Studies on Removal of Dyes from wastewater using Electro-coagulation Process

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Abstract—**Electro-coagulation (EC) is one of the effective techniques to remove colour, COD and organic compounds from wastewater. In this paper electro coagulation technique has been used for the removal of colour and COD from dye solutions containing Direct Black 22 and Acid Red 97 using batch process. For batch the process effect of operational parameters such as current density, initial pH of the solution, time of electrolysis and electrode materials were studied to attempt maximum reduction of COD and colour. Different electrodes used in practical work were iron, and aluminium with D.C. current.**

I. INTRODUCTION

ON e of the major challenges facing mankind today is to provide clean water to a vast majority of the population around the world. The need for clean water is particularly critical in Third-World Countries. Rivers, canals, estuaries and other water-bodies are being constantly polluted due to indiscriminate discharge of industrial effluents as well as other anthropogenic activities and natural processes. The reuse of wastewater has become an absolute necessity. There is, therefore, an urgent need to develop innovative, more effective and inexpensive techniques for treatment of wastewater. A wide range of wastewater treatment techniques are known which includes biological processes as well as a range of physico-chemical processes that require chemical additions. A host of very promising techniques based on electrochemical technology are being developed that do not require chemical additions. These include electro coagulation, electro-flotation and electro-oxidation. Electro coagulation, has reached profitable commercialization. This process has the potential to extensively eliminate the disadvantages of the classical treatment techniques. [1] In addition, this process has been applied to treat water [26-42] containing Foodstuff wastes, oil wastes, dyes, suspended particles, chemical and mechanical polishing waste, and organic matter from landfill leachates, defluorination of water, synthetic detergent effluents, mine wastes and heavy metal-containing solution.

Dyeing and finishing are the two important processes generally applied in most of the textile manufacturing industries. These two processes generate considerable amount of wastewater, which may contain strong colour, suspended particles, high pH and high chemical oxygen demand (COD) concentration. [2]It is estimated that 1-15% of the dye is lost during dyeing and finishing processes and is released into wastewaters. As a result, a high amount of these dyes can exist in effluents of dyeing processes. Most kinds of synthetic dyes are toxic substances to human and aquatic life, which [3, 4] can be removed by adsorption, precipitation, chemical degradation, photo-degradation, biodegradation, [5] chemical coagulation and electro-coagulation. [6, 7, 8]

The objective of these Studies is Removal of Dyes from wastewater using electro-coagulation process at different pH, dyes concentration, electrode material, conductivity, current density, residence time and effects of A.C. and D.C. current.

II. THEORIES OF ELECTRO-COAGULATION

A. *Electro-coagulation mechanism:*

As shown in Fig, 1, Electro-coagulation technology offers an alternative to conventional coagulation process, where the metal salts are added to break the stable suspensions of the colloidal particles. EC process involves three successive stages: (a) formation of coagulants by electrolytic oxidation of the 'sacrificial electrode'; (b) destabilization of the contaminants, particulate suspension, and breaking of emulsions; (c) aggregation of the destabilized phases to form flocs. In electro-coagulation, coagulants are produced in situ within the reactor without direct addition of any chemicals. Coagulants are produced by electrolytic oxidation of appropriate anode materials, such as stainless steel, Aluminium, carbon, graphite, iron etc. Which result in formation of highly charged polymeric metal hydroxyl species. These species neutralize the electrostatic charges on the suspended solids and facilitate agglomeration resulting in separation from the aqueous phase. The technology removes metals, colloids particles and soluble organic pollutants from aqueous media by introducing highly charged polymeric hydroxide species. The treatment prompts the precipitation of certain metals and salts [7].
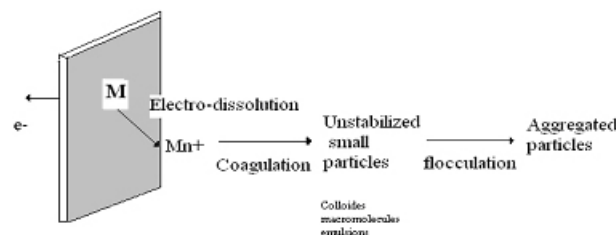

Fig. 1. Electrocoagulation step wise process[8]

An electrochemical reactor when connected to an external power source, the anode material will electrochemically corrode due to oxidation, while the cathode will be subjected to passivation. But, for wastewater treatment, due to metal dissolution, the use of electrodes with large surface area is required. This has been achieved by using cells with monopolar

electrodes either in parallel or series connections. It essentially consists of pairs of conductive metal plates placed between two parallel electrodes and a DC power source.

The mechanism of EC is highly dependent on the chemistry of the aqueous medium, especially conductivity; pH, particle size, and chemical constituent concentrations. Following are the different mechanism of electro-coagulation with Iron and aluminium as an electrode.

B. Aluminium as electrode:

The electrolytic dissolution of the aluminium anode produces the cationic monomeric species such as Al^{3+} 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:
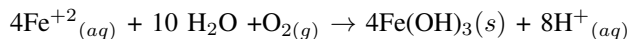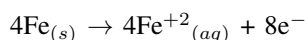
$$Al \rightarrow Al^{3+}{}_{(aq)}$$

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

$$nAl(OH)_3 \rightarrow Al_n(OH)_{3n}$$
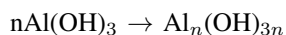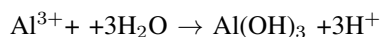
However, depending on the pH of the aqueous medium other ionic species, such as $Al(OH)^{2+}$, $Al_2(OH)_2{}^{4+}$ and $Al(OH)^{4-}$ may also be present in the system.
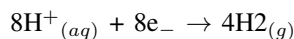
C. Iron as electrode:

Iron upon oxidation in an electrolytic system produces iron hydroxide, $Fe(OH)_n$, where n = 2 or 3. Two mechanisms have been proposed for the production of $Fe(OH)_n$ [7].
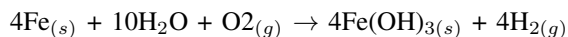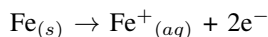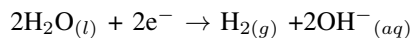
Mechanism 1

Anode:

$$4Fe_{(s)} \rightarrow 4Fe^{+2}{}_{(aq)} + 8e^-$$

$$4Fe^{+2}{}_{(aq)} + 10 H_2O +O_{2(g)} \rightarrow 4Fe(OH)_3(s) + 8H^+{}_{(aq)}$$

Cathode:

$$8H^+{}_{(aq)} + 8e_- \rightarrow 4H2_{(g)}$$

Overall:

$$4Fe_{(s)} + 10H_2O + O2_{(g)} \rightarrow 4Fe(OH)_{3(s)} + 4H_{2(g)}$$

Mechanism 2

Anode:

$$Fe_{(s)} \rightarrow Fe^+{}_{(aq)} + 2e^-$$

Cathode:

$$Fe^{2+}{}_{(aq)} + 2OH^-{}_{(aq)} \rightarrow Fe(OH)_{2(s)}$$

$$2H_2O_{(l)} + 2e^- \rightarrow H_{2(g)} +2OH^-{}_{(aq)}$$

Overall:

$$Fe_{(s)} +2H_2O {}_{(l)} \rightarrow Fe(OH)_2(s) + H_{2(g)}$$
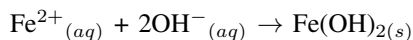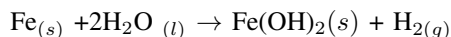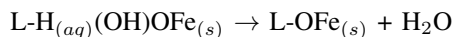
The $Fe(OH)_{n(s)}$ formed remains in the aqueous stream as a gelatinous suspension, which can remove the pollutants from wastewater either by complexation or by electrostatic attraction, followed by coagulation. In the surface complexation mode, the pollutant acts as a ligand (L) to chemically bind hydrous iron:

$$L-H_{(aq)}(OH)OFe_{(s)} \rightarrow L-OFe_{(s)} + H_2O$$

D. Mechanism of COD removal:

Chemical oxygen demand (COD) is a measure of the amount of the oxygen used in the chemical oxidation of inorganic and organic matter present in wastewater. Degradation of pollutants in wastewater in its simple form decreases oxygen requirement for chemical oxidation hence decreases COD. [8, 9, 10]

E. Batch vs continuous electro-coagulation process:

Present study includes practical work done with only batch process. Continuous electro coagulation process with different metals or metals coated with metal oxides are also possible. [11,12]

F. A.C. versus D.C .Electro-coagulation Process:

The direct current electro-coagulation (DCE) technology is inherent with the formation of an impermeable oxide layer on the cathode as well as deterioration of the anode due to oxidation. This leads to the loss of efficiency of the EC unit. These limitations of the DCE process have been minimized to some extent by the addition of parallel plate sacrificial electrodes in the cell configuration.

However, many have preferred [13] the use of alternating current electro-coagulation (ACE) technology. It is believed that the AC cyclic energization retards the normal mechanisms of electrode attack that are experienced in DCE system, and thus, ensure reasonable electrode life.

III. EXPERIMENTAL PROCEDURE

A. Apparatus:

The study is based on experimental work conducted at laboratory scale in batch process. The batch unit consist of a 0.5 L electrochemical reactor with 75mm X 65mm X 3mm (iron, and aluminium) anode and cathode kept a part 8mm.The applied potential was maintained constant by means of a precision D.C. power supply and A.C.

characteristic	Direct Black 22
max (nm)	482
Chemical class	Tetraazo
Mw	1068.89

TABLE I
CHARACTERISTIC OF DIRECT BLACK 22

characteristic	Acid Red 97
max (nm)	496
Chemical class	Diaazo
Mw	698.63274

TABLE II
CHARACTERISTIC OF ACID RED 97

power supply. Dyes were collected from dyes manufacture industries named Gayatri colour chemical, GIDC, Ahmadabad.

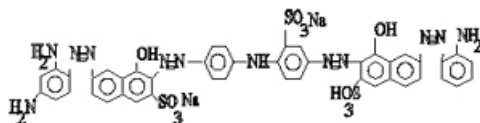*B. Characteristics of treated effluent: [14, 15, 16, 17]*

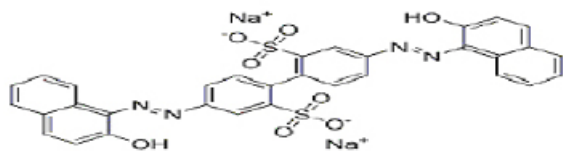Fig. 2. Structure of Direct Black 22

1) Direct Black 22[21, 22, 23, 24, 25]:

Fig. 3. Structure of Acid Red 97

2) characteristic of Acid Red 97:

C. Laboratory procedure:

1) Batch Process: : The commercial dyes used in this project were purchased from industry. Dye solutions were prepared by dissolving dyes in distilled water to prepare 100ppm of dye solution. The conductivity of solutions was raised up and adjusted in different values by the addition of Nacl. The conductivity measurement was [18, 19,20] performed using a Philips conductivity meter. The pH of the solutions was measured by pH meter and adjusted by adding NaoH or H2SO4 solutions. Iron and Aluminium were used as electrode. Dimensions of electrodes were 75mm X 65mm X 3mm and the distance between two electrodes in EC cell was 8mm in all experiments. The electrodes were connected to a DC power supply with galvanostatic

operational options for controlling the current density. All the runs were performed at room temperature. In each run, 500 ml of the dye solution was decanted into the electrolytic cell. At the end of electro-coagulation, all samples were allowed to settle for 20 min in a 500 ml vessel before any analysis. Neither centrifuging nor filtration was performed [18].
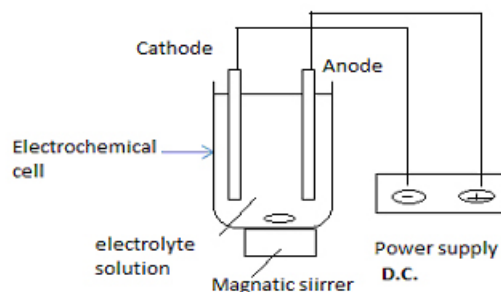

Fig. 4. Experimental setup for batch process

IV. ANALYSIS

A. Methods:

dyes concentrations were determined from their absorbance characteristics in the UV- vis range (200-800 nm) with the calibration method. The calculation of colour removal efficiency after electro coagulation treatment was performed using this formula: $CR(\%) = (C0 - C) / C0 \times 100$ where C0 and C are concentrations of dye before and after electro coagulation in mg/l respectively.

The chemical oxygen demand (COD) of dye solutions was measured according to the standard methods for examining water and wastewater [17].

V. RESULT AND DISCUSSION

Results obtained for batch process by laboratory experiments changing various parameters are tabulated as follows. (With D.C. supply)

A. Discussion:

1) Effect of Initial pH: : The effect of initial pH on the COD and colour removal efficiencies is presented in Fig 5 and 6. For Aluminium and Iron electrode, for pH 8, the colour removal efficiency is as high as 90.88, 89.55 and COD removal efficiency is 68.33, 58.33% respectively. Colour removal efficiency and COD removals drop dramatically at pH > 8. There was minimum removal efficiency at the pH = 2. Since hydroxide ions, which were generated at the cathode, were neutralized by H+ ions, sufficient amount of iron hydroxide complexes were not formed at this pH. At the pH > 8, colour and COD removal efficiency decreased up to 39.68,

Electrode	Current Density A/m2	pH	Reduction of COD%	Reduction of colour %	Time Min
Fe-Fe	50	2	19	58.09	5
			19	53.35	10
			25	40.86	15
Fe-Fe	100	2	19	62.16	5
			19	54.36	10
			25	45.35	15
Al-Al	50	2	15	62.54	5
			15	54.08	10
			22	41.87	15
Al-Al	100	2	15	65.89	5
			15	59.13	10
			22	54.21	15
Fe-Fe	50	5	62.5	86.33	5
			50	85.00	10
			35.5	79.26	15
Fe-Fe	100	5	62.5	90.67	5
			50	87.00	10
			35.5	81.57	15
Al-Al	50	5	48	88.97	5
			48	87.58	10
			62.5	84.90	15
Al-Al	100	5	62.5	65.89	5
			62.5	59.13	10
			68	54.21	15

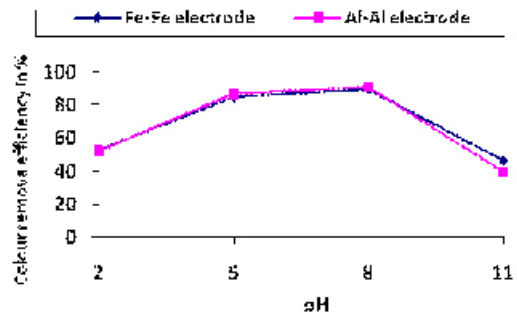| Electrode	Current Density A/m2	pH	Reduction of COD%	Reduction of colour %	Time Min
Fe-Fe	50	8	50	91.76	5
			62.5	91.13	10
			62.5	90.00	15
Fe-Fe	100	8	50	92.30	5
			62.5	80.35	10
			62.5	45.35	15
Al-Al	50	8	32	91.67	5
			48	92.54	10
			48	90.47	15
Al-Al	100	8	48	65.89	5
			48	48.56	10
			60	46.39	15
Fe-Fe	50	11	19	44.22	5
			19	48.56	10
			25	39.05	15
Fe-Fe	100	11	19	41.08	5
			25	50.72	10
			32.5	81.57	15
Al-Al	50	11	19	88.97	5
			19	37.96	10
			25	39.05	15
Al-Al	100	11	19	38.89	5
			25	41.08	10
			25	43.74	15

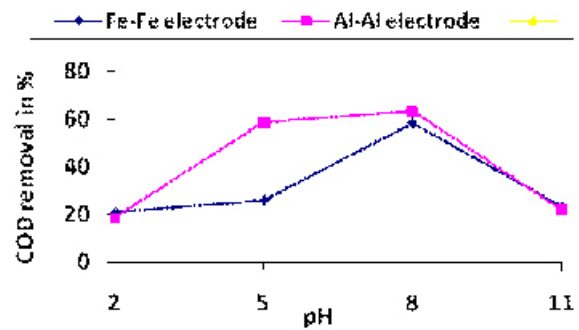Fig. 5. Effect of Initial pH on Colour removal efficiency (%)

Fig. 6. Effect of Initial pH on COD removal efficiency (%)

because of degradation of iron electrode. And COD removal efficiency increases from 50 to 62.5%.
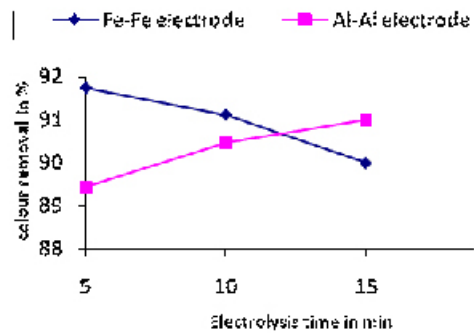

Fig. 7. Effect of Time of Electrolysis on Colour removal efficiency (%) at 50 A/m2 , 8 pH

48.48 % for Aluminium and Iron electrodes because above this pH $Fe(OH)_{4-}$ and $Al(OH)_{4-}$ are the dominant species and they are unable to form flocs [19].

2) Effect of Time of Electrolysis: : The colour and COD removal efficiency depends directly on the concentration of ions produced by the electrodes. When the electrolysis period increases, concentration of Aluminium ion increases but for iron maximum ion production is up to 5 minutes and then after concentration of it decreases. According to Fig 7 and 8 an increase in the time of electrolysis from 5 to 15 min yields in increase in the efficiency of colour and COD removal up to 91% and 48% for Aluminium electrode. For iron electrode, efficiency of colour removal decreases from 91.76 to 90%

3) Effect of Current Density: : It is well known that the amount of current density determines the coagulant production rate, and adjusts the rate and size of the bubble production, and hence affects the growth of flocs. Fig. 9, 10 shows the colour removal percentage against current density applied to the electrodes in the EC process for Aluminium and iron electrodes. Raising current density causes a corresponding increase in the oxidized ion production from electrodes. Results show that increase in current density is favourable for Aluminium and Iron electrode. It is also clear that Fe-Fe electrodes perform better than the same size of Aluminium
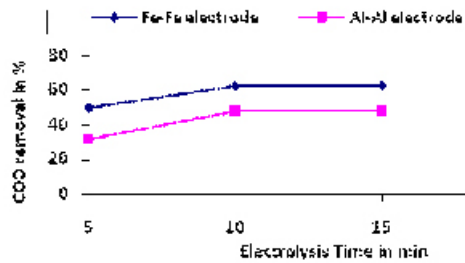
Fig. 8.   Effect of Time of Electrolysis on COD removal efficiency (%) at 50 A/m2 , 8 pH
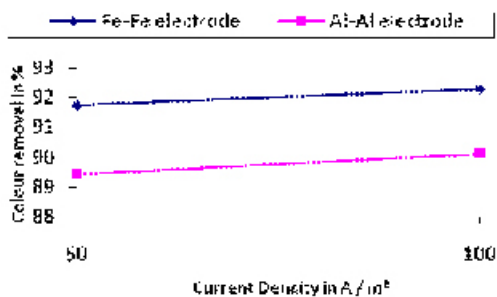
electrodes giving higher efficiency.

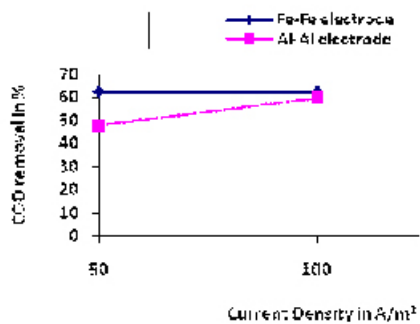Fig. 9.   Effect of Current Density on Colour removal efficiency (%) at 8 pH, 5 min electrolysis time

Fig. 10.   Effect of Current Density on COD removal efficiency (%)at 8 pH, at 15 min of electrolysis time

VI. CONCLUSION

From the experiment it was found that

1) Treatment of degradation of dyes by electro-coagulation has been found to be pH dependent.
2) In almost neutral medium, pH 8, COD and Colour removal efficiencies of Aluminium and iron electrodes are higher than those of all pH
3) Comparing overall performance of Iron and Aluminium electrodes, Iron electrodes give higher efficiency for COD and colour removal.
4) For Iron electrodes %colour removal decreases with electrolysis time.

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