containing chlorides, as given in Eq. (12) [26,27]:



In this study, electrocoagulation using iron and aluminium electrodes was conducted to investigate decolorization of dye solutions containing Tartrazine. Tartrazine (C.I. Acid Yellow 23) is a synthetic yellow azo dye often used in textile and foodstuff and pharmaceutical industries [28–30]. Table 1 displays the characteristics and structure of this dye.

2. Experimental

2.1. Materials and methods

The experiments were performed on Tartrazine solutions with concentrations of 40 mg l^{−1}. At the beginning of a run, 250 ml solution of the desired concentration of the dye was

Table 1
Characteristics of Tartrazine

Absorption maximum (nm)	Formula weight (g mol ^{−1})	C.I. name	Empirical formula	Structural formula
428	534.385	Acid Yellow 23	C ₁₆ H ₉ N ₄ O ₉ S ₂ Na ₃	

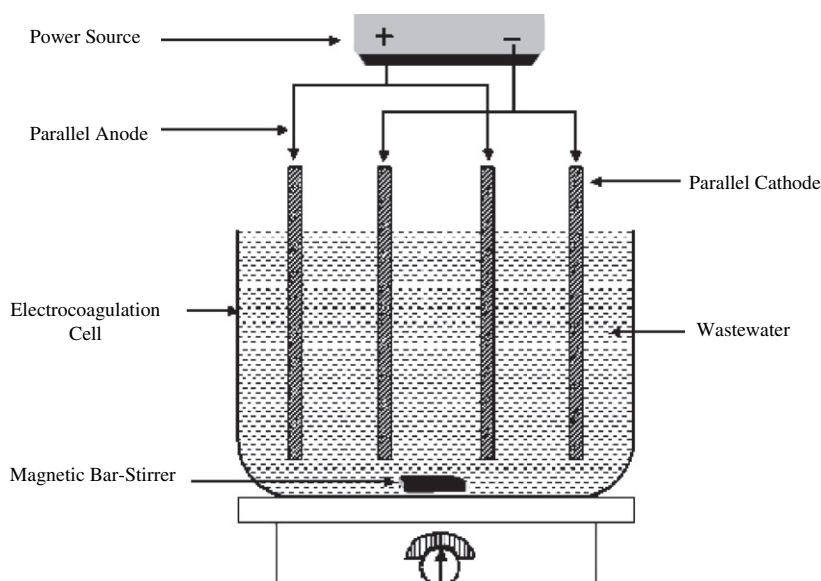


Fig. 1. Bench-scale EC reactor with monopolar electrodes in parallel connections.

fed into the reactor and NaCl salt as electrolyte was added to the solution to adjust a fixed strength ionic solution. The electrodes are placed inside a Pyrex glass reactor with a distance of 1.5 and 1 cm between anode and cathode in a simple EC cell and an EC cell with sacrificial electrodes, respectively. All experiments were performed in the above reactor as shown in Figs. 1–3, and a DC power supply (REC-P-6, 0–50 V/0–5 A), magnetic stirrer, flat-plate anode and cathode were employed. The total submerged surface area of each electrode was 18 cm² and there was a 2 cm distance between the bottom of the electrodes and the bottom of the cell, which allowed easy stirring. Before each experiment, the electrodes were abraded with sand-paper to remove scale and then cleaned with successive rinses of water, 0.1 N NaOH, HNO₃ solution

(10%) and distilled water. In order to find out the effect of the sludge contact time, after electrolysis of the dye solution, it was filtered and allowed to precipitate, after reading the absorbance, the sludge was mixed with the filtered solution for 5 min and then the above process was repeated for five times.

2.2. Chemical analysis

The dye concentration was determined by spectrophotometry at $\lambda_{\text{max}} = 254 \text{ nm}$ and $\lambda_{\text{max}} = 428 \text{ nm}$, according to Beer–Lambert law, using a Ultrospec 2000, Biotech Pharmacia UV/vis spectrophotometer. It has to be mentioned that at 254 nm and 428 nm aromatic rings and azo bands could be

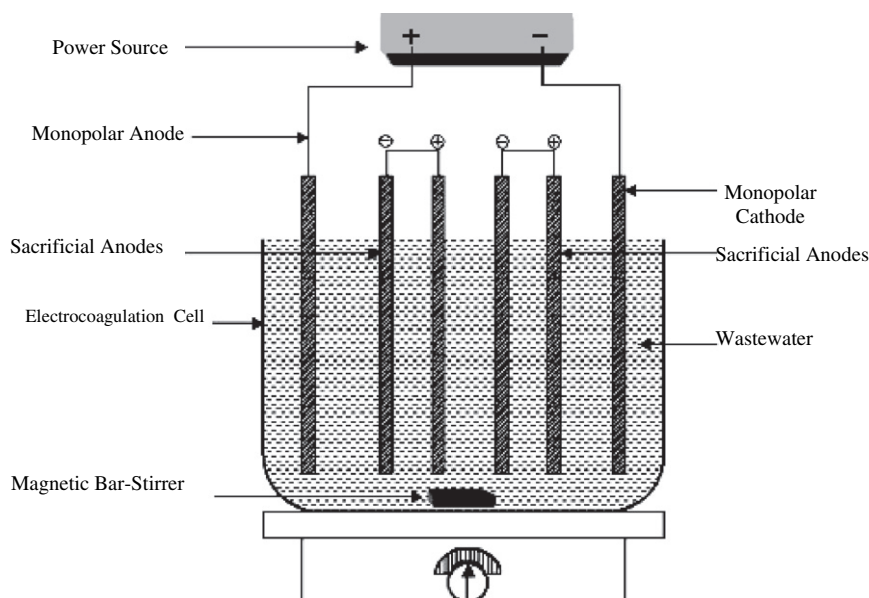


Fig. 2. Bench-scale EC reactor with monopolar electrodes in series connections.

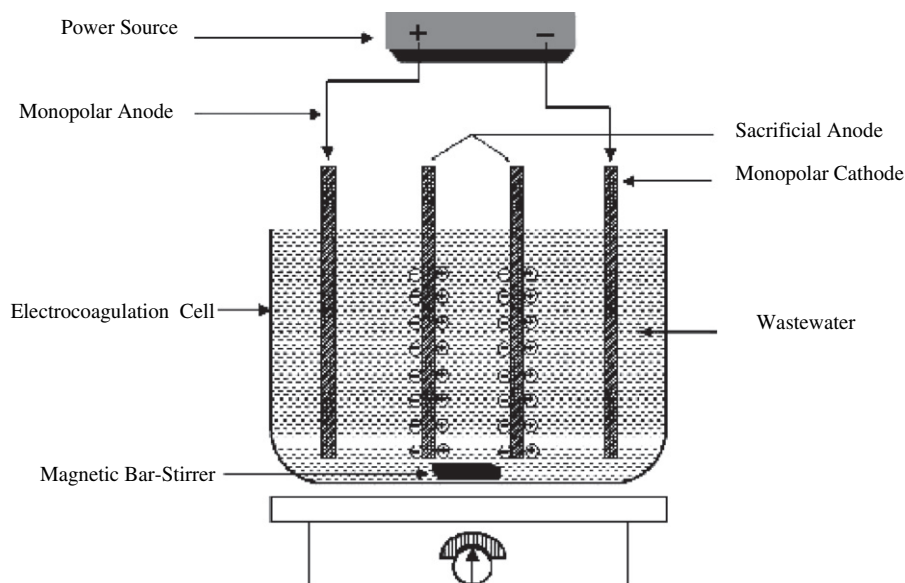


Fig. 3. Bench-scale EC reactor with bipolar electrodes in parallel connections.

degraded, respectively. The equation used to calculate the color removal efficiency in the experiments was:

$$\text{CR}\% = \frac{C_0 - C}{C_0} \times 100$$

where C_0 and C were the initial and present concentrations of the dye in the solution (mg l^{-1}), respectively.

The chemical oxygen demand (COD) was measured by a special kit (C1/25 COD 160) which contains potassium dichromate in sulphuric acid and silver sulphate as a catalyst.

3. Results and discussion

3.1. Effect of the current density

It is well-known that current not only determines the coagulant dosage rate but also the bubble production rate, size and the floc's growth [31,32], which can influence the treatment efficiency of the electrocoagulation. Therefore, the effect of current density on the pollutants removal was investigated. As shown in Fig. 4, an increase in current density from 40

to 120 A m^{-2} yields an increase in the efficiency of color removal from 16.38 to 99.18% because when the current density increases, the efficiency of ion production on the anode and cathode increases. Therefore, there is an increase in floc production in the solution and hence an improvement in the efficiency of color removal. For a solution with a dye concentration of 40 mg l^{-1} , the optimum current density was 120 A m^{-2} .

3.2. Effect of the time of electrolysis

To explore the effect of the operating time, the current density was held constant at 120 A m^{-2} . The color removal efficiency depends directly on the concentration of ions produced by the electrodes. When the electrolysis period increases, an increase occurs in the concentration of ions and their hydroxide flocs. Accordingly, as shown in Fig. 5, an increase in the time of electrolysis from 2 min to 5 min yields an increase in the efficiency of color removal from 49.53 to 99.21%. For a solution

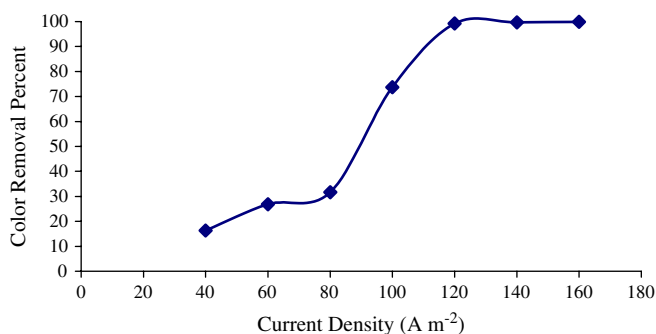


Fig. 4. Effect of the current density on the removal efficiency of Tartrazine with Fe/Fe electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $d = 1.5 \text{ cm}$, retention time = 20 min, and $\lambda_{\text{max}} = 428 \text{ nm}$.

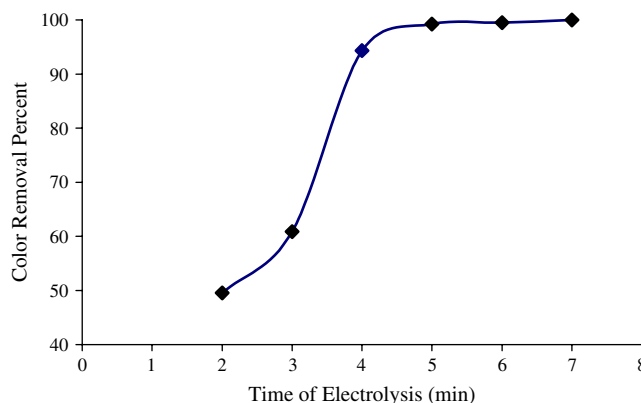


Fig. 5. Effect of the time of electrolysis on the removal efficiency of Tartrazine with Fe/Fe electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, retention time = 20 min, and $\lambda_{\text{max}} = 428 \text{ nm}$.

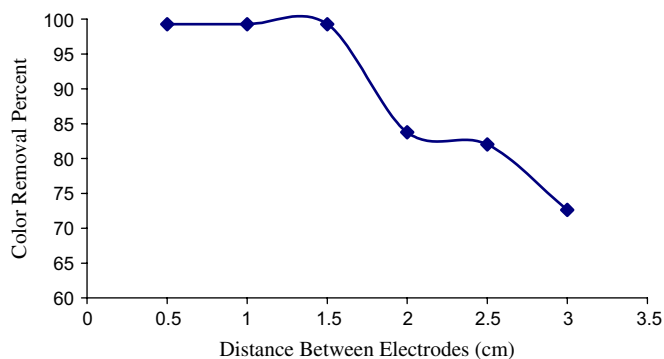


Fig. 6. Effect of the distance between the electrodes on the removal efficiency of Tartrazine with Fe/Fe electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, retention time = 20 min, and $\lambda_{\text{max}} = 428 \text{ nm}$.

having a dye concentration of 40 mg l^{-1} , and a treatment unit with current density of approximately 120 A m^{-2} , the optimum time of electrolysis was 5 min.

3.3. Effect of the distance between the electrodes

Once distance between the electrodes increases, the electrical current decreases. To achieve a certain current density, the voltage must be increased. On the other hand the IR-drop increases as the distance between electrodes increases. With increasing distance, less interaction of ions with hydroxide polymers is expected. In other words, decreasing both local concentration and electrostatic attraction are the reasons for decreasing the removal of Tartrazine. When the distance of the electrodes is increased from 0.5 to 3 cm, the removal efficiency decreases by about 26.64%. These results are shown in Fig. 6.

3.4. Effect of the initial concentration of the dye

As shown in Fig. 7, the removal efficiency decreases with the increase in the initial concentration of the dye. This could be due to the presence of intermediate products produced during the electrolysis process at higher concentrations, which then could compete with Tartrazine and water for the active sites on the electrode, or these intermediate products may be insoluble in water and would block the electrode's active sites.

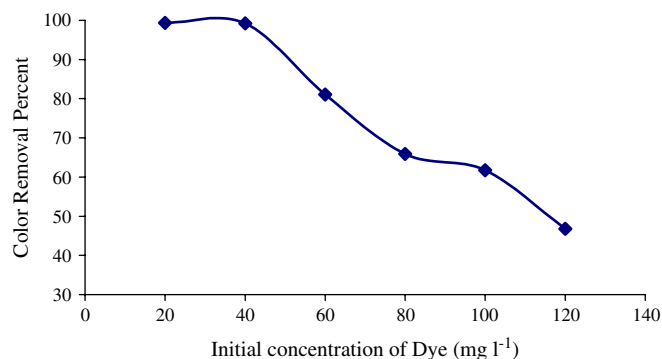


Fig. 7. Effect of the initial concentration of the dye on the removal efficiency of Tartrazine with Fe/Fe electrode pair. $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, and $\lambda_{\text{max}} = 428 \text{ nm}$.

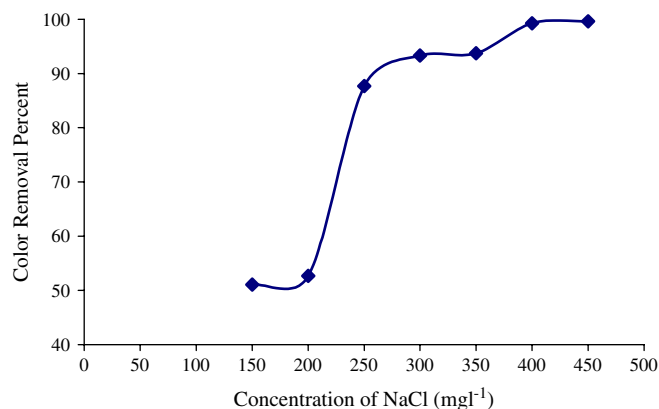


Fig. 8. Effect of the concentration of NaCl on the removal efficiency of Tartrazine with Fe/Fe electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, retention time = 20 min, and $\lambda_{\text{max}} = 428 \text{ nm}$.

On the other hand one of the most important pathways of color removal by EC process is adsorption onto metallic hydroxide flocs and the adsorption capacity of flocs is limited.

3.5. Effect of the electrolyte type and concentration

The influence of sodium sulphate concentration as an electrolyte on the color removal was tested for four different solutions (300, 400, 650 and 3000 mg l^{-1}) with an initial dye concentration of 40 mg l^{-1} and a current density of 120 A m^{-2} . At this condition the maximum removal efficiency was observed with 3000 mg l^{-1} sodium sulphate (CR% = 20.53%). However, the dye removal is faster when NaCl is used as an electrolyte. Because the existence of the sulphate ions would lead to the precipitation of Ca^{2+} or Mg^{2+} ions that form an insulating layer on the surface of the electrodes [20]. It should be mentioned that the presence of Cl_2 molecules which are formed could also act as bleaching agent. When the concentration of NaCl salt in solution increases, solution conductivity and current density are raised and the necessary voltage for reaching to a certain current density will be reduced, so the consumption of electrical energy is decreased. For lower concentrations, there is a decrease of that rate, probably because there are not enough ions to

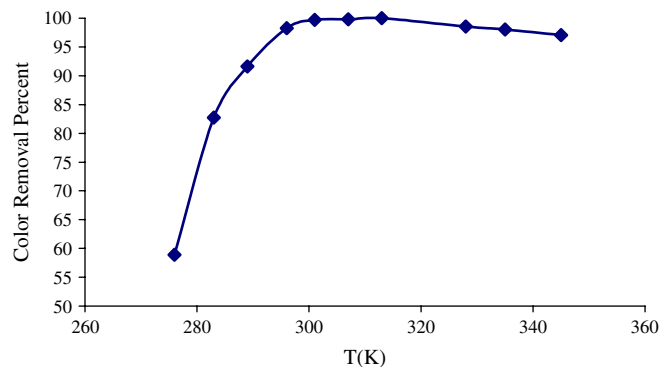


Fig. 9. Effect of the temperature on the removal efficiency of Tartrazine with Fe/Fe electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, retention time = 20 min, and $\lambda_{\text{max}} = 428 \text{ nm}$.

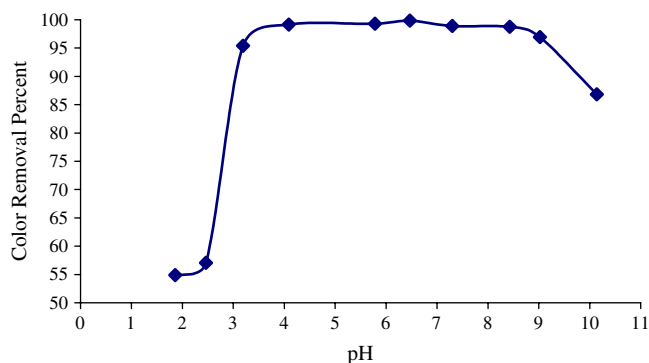


Fig. 10. Effect of the initial pH on the removal efficiency of Tartrazine with Fe/Fe electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, retention time = 20 min, and $\lambda_{\text{max}} = 428 \text{ nm}$.

conduct the current. Thus it may be concluded that increasing the electric resistance of the solution would decrease the efficiency of the process. The effect of NaCl concentration on the removal efficiency is shown in Fig. 8. For a solution having a dye concentration of 40 mg l^{-1} , and a treatment unit with current density of approximately 120 A m^{-2} , the optimum concentration of sodium chloride was 400 mg l^{-1} .

3.6. Effect of the temperature

As shown in Fig. 9, raising the temperature up to 28°C (301 K), causes an increase in the removal efficiency of Tartrazine. The reason could be due to increase in mobility and collisions of ions with hydroxide polymer. On the other hand, when the temperature is increased above 301 K, the removal efficiency decreases, because of the formation of unsuitable flocs or an increase in the solubility of precipitates.

3.7. Effect of the initial pH

It has been established that pH is an important parameter which influences the performance of the electrochemical process [33]. To examine its effect, the sample was adjusted to a desired pH for each experiment by using sodium hydroxide

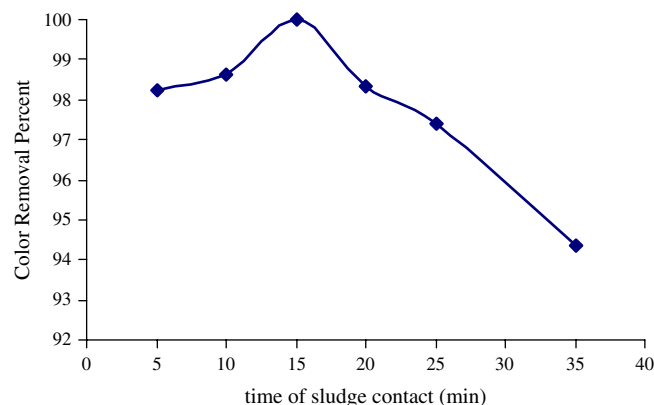


Fig. 12. Effect of the time of sludge contact on the removal efficiency of Tartrazine with Fe/Fe electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, and $\lambda_{\text{max}} = 428 \text{ nm}$.

or sulphuric acid. Fig. 10 demonstrates the removal efficiencies of Tartrazine as a function of pH. The maximum removal of Tartrazine was observed at a pH about 6.47. When the pH is increased above 6.47, the amount of hydroxide ions in solution increases. Consequently, some of the hydroxide ions are probably oxidized at the anode. This action prevents the production of the same proportion of iron ions [34] and therefore the removal efficiency of Tartrazine decreases. At low pH, the solution protons were reduced at the cathode to H_2 and the same proportion of hydroxide ions cannot be produced.

3.8. Effect of the sedimentation time

At the end of the electrocoagulation, the sample was poured into a 250 ml graduated cylinder for precipitation of flocs. As Fig. 11 shows, when sedimentation time is increased (up to 20 min), the removal efficiency of Tartrazine increases but after 20 min, the rate of color removal is relatively constant, since the metallic hydroxide flocs can attract contaminated particles during settling, but after 20 min the adsorption capacity of flocs becomes exhausted.

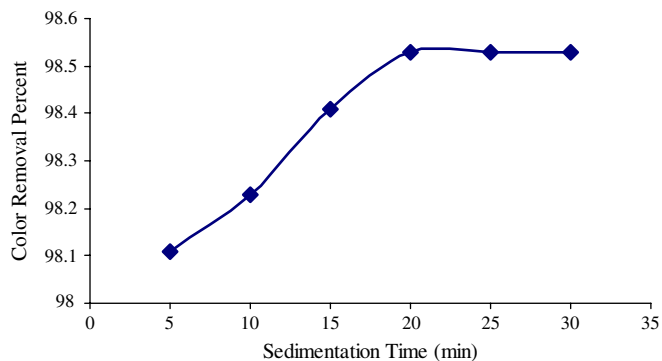


Fig. 11. Effect of the sedimentation time on the removal efficiency of Tartrazine with Fe/Fe electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, and $\lambda_{\text{max}} = 428 \text{ nm}$.

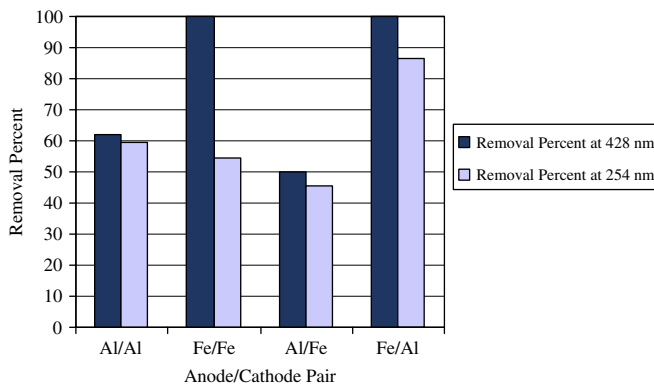


Fig. 13. Effect of the electrode material on the removal efficiency of Tartrazine. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 6 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, and retention time = 20 min.

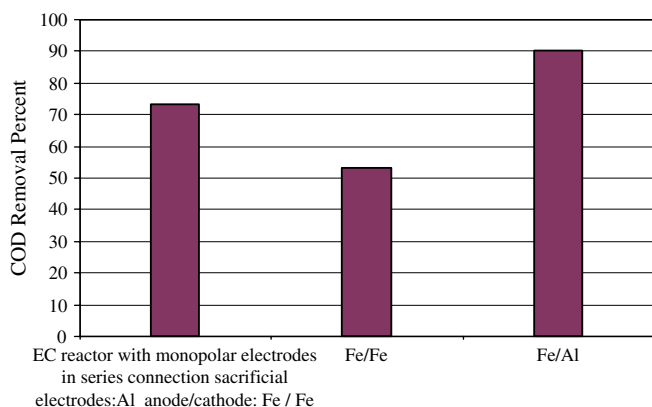


Fig. 14. Effect of cathode material and Al sacrificial electrodes on the COD removal. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 6 \text{ min}$, $i = 0.43 \text{ A}$, and retention time = 20 min.

3.9. Effect of the time of sludge contact

As Fig. 12 shows, when the time of sludge contact is increased (up to 15 min) the removal efficiency of Tartrazine increases. But, if the contact time is increased to more than an optimum amount, the removal efficiency decreases. This is because the capacity of the sludge for removal will be saturated and the removed dye in the former process would be gradually released from the sludge.

3.10. Effect of the electrode material

The most widely used materials, as sacrificial anode for electrocoagulation, are aluminium and iron. For comparative purpose, electrocoagulation has been carried out with both materials under exactly the same conditions. As Figs. 13 and 14

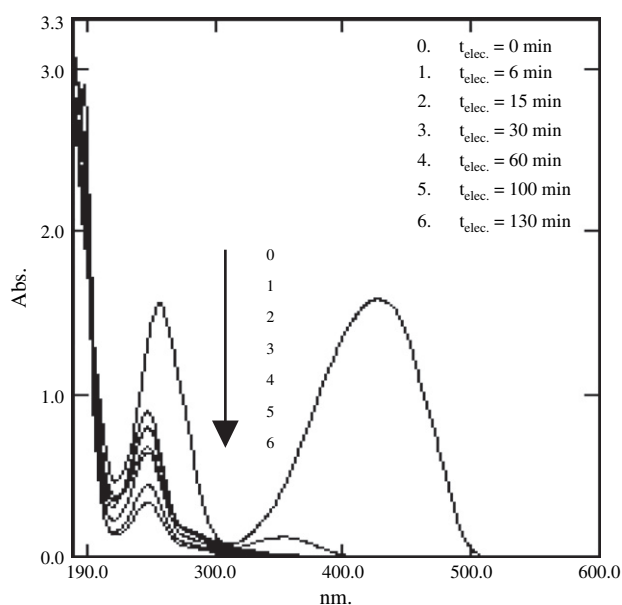


Fig. 15. Spectral changes of Tartrazine during electrolysis with Fe/Fe electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, and retention time = 20 min.

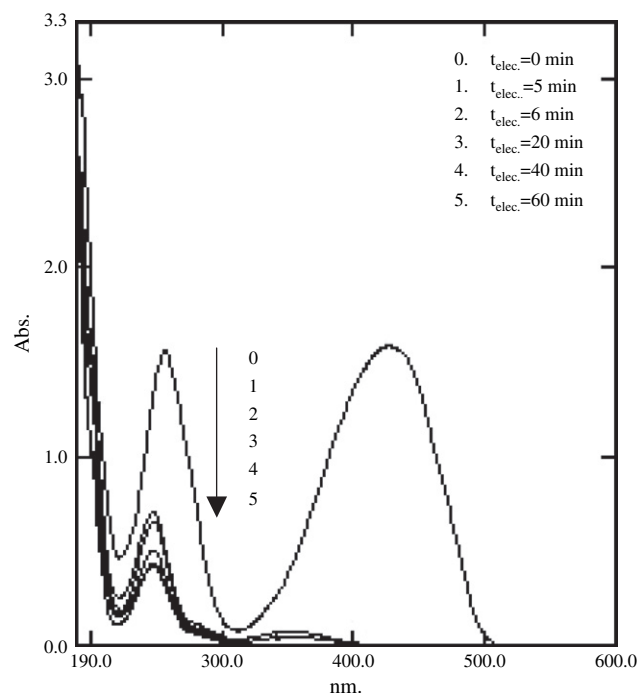


Fig. 16. Spectral changes of Tartrazine during electrolysis with Fe/Al electrode pair. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 120 \text{ A m}^{-2}$, $d = 1.5 \text{ cm}$, and retention time = 20 min.

show, electrocoagulation with Fe/Al (anode/cathode) was to form a better electrode pair due to high COD reduction and color removal. This probably results from the differences in the mechanisms of decolorization of the dye for iron and aluminium electrodes. The dye from solutions using aluminium electrodes is mainly removed by electrocoagulation,

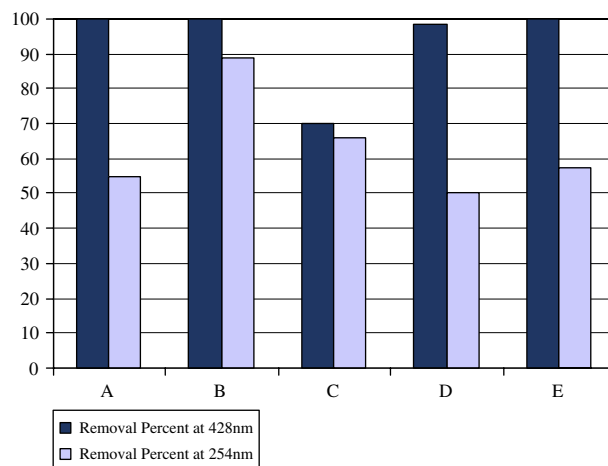
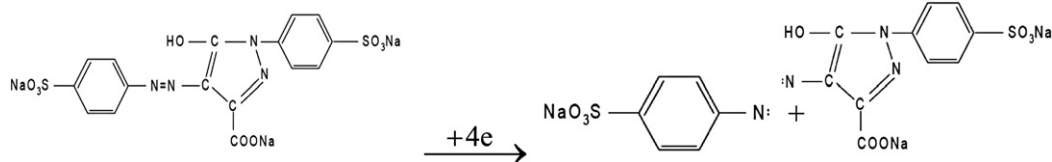


Fig. 17. Effect of the different electrode connections on the removal efficiency of Tartrazine. $C_{0[\text{Dye}]} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 60 \text{ A m}^{-2}$, $d = 1 \text{ cm}$, and retention time = 20 min. (A) EC reactor with monopolar electrodes in series connection; four Fe electrodes, (B) EC reactor with monopolar electrodes in series connection; sacrificial electrodes: Al; anode/cathode: Fe/Fe, (C) EC reactor with monopolar electrodes in series connection; four Al electrodes, (D) EC reactor with monopolar electrodes in parallel connection; four Fe electrodes, and (E) EC reactor with bipolar electrodes in parallel connection; four Fe electrodes.

while the dye removal by iron electrodes is due to the collective effect of electrocoagulation and electrooxidation. And another possible reason for this behavior could be the adsorption capacity of hydrous aluminium oxide for contaminants which is much lower than hydrous ferric oxides. The changes in the UV/vis absorption spectra of Tartrazine solutions as a function of time for EC treatment with Fe/Fe and Fe/Al electrodes pair are shown in Figs. 15 and 16, respectively. The color removal process may be due to the interaction of the dye molecules with the iron hydroxides which can remove the dye from water either by surface complexation or electrostatic attraction. Another mechanism may be a reductive process for example:



The COD of the initial dye solution was 30 mg l^{-1} . After decolorization process in the optimized condition, COD dropped to 14 mg l^{-1} for Fe/Fe pair, 3 mg l^{-1} for Fe/Al pair and 8 mg l^{-1} for an EC cell with monopolar electrodes in series connection with sacrificial aluminium electrodes and iron anode and cathode.

3.11. Effect of the different electrode connections

The effect of different electrode connections on the efficiency of Tartrazine is shown in Fig. 17. The experimental results showed that aluminium electrodes which were placed in series connection with iron electrodes were more effective and the changes in the UV/vis absorption spectra of Tartrazine solutions as a function of time for EC treatment with Fe–Al hybrid electrodes are shown in Fig. 18. This result is probably due to the higher potential required to achieve a certain current density in the series connection mode. Hence, this arrangement produces more flocs than the parallel connection mode, so the efficiency of color removal is higher. And on the other hand, the nascent Al^{3+} ions are more effective in coagulation process and it is well-known that aluminium hydroxide flocs are relatively large and having low density, which can be easily floated and separated [35].

4. Conclusions

Based on the results of experiments, the following conclusions may be obtained:

1. EC requires simple equipment and wastewater treatment by this method gives clear, colorless and odorless water.

2. The rate of decolorization of the dye solution (Tartrazine) by means of electrocoagulation was affected by the current density, initial pH of the solution, interelectrode distance, time of electrolysis, electrolyte type and concentration, solution temperature, different electrode connections and materials.
3. In this work it was shown that the electrocoagulation treatment achieves a fast and effective removal of Tartrazine. For 250 ml dye solution with COD of approximately 30 mg l^{-1} , dye concentration of 40 mg l^{-1} and NaCl concentration of 400 mg l^{-1} , color and COD elimination of 100% and 90% were obtained, respectively, when the pH was about 5.78, time of electrolysis was approximately

6 min, current density was approximately 120 A m^{-2} , iron anode and aluminium cathode and interelectrode distance was 1.5 cm.

4. The experimental results showed that an EC cell with monopolar electrodes in series connection with sacrificial aluminium electrodes and iron anode and cathode was more effective for the treatment process than parallel connection and a simple EC cell with iron anode and cathode.
5. For electrocoagulation of Tartrazine in aqueous solution, iron anode and aluminium cathode were found to form a better electrode pair due to high COD reduction and color removal.

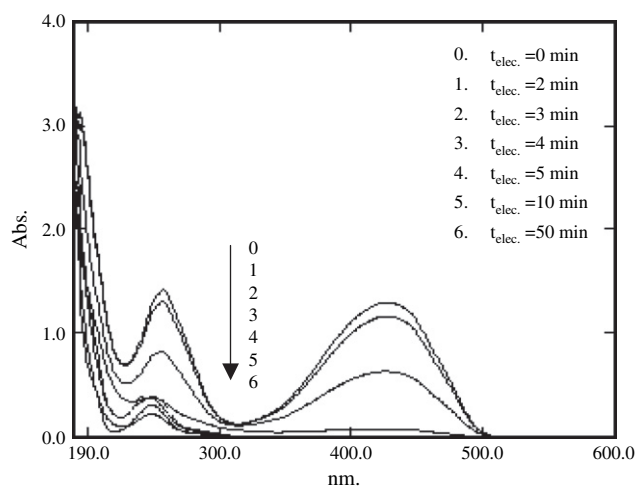


Fig. 18. Spectral changes of Tartrazine during electrolysis with monopolar electrodes in series connections of sacrificial Al electrodes; anode/cathode Fe/Fe. $C_{0(\text{Dye})} = 40 \text{ mg l}^{-1}$, $C_{\text{NaCl}} = 400 \text{ mg l}^{-1}$, $t_{\text{elec.}} = 5 \text{ min}$, $[i] = 60 \text{ A m}^{-2}$, $d = 1 \text{ cm}$, and retention time = 20 min.

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