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

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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/cm²), temperature and mixing speed etc along with electrolyte concentration and type (NaCl, HCl, H₂SO₄). 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 as used in conventional wastewater plant. Coagulation/flocculation can be effective to enhance nanofiltration performance towards water reuse and minimisation of fouling textil effluent using different process coagulation/flocculation, enzymatic catalysis by commercial laccase and nanofiltration. Textile industry daily basis membrane process for dye wastewater treatment and fouling control. Compound bioflocculant used as a coagulation and dye wastewater treatment effect of pH solution. Treatment of highly concentrated wastewater containing multiple synthetics dyes combined process.

**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. 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](image1)

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

![Fig. No. 2 Chemical representation of Methylene blue](image2)

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. Used mostly in leather, paper and textile industry.

![Fig. No. 3 Chemical representation of Congo red](image3)
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 mandatory to develop 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 coagulation and electrogulation, Enzymatic catalysis, coagulation/flocculation and nanofiltration processes, membrane filtration, Compound bioflocculant, combined process of coagulation/flocculation and nanofiltration, dicyanidiamide fixer in the presence of ferric chloride. However, many of these techniques are often associated with limitations like high 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. 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. A period of 1-6 hours is required for settling.

### Table 1 Coagulant used in Conventional Coagulation 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 mixtures 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        |
| No. | Case Study                                                                 | Details                                                                                                                                  |
|-----|-----------------------------------------------------------------------------|-----------------------------------------------------------------------------------------------------------------------------------------|
| 3.  | Natural-based coagulants                                                   | Direct Blue pH 4, tannin-coagulant, while dosages up to 240 mg L\(^{-1}\)                                                             |
| 4.  | Natural coagulant                                                          | leachate treatment using 40 g/L of *Salvia hispanica* at pH 7                                                                         |
| 5.  | Polysilicate ferric manganese                                              | Active dye removal rate of the active yellow dye reached 86% at pH 5.0                                                                   |
| 6.  | polyaluminum chloride and flat-sheet cross-flow ultrafiltration            | deinking wastewater polyaluminum chloride dosage from 0 to 2,000 mg L\(^{-1}\)                                                        |
| 7.  | MgSO\(_4\) as a coagulant                                                 | Poly(vinyl alcohol) and reactive dye color attained 88.9%, 86.3%, and 99.2%                                                             |
| 8.  | Alkaline lignin (AL)                                                       | disperse dye high molecular weight like AL-g-DMC\(_1\) and AL-g-DMC\(_2\) exerted excellent color removals |
| 9.  | PFS/FeSO\(_4\) coagulation                                                | Sb(V) textile wastewater Temperature of 35 °C, pH ranged from 5 to 6                                                                   |
| 10. | Hybrid coagulant                                                          | reactive dye wastewaters treatment reduced by PFTS compared to T-PSF in reactive dye treatment                                         |
| 11. | Natural coagulant                                                         | water and wastewater treatment high purity of natural coagulants; and the synthesis of multifunctional natural coagulants        |
| 12. | Natural extract coagulant                                                  | dairy industry Calcium chloride (CaCl\(_2\)) good alternative for the primary treatment of dairy wastewater and remove 90% of turbidity. |
| 13. | Phytogenic aluminum sulfate nano coagulant                                | Congo red dye temperature from 30 °C to 70 °C                                                                                        |
| 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). |
| 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 %) |
| 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. |
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 production, ease of operation, and environmental compatibility. 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. 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.

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 for determining the effectiveness of synthetic wastewater treatment by electrocoagulation. Studies with effect of current density for electrocoagulation 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 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\(_2\)SO\(_4\) 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. 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 ELECTROCOAGULATION

| Sl. No. | Current Density | Dye | Remarks | Reference |
|---------|----------------|-----|---------|-----------|
| 1.      | 1.04, 2.08, 4.17 and 10.42 A/m² | 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/m² | 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/cm² (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/m² | 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/m² | brilliant green dye | Dye removal increased with increase in current density. However, for all current densities above 41.7 A/m², the removal efficiency was more than 99% for residence time of 30min. and above. | 37 |
| 7.      | 100-250 A/m² | 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/m² | 12 |
| 8.      | 13.9, 20.8, 27.8, 34.7, 41.7, 69.4, 138.9 A/m² | brilliant green dye | Optimum pH4.0 and highest dye removal salt concentration reduce the specific electrical energy consumption | 38 |
| 9.      | 1mA/cm² | Reactive Black 5 dye | Color 97% removal, TOC 97.3 Removal and natural pH | 39 |
| 10.     | 26, 53, 79, 106, 132 A/m² | Diazdye | The high efficiency electrocoagulation and anodic oxidation large amount wastewater treatment | 40 |
| 11.     | 5, 10,15 Am/cm² | Acid dye | Complete decolorization in 30 min | 41 |
| 12.     | (0.5, 2.5, 5.0 and 10.0 Am /cm² | Acid black 194 dye | Removing practically 100% total organic carbon | 42 |
| 13.     | 4.17 A/m² and 8.33 A/m² | 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/cm² | 44 |
and rapidly removed

| Sl. No | Electrolyte | Dye | Remark | Reference |
|-------|-------------|-----|--------|-----------|
| 10    | HCL         |      |        |           |
| 11    |             |      |        |           |
| 12    |             |      |        |           |
| 13    |             |      |        |           |
| 14    |             |      |        |           |
| 15    |             |      |        |           |
| 16    |             |      |        |           |
| 17    |             |      |        |           |
| 18    |             |      |        |           |
| 19    |             |      |        |           |
| 20    |             |      |        |           |

Table 3 ELECTROLYTE USED IN ELECTROCOAGULATION

| Electrolyte | Dye | Remark | Reference |
|-------------|-----|--------|-----------|
| NaCl        | Diazo dye | Electrocoagulation and anodic oxidation operation condition same but electrocoagulation more effective, EC(66.67), AO (44.96) | 40 |
| HCl         | Acid dye | Ozonation and electrocoagulation performed simultaneously decolorization acid dye 94.43 | 34 |
| HCl         | Acid black 194 dye (1.0L) | Electrocoagulation | 42 |
| 0.1 M NaOH, NaCl solution | Reactive red 241 | Compare between electrocoagulation, ultrasound and ultrasound assisted electrocoagulation 98% | 48 |
| NaOH, H2 SO4, HCl | Congo red dye | Electrocoagulation (89%) and response surface methodology (97%) | 36 |
| KCl         | Brilliant green dye | Electrocoagulation 96.1% | 49 |
| Na2SO4      | Real indigo dyeing | Response surface methodology 93.77% and electrocoagulation 92.07% | 50 |
| NaCl        | Azo dye | Peroxi- coagulation process | 44 |
| Electrolytes, With Cl-, SO42-, NO anions and Na+, K+, NH4+ | Reactive black 5 | Electrocoagulation process 98.0% | 42 |
| HCl         | Diazo dye i.e. Congo Red dye (CR) and a basic dye Methylene Blue (MB) | Electrocoagulation – flotation (86.04) and pulsed power technology (50.55) toxic dye removal | 51 |
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| Sl. No. | pH     | Dye                          | Remarks                                                                 |
|--------|--------|------------------------------|------------------------------------------------------------------------|
| 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. |
| 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 |
| 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 |
| 4.     | 6.63 (optimum) | Reactive Black 5 dye (RB5) | 6.63-7 pH resulted in 80.9% RB5 removal |
| 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. |
| 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. |
| 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 |
| 8.     | 2.5 to 5 | Acid black 52 and acid yellow 220 | Current density 40A/m² and electrolyte concentration 0 to 8 g/L |
| 9.     | 5 to 8  | Disperse dye | Electrocoagulation 95% and chemical coagulation 90% color removal at pH 8 |
| 10.    | 2 to 9  | Disperse red 167, azo dye | Decolorization 100% bipolar electrocoagulation |
| 11.    | 5 to 12 | Synthetic C.I reactive | Optimum current density 79A/m² and color |

Table 4 pH variation in ELECTROCOAGULATION

| Sl. No. | pH     | Dye | Electrocoagulation process | Remarks |
|---------|--------|-----|-----------------------------|---------|
| 11      | sodium chloride | Reactive Dye | 99.6% decolourization achieved and using XRD, FTIR |         |
| 12      | H₂SO₄ | Reactive dye (methylene blue) | Turbidity and colour removal electrocoagulation and Electro - Fenton |         |
| 13      | NaCl  | Blue SI dye | Electrocoagulation, XPS analysis, phyto and ecotoxicity 97.9% |         |
| 14      | HCL   | Methyl orange | Electrocoagulation and magnetic field (EC – MF) lower energy consumption 95% |         |
| 15      | NaCl  | Synthetic C. I reactive violet 2 | Electrocoagulation color removal efficiencies 94.1% |         |
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 H₂SO₄ current density 5, 10, 15.

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References

1. Dia, O., Drogui, P., Buelna, G. & Dubé, R. Hybrid process, electrocoagulation-biofiltration for landfill leachate treatment. Waste Manag. 75, (2018).
2. Zahrim, A. Y., Tizaoui, C. & Hilal, N. Coagulation with polymers for nanofiltration pre-treatment of highly concentrated dyes: A review. Desalination 266, 1–16 (2011).
3. Zahrim, A. Y., Tizaoui, C. & Hilal, N. Coagulation with polymers for nanofiltration pre-treatment of highly concentrated dyes: A review. Desalination 266, 1–16 (2011).
4. Khouni, I., Marrot, B., Moulin, P. & Ben Amar, R. Decolourization of the reconstituted textile effluent by different process treatments: Enzymatic catalysis, coagulation/flocculation and nanofiltration processes. Desalination 268, 27–37 (2011).
5. Thamaraiselvan, C. & Noel, M. Membrane processes for dye wastewater treatment: Recent progress in fouling control. Crit. Rev. Environ. Sci. Technol. 45, 1007–1040 (2015).
6. Huang, X. et al. Characterization and application of poly-ferric-titanium-silicate-sulfate in disperse and reactive dye wastewaters treatment. Chemosphere 249, 126129 (2020).
7. Liang, C. Z., Sun, S. P., Li, F. Y., Ong, Y. K. & Chung, T. S. Treatment of highly concentrated wastewater containing multiple synthetic dyes by a combined process of coagulation/flocculation and nanofiltration. J. Memb. Sci. 469, 306–315 (2014).
8. Zazou, H. et al. Treatment of textile industry wastewater by electrocoagulation coupled with electrochemical advanced oxidation process. J. Water Process Eng. 28, (2019).
9. Daud, M. et al. A review on the recent advances, challenges and future aspect of layered double hydroxides (LDH)–Containing hybrids as promising adsorbents for dyes removal. J. Mol. Liq. 288, 110989 (2019).
10. Moussa, D., El-Naas, M., Nasser, M. & Al-Marri, M. A comprehensive review of electrocoagulation for water treatment: Potentials and challenges. j 186, (2017).
11. Verma, A. K. Treatment of textile wastewaters by electrocoagulation employing Fe-Al composite electrode. J. Water Process Eng. 20, (2017).
12. Gautam, K., Kumar, S. & Kamsonlian, S. Decolourization of Reactive Dye from Aqueous Solution using Electrocoagulation: Kinetics and Isothermal Study. Zeitschrift fur Phys. Chemie (2019) doi:10.1515/zpch-2017-1044.
13. Liang, R., Chiu, E. & Loke, S. L. Secondary central nervous system involvement by non-Hodgkin’s lymphoma: The risk factors. Hematol. Oncol. 8, 141–145 (1990).
14. An, C., Huang, G., Yao, Y. & Zhao, S. Emerging usage of electrocoagulation technology for oil removal from wastewater: A review. Science of the Total Environment vol. 579 (2017).
15. Bener, S. et al. Electrocoagulation process for the treatment of real textile wastewater: Effect of operative conditions on the organic carbon removal and kinetic study. Process Saf. Environ. Prot. 129, 47–54 (2019).
16. Zhou, L., Zhou, H. & Yang, X. Preparation and performance of a novel starch-based inorganic/organic composite coagulant for textile wastewater treatment. Sep. Purif. Technol. 210, 93–99 (2019).
17. Dotto, J., Fagundes-Klen, M. R., Veit, M. T., Palácio, S. M. & Bergamasco, R. Performance of different coagulants in the coagulation/flocculation
process of textile wastewater. *J. Clean. Prod.* **208**, 656–665 (2019).

18. Lopes, E. C., Santos, S. C. R., Pintor, A. M. A., Boaventura, R. A. R. & Botelho, C. M. S. Evaluation of a tannin-based coagulant on the decolorization of synthetic effluents. *J. Environ. Chem. Eng.* **7**, 103125 (2019).

19. Tawakkoly, B., Alizadehdakhel, A. & Dorosti, F. Evaluation of COD and turbidity removal from compost leachate wastewater using Salvia hispanica as a natural coagulant. *Ind. Crops Prod.* **137**, 323–331 (2019).

20. Tang, L. *et al.* Removal of active dyes by ultrafiltration membrane pre-deposited with a PSFM coagulant: Performance and mechanism. *Chemosphere* **223**, 204–210 (2019).

21. Wu, Y. *et al.* Membrane fouling in a hybrid process of enhanced coagulation at high coagulant dosage and cross-flow ultrafiltration for drinking wastewater tertiary treatment. *J. Clean. Prod.* **230**, 1027–1035 (2019).

22. Shen, C. *et al.* A crosslinking-induced precipitation process for the simultaneous removal of poly(vinyl alcohol) and reactive dye: The importance of covalent bond forming and magnesium coagulation. *Chem. Eng. J.* **374**, 904–913 (2019).

23. Guo, K., Gao, B., Wang, W., Yue, Q. & Xu, X. Evaluation of molecular weight, chain architectures and charge densities of various lignin-based flocculants for dye wastewater treatment. *Chemosphere* **215**, 214–226 (2019).

24. Liu, Y. *et al.* Coagulation removal of Sb(V) from textile wastewater matrix with enhanced strategy: Comparison study and mechanism analysis. *Chemosphere* **237**, 124494 (2019).

25. Ang, W. L. & Mohammad, A. W. State of the art and sustainability of natural coagulants in water and wastewater treatment. *J. Clean. Prod.* **262**, 121267 (2020).

26. Triques, C. C. *et al.* Influence evaluation of the functionalization of magnetic nanoparticles with a natural extract coagulant in the primary treatment of a dairy cleaning-in-place wastewater. *J. Clean. Prod.* **243**, 118634 (2020).

27. Garvasis, J., Prasad, A. R., Shamsheera, K. O., Jaseela, P. K. & Joseph, A. Efficient removal of Congo red from aqueous solutions using phytopgenic aluminum sulfate nano coagulant. *Mater. Chem. Phys.* **251**, 123040 (2020).

28. Yang, S., Li, W., Zhang, H., Wen, Y. & Ni, Y. Treatment of paper mill wastewater using a composite inorganic coagulant prepared from steel mill waste pickling liquor. *Sep. Purif. Technol.* **209**, 238–245 (2019).

29. Chen, J. *et al.* High-efficiency extraction of aluminum from low-grade kaolin via a novel low-temperature activation method for the preparation of poly-aluminum-ferric-sulfate coagulant. *J. Clean. Prod.* **257**, 120399 (2020).

30. Xue, M., Gao, B., Li, R. & Sun, J. Aluminum formate (AF): Synthesis, characterization and application in dye wastewater treatment. *J. Environ. Sci.* **74**, 95–106 (2018).

31. Lemlikchi, W., Khaldi, S., Mecherri, M. O., Lounici, H. & Drouiche, N. Degradation of Disperse Red 167 Azo Dye by Bipolar Electrocoagulation. *Sep. Sci. Technol.* **47**, 1682–1688 (2012).

32. Ahangarnokolaei, M. A., Ganjidoust, H. & Ayati, B. Optimization of parameters of electrocoagulation/flotation process for removal of acid red 14 with mesh stainless steel electrodes. *J. Water Reuse Desalin.* **8**, 278–292 (2018).

33. Graça, N. S., Ribeiro, A. M. & Rodrigues, A. E. Modeling the electrocoagulation process for the
treatment of contaminated water. Chem. Eng. Sci. 197, (2019).
34. Behin, J., Farhadian, N., Ahmadi, M. & Parvizi, M. Ozone assisted electrocoagulation in a rectangular internal-loop airlift reactor: Application to decolorization of acid dye. J. Water Process Eng. 8, 171–178 (2015).
35. Bazrafshan, E., Alipour, M. R. & Mahvi, A. H. Textile wastewater treatment by application of combined chemical coagulation, electrocoagulation, and adsorption processes. Desalin. Water Treat. 57, 9203–9215 (2016).
36. Akhtar, A., Aslam, Z., Asghar, A., Bello, M. M. & Raman, A. A. A. Electrocoagulation of Congo Red dye-containing wastewater: Optimization of operational parameters and process mechanism. J. Environ. Chem. Eng. 8, 104055 (2020).
37. El-Ashtoukhy, E. S. Z., Amin, N. K., Abd El-Latif, M. M., Bassyouni, D. G. & Hamad, H. A. New insights into the anodic oxidation and electrocoagulation using a self-gas stirred reactor: A comparative study for synthetic C.I Reactive Violet 2 wastewater. J. Clean. Prod. 167, 432–446 (2017).
38. Syam Babu, D., Anantha Singh, T. S., Nidheesh, P. V. & Suresh Kumar, M. Industrial wastewater treatment by electrocoagulation process. Sep. Sci. Technol. 00, 1–33 (2019).
39. Yavuz, Y. & Shahbazi, R. Anodic oxidation of Reactive Black 5 dye using boron doped diamond anodes in a bipolar trickle tower reactor. Sep. Purif. Technol. 85, 130–136 (2012).
40. Bassyouni, D. G., Hamad, H. A., El-Ashtoukhy, E. S. Z., Amin, N. K. & El-Latif, M. M. A. Comparative performance of anodic oxidation and electrocoagulation as clean processes for electrocatalytic degradation of diazo dye Acid Brown 14 in aqueous medium. J. Hazard. Mater. 335, 178–187 (2017).
41. Behin, J., Farhadian, N., Ahmadi, M. & Parvizi, M. Ozone assisted electrocoagulation in a rectangular internal-loop airlift reactor: Application to decolorization of acid dye. J. Water Process Eng. 8, 171–178 (2015).
42. Vidal, J., Villegas, L., Peralta-Hernández, J. M. & Salazar González, R. Removal of Acid Black 194 dye from water by electrocoagulation with aluminum anode. J. Environ. Sci. Heal. - Part A Toxic/Hazardous Subst. Environ. Eng. 51, 289–296 (2016).
43. Keyikoglu, R., Can, O. T., Aygun, A. & Tek, A. Comparison of the effects of various supporting electrolytes on the treatment of a dye solution by electrocoagulation process. Colloids Interface Sci. Commun. 33, 100210 (2019).
44. do Vale-Júnior, E., da Silva, D. R., Fajardo, A. S. & Martínez-Huitle, C. A. Treatment of an azo dye effluent by peroxy-coagulation and its comparison to traditional electrochemical advanced processes. Chemosphere 204, 548–555 (2018).
45. El-Ashtoukhy, E.-S. Z., Amin, N. K., Abd El-Latif, M. M., Bassyouni, D. G. & Hamad, H. A. New insights into the anodic oxidation and electrocoagulation using a self-gas stirred reactor: A comparative study for synthetic C.I Reactive Violet 2 wastewater. J. Clean. Prod. 167, 432–446 (2017).
46. Khosravi, R., Hazrati, S. & Fazlzadeh, M. Decolorization of AR18 dye solution by electrocoagulation: sludge production and electrode loss in different current densities. Desalin. Water Treat. 57, 14656–14664 (2016).
47. Zodi, S., Merzouk, B., Potier, O., Lapicque, F. & Leclerc, J.-P. Direct red 81 dye removal by a continuous flow electrocoagulation/flotation reactor. Sep.
41. Singh and Ransingh, 2020, Coagulation and Electrocoagulation Process for Dye Removal from Textile Wastewater: A Review

42. Özyonar, F., Gökkuş, Ö. & Sabuni, M. Removal of disperse and reactive dyes from aqueous solutions using ultrasonic-assisted electrocoagulation. *Chemosphere* **258**, (2020).

43. Mariah, G. K. & Pak, K. S. Removal of brilliant green dye from aqueous solution by electrocoagulation using response surface methodology. *Mater. Today Proc.* **20**, 488–492 (2020).

44. Hendaoui, K. *et al.* Real indigo dyeing effluent decontamination using continuous electrocoagulation cell: Study and optimization using Response Surface Methodology. *Process Saf. Environ. Prot.* **116**, 578–589 (2018).

45. Nippatla, N. & Philip, L. Electrocoagulation-floatation assisted pulsed power plasma technology for the complete mineralization of potentially toxic dyes and real textile wastewater. *Process Saf. Environ. Prot.* **125**, 143–156 (2019).

46. Hosseinifard, S. M., Aroon, M. A. & Dahrazma, B. Application of PVDF/HDTMA-modified Clinoptilolite Nanocomposite Membranes in Removal of Reactive Dye from Aqueous Solution. *Sep. Purif. Technol.* 117294 (2020) doi:https://doi.org/10.1016/j.seppur.2020.117294.

47. Irki, S., Ghernaout, D. & Naceur, M. W. Decolourization of methyl orange (MO) by electrocoagulation (EC) using iron electrodes under a magnetic field (MF). *Desalin. Water Treat.* **79**, 368–377 (2017).

48. El-Ashtoukhy, E.-S. Z., Amin, N. K. & Abdelwahab, O. Removal of lead (II) and copper (II) from aqueous solution using pomegranate peel as a new adsorbent. *Desalination* **223**, 162–173 (2008).

49. Kłodowska, I., Rodziewicz, J., Janczukowicz, W., Cydzik-Kwiatkowska, A. & Parszuto, K. Effect of citric acid on the efficiency of the removal of nitrogen and phosphorus compounds during simultaneous heterotrophic-autotrophic denitrification (HAD) and electrocoagulation. *Ecol. Eng.* **95**, 30–35 (2016).

50. Pajootan, E., Arami, M. & Mahmoodi, N. M. Binary system dye removal by electrocoagulation from synthetic and real colored wastewaters. *J. Taiwan Inst. Chem. Eng.* **43**, 282–290 (2012).

51. Merzouk, B., Gourich, B., Madani, K., Vial, C. & Sekki, A. Removal of a disperse red dye from synthetic wastewater by chemical coagulation and continuous electrocoagulation. A comparative study. *Desalination* **272**, 246–253 (2011).

52. Azarian, G. *et al.* Monopolar Electro-Coagulation Process for Azo Dye C. I. Acid Red 18 Removal from Aqueous Solutions. *Avicenna J. Environ. Heal. Eng.* **1**, (2014).

53. Othmani, A., Kesraoui, A., Hanene Akrout, Elaissaoui, I. & Seffen, M. Coupling anodic oxidation, biosorption and alternating current as alternative for wastewater purification. *Chemosphere* **249**, 126480 (2020).

54. Naraghi, B., Baneshi, M. M., Amiri, R., Dorost, A. & Biglari, H. Removal of Reactive Black 5 dye from aqueous solutions by coupled electrocoagulation and bio-adsorbent process. *Electron. Physician* **10**, 7086–7094 (2018).

55. Kalivel, P. *et al.* Elucidation of electrocoagulation mechanism in the removal of Blue SI dye from aqueous solution using Al-Al, Cu-Cu electrodes - A comparative study. *Ecotoxicol. Environ. Saf.* **201**, 110858 (2020).