Treatment of pulp and paper industry bleaching effluent by electrocoagulant process

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ABSTRACT

The experiments were carried out in an electrocoagulation reactor with aluminum as sacrificial electrodes. The influence of electrolysis time, current density, pH, NaCl concentration, rotational speed of the stirrer and electrode distance on reduction of color, COD and BOD were studied in detail. From the experimental results, 15 mA/cm² current density, pH of 7, 1 g/l NaCl, 100 rpm, 28 °C temperature and 3 cm electrode distance were found to be optimum for maximum reduction of color, COD and BOD. The reduction of color, COD and BOD under the optimum condition were found to be 94%, 90% and 87% respectively. The electrode energy consumption was calculated and found to be varied from 10.1 to 12.9 kWh/m³ depending on the operating conditions. Under optimal operating condition such as 15 mA/cm² current density, pH of 7, 1 g/l NaCl, 100 rpm, 28 °C temperature and 3 cm electrode distance, the operating cost was found to be 1.56 US \$/m³. The experimental results proved that the electrocoagulation is a suitable method for treating bleaching plant effluents for reuse.

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1. Introduction

The pulp and paper making industry is a water intensive industry; it is ranked third based on water consumption after metals and chemical industries. The Indian pulp and paper industry consuming 100–250 m³ freshwater/ton of paper production depend on the process [1]. The volume of wastewater generated by pulping and bleaching operations is approximately in the range of 75–225 m³/ton of product [2]. The volume of effluent generated and its characteristics are normally dependent on process used for paper making and the effectiveness of the process. The discharge of highly colored wastewater into the ecosystem involves environmental problems like aesthetic pollution and perturbation of aquatic life. The State Pollution Control Board of Tamil Nadu, India, has directed the industries to implement zero discharge facilities [3]. The continuous depletion of groundwater, shortage in rainfall and huge consumption of water by paper and pulp

industries has necessitated the recycling and reuse of processed water.

The characteristics of the wastewater generated from the pulp and paper industry depend upon the types of raw material, types of process technology applied, internal recirculation of the effluent and management practices. In order to select and design a suitable treatment technique for reuse of effluent in the process application, a complete knowledge of characteristics of the particular industrial effluent is necessary. The general characteristics of the wastewater produced at various process stages and pollution sources of various industries located in and around Erode district, Tamil Nadu India, where samples were collected are given in Fig. 1.

In wood preparation section, the raw materials are chopped into small pieces using suitable techniques. Then the pieces are sent to the pulping section where sulfate process method is applied to produce pulp. In pulp bleaching section, pulps are bleached with liquid chlorine and liquid oxygen followed by a second bleaching with chlorine dioxide and third bleaching with hydrogen peroxide. The bleached pulp is then passed to paper making section. The waste water from wood preparation sections contains mostly suspended solids, dirt, fibers, etc., so this waste water is filtered and reused again. The effluent from pulping section is called black liquor effluent, which is approximately 20–25 m³/ton of paper production. The black liquor effluent contains more amounts of cooking chemicals such as sodium hydroxide and sodium sulfate, lignin and other extractives. In industries the effluent from pulping section is sent to chemical recovery plant where chemicals are recovered and reused
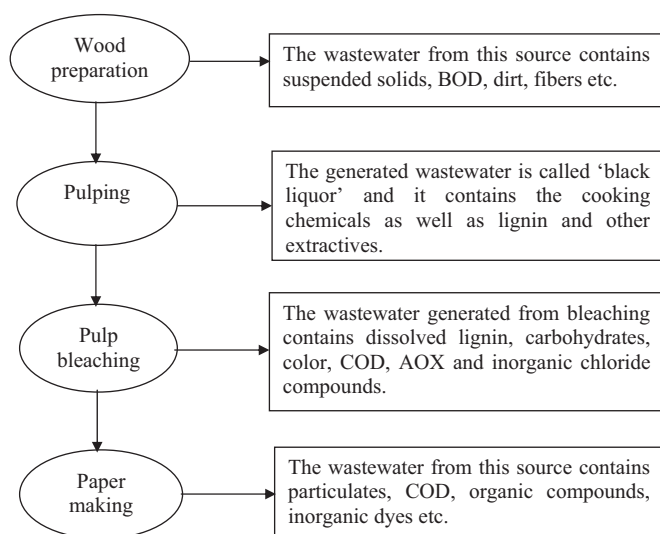
Fig. 1. Sources that contribute in the effluent from pulp and paper industry.

for process. Effluent from pulp bleaching units (80–100 m^3/ton paper production) contains dissolved lignin, carbohydrate, color, COD, AOX and inorganic chloride compounds. The wastewater from paper making section contains particulates, organic compounds, inorganic dyes, etc. It is characterized depending on the dyes used and the effluent generation is very low when color papers are produced. Development of unified method for treating effluent from paper making process for reuse is more difficult and uneconomical, whereas a large volume of bleaching effluent has been generated

and having the uniform characteristics, it is economically feasible to develop new techniques to recover water from bleaching effluent for reuse.

Physical and chemical processes are quite expensive and generate a considerable amount of sludge which itself needs further treatment [4,5]. The low biodegradability index of pulp and paper effluent from bleaching process clearly shows that the biochemical method cannot be used [6,7]. Various treatment methods used for bleaching effluent reported in the literature are given in Table 1. In recent years, electrochemical technologies such as electrocoagulation, electrofloatation and electrooxidation have been used for the treatment of pulp and paper effluent. Among these methods, electrocoagulation emerges as one of the promising techniques due to its unique feature such as complete degradation of pollutants, less sludge generation and ease in operation. Electrocoagulation is a complex and interdependent process where the generation of coagulants takes place in situ by dissolving sacrificial anode. From the literature it is found that most of the researchers successfully used electrocoagulation method for the treatment of black liquor from pulp and paper industries (Table 2). However, very limited works have been reported for the treatment of pulp bleaching effluent using electrocoagulation method and hence in the present paper an attempt is made to study the influence of operating parameters such as time of electrolysis, current density, initial pH, electrolyte concentration, agitation speed and electrode distance on color, COD and BOD reduction and the fundamental and operating variables were optimized.

2. Experimental set-up and measurements

Bleaching effluent used in this study was obtained from pulp and paper mill located in Erode, Tamil Nadu. Experiments were car-
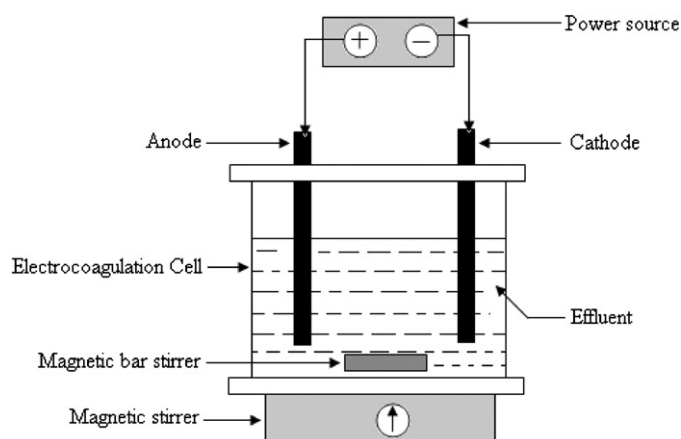
Table 1
Various methods reported in the literature for treatment of bleaching effluent.

Treatment process	Waste water	% color removal	% COD removal	% AOX removal	Energy consumption	Reference
Electrochemical	Decker filtrate effluent	87 for iron electrode	57 for iron electrode	–	1.19 kWh/kg COD	Soloman et al. [8]
Electrochemical	Bleaching effluent	90	90	90	–	Patel and Suresh [9]
Advanced oxidation	Bleaching effluent	66–72	–	–	–	Wang et al. [10]
Biological – fungal treatment	Bleaching effluent	92–95	50	72	–	Nagarathnamma and Bajpai [11]
Chemical precipitation	Bleaching effluents	–	90	–	–	Eskelinen et al. [12]
Ultrasonic irradiation + Fenton-like oxidation		–	12	–	–	
Photo-Fenton's oxidation		–	20	–	–	
Electro-oxidation treatment		–	28	–	–	
Biological – fungi treatment	Bleaching effluents	38.9–43.9	65–67	–	–	Belém et al. [13]

Table 2
Electrochemical and electro coagulation method used for treatment of black liquor.

Treatment process	Waste water	% Color removal	% COD removal	% AOX removal	Energy consumption	Reference
Electrocoagulation	Black liquor	100	91	–	–	Mahesh et al. [14]
Electrocoagulation	Black liquor	92 for mild steel and 84 for aluminum electrode	95 for mild steel and 89 for aluminum electrode	–	–	Parama Kalyani et al. [15]
Electrochemical	Unbleached kraft pulp effluent	84 for stainless steel and 98 for aluminum electrode	82 for stainless steel and 67 for aluminum electrode	–	14–20 Wh/l	Buzzini et al. [16]
Electrochemical	Black liquor	53–100	80–97.3	–	4–29 kWh/m^3	El-Ashtoukhy et al. [17]
Electrocoagulation	Black liquor	99	98		1.6–1.8 kWh/kg COD	Zaied and Bellakhal [18]
Electrochemical	Black liquor	97	87	–	1.2 kWh/m^3	Khansorthong and Hunsom [19]
Electrocoagulation	Black liquor	70	70	–	–	Ugurlu et al. [20]
Electrochemical	Black liquor	75	75	–	–	Wang et al. [21]

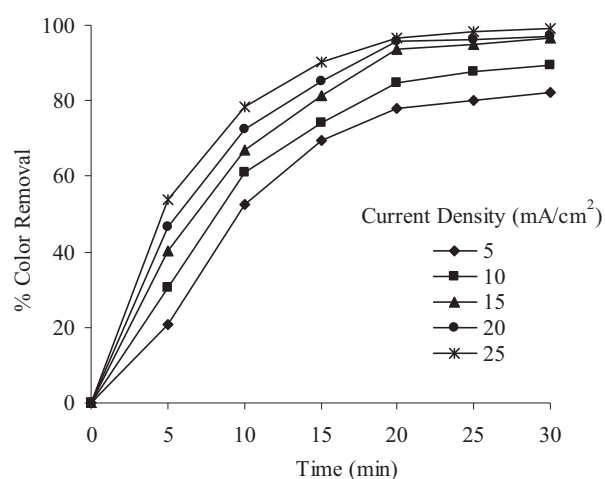**Fig. 2.** Schematic diagram of the experimental set-up.

Fig. 3. Effect of current density on percentage color removal (rpm = 100, pH 7, temperature = 28 °C, NaCl = 1 g/l, electrode distance = 3 cm).

ried out in a 500 ml reactor made up of glass. Aluminum plates of thickness 3 mm were used as electrodes. Dimensions of electrodes were 50 mm × 60 mm; and area of electrodes dipped into the solution was 50 mm × 50 mm. The total effective surface area of each electrode was 25 cm^2; and 1 cm gap was maintained between the bottom of the electrodes and the bottom of the cell is allowed for easy stirring. The lab-scale batch experimental setup used for the electrocoagulation studies is shown in Fig. 2. The spacing between two electrodes in EC cell was varied from 1 to 4 cm. Magnetic stirrer was used to agitate the solution. The electrodes were washed with HCl solution (15%, w/v) before each run. Following each run, the electrodes were washed with distilled water, dried and used again [20]. The current density was maintained constant by means of a precision digital direct current power supply (0–30 V, 0–2 A). All experiments were performed at a room temperature. In each run, 300 ml of wastewater solution was used. The current density was changed by using regulator. At the end of the experiments, sample was filtered to remove sludge. The filtered liquid was used for color, BOD and COD analysis. The characteristics of effluent are analyzed using the following technique and are reported in Table 3.

2.1. Determination of color

The sample was centrifuged at 10,000 rpm for 30 min to remove all the suspended matter. The pH of supernatant was adjusted to 7.6 by the addition of 2 M NaOH and absorbance was measured at 465 nm. Color units (CUs) were determined from UV absorbance using the following equation [22].

$$CU \ (Pt–Co) = 500 \times \frac{A_2}{A_1} \tag{1}$$

where A_1 is the absorbance of 500-CU standard platinum–cobalt solution (Pt–Co; 0.1214) and A_2 is the absorbance of the effluent samples.

Table 3
Characteristics of bleaching effluent.

Characteristics	Value
pH	6.9–7.2
COD (mg/l)	620
BOD (mg/l)	210
BOD/COD	0.33
Suspended solids (mg/l)	1200
Color (CU (Pt–Co))	255

2.2. Determination of biochemical oxygen demand

The 5-day BOD of sample was determined at 20 °C using standard dilution technique according to the American Public Health Association (APHA) [23]. 300 ml of sample was taken in airtight BOD bottle and kept it in incubator for 5 days at 20 °C. Dissolved oxygen concentration (DO) was measured initially and after 5 days of incubation. BOD was calculated from the difference between initial and final value of dissolved oxygen.

2.3. Chemical oxygen demand

COD was determined by the open reflux method according to APHA [23]. In this method, the sample was refluxed with potassium dichromate and sulfuric acid for 2 h and then titrated with ferrous ammonium sulfate and COD was calculated.

3. Results and discussion

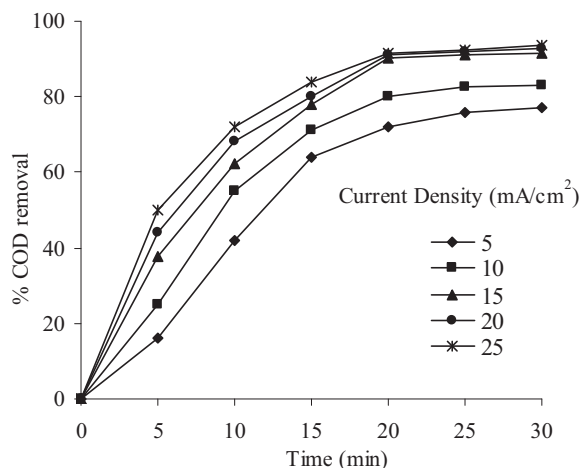### 3.1. Effect of current density

Several authors have reported [14,24] that the current density has significant influence on the efficiency of the electrocogulation process. Experiments were carried out by varying current density and the observations are shown in Figs. 3–5. The removal of color, COD and BOD were increased with increasing current density. This can be attributed due to the fact that the applied current density determines the rates of coagulant and bubble production, which in turn can influence the process efficiency. An increase in the gas bubbles density with reduction in their size enhances upwards flux resulting increased pollutant degradation and sludge floatation [25]. The removal of color, COD and BOD were increased with increasing current density up to 20 min. After 20 min, increasing current density beyond 15 mA/cm^2 did not show any significant improvement on the percentage of color, COD and BOD removal.

3.2. Effect of pH

In electrocoagulation, the pH plays an important role on the performance of the process. When potential is charged through an external power source, the aluminum sacrificial electrodes undergo oxidation in acid and alkali medium as follows [18].
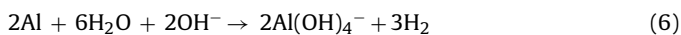
For acid condition

$$Al \rightarrow Al^{3+} + 3e^- \tag{2}$$

Fig. 4. Effect of current density on percentage COD removal (rpm = 100, pH 7, temperature = 28 °C, NaCl = 1 g/l, electrode distance = 3 cm).

$$Al^{3+} + 3H_2O \rightarrow Al(OH)_3 + 3H^+ \quad (3)$$

For alkali condition

$$Al \rightarrow Al^{3+} + 3e^- \quad (4)$$

$$3H_2O + 3e^- \rightarrow (3/2)H_2(g) + 3OH^- \quad (5)$$

$$2Al + 6H_2O + 2OH^- \rightarrow 2Al(OH)_4^- + 3H_2 \quad (6)$$

From Figs. 6–8 it was observed that the percentage of color, COD and BOD removal increases with the increasing pH up to 7. Freshly formed amorphous $Al(OH)_3$ has large surface areas, which are beneficial for a quick adsorption of soluble organic compounds and entrapping of colloidal particles and hence more reduction of color, COD and BOD. Further increasing pH beyond 7 decreases in the reduction of color, COD and BOD and it is mainly due to the formation of $Al(OH)_4^-$ which does not contribute to the reduction of color, COD and BOD [18].

3.3. Effect of rotational speed

Fig. 9 shows that the percentage color removal increases from 80% to 99% as the rotational speed increases from 50 to 200 rpm. From Figs. 10 and 11, it was observed that the increasing rotation speed significantly increases the percentage reduction of COD
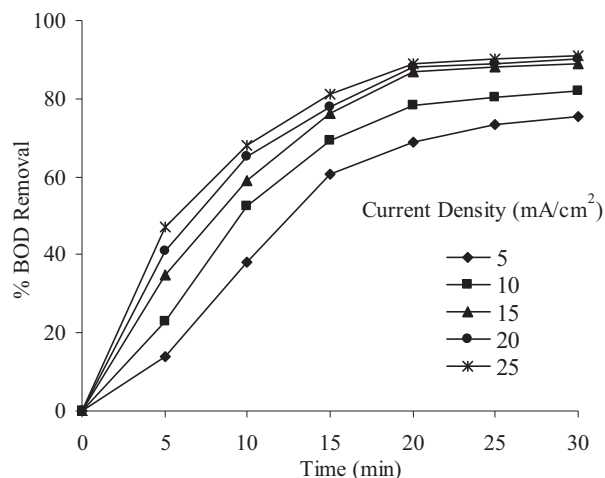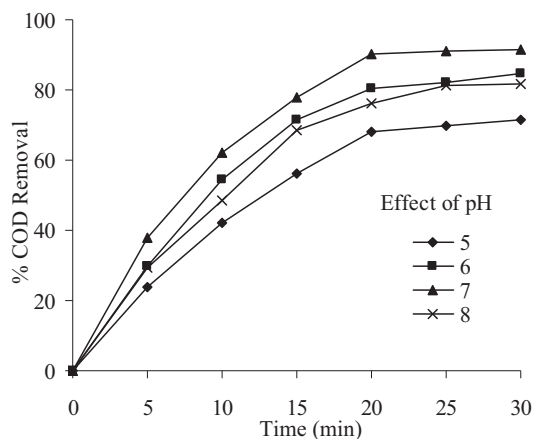

Fig. 5. Effect of current density on percentage BOD removal (rpm = 100, pH 7, temperature = 28 °C, NaCl = 1 g/l, Electrode distance = 3 cm).
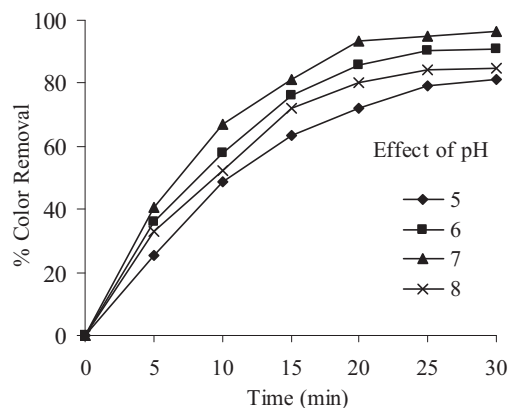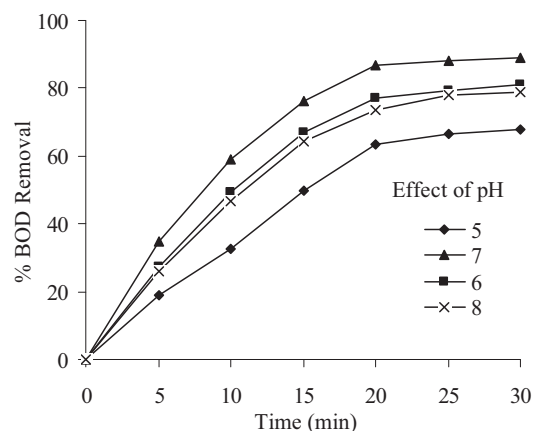

Fig. 6. Effect of pH on percentage color removal (rpm = 100, current density = 15 mA/cm², temperature = 28 °C, NaCl = 1 g/l, electrode distance = 3 cm).

and BOD up to 100 rpm. It is mainly due to that increasing rotational speed increases the intensity of turbulence and reduces the diffusion layer thickness at the electrode surface. No significance increase in COD and BOD removal was observed by increasing rotation speed beyond 100 rpm and it is mainly due to complete mixing [17].
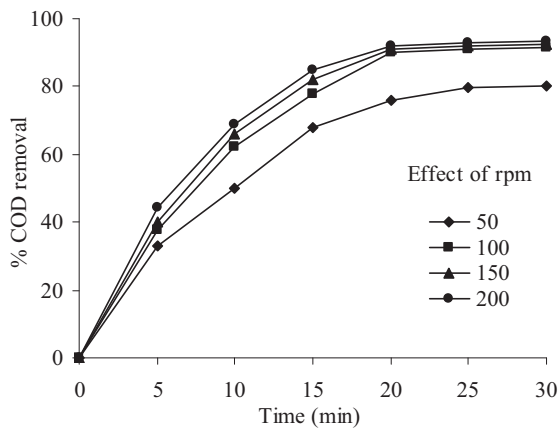

Fig. 7. Effect of pH on percentage COD removal (rpm = 100, current density = 15 mA/cm², temperature = 28 °C, NaCl = 1 g/l, electrode distance = 3 cm).
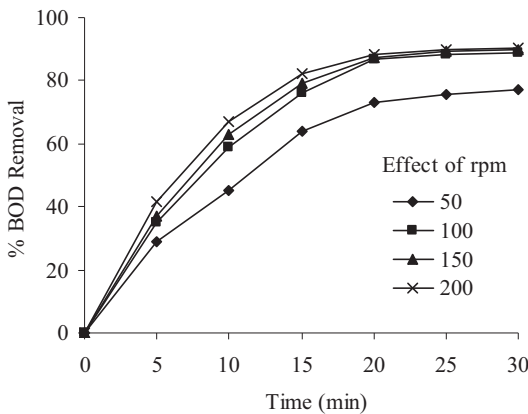

Fig. 8. Effect of pH on percentage BOD removal (rpm = 100, current density = 15 mA/cm², temperature = 28 °C, NaCl = 1 g/l, electrode distance = 3 cm).
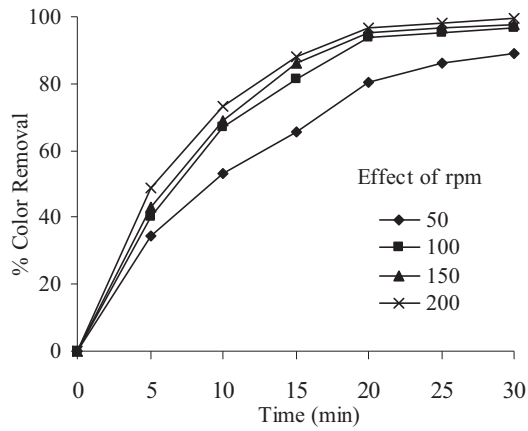
Fig. 9. Effect of rotational speed (rpm) on percentage color removal (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, NaCl = 1 g/l, electrode distance = 3 cm).

Fig. 10. Effect of rotational speed (rpm) on percentage COD removal (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, NaCl = 1 g/l, electrode distance = 3 cm).

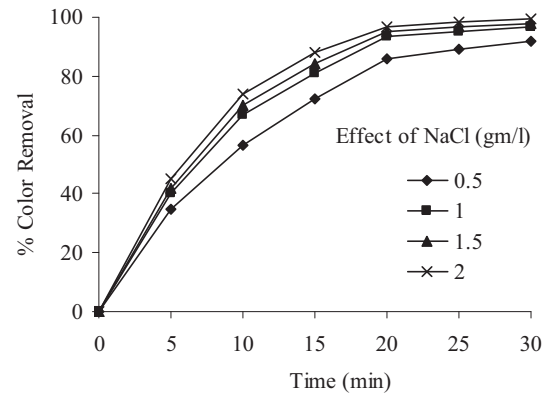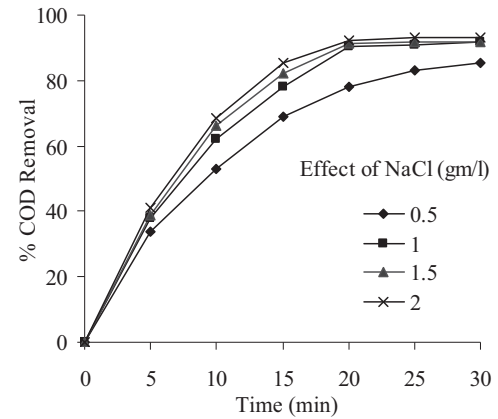

Fig. 11. Effect of rotational speed (rpm) on percentage BOD removal (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, NaCl = 1 g/l, electrode distance = 3 cm).

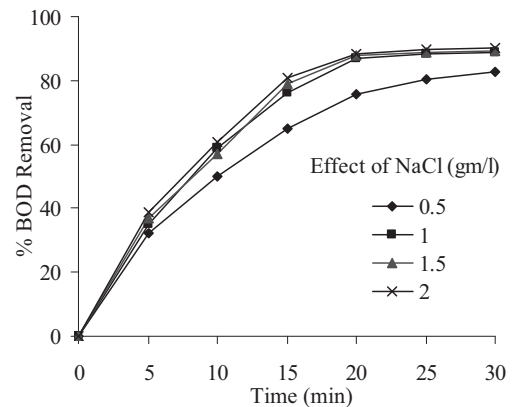### 3.4. Effect of NaCl concentration

The effect of NaCl concentration on color removal efficiency is given in Fig. 12. Color removal increased from 86% to 99% as NaCl concentration increased from 0.5 to 2 g/l. When the chloride is present in the solution, the products of anodic discharge are Cl$_2$ and OCl$^-$. This OCl$^-$ acted as a strong oxidant agent to remove organic molecules. NaCl not only increases the conductivity, but also contributes as strong oxidizing agents and hence color removal

Fig. 12. Effect of NaCl concentration on percentage color removal (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, rpm = 100, electrode distance = 3 cm).

Fig. 13. Effect of NaCl concentration on percentage COD removal (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, rpm = 100, electrode distance = 3 cm).

Fig. 14. Effect of NaCl concentration on percentage BOD removal (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, rpm = 100, electrode distance = 3 cm).

increases [15,26]. The presence of NaCl has a considerable effect on the percentage COD and BOD removal up to 1.0 g/l NaCl, beyond that there is no significant reduction of COD and BOD (Figs. 13 and 14).

3.5. Effect of electrode distance

Experiments were carried out by varying electrode distance and the observations are given in Figs. 15–17. The percentage removal of color, COD and BOD increases with the increase in electrode distance between 2 and 3 cm. Further increase in the distance beyond 3 cm did not yield any improvement on the percentage color, COD
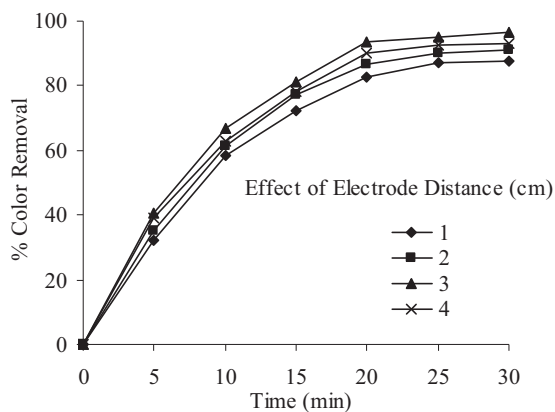
Fig. 15. Effect of Electrode distance on percentage color removal (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, rpm = 100, NaCl = 1 g/l).

Fig. 16. Effect of electrode distance on percentage COD removal (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, rpm = 100, NaCl = 1 g/l).

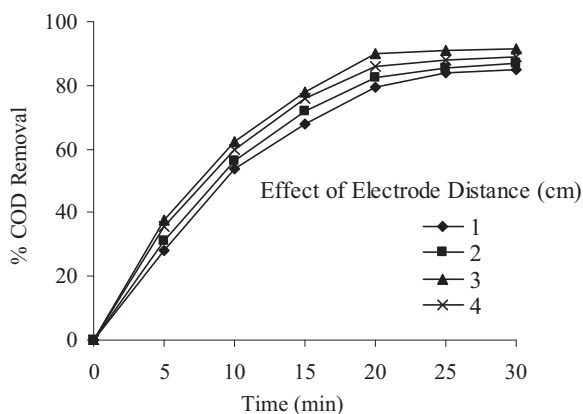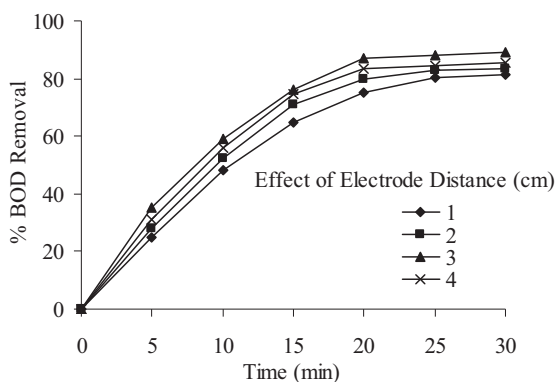**Fig. 17.** Effect of electrode distance on percentage BOD removal (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, rpm = 100, NaCl = 1 g/l).

and BOD removal because the electric potential between the electrodes decrease while increasing electrode distance. The electrode distance of 1–2 cm removal is lower than 3 cm because the gap between anode and cathode is too closed and solid and fluid transfer is obstructed. The accumulated solid particles and bubbles between the anodes and cathodes caused a consequent higher electrical resistance [27].
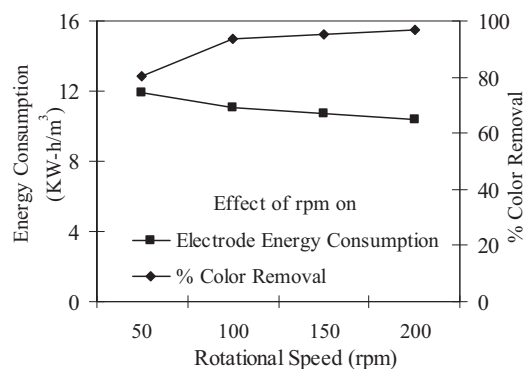

Fig. 18. Effect of rotational speed (rpm) on percentage color removal and electrode energy consumption (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, NaCl = 1 g/l, electrode distance = 3 cm, time = 20 min).
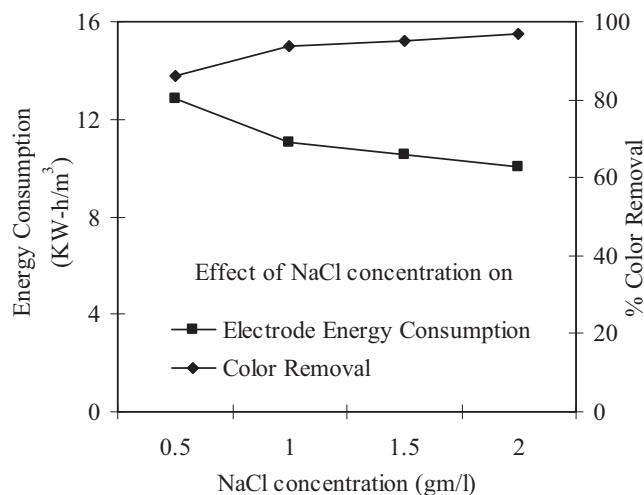

Fig. 19. Effect of NaCl concentration on percentage color removal and electrode energy consumption (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, rpm = 100, electrode distance = 3 cm, time = 20 min).

3.6. Energy consumption

The electrical energy consumption was calculated under different operating conditions using the following equation [17].

$$\text{Energy consumption } (\text{kWh/m}^3) = \frac{VIt}{\text{Treated volume (l)}} \quad (7)$$

where V is the cell voltage in volt, I is the current in ampere (A) and t is the treatment time (h).

Figs. 18 and 19 show that the energy consumption under optimized conditions (pH 7, current density = 15 mA/cm^2, temperature = 28 °C, NaCl = 1 g/l, Electrode distance = 3 cm, time = 20 min and pH 7, current density = 15 mA/cm^2, temperature = 28 °C, rpm = 100, Electrode distance = 3 cm, time = 20 min). From Figs. 18 and 19, it is observed that the energy consumption varies from 10.1 to 12.9 kWh/m^3 depending on the operating conditions. The energy consumption decreases with increasing rotational speed and NaCl concentration. It is mainly due to that increasing the degree of stirring tends to reduce energy consumption via reducing the cell voltage by assisting in the early release of gas bubbles adhering to the electrode surface which increase the cell resistance appreciably [28].

Table 4
Economical data used for calculating the operating cost.

Item	Cost ($)
Rectifier installing cost	800
Electrocoagulation tank installing cost	100
Maintenance (m^{-3})	0.003
Electricity (kWh^{-1})	0.085
Labor costs (m^{-3})	0.05
Sludge transportation and disposal cost (kg^{-1})	0.01
Material and chemical cost	
Al electrode (kg^{-1})	2.23
Electrolyte (NaCl) (m^{-3})	0.043

3.7. Economic analysis

Economic cost analysis has been done for a plant capacity of 100 m^3 wastewater/day. Economic data used for the evaluation of operating cost are given in Table 4. The operating cost was calculated using the following equation [29,30].

$$\text{Operating cost} \quad (\text{US } \$/\text{m}^3) = aC_{energy} + bC_{electrode} + cC_{chemicals} \quad (8)$$

where C_{energy} is the energy consumption (kWh/m^3), $C_{electrode}$ is the electrode consumption (kg/m^3) and $C_{chemicals}$ is the chemical consumption (kg/m^3) of wastewater treated. Unit prices a, b and c given for the India market, are as follows: (a) electrical energy price 0.085 US $/kWh, ($b$) electrode material (Al) price 2.23 US $/kg and ($c$) electrolyte (NaCl) price 0.043 US $/kg.

It is evident from Fig. 18, the color removal varied from 80.5 to 96.6% and energy consumption varied from 11.9 to 10.4 kWh/m^3 with respect to rotational speed and hence operating costs decreased from 1.72 to 1.35 $/m^3. Similarly with reference to Fig. 19, the color removal, energy consumption and operating costs varied from 86.0 to 96.8%, 12.9 to 10.1 kWh/m^3 and 1.69 to 1.52 $/m^3 respectively with respect to electrolyte concentration. Under optimum condition such as 15 mA/cm^2 current density, pH of 7, 1 g/l NaCl, 100 rpm, 28 °C temperature and 3 cm electrode distance, the energy, electrode and electrolyte consumptions were 11.055 kWh/m^3, 0.26 kg/m^3 and 1 kg/m^3 respectively. The operating cost was calculated under optimum condition and it was found to be 1.56 $/m^3 [31,32].

4. Conclusion

The influences of current density, pH, rotational speed and NaCl concentration on electrocoagulation using aluminum electrodes have been examined. The results showed that optimal operating conditions were found to be an initial pH of 7, current density of 15 mA/cm^2, rotational speed of 100 rpm, NaCl concentration of 1 g/l and electrolysis time of 20 min. This experimental study clearly showed that under the optimal conditions, about 94% color, 90% COD and 87% BOD were successfully removed. The decolorization, COD and BOD percentage removal were found to increase with the increase in sodium chloride concentration, current density, impeller rotational speed. Energy consumption was found to decrease with increasing impeller rotational speed and NaCl concentration. Under the optimal operating conditions the operating cost was found to be 1.56 US $/m^3. From the experimental result it is found that electrocoagulation technique could be successfully used for the recovery of water from pulp and paper industries bleaching effluent for reuse.

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