# Elimination of Anionic Dye from Wastewater through Coagulation/Flocculation process

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***Abstract - Effluents discharged from various textile industries are responsible for one of the major environmental pollution. Textile wastewater contains a variety of components some of which are exceedingly toxic to the environment. Several wastewater treatment technologies have been used from time to time by the researchers. Coagulation-flocculation has always been proven an effective treatment technology for removing dye pollutants. This research was conducted to study the maximum removal efficiency of the anionic dye from synthetic wastewater for the best coagulant or coagulant-flocculant combination. Comparing the results among the different coagulants and flocculants with same dosages it was observed that at 400 ppm concentration and at pH 7, polyaluminium chloride alone gave the best possible colour removal efficiency of 99.7%. Although both sodium alginate and sodium aluminate showed significantly good results when used as flocculants. Colour removal efficiency was also investigated for pH variation. Congo Red being an anionic dye showed maximum colour removal efficiency at acidic to neutral pH i.e., pH 5-7.***

***Keywords: Textile industry; wastewater; Congo Red dye; coagulation; flocculation; Jar Apparatus.***

## 1.0 Introduction

Textile, tannery, paint, paper and pulp industries are some of the major consumers of synthetic dyes. Among them, the Textile industry has the largest contribution of releasing hazardous dye effluents into the water. These pollutants greatly affect the water and soil and create severe threats to the ecosystem with which they are associated (Popuri *et al.*, 2019). Carcinogenic Synthetic dyes are most desirable due to their availability and it causes in an increase in biochemical oxygen demand (BOD) and Chemical oxygen demand (COD) in the effluent (Sarayu et al., 2012). High levels of biochemical oxygen demand (BOD) and chemical oxygen demand (COD) in untreated wastewater from textile industries may result in rapid depletion of dissolved oxygen in the water bodies creating a toxic environment for aquatic flora and fauna. (Patel et al., 2010). Thus, it is of great importance for the textile factories to oblige by the environmental norms directed to control the harmful impacts of the dye effluents.

There are plenty of techniques can be used like precipitation, microbial degradation, chemical oxidation, filtration, adsorption, , membrane separation, coagulation-flocculation, reverse osmosis electrochemical treatment, ozonation, hydrogen peroxide catalysis, flotation, and biological techniques for the textile wastewater treatment. In most of these technologies, the most essential step is to characterize the wastewater by analysing characteristics such as pH, BOD, COD, sulphate, colour, heavy metals and total suspended solids (TSS), (Al-Tohamy et al., 2022).

Coagulation-flocculation is the most recommended method of treating wastewater (Islam and Mostafa 2018a) as it is most efficient and simple to operate. This process is also less temperature sensitive, low costlier and effective in colour removal efficiency with considering reducing parameters such as TDS, BOD and COD (Hu et al., 2017). Coagulation is a process of addition of an external chemical agent that is capable of destabilizing the charges contained by the dye particles suspended in wastewater. Once the charges are destabilized the suspended particles form micro flocs. Coagulation is succeeded by flocculation. The micro flocs formed collide with each other resulting bigger and heavier flocs. This process is aided by some coagulant aid or flocculants that binds and strengthens the flocs together. The heavier flocs thus settle down forming sediments and finally separated as the suspended solid dye particles present in the effluent stream. (The *et al.*, 2016). The dye pollutants in textile industry wastewater are classified on the basis of ionic nature, dye properties, toxicity, chemical type and chemical structure of the dyes. Ionic dyes are classified into two types; cationic and anionic dyes. Cationic dyes give positively charged ion in an aqueous solution. Cationic dyes mostly contain the azo group, triarylmethane group and anthraquinone groups. Examples of Cationic dyes are Methylene blue (MB), Crystal Violet (CV), Rhodamine B etc. Dyes that give negatively charged ions when dispersed in an aqueous solution are called anionic dyes. Anionic dyes generally contain sulphonic acid or carboxylic acid group. Examples of anionic dyes include Acid yellow, Methyl Orange (MO), Congo Red (CR) etc. (Benkhaya *et al.*, 2020).

Congo Red is an anionic azo dye that is widely used in the textile industry. Such azo dyes are popularly used in textile manufacturing industries because they provide a variety of colour shades and have longer colour retention properties. They don't get decolourised easily and can easily stain textiles. Congo Red is a dye belonging to this category and it has widespread application in textile manufacturing industries because of its easy availability and quick staining property (Harja et al., 2022).

Our aim is to study the best possible combination of coagulant-flocculant that causing maximum anionic dye removal from the synthetic wastewater. And also to determine the optimum conditions of pH resulting in most efficient dye removal.

## 2.0 Methodology

The coagulation-flocculation processes were conducted with Jar test apparatus to get the highest anionic dye removal efficiency from synthetic wastewater.

### 2.1 Preparation of Simulated wastewater

Congo red dye was purchased from Loba Chemicals. Deionised water was used to prepare the simulated wastewater. 1000ppm stock solution was prepared to obtain 50ppm solution for further sets of experiment.

### 2.2 Chemicals

The chemicals used as coagulant for conducting the experiments were Polyaluminium Chloride (PAC) and Aluminium Sulfate (alum, $Al_2(SO_4)_3$) obtained from Merck Chemicals. Sodium Aluminate and Sodium Alginate were used as coagulant aid (from Loba Chemicals). Maintaining of the pH and optimum temperature is very important for successfully conduction of the experiment and obtaining optimum results. So, sulfuric acid [$H_2SO_4$] and sodium hydroxide (NaOH) were used to maintain the pH of the dye solution.

### 2.3 Method and Analysis

To study the best combination of coagulant and flocculant a known amount of coagulants and flocculants were added in 50 ppm synthetic wastewater of a series of 1000 mL beakers and they were placed properly in Jar test apparatus (Make-KPH-143 TEMPSTAR) showed in Fig. 1. The coagulant or coagulant aid and flocculant were added to the synthetic wastewater solution depending on the treatment requirements. Basically, 1:1 dosage ratio of coagulant and flocculant was maintained throughout the experiment. 8 such sets were prepared for each experiment with varying amount of coagulant or coagulant and flocculant ranging from 50 ppm – 1000 ppm. The pH of the solution was maintained adding sulphuric acid and sodium hydroxide. The pH optimization was done by varying the pH from 4-8.


Fig 1 Maximum Colour removal of Congo Red using PAC in Jar Apparatus

The experiment was conducted initially maintaining the stirrer speed at 200 rpm for 2 minutes for rapid mixing. Then slow mixing was conducted by setting the stirrer speed at 50 rpm for 15 minutes. Finally the solution was kept at rest for 30 minute for sedimentation. Final pH of the solution was measured. After completion of the coagulation–flocculation process total amount of the final dye solution was measured by the UV-VIS-NIR spectrophotometer (Make- U-4100 HITACHI) at the maximum wavelength of 499 nm ($\lambda_{max}$). The corresponding concentration of total amount of anionic dye (congo red) in aqueous solution is determined by using the standard curve method. All calibrations were performed using standard curve over the used dye range of 10–90.0 mg/L and the obtained standard curve had linear correlation coefficients ($R^2$) of 0.996. The removal percentage of dye were calculated by the equation,

$$\text{Removal percentage} = \frac{(C_0 - C)}{C_0} \times 100 \qquad (1),$$

Where, $C_0$ and C are the initial and final concentration of dye (mg/*l*)

## 3.0 Results and Discussion

In this section, estimation of maximum dye removal efficiency for the best combination of coagulant and flocculant was done. It was investigated comparing the results using different coagulants and flocculants at different concentrations (ppm) maintaining 1:1 dosage ratio of coagulant and flocculant. Among all the coagulant and flocculant combinations both the coagulant and the coagulant aids showed impressive results as showed in Fig. 2. Among them, polyaluminium chloride with 400 ppm alone gave maximum removal efficiency of about 99.7% showed in Fig. 2. The values of removal percentages of dye in synthetic wastewater were determined using equation (1) and presented in Table 1.

Table 1: Removal efficiency of different coagulants and flocculants at different concentrations (ppm) for Congo Red Dye

| Coagulant Dose (ppm) | Removal Efficiency of ALUM(%) | Removal Efficiency of PAC(%) | Removal Efficiency of ALUM + SA(%) | Removal Efficiency of ALUM + SALG(%) | Removal Efficiency of PAC + SA(%) | Removal Efficiency of PAC + SALG(%) |
|---|---|---|---|---|---|---|
| 50 | 22.83 | 20.63 | 82.24 | 49.67 | 61.85 | 31.71 |
| 100 | 25.51 | 79.85 | 91.02 | 51.76 | 94.97 | 38.29 |
| 200 | 26.63 | 98 | 98.68 | 56.24 | 98.1 | 44.58 |
| 300 | 88.54 | 99.66 | 98.78 | 59.41 | 99.36 | 99.17 |
| 400 | 93.32 | 99.7 | 98.83 | 99.17 | 99.07 | 99.41 |
| 500 | 98.15 | 99.6 | 97.95 | 99.46 | 98.2 | 99.41 |
| 750 | 98.34 | 98.97 | 99.12 | 89.36 | 96.76 | 97.71 |
| 1000 | 98.15 | 98.68 | 99.02 | 69.85 | 98.78 | 69.85 |

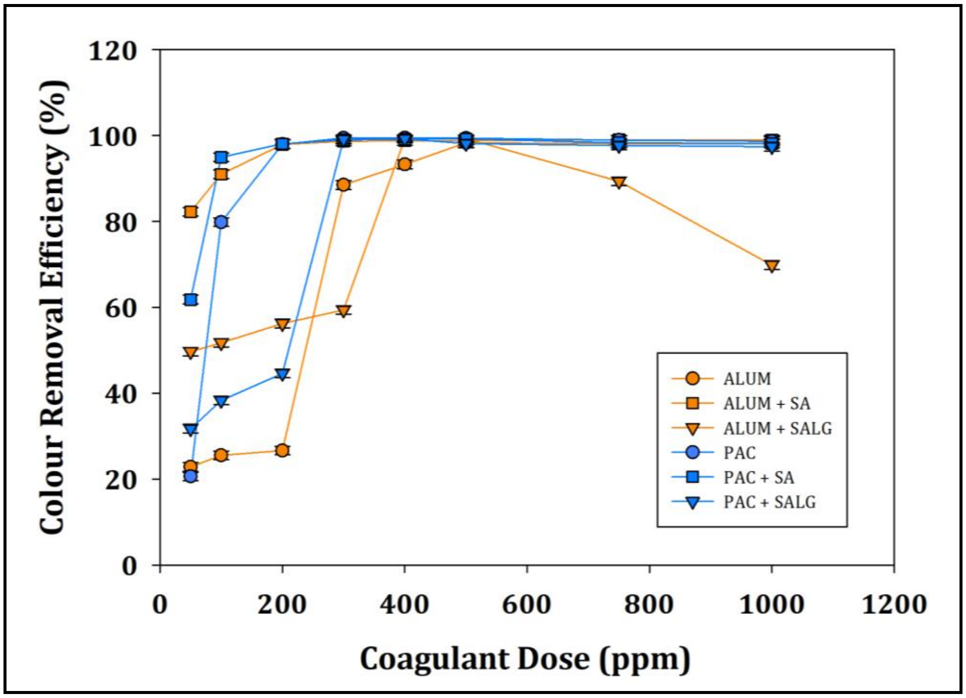


Fig. 2 Comparison of removal efficiency (%) of different coagulants and flocculants at different dosages (ppm)for 1:1 Coagulant and Flocculant Ratio for Congo Red Dye

It was also observed from the values of Table 2 and comparison showed in Fig. 3 that from pH 4 – 6 the trend of removal efficiency was increasing and after pH 6 it was gradually going downward for some of the combinations of coagulant and flocculant.

Table 2: Colour removal efficiency (%) with pH variation for Congo Red Dye

| pH | Removal Efficiency of ALUM(%) | Removal Efficiency of ALUM + SA(%) | Removal Efficieny of ALUM + SALG(%) | Removal Efficiency of PAC(%) | Removal Efficiency of PAC + SA(%) | Removal Efficiency of PAC + SALG(%) |
|---|---|---|---|---|---|---|
| 4 | 96.68 | 99.07 | 99.51 | 60.29 | 99.07 | 99.8 |
| 5 | 99.56 | 98.78 | 99.41 | 60.39 | 99.56 | 99.8 |
| 6 | 99.36 | 98.97 | 88.24 | 81.07 | 99.56 | 99.12 |
| 7 | 93.32 | 98.83 | 99.41 | 99.71 | 99.07 | 99.41 |
| 8 | 54.39 | 72.29 | 42.54 | 45.32 | 98.05 | 98.97 |

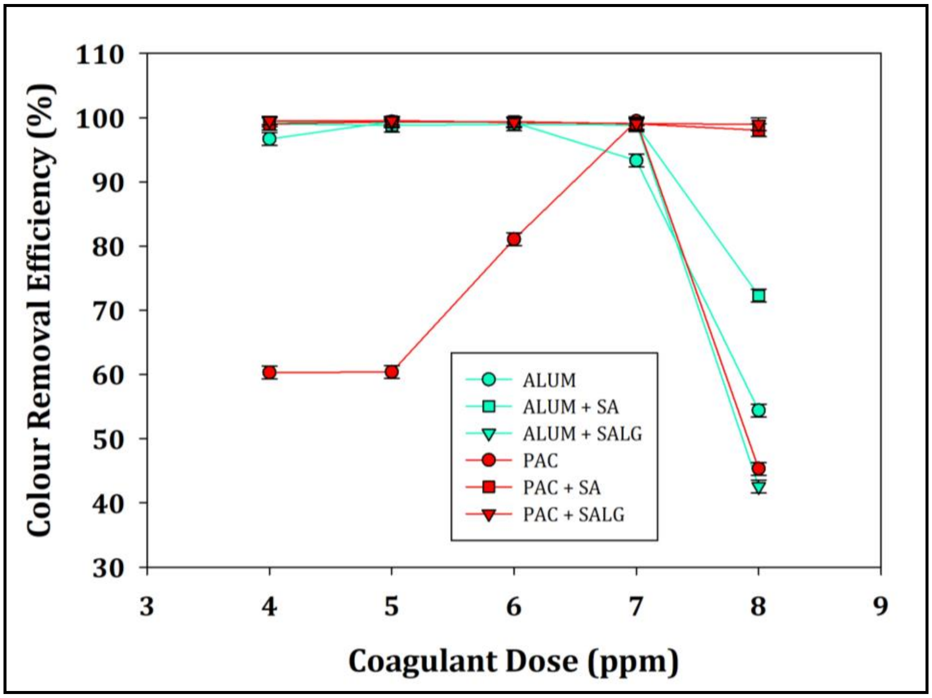


Fig. 3 Comparison of colour removal efficiency (%) at different pH for Congo Red Dye

Comparing all results it was investigated that polyaluminium chloride (PAC) alone showed better results than Alum for the removal of Congo Red dye. Both sodium alginate and sodium aluminate showed significantly good results when used as flocculants. However, at higher coagulant concentration ranges of 400-1000 ppm, the addition of flocculants did not bring any significant improvement in the colour removal efficiency. From the experiments, it was also observed that the colour removal efficiency initially increased with an increase in coagulant-flocculant dosage but after reaching the maximum, the colour removal efficiency did not increase any further with an increase in coagulant-flocculant dosage and thus attained saturation. The colour removal efficiency also varied significantly with pH variation. It was observed that the maximum colour removal efficiency was found for acidic to neutral pH (5-7). Thus anionic dyes can be easily removed at lower pH. Whereas at basic pH (8), the colour removal efficiency obtained were not satisfactory.

Cost analysis of wastewater treatment for each coagulant-flocculant combination per kg is presented below in table 3. From the above discussions, it is observed that PAC alone can remove colour of anionic dye about 99.7% and also its cost analysis showed that PAC is much more economically feasible than other coagulant or with any coagulant-flocculant combinations.

Table 3: Cost analysis of different coagulants and flocculants

| Items | Cost per Kg for Alum + SA (in USD) | Cost per Kg for Alum + SALG (in USD) | Cost per Kg for PAC + SA (in USD) | Cost per Kg for PAC + SALG (in USD) |
|---|---|---|---|---|
| Aluminium Sulfate | $9.78 | $9.78 | × | × |
| Polyaluminim Chloride | × | × | $6.47 | $6.47 |
| Sodium Aluminate | $2.72 | × | $2.72 | × |
| Sodium Alginate | × | $59.59 | × | $59.59 |
| Sodium Hydroxide | $9.63 | $9.63 | $9.63 | $9.63 |
| Total | $22.13 | $79 | $18.81 | $75.69 |

## 5.0 Conclusion

The removal of anionic dye from synthetic wastewater was investigated by coagulation-flocculation process. The experimental data was used to estimate the removal percentage of dye using various coagulants and flocculants. The laboratory studies proved a good efficiency of the coagulation–flocculation process for removal of the anionic dye such as congo red; which is mostly used in various textile industries. It also outlined the best and economically feasible coagulant or combined coagulant-flocculant as its future applicability in large scale in wastewater treatment technologies.
Although, more extensive research and developmental work is needed to achieve the best removal efficiency by performing more experimentation for dosages ratios of 1:2 and 2:1 of coagulant with flocculant for the same dye and also for a cationic dye as most of the textile industries wastewater contains various types of dyes. Further, characterization of the sediment formed during coagulation-flocculation process can be performed using FTIR, XRD, SEM and EDX analysis to understand reaction mechanism during coagulation-flocculation process. In future, it can be applied to raw industrial wastewater from textile manufacturing industries.

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