Comparison between coagulation and electrocoagulation processes for the removal of reactive black dye RB-5 and COD reduction

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ABSTRACT

This study examined the possibility of removing reactive black-5 (RB-5) dye and chemical oxygen demand (COD) from simulated wastewater using chemical coagulation technique, in comparison with electrocoagulation technique. Ferric sulfate $Fe_2(SO_4)_3$, ferrous sulfate ($FeSO_4$), ferric chloride ($FeCl_3$), polyaluminum chloride (PACl) and alum ($Al_2SO_4 \cdot 5H_2O$) were used as coagulants. Optimum pH, coagulant dosages, and initial concentration were determined by jar tests, and the maximum dye and COD removal were obtained. The maximum removal efficiency was found to be 98% and 90% for dye and COD, respectively, where the best coagulant was $Fe_2(SO_4)_3$ at optimum pH and dosage of 4,200 mg/L. On the other hand, the electrochemical cell was designed using aluminum plates as the anode and cathode with dimensions of (7 cm³ × 4 cm³ × 0.3 cm³). Many experimental runs were done at a different operating condition (pH, NaCl concentration, initial concentration, and electrical supply voltage) to study the removal performance of electrocoagulation for RB-5 dye and COD removal. The results indicated that at an optimum pH of 2 and NaCl concentration 1,000 mg/L the maximum removal was 100% and 93% for dye and COD, respectively. The study proved that the percentage removal of RB-5 and COD better by electrochemical than by chemical coagulation.

Keywords: Chemical coagulation; $Fe_2(SO_4)_3$; Electrocoagulation; NaCl; Reactive black-5 (RB-5); Chemical oxygen demand (COD)

1. Introduction

The reuse and recycle are the main factors to face the challenges of depletion of our reserves and environmental troubles in the new world age where. Wastewater is not only considered one of the sources of the main pollutants that damage the environment but also leads to the deficit of the earth's water. A lot of industrial processes are managed at the spend of plenty of freshwaters, which is haggard as wastewater and needs to be treated to eliminate or uproot the pollutants and to produce pure treated water to be reused as a sustainable environment natural resource [1].

Textile industry wastewater is strongly polluted by many chemical pollutants, its percentage emission to nature is 70% the wastewater from textile processing as follows: water bath from preparation, dyeing, finishing, slashing, printing, coloring, and other operations, In the textile industries and during the dyeing process a lot of freshwaters are used which are the main consumers of synthetic dyes, and the industrial wastewater contains many kinds of pollutants chemical oxygen demand (COD), dyes, suspended solids, high temperature, salts, and variable pH. These pollutants damage the environment, so, the wastewater should be treated before it is drained to the rivers or seas. However, these pollutants could be treated for minimum harmful for the environment or better behavior after they flow to nature, and the textile industry utilizes about 10,000 dyes and pigments [2].

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During the dyeing process the pollutants are so barely removed from effluent waste discharge, as a result there should be a concern with these pollutants. Dyes may enter into the environment through their production, and in this case, only one method of treatment is not sufficient for a perfect treatment. Combined treatment with physical, chemical, and biological treatment is used to remove the heavy pollutants from textile wastewater [3,4].

More recently, studies have focused on color and COD removal from real combined textile wastewater and reactive dye bath effluent [5] as well as aqueous textile dyes [6].

Textile wastewater treatment conventional methods are the combination of chemical, physical, and biological methods. Because of the high molecular weight dyestuffs and the presence of aromatic rings, the biological method reveals slow and low degradation [7]. Adsorption and precipitation are relatively time-consuming and costly physical methods, while most chemical methods such as additions of aluminum sulfate and chlorine can generate secondary pollutants. Some other advanced technologies such as photo-oxidation by UV/H_2O_2 or UV/TiO_2 also generate secondary pollutants [8].

Chemical coagulation is an essential part of drinking water and wastewater treatment. In drinking water, it is used for the clarification of water, using coagulant agents mainly for municipal water treatment. Physical separation techniques for treating colloidal wastewater, such as sedimentation, are not adequate since colloidal systems contain particles from 0.1 μm to 1 nm. This makes the settling velocity range from 0.3 to 3 m/y and implies that the processing time will be extremely long. Chemicals such as lime ($Ca(OH)_2$, most commonly used for drinking water treatment), soda ash (Na_2CO_3) and organic polyelectrolytes (starch, polysaccharide gums or synthetic polyelectrolytes) are used in drinking water processing to produce quick-forming, dense and rapid-settling flocs to enhance the removal efficiency of suspended solids [9].

Electrocoagulation (EC) is an electrochemical technology closely related to chemical coagulation. EC is based on the application of an electric current to a sacrificial anode for example, Al or Fe. This produces the electro-dissolution of the anodes, generating a clot formed by a wide variety of hydroxides, oxy-hydroxides, and polymeric hydroxides of Fe or Al during the wastewater treatment. This clot is capable of aggregating suspended particles as well as destabilizing and adsorbing dissolved or emulsified contaminants.

Other phenomena involved in the process of electrocoagulation are associated with the direct or indirect oxidation of organic matter and the generation of hydrogen bubbles in the cathode that helps the flotation of flocs formed. The formed sludge is usually separated by sedimentation and subsequent filtration. There are clear advantages in terms of (i) use of a smaller amount of chemical reagents, which does not increase the water salinity of treated wastewater, (ii) generation of a smaller quantity of sludge, and (iii) ease of operation, automation, and lower space requirements [10].

This research seeks to perform an experimental investigation using chemical coagulation and electrocoagulation techniques on the removal of a reactive textile dye reactive black-5 (RB-5) and COD from the wastewater. Several variables were studied concerning the impacts of coagulant type, pH, coagulant dosage, electrolyte type, pH, current density, electrolyte concentration, COD, and initial dye concentration on the effectiveness of dye removal and COD removal.

2. Materials and methods

2.1. Preparation of RB-5 dye and COD solutions

RB-5 with molecular weight 991.82 g/mol, molecular formula $C_{26}H_{21}N_5Na_4O_{19}S_6$ was used in this study as a model pollutant for textile dyes. The molecular structure is shown in Fig. 1. Dye stock solution was prepared by dissolving 1 g of dye in 1 L distilled water, the final concentration is 1,000 mg/L. The experimental solutions were obtained by diluting the dye stock solutions in accurate proportions to obtain different initial concentrations. Simulated wastewater polluted with COD was prepared using sucrose $C_{12}H_{22}O_{11}$, molecular weight 342 g/mol. When the concentration of sucrose is 500 mg/L, the COD concentration is 560 mg/L [11].

2.2. Chemicals

Ferrous sulfate [$FeSO_4$], ferric sulfate [$Fe_2(SO_4)_3$], ferric chloride [$FeCl_3$], aluminum sulfate or alum [$Al_2(SO_4)_3 \cdot 14H_2O$] and polyaluminium chloride or PACl [$AlCl_3 \cdot 6H_2O$], were used as a coagulant using the stock solution (1,000 mg/L). Sodium chloride NaCl was used as the electrolyte. For pH adjustment, sodium hydroxide, NaOH, and hydrochloric acid were used. All the chemicals used were supplied from BDH Company, England.

2.3. Chemical coagulation technique

Jar tests were used in the coagulation process to find the optimum pH, initial concentration, coagulant doses (for each coagulant). Jar tests were done with six bakers size 1,000 ml. RB-5 and COD solutions were prepared previously. pH was adjusted to the desired value by the addition of 0.1 M HCl or/and 0.1 M NaOH solutions. The samples were mixed for 2 min, 200 rpm (rapid mixing), then slow mixing for 30 min and 50 rpm. The coagulation process was taken place, with the consistency of flocculants, precipitation, and settlements. The mixers were left for 30 min to allow the agglomerates to settle down. Samples were taken from 1–2 mm below the solution surface. Ali et al. [11] followed the same procedures.

2.4. Electrocoagulation cell

The experiments were conducted in 1L beaker capacity. The anode and cathode were positioned vertically and in parallel to each other with an inter-electrode gap of 3.5 cm. Both anode and cathode were aluminum plates 7 cm² × 4 cm², the thickness of the plate was 0.3 cm. These electrodes were dipped in the electrolyte solution. Stabilized DC power supply was used as the source of electrical current. A schematic illustration of the experimental setup is shown in Fig. 2. The solution was constantly stirred at 200 rpm using a magnetic stirrer to maintain a uniform concentration of

the electrolyte solution, a stirrer Model LMS-1003 Korea was used.

2.5. Analytical methods

The mixture of RB-5 dye and COD of the samples were drawn, centrifuged using an electrical centrifuge
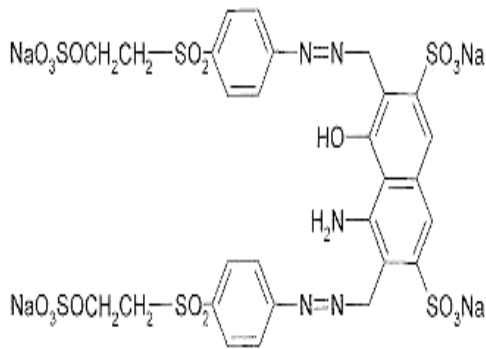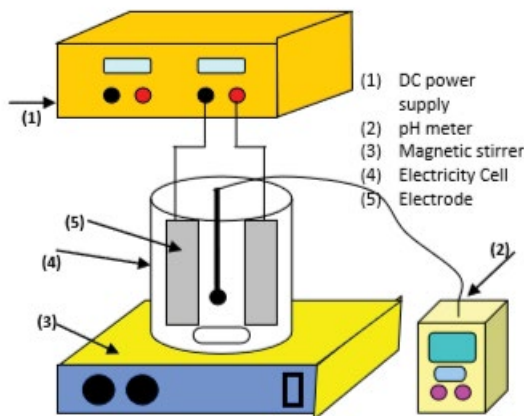

Fig. 1. RB-5 dye molecular structure.

Fig. 2. Experimental setup.

at 5,000 rpm for 20 min and the unabsorbed supernatant liquid was analyzed for the residual dye concentration using ELICO UV-Vis Spectrophotometer (CI 73) at a max wavelength of 595 nm. The COD of the samples was determined according to the dichromate digestion method [12]. RB-5 dye and COD removal efficiencies were calculated by:

$$\text{Removal}\left(\%\right) = \frac{C_r - C_t}{C_r} \times 100 \tag{1}$$

where C_r and C_t are the concentration in raw and treated solutions, respectively.

All the experiments were repeated triple and the average concentration was taken.

3. Results and discussion

3.1. For chemical coagulation

3.1.1. pH effects

To study the effect of pH on RB-5 dye and COD removal efficiency, the dosage of each coagulant was kept constant at 150 mg/L, while varying pH of samples from 2 to 12 using 0.1 M HCl or/and 0.1 M NaOH. RB-5 dye and COD concentrations were fixed during the experiment at 50 and 500 mg/L, respectively.

The maximum removal of RB-5 is 90%, 85%, 91%, 20% and 92% at pH 8, 10, 4, 9 and 4 for alum, PACl, $FeCl_3$, $FeSO_4$ and $Fe_2(SO_4)_3$, respectively as shown in Fig. 3a. However, for COD, the percentage removal was 71%, 50%, 76%, 12% and 81% at pH 8, 10, 4, 9 and 4 for alum, PACl, $FeCl_3$, $FeSO_4$ and $Fe_2(SO_4)_3$, respectively as shown in Fig. 3b.

It could be noticed that when alum was used; the percentage removal of dye increased by a large value from 0 to 90% when pH increased from 2 to 8, however, it decreased when pH was more than 8. For PACl, the percentage of removal increased from 15% to 85% with pH from 2 to 10 then it decreased. When $FeCl_3$ used the removal was 29% to 91% at pH 2 and 4, respectively. For $FeSO_4$ the removal was 0% at pH 2, whereas the removal was 20% when pH was 9.
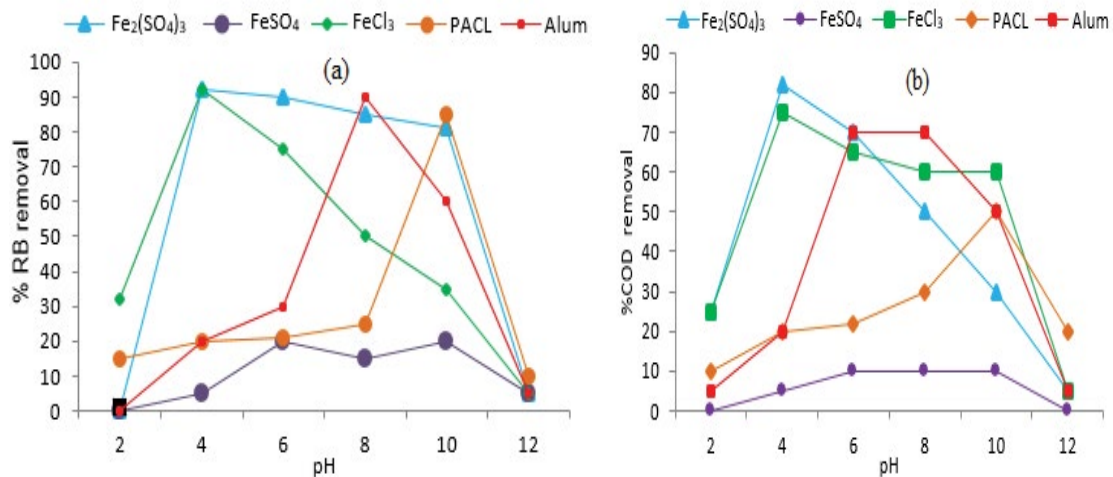

Fig. 3. Removal of (a) RB-5 and (b) COD as a function of pH.

Finally, when $Fe_2(SO_4)_3$ was used the removal increased from 0% to 92% when pH increased just from 2 to 4. COD removing was as follows 82%, 10%, 75%, 50%, and 70% for $Fe_2(SO_4)_3$, $FeSO_4$, $FeCl_3$, PACl and alum, respectively. Optimum pH was 4, 9, 4, 10, and 8 as in dye removing.

As a result, the maximum removal efficiency (%) using different coagulants were as follow:

$Fe_2(SO_4)_3$ (92 ± 0.01) = $FeCl_3$ (90 ± 0.01) > alum (90 ± 0.002) > PACl (85 ± 0.001) > $FeSO_4$ (20 ± 0.003).

For COD the removal efficiency follows the below sequence:

$Fe_2(SO_4)_3$ (82 ± 0.001) = $FeCl_3$ (75 ± 0.01) > alum (70 ± 0.003) > PACl (50 ± 0.002) > $FeSO_4$ (10 ± 0.003).

Daud et al. [14] studied the removal of suspended solids (SS), color, COD and oil and grease from wastewater. The result shows that, at the optimum dose of PACl (300 mg/L), the percentage removal of color, COD, and oil and grease respectively 95%, 75%, and 97% compared to only 92%, 92%, 53%, and 99% at the optimal dose alum (500 mg/L), 95%, 93%, 63%, and 97% at the optimum dose of ferric chloride (350 mg/L) and 88%, 88%, 54%, and 94% at the optimum dose of ferric sulfate (450 mg/L) [13–15].

3.1.2. Coagulant dosage effects

To study the effect of coagulant dose, the pH was fixed at optimum value from the previous experiment and the initial concentrations of RB-5 and COD were 50 and 500 mg/L, respectively. The coagulant doses were varied in the range (5, 25, 50, 100, 150, 200, and 250 mg/L). The optimum coagulant dose was found to be 200 mg/L. Above this value, there is no increase in the percentage removal of RB-5 and COD. The maximum removal efficiency was 95%, 20%, 88%, 90%, and 92% for $Fe_2(SO_4)_3$, $FeSO_4$, $FeCl_3$, PACl and alum, respectively. COD removing reported the following results 82%, 10%, 78%, 75%, and 68% for $Fe_2(SO_4)_3$, $FeSO_4$, $FeCl_3$, PACl and alum respectively. The same results were reported by some other researchers (Verma et al. [16]). Fig. 4a and b reveal the maximum dye and COD removing vs. the optimum coagulant dosage. For the dosage more than the best dosage, the efficiency curve decreased which is probably

due to the re-stabilization phenomenon. This is consistent with the findings reported by Ali et al. [17].

The maximum removal efficiency (%) for RB-5 and COD follow in term of coagulant type and dose follow the below sequence:

For RB-5: $Fe_2(SO_4)_3$ (95 ± 0.001) > alum (90 ± 0.003) > PACl (90 ± 0.003) > $FeCl_3$ (88 ± 0.001) > $FeSO_4$ (20 ± 0.003).

For COD: $Fe_2(SO_4)_3$ (82 ± 0.003) > $FeCl_3$ (78 ± 0.004) > PACl (75 ± 0.01) > alum (62 ± 0.003) > $FeSO_4$ (10 ± 0.005).

3.1.3. Initial concentration effects

The experiments were performed to determine the effect of initial RB-5 and COD concentration on percentage removal, using a constant coagulant dosage (the optimum is 200 mg/L). The results are illustrated in Figs. 5a and b. For RB-5 with increased of initial concentration from 25 to 75 mg/L the removal efficiency increased from 90% to 98%; 18% to 20%; 80% to 92%; 80% to 90% and 80% to 92% 88% for $Fe_2(SO_4)_3$, $FeSO_4$, $FeCl_3$, PACl and alum, respectively. While for dye concentrations more than 75 mg/L the efficiency decreased to 86%, 9%, 65%, 58%, and 60% for the same sequence, for initial concentration of 150 mg/L. In the case of COD the removal curves changed in the same trend as shown in Fig. 5a where the maximum percentage removal which is 90% is obtained at an initial concentration of 1,000 mg/L for $Fe_2(SO_4)_3$.

The consequence of removal efficiency (%) in term of initial concentration using different coagulants follow the below pattern:

For RB-5: $Fe_2(SO_4)_3$ (98 ± 0.004) > $FeCl_3$ (92 ± 0.002) = alum (92 ± 0.002) > PACl (90 ± 0.005) > $FeSO_4$ (20 ± 0.006).

For COD: $Fe_2(SO_4)_3$ (90 ± 0.002) > $FeCl_3$ (78 ± 0.02) > alum (76 ± 0.006) > PACl (60 ± 0.005) > $FeSO_4$ (20 ± 0.003).

According to the data of Hoong and Ismail [18], the removal degree was associated with the initial concentration of the selected dyes, and the smallest removal degree was obtained for the smallest concentrations of the dyes. This result is inconsistent with the results obtained for the used coagulants in this study, which showed the smallest values of removal efficiency for smaller initial concentration [18].
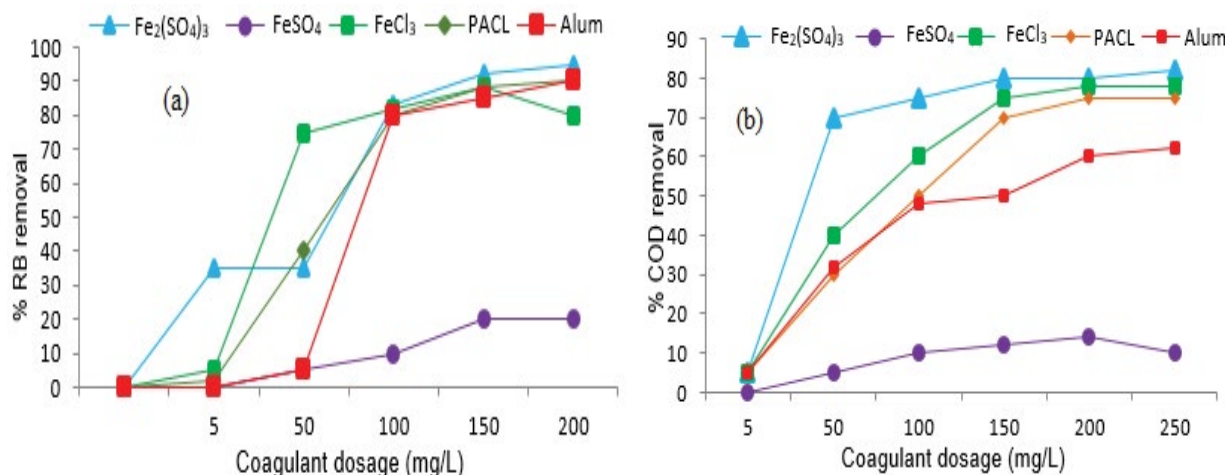

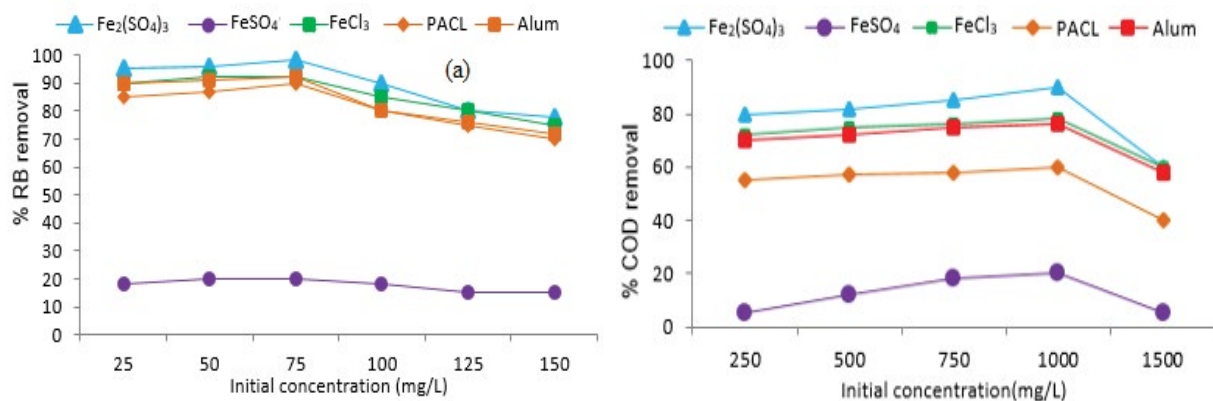Fig. 4. Removal of (a) RB-5 and (b) COD as a function of coagulant dosages.

Fig. 5. Removal of (a) RB-5 and (b) COD as a function of initial concentration at optimum pH and dosage.

3.2. For electrocoagulation

3.2.1. pH effects

The effects of pH are given in Fig. 8, where the removal of RB-5 and COD increased with decreased the pH of the solution. The reason may be due to the increase of the hypochlorite ion in an acidic medium and in low pH solutions the presence of hypochlorous acid (HOCl), which possesses higher oxidation potential than that of hypochlorite ion. This behavior is attributed to the aluminum hydroxide product being soluble in acidic or basic solution due to its amphoteric nature.

It can be ascertained from Fig. 9 that, the percentage of RB-5 reduction had increased from 42% at pH 4 to 98% ± 0.007% at pH 2 for NaCl concentration of 500 mg/L and voltage of 5 V after about 30 min reaction time. The maximum removal percentages reached 65% ± 0.005% for COD at pH = 2 as shown in Fig. 6. Some researchers achieved maximum removal of color and COD in the pH range of 1.5–4 [19].

3.2.2. NaCl concentration effects

The effect of changing the electrolyte concentration (250, 500, and 1,000 mg/L) on dye and COD removal efficiency at optimum pH, fixed dye, and COD initial concentration of 50 and 500 mg/L, respectively, and voltage of 5 V is illustrated in Fig. 7. It is clear from the figure that, the RB-5 removal was increased when NaCl concentration increased from 250 to 500 mg/L, and when NaCl concentration increased up to 1,000 mg/L, dye removing increased and equal 97% ± 0.02%, because the conductivity of solutions rise when NaCl concentration increased and resistance decreased. So the passing current increased and the produced amount of metallic hydroxide and dye removal increased, also hypochlorite ions produced at the anode causes the increase in dye removal by oxidation of dye molecules [20]. COD removed by 85% ± 0.001% when NaCl concentration increased to 1,000 mg/L at pH 2 and current density 44 mA/cm², respectively. It is clear that increasing the chloride concentration increases the removal due to increased mass transport of chloride ions to the anode surface and also increased diffusion in the diffusion

layer of the anode. As a result, more amount of chlorine/hypochlorite will be generated. Hence the rate of removal was increased [21].

3.2.3. Initial concentration effects

The RB-5 and COD percentage removal decreased with an increase in the initial concentration, because the number of metal hydroxide flocs formed may be insufficient to absorb all of the pollutant molecules when the initial pollutant concentrations are rather high [22]. Dye solutions with the following concentrations (50, 75, 100, 200, and 300 mg/L) and five COD concentrations of (250, 500, 750, 1,000 and 1,500 mg/L) were used to test the effect of initial dye and COD concentration on degradation efficiency at optimum pH and NaCl concentration. Results showed that, when dye concentration increased from 50 to 100 mg/L, dye removals increased a little bit from 97% ± 0.002% to 98% ± 0.001% and from 85% ± 0.01% to 89% ± 0.03% for COD when the initial concentration increased from 500 to 750 mg/L (Fig. 8). However, when the concentration raised to those concentrations, the removal decreased to 55% ± 0.008% and 50% ± 0.04% for dye and COD, respectively because of dye molecule adsorption on metallic hydroxide flocs. At the same voltage, the amount of Al^{3+} that released from anode to the solution was constant, thus, the amount of flocs is also constant, therefore, a specific amount of dye molecules would adsorb into the flocs, no more dye molecule adsorption, therefore, the efficiency of dye and COD removal decreased when concentration increased. This may be also explained by the fact that, under galvanostatic condition, the production rate of chlorine/hypochlorite is constant; however, the active chloro species have a nonselective property, which results in an attack on intermediate compounds produced rather than on the chromophore of the initial dye molecule while increasing the initial dye concentration [23].

3.2.4. Voltage variation effects

The effect of varying the voltage in ranges 3, 4, 5, and 10 V at optimum operation conditions were investigated. RB-5 removal increased when the voltage increased. The removal efficiency was 90%, 93%, 95%, and 98% when the
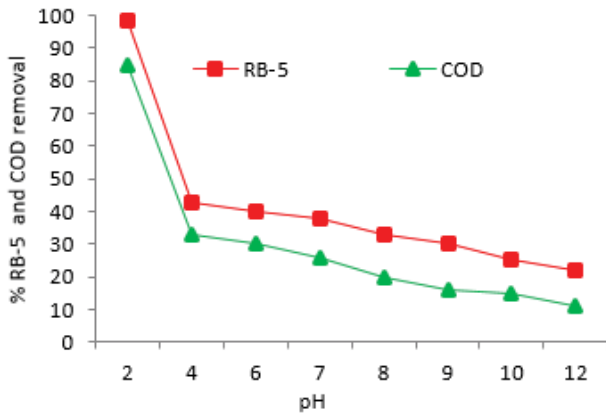
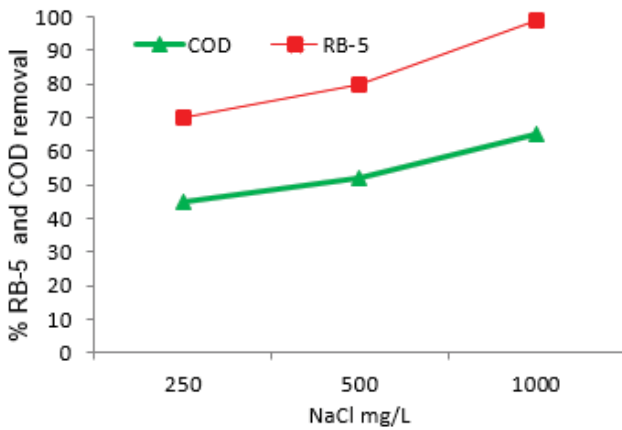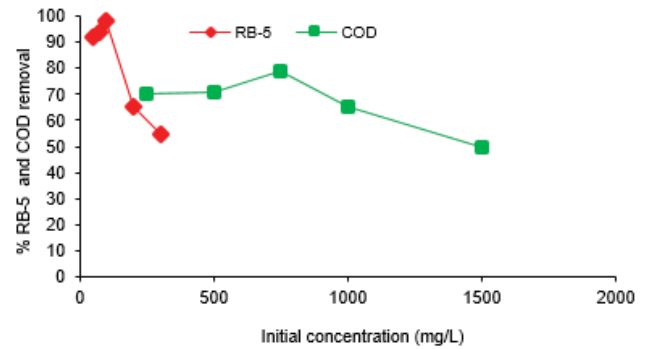Fig. 6. The percentage removal of dye and COD, as a function of pH.

Fig. 8. The percentage removal of RB-5 and COD as a function of initial concentration.

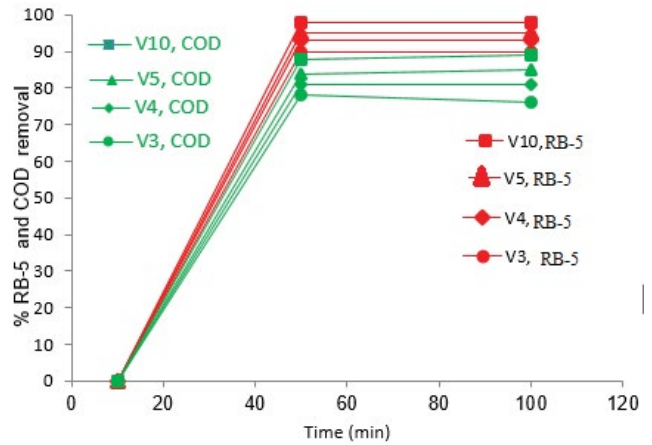Fig. 7. The percentage removal of RB-5 and COD, as a function of NaCl concentration.

Fig. 9. The percentage removal of RB-5 and COD as a function of voltage.

voltage is 3, 4, 5, and 10 V, respectively as shown in Fig. 9. The removal of COD was greatly increased with increasing voltage from 3 to 10. The maximum removal is registered at 10 V with percentage removal 100% and 93% ± 0.008% for dye and COD, respectively. However, after 60 min the removal was at a constant rate. There was no significant difference in the results at 3 to 10 V. It is clear that dye and COD removals depend on many factors first of all aluminum dosage which consequently depended on the voltage and time of reaction. Because of that, when the voltage increase, coagulants, and flocs amount increase so dye and COD are removed. The results clearly showed that increasing voltage decreases the charge loading for the degradation of RB-5 and COD because of the increased production of chlorine at a higher voltage. The same results were obtained by Govindaraj et al. [24].

3.3. Measuring of sludge produced

For industrial applications, it is necessary to optimize the process using reasonable consumption of coagulant and quantity of obtained sludge. In general, the volume and the characteristics of the sludge produced during the coagulation/flocculation process are highly dependent on the specific coagulant used and on the operating conditions [25]. The wet sludge volume at the bottom of the jar test beakers after the coagulation process was used to quantify the volume of sludge generated in the present study. The sludge resulting from the coagulation process is measured using Imhoff cone at optimum pH resulting from previous experiments, initial dye and COD concentrations of 50 and 500 mg/L, respectively, and coagulants dosage of 200 mg/L as shown in Fig. 10a and b. A maximum of 60, 40.1, 29, 24, 10 and 6 mL settled sludge/1 L of the sample was observed in the case of PACl, alum, $Fe_2(SO_4)_3$, Fe_2SO_4, $FeCl_3$, and EC respectively for COD. In the case of RB-5, the volume of sludge is 40, 36.5, 18, 15, 8, and 4, respectively.

For coagulants, the volume of sludge produced was less when $FeCl_3$ used. Significantly reduced sludge production using $FeCl_3$ may be explained by the fact that when dissolved it produces Fe^{3+} and Cl^- which they considered as strong oxidizing agents that work together to remove RB-5 and COD from wastewater. However, the EC process produces less sludge compared to all chemical coagulants,
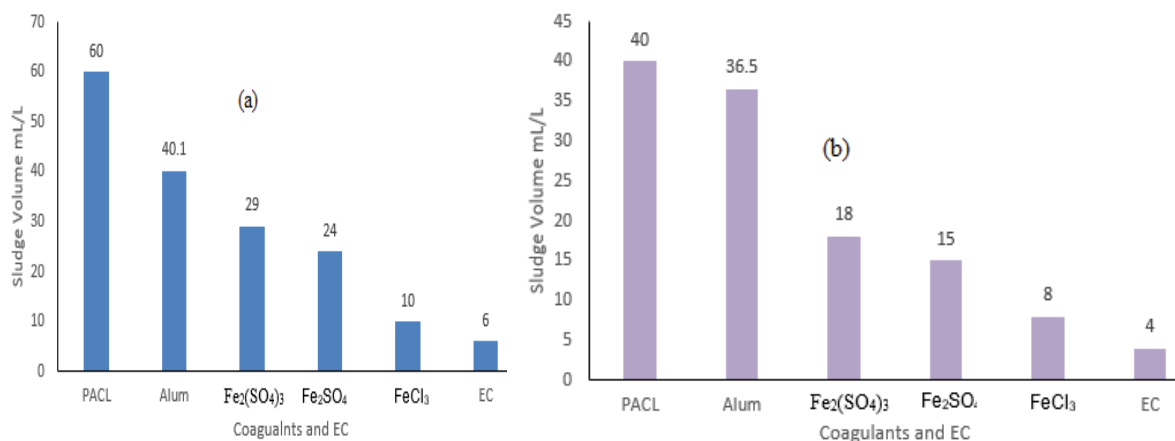
Fig. 10. Volume of sludge produced from (a) COD and (b) RB-5 removal at coagulant dose = 200 mg/L and dye initial concentration = 500 mg/L.

and since this process involves no addition of a chemical to form the flocs [26].

4. Conclusions

After treatment by both electrocoagulation and chemical coagulation processes, the obtained removal efficiency of RB-5 and COD are very high, especially for dye where the values are 98% ± 0.007% and 90% ± 0.002% for dye and COD using chemical coagulation while it's 100% and 93% ± 0.008% using electrocoagulation. The optimum pH for both processes occurred under the acidic conditions where the value is 4 and 2 for chemical coagulation and electrocoagulation, respectively. The removal efficiency increased with increased dosages of coagulants and electrolytes until they reach some values, then the removal efficiency remained constant. The removal efficiency, increased with an increase in the initial concentration to certain values then decreased.

The performance of electrocoagulation is relatively better than chemical coagulation. To overcome problems associated with chemical coagulation, EC could be advocated as a novel approach in removing a wide range of wastewater pollutants such as suspended solids and color. Parameters such as pH, current density, applied voltage, wastewater type, type of electrodes, number of electrodes, and size of the electrodes as well as configurations of metals used could be considered for future studies.

Acknowledgments

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