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Coagulation and Electrocoagulation Process for Dye Removal from Textile Wastewater: A Review

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Research Article

Abstract

Dye components like azo dye, Congo red dye, methylene blue is common in the textile wastewater and challenges often arises as soluble dyes are the most problematic for removal. Conventional coagulation and Electrocoagulation often provide suitable outcome while treating these dye wastewaters. Coagulation process is a conventional treatment process where chemicals for binding the dye colloids are applied for floc formation and removed the pollutant following settling. In electrocoagulation, electrode of required metals ion is provided to generate current density instead chemical salt resulting in low amount of sludge generation. Mostly, Aluminum salt and iron salt is used as coagulant and similarly, electrode made of Al and Fe is used as anode for electrocoagulation process. In present study, articles are reviewed to have clear idea on the basis of initial pH (6-8), electrolysis time (2-30min), initial dye concentration (50 to 400mg/L), current density (5, 10,15 mA/cm^2), temperature and mixing speed etc along with electrolyte concentration and type (NaCl, HCL, H_2SO_4). The review from various articles suggested that final outcome of electrocoagulation process in dye removal is more effective as compared to the conventional coagulation.

Key Words- Conventional Coagulation; Electrocoagulation; Dye wastewater, Textile Wastewater, Electrolyte, Current density

*Broad Area- **Environmental Engineering.***

1. Introduction

Huge amount of dye wastewater is generated from the various steps such as colouring and processing of textile substrates (fiber, yarns, fabrics, garments). These wastewaters may generate in batch wise while treating the textile substrates to retain its market level quality in terms of acceptable durability of the required colour/ shade during production as well as normal end use. Dyes are the basic raw material for textile industries and many times dyes used in coloring textile fiber are synthetic compounds [1]. When these components are directly released to the aquatic system without any treatment, it obviously can cause havoc to the ecosystem and therefore treatment unit should be placed before releasing the wastewater to the receiving water to remove hazardous materials, reduce the BOD, and disinfection as well as dye removal. The common dyes used in textiles are azo dye, congo red dye and methylene blue. Dyes especially acid and reactive dye can escape from conventional wastewater treatment because they are generally designed to withstand microbial, chemical and photolytic degradation [2] as used in conventional wastewater plant. Coagulation/flocculation can be effective to enhance nanofiltration performance towards water reuse and minimisation of fouling[3].textile effluent using different process coagulation/flocculation , enzymatic catalysis by commercial laccase and nanofiltration [4]. Textile industry daily basis membrane process for dye wastewater treatment and fouling control[5]. Compound bioflocculant used as a coagulation and dye wastewater treatment effect of pH solution[6]. Treatment of highly concentrated wastewater containing multiple synthetics dyes combined process[7].

Description of dyes considered in present study

***Synthetic dye**:* Colorful substance which when applied to a fabric imparts a permanent color and the color is not removed by washing with soap or any exposure to light.

Azo dye: Azo dyes are organic compounds bearing the functional group R-N=N-R', in which R and R', are usually aryl [8]. They are a commercially important family of azo compounds. Compounds containing the linkage C-N=N-C azo dye used to treat textiles, leather articles, and some foods. Acid dyes are sodium salts of sulphonic acids and nitrophenoles. These dyes applied to animal fibers directly not to vegetable fibers. Acid dye has a higher affinity for nylon because polyamide fibers contains a large proportion of free amino groups. Such dyes are used for colouring wool, silk and nylon but not cotton.

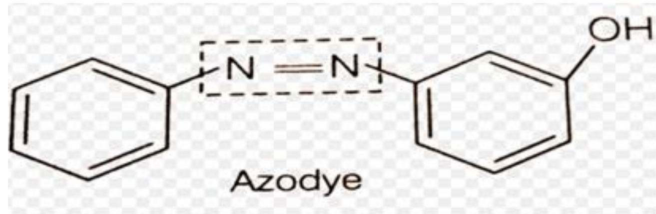

Fig. No.1 Chemical representation of Azo dye[9]

Methylene blue: methylene blue is a thiazine dye and works by converting the ferric iron in hemoglobin to ferrous iron [10].

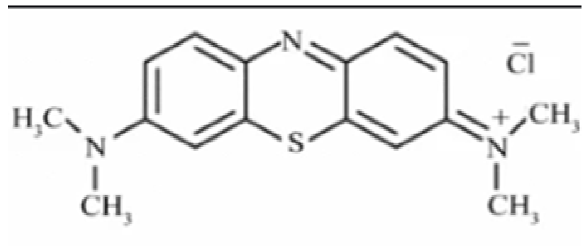

Fig. No. 2 Chemical representation of Methylene blue [9]

Congo red: Congo red is an organic compound the sodium salt of 3-3bis. It is an azo dye. Congo red is water- soluble, yielding a red colloidal solution; its solubility is greater in organic solvents.[11] Used mostly in leather, paper and textile industry.

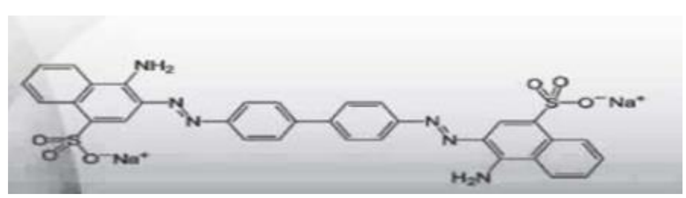

Fig. No. 3 Chemical representation of Congo red [9]

1.1 Treatment process used for dye removal

The precious molecule water is highly used in industries and textile industry is a huge consumer of it. Therefore it is always m,andatory to sdvelop a better treatment process for the recovery of the used water for further use or release to the receiving water body in good quality. Therefore, researchers and scientists groups are always working for development of a better, profound and suitable technique and treatment process for the challenging task of dye removal. In this regards some studies are to mentioned as various techniques are used for dye removal nanofiltration coupled with coagulationand electrogulation, Enzymatic catalysis, coagulation/flocculation and nanofiltration processes [4] membrane filtration [5], Compound bioflocculant [12]], combined process of coagulation/flocculation and nanofiltration [13] dicyanidiamide fixer in the presence of ferric chloride. However, many of these techniques are often associated with limitations like hish cost, operating and maintenance issues like membrane fouling when the membrane process is considered for example. Therefore, in this study we have focused mostly on coagulation and electrocoagulation process for dye removal and discussed in detail with their operating conditions in this article.

1.2 Conventional coagulation

Coagulation means treatment of water with coagulants to remove colloidal and fine suspended impurities. The particles are destabilized and aggregated to form floc by the addition of some chemicals. The formed flocs adsorb and entrap the suspended particles and settles rapidly. Most commonly used coagulants for the treatment of textile effluents are alum, ferric sulphate, ferrous sulphate, chlorinated copperas, ferric chloride and calcium chloride. To enhance the process of coagulation, some chemicals are added in smaller quantities such as activated silica and poly electrolytes that promote the rapid settling of flocs. These chemicals are called as coagulant aids. They act by reducing the charge on the colloid.[14] The coagulants, coagulant aids, and chemicals for pH adjustments are rapidly mixed for 4-6 minutes in a smaller tank for greater dispersion. The process of floc formation is called flocculation. The flash mixing is followed by gentle stirring of wastewater and coagulants in a large tank for 15-45 minutes for formation of flocs without being broken down by the turbulence. Settle quickly leaving a clear supernatant liquid [15] A period of 1-6 hours is required for settling.

Table 1 COAGULANT USED IN CONVENTIONAL COAGUALATION PROCESS FOR DYE WASTEWATER

S.No.	**Coagulant**	**Dye**	**Remarks**	**Reference**
1.	Natural polymer composite coagulant	Reactive dye	Chemical dosage by more than 50% pH 0.1 mixutures were stirred at 120 rpm.	16
2.	Natural coagulants	Reactive dyes	reaching removals of 82.2 % for the apparent colour, 83.05 % for COD, 78.4 %, pH 10.9,	17

3.	Natural-based coagulants	Direct Blue	pH 4, tannin-coagulant, while dosages up to 240 mg L^{-1}	18
4.	Natural coagulant	leachate treatment	using 40 g/L of *Salvia hispanica* at pH 7	19
5.	Polysilicate ferric manganese	Active dye	removal rate of the active yellow dye reached 86% at pH 5.0	20
6.	polyaluminum chloride and flat-sheet cross-flow ultrafiltration	deinking wastewater	polyaluminum chloride dosage from 0 to 2,000 mgL^{-1},	21
7.	$MgSO_4$ as a coagulant	Poly(vinyl alcohol) and reactive dye	color attained 88.9%, 86.3%, and 99.2%,	22
8.	Alkaline lignin (AL)	disperse dye	high molecular weight like AL-g-DMC_1 and AL-g-DMC_2 exerted excellent color removals	23
9.	PFS/$FeSO_4$ coagulation	SB(V) textile wastewater	Temperature of 35 °C, pH ranged from 5 to 6	24
10.	Hybrid coagulant	reactive dye wastewaters treatment	reduced by PFTS compared to T-PSF in reactive dye treatment	6
11.	Natural coagulant	water and wastewater treatment	high purity of natural coagulants; and the synthesis of multifunctional natural coagulants	25
12.	Natural extract coagulant	dairy industry	Calcium chloride ($CaCl_2$) good alternative for the primary treatment of dairy wastewater and remove 90% of turbidity.	26
13.	Phytogenic aluminum sulfate nano coagulant	Congo red dye	temperature from 30 °C to 70 °C	27
14.	Inorganic coagulant	Paper mill wastewater	COD and Chroma removed by 65.3% and 71.2%, respectively (initial pH 7.5, 1 ml/L PFASC, 1.0 ppm PAM).	28
15.	Poly-aluminum-ferric-sulfate coagulant	Water leaching	sulfuric acid solution was stirred at 25 °C for 3 h and further heated at 150 °C for 2 h, the optimal extraction efficiencies of Al (98.8 %) and Fe (98.1 %)	29
16.	Polyamidine (PA)	Dye wastewater treatment	optimized dosage of AF/PA 18.91/0.71 mg/L, and for PAC/PA was 21.19/0.91 mg/L.	30

1.3 Electrocoagulation (EC)

Electrochemical treatment is an emerging technology, and its application to dye decontamination has received increasing attention recently due to advantages as high efficiency, short reaction time, low sludge pro- duction, ease of operation, and environmental compatibility [31]. The electrochemical method is reported to be a better treatment method with high efficiency for treating textile wastewaters which contain a high concentration of dye [32]. EC is stable in settle down the pollution and two electrodes are used Al and Fe to neutralize. The EC process is influenced by operating pH, current density, and electrolysis time. Two type's electrodes are used aluminium and iron plate as Anode and cathode. [33].

4.0 Basic parameters considered in electrocoagulation

For successful operation of the treatment process of electrocoagulation pH, current density, electrode type and electrolyte are major factor to be considered. These parameters are considered as tool for success of the study and various observations by the earlier researchers are presented in the following tables.

4.1 Effect of current density

The magnitude of current density determines the amount of anode metal ions dissolved during electrolysis, and the rate at which bubbles are generated at the cathode, so the current density is the main factor [34] for determining the effectiveness of synthetic wastewater treatment by electrocoagulation. Studies with effect of current density for electrocoagualation is presented in Table 2.

4.2 Electrode

The two sacrificial anode and cathode (Al and Fe) electrode are used in treatment. The result of wastewater treatment by electrocoagulation was as follows: Al (anode) - Al (cathode)>Al (anode) - Fe (cathode)>Fe (anode) - Al (cathode)>Fe (anode) - Fe (cathode). Specifically, after 40min of electrolysis [35] the oil removal fractions reach 72.9%, 54.7%, 35.4% and 33.2%, and the turbidity removal rates are 62.5%, 60.4%, 52.4% and 49.8%, for these material configurations respectively.

4.3 Effect of Electrolyte

During electrocoagulation, highly charged cations such as Al^{3+} and Fe^{2+} formed at the anode destabilize colloidal particles to be removed, for example dye, by the formation of monomeric and polymeric hydroxo complex species. These metal hydroxo complexes have high adsorption properties, forming strong aggregates with pollutants. The electrocoagulation process uses in electrolyte NaCl, HCL, H_2SO_4 etc the treatment EC dye colour removal in the treatment is given in Table 3.

4.4 Effect of pH

pH has great influence on wastewater treatment and it is same for electrocoagulation process [Table 4]. It also depends on the wastewater to be treated. If an EC system has an acidic influent, the pH of the effluent increases during the treatment process; conversely, if such a system has an alkaline influent, the pH of the effluent decreases during the treatment process. The wastewater from textile industries normally have pH higher than 8.5 and in alkaline condition making it ideal to metal precipitation using electrocoagulation.

Table 2 EFFECT OF CURRENT DENSITY IN ELECTROGUALATION

Sl. No.	Current Density	Dye	Remarks	Reference
1.	1.04, 2.08, 4.17 and 10.42 A/m^2	Reactive Blue 198 (RB198), Reactive Yellow (RY145) and Reactive Blue 19 (RB19)	Dye removal was found to be directly proportional to current density, at highest current density, max. removal of 98.8% was obtained.	13
2.	0.12 to 0.59 A/m^2	Bromophenol Blue (BPB).	the percentage colour removal increases from 60 to 99.31 % as the current density increases from 0.12 to 0.59 Am2.	36
3.	0.011 A/cm^2 (optimum)	Acid Red 1 (AR1) (Synthetic Azo dye)	Ti was observed that with an increase in current density and electrolyte concentration,	9
4.	0.075 A (optimum) (not current density)	Reactive Black 5 dye (RB5)	Higher current (0.075 A), longer treatment duration (50.3 min.) and electrolyte (0.11g/L) resulted in 80.9% RB5 removal	17
5.	100-400 A/m^2	Acid Red 336	Higher current are more efficient but also generates sludge with more Al concentration and leads to higher energy consumption.	27
6.	13.9-138.9 A/m^2	brilliant green dye	Dye removal increased with increase in current density. However, for all current densities above 41.7 A/m^2, the removal efficiency was more than 99% for residence time of 30min. and above.	37
7.	100-250 A/m^2	Crystal violet dye	Under residence time of 15 min., the removal efficiency increases from 19% to 65% when the current density increased from 100 to 250 A/m^2	12
8.	13.9, 20.8, 27.8, 34.7, 41.7, 69.4, 138.9 A/m^2	brilliant green dye	Optimum pH4.0 and highest dye removal salt concentration reduce the specific electrical energy consumption	38
9.	1mA/cm^2	Reactive Black 5 dye	Color 97% removal, TOC 97.3 Removal and natural pH	39
10.	26, 53, 79, 106, 132 A/m^2	Diazo dye	The high efficiency electrocoagulation and anodic oxidation large amount wastewater treatment	40
11.	5, 10,15 Am/cm^2	Acid dye	Complete decolorization in 30 min	41
12.	(0.5, 2.5, 5.0 and 10.0 Am /cm^2	Acid black 194 dye	Removing practically 100% total organic carbon	42
13.	4.17 A/m2 and 8.33 A/m^2	Reactive black 5	Electrocoagulation 98% removal efficiencies, 10 min treatment time duration	43
14.	33.33,66.7,16.7	azo dyes	The optimum current density 33.33mA/cm$^-$	44

	mA/cm^2		^{2}and rapidly removed	
15.	5 to 25A/m^2	Textile wastewater	COD and colour 90% removal and reaction time 180 min	11
16.	4.14, 8.29,12.4,16.58 mA/cm^2	Reactive blue 19	Color removal 97.4 and COD removal 93% optimum operating condition	45
17.	26 to 331 mA/cm^2	AR18 dye	Critical point 26 mA/cm^{2}current density	46
18.	100 to 200 A/m^2	Direct red 81	Maximum current dendity 200A/m^2	47
19.	40 mA/cm^2	Acid red 14	Intial pH 7 and 20 min reaction time	32
20.	2,8,12,16,20 mA/cm^2	Acid red 73	Using polyalunminum chloride coagulant and electrocoagulation 99% dye remove ,COD 88% remove	41

Table 3 ELECTROLYTE USED IN ELECTROCOAGULATION

Sl. No	Electrolyte	Dye	Remark	Reference
1	NaCl concentration (1gL^{-1})	Diazo dye concentration (50 to 300 mg/L)	Electrocoagulation and anodic oxidation operation condition same but electrocoagulation more effective. EC(66.67), AO (44.96)	40
2	HCL concentration	Acid dye concentration (50-150 mg/L)	Ozonation and electrocoagulation performed simultaneously decolorization acid dye 94.43	34
3	HCL concentration	Acid black 194 dye (1.0L)	Electrocoagulation	42
4	0.1 M NaOH, NaCl solution	Reactive red 241	Compare between electrocoagulation, ultrasound and ultrasound assisted electrocoagulation 98%	48
5	NaOH, H_2 SO_4, HCL	Congo red dye	Electrocoagulation (89%) and response surface methodology (97%)	36
6	KCl	brilliant green dye	Electrocoagulation 96.1%	49
7	Na_2SO_4	Real indigo dyeing	Response surface methodology 93.77% and electrocoagulation 92.07%	50
8	NaCl	Azo dye	peroxi- coagulation process	44
9	Electrolytes, With Cl^-, SO_4^2, NO^- anions and Na^+, K^+, NH_4^+	Reactive black 5	Electrocoagulation process 98.0%	42
10	HCL	diazo dye i.e. Congo Red dye (CR) and a basic dye Methylene Blue (MB)	Electrocoagulation – floatation (86.04) and pulsed power technology (50.55) toxic dye removal	51

11	sodium chloride	Reactive Dye	Electrocoagulation process 99.6 % decolourization achieved and using XRD, FTIR	12
12	H_2SO_4	Reactive dye (methylene blue)	Turbidity and colour removal electrocoagulation and Electro - Fenton	52
13	NaCl	Blue SI dye	Electrocoagulation, XPS analysis, phyto and ecotoxicity 97.9%	8
14	HCL	Methyl orange	Electrocoagulation and magnetic field (EC – MF) lower energy consumption 95%	53
15	NaCl	Synthetic C. I reactive violet 2	Electrocoagulation color removal efficiencies 94.1%	54

Table 4 pH variation in ELECTROCOAGUALATION

Sl. No.	pH	Dye	Remarks	Authors
1.	7 (optimum)	Acid Red 1 (anionic), Basic Violet 3 (cationic) and Disperse Blue 14 (nonionic)	In the high and low pH, $Al(OH)_3$ is in its charged form and is soluble in water, hence, cannot be used for electro-coagulation-flotation. But in neutral pH, $Al(OH)_3$ is stable and insoluble in the water and available for pollutant adsorption from water.	55
2.	7 (optimum)	Bromophenol Blue (BPB).	The colour removal efficiency is optimum at pH 7 with roughly 100 % colour removal efficiency. The decrease in removal efficiency at more acidic and alkaline pH had been attributed to amphoteric behaviour of $Al(OH)_3$	36
3.	6.8 (optimum)	Acid Red 1 (AR1) (Synthetic Azo dye)	It was observed that with increase in pH, there was decrease in pollutant removal	9
4.	6.63 (optimum)	Reactive Black 5 dye (RB5)	6.63-7 pH resulted in 80.9% RB5 removal	37
5.	7	Acid Red 336	At inlet pH of 3.46 minimum removal was obtained, maximum removal was obtained in pH range 6.9-9 with a residence time of 35 min.	27
6.	4-10	brilliant green dye	Initial pH has significant effect on dye removal. In pH range of 4.5-8.5 max. removal of about 99% was observed. Beyond 8.5 there was no change in removal efficiency.	37
7.	5.4 (optimum)	Crystal violet dye	For initial pH lower than 5 and above 6, the removal efficiency was less. Max. removal was observed at pH 5.4	12
8.	2.5 to 5	Acid black 52 and acid yellow 220	Current density $40A/m^2$and electrolyte concentration 0 to 8 g/L	56
9.	5 to 8	Disperse dye	Electrocoagulation 95% and chemical coagulation 90% color removal at pH 8	57
10.	2 to 9	Disperse red 167, azo dye	Decolorization 100% bipolar electrocoagulation	54
11.	5 to 12	Synthetic C.I reactive	Optimum current density $79A/m^2$ and color	54

		violet 2 wastewater	removal efficiency 98%	
12.	6.50 to 11.35	Reactive black 5	Removal of KNO_3 49.1% and complete decolorization 99.9%	43
13.	3 to 4	Congo red dye	Response surface methodology and color 97% chemical oxygen demand 90% removal	36
14.	5 to 11	brilliant green dye	Dye removal 96.1% and pH 7	49
15.	3.0, 5.0, 7.0, 9.0, 11.0	Azo dye, acid red 18	Optimum condition pH 7.6 reaction time 60 min	58
16.	2 to 10	Methylene blue and indigo carmine	Optimum pH 10	59
17.	4.0 to 8.7 (Initial)	Acid black 194 dye	Aluminum anodes and initial pH 8.7 better removal	42
18.	3 to 8.75	Reactive dye (methylene blue)	Electrocoagulation and electro- Fenton 97% color removal and turbidity	8
29.	2 to 9	Reactive black 5	Reaction time 80 min, pH 6	60
20.	3 to 9	Reactive black B	High decolourization 99.6%	12
21.	5 to 10	Blue SI dye	Al- Al and Cu – Cu electrodes and optimum current density	61

The advantages and limitations are enlisted in table 5.

Table 5. Electrocoagulation advantages and disadvantage

Sl. No.	Advantage	Disadvantage
1.	Maximum good result aluminium and iron electrode	Electrode replaced regular
2.	Treat almost all type of wastewater like pH, turbidity.	More amount of wastewater produce industry
3.	Before process addition of chemical	Maintenance is most important
4.	Effective for very small size colloid particle	Requirement of Trained professional
5.	Less amount of sludge generates and settle down the process.	----

Conclusion

The study compared the electrocoagulation process and the conventional coagulation process. In electrocoagulation process color were removed to much greater extent and hence the pollution of the synthetic wastewater was reduced. Convention Coagulation is not as effective method as electrocoagulation for synthetic textile wastewater treatment. In the electrocoagulation process the pH effects the turbidity to a large extent. Optimum condition of Congo red dye pH 7 and electrolyte H_2SO_4 current density 5, 10, 15.

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