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# Effect of reactional parameters on the elimination of Congo Red by the combination of coagulation–floculation with aluminum sulfate

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**ABSTRACT**

The main objective of our study is to test the performance of aluminum sulfate (AS) and the coagulation–flocculation process in removing a dye widely used in the textile industry called Congo Red (CR). In order to achieve our objective, the jar-test tests were carried out in the laboratory on synthetic solutions. Various reaction parameters were tested such as the effect of variation in coagulant dose, variation in initial CR content, and variation in pH. A combination between the AS and coagulants aid such as powder activated carbon (PAC), polymers, and lime ($Ca(OH)_2$) has been carried out to improve this process. The results obtained indicated for 30 mg/L concentration of CR, the AS optimal dose is found 40 mg/L. This dose has an elimination yield around to 97.24%. For the pH effect, the best performance was achieved in the range of pH between 4 and 6. Similar efficiency trends were obtained for 50 mg/L of the initial content of the CR. The combination of AS with coagulants aid has also shown that there is a very strong elimination marked by the small amounts of polymer, where the yield exceed 99%, in comparison with PAC and lime.

*Keywords:* Congo red; Aluminum sulfate; Powder activated carbon; Polymer; Lime; Coagulation–flocculation

## 1. Introduction

Water pollution is considered as the greatest danger to the environment and the degradation of ecosystems. Whether accidental or anthropogenic, this pollution is the result of the massive use of organic and mineral pollutants of agricultural, urban, and industrial origin. So, the textile industry is an important source of liquid effluents charged with polluting, because of the large volume of water and coloring and non-coloring substances used in the printing operations, dyeing, and finishing [1,2]. These waters are globally characterized by high values of turbidity, biological oxygen demand (BOD), carbon organic dissolved chemical oxygen demand (COD), and other pollutants.

Among the dyes, we find the synthetic type that is used in the textile industry. Most synthetic dyes are made up of azo, anthraquinone dyes (65%–75% of dyes), and that represent this type of contaminant [3]. Currently, synthetic dyes represent a relatively large group of organic chemical compounds found in virtually every sphere of our daily life. They are more popular than natural dyes because the former have greater durability and shine [3,4].

It is important to note that, the disposal of wastewater containing industrial dyes into rivers, and lakes without proper treatment has caused many problems. Therefore, it does not only affects the aesthetic quality but, also decreases the transmittance of light into the water, as well as reduces photosynthesis, which contributes to the degradation of the water environment. Indeed, the presence of large amounts of dyes is toxic which can harm aquatic ecosystems, human health. They can be responsible for acute or chronic respiratory pathologies (asthma), urinary tract and lung

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cancer, harmful effects on red blood cells, skin effects allergic (eczema, urticaria), and the horrible effects like carcinogenic or teratogenic properties, mutagenic and so on. Because, of its own color exposure and toxicity even at low concentrations, therefore, one of the biggest environmental problems today is eliminating dyes from the wastewater [5,6].

The discoloration of these rejects is often difficult since conventional treatments do not easily degrade organic dyes [7]. Therefore, different treatment processes such as chemical oxidation [1,8,9], electrocoagulation [10–12], biodegradation [13–15], adsorption [2,15–19], chemical coagulation [20–27], and membrane processes [28,29] can achieve dyes removal from water and industrial wastewater.

However, the most of the techniques mentioned above are limited by methods, costs or difficulties during the operation, unlike the coagulation–flocculation process, which is a traditional technique, but it is one of the most effective and easiest technical methods that has been used to eliminate dyes from industrial wastewater [30]. At this time, it is important to point out that, in Algeria, at drinking water production or wastewater treatment plants; aluminum sulfate (AS) is very often used as a coagulant. Because, it is a very efficient chemical reagent at removing organic and inorganic pollutants, although the purchase cost is significant. A comparative study between the purchase price of aluminum sulfate (AS) and lime carried out by Bouchahm et al. [31], they clearly indicated that lime is cheaper and more economical (approximately 2 euros/10 kg) compared to the cost of treatment by the aluminum sulfate (approximately 15 euros/10 kg). Therefore, the combination with coagulants aid such as powder activated carbon (PAC) or polymers is considered to be one of the most effective methods of removing suspended solids and decreases the amount of coagulant. This automatically translates into a reduction in the cost of coagulant. The combination is widely used in various domestic wastewater treatment plants, and even in several industries for removing dyes from industrial wastewater, due to its important role in the primary treatment of wastewater from [4,25,27].

It is interesting to note that the effectiveness of flocculation coagulation depends on the characteristics of the water to be treated, the dose of coagulant, and the pH; several mechanisms may be involved in the treatment process such as charge neutralization followed by precipitation of the insoluble dye-coagulant complexes, and their adsorption into the aluminum hydroxides. In the treatment of drinking water from surface water, coagulation–flocculation is the most important step in water clarification. In this study, we will determine the favorable conditions, study the performance of aluminum sulfate, and the process of coagulation–flocculation in removing color from a synthetic solution containing a dye belonging to the diazo family group (direct dye) named Congo Red (CR). This process will be followed by a combination with three coagulants aid such as PAC, polymer, and lime in order to improve the removal of the CR dye.

## 2. Materials and methods

All experiments were performed at Research Laboratory of Underground and Surface Hydraulics (LARHYSS) at Biskra University in Algeria. The CR and the Lime were purchased from Sigma-Aldrich Company and product respectively of India and Germany. Wille the AS was provided by Fluka and product of Germany. Noting that all, chemicals (CR, AS, and the Lime) are used in our study characterized by purity greater than 97% and used without undergoing any treatment.

### 2.1. Solutions and reagents

#### 2.1.1. Description and preparation of the CR dye

The CR used in this work is an anionic dye, also called benzidinediazo-bis-1-naphthylamine-4-sulfonic acid is a diazo molecule. That is to say, which has twice the structure of azobenzene [7]. It is widely used in the production of papers, plastics, leather, and in particular in the textile industry [32]. Several studies have shown that it is toxic like many azo dyes, which requires the application of an effective and feasible method to treat this dye in effluents [5,33,34]. The main physicochemical properties of this dye are shown in Table 1and Fig. 1 [35].

The coagulation–flocculation tests were carried out on solutions of the CR dye obtained by dilution from stock solution at 1 g/L. This concentration is lower than the solubility limits generally accepted by CR.

#### 2.1.2. Description and preparation of the coagulant

In this study, we chose to inject AS as a coagulant. SA is a white, odorless chemical powder with the formula $Al_2(SO_4)_3$, $18H_2O$ and a molar mass of 666.6 g/mol. The coagulant solution was prepared periodically using distilled water. We have prepared in a large volume flask (1 L) a stock solution of coagulant at a concentration of 20 g/L.

#### 2.1.3. Coagulants aid

In order to assess the effect of the coagulants aid on the elimination of CR dye, three coagulants aid were used which are very useful in the field of water treatment and research. In order to test their performances in combination with the coagulant in the elimination of CR. The coagulants aid tested are PAC, a commercial product. It is a mesoporous adsorbent characterized by a particle size of 20 µm and a specific surface of 658 $m^2$/g. The polymer is an agriculture food product used in a treatment plant of drinking water and the lime ($Ca(OH)_2$), which is a commercial chemical.

Table 1
Characteristics of CR

| Name | Description of characteristics |
|---|---|
| Chemical name | Congo Red (direct Red 28) |
| Appearance | Dark Red powder |
| Molecular formula | $C_{32}H_{22}N_6Na_2O_6S_2$ |
| Molar mass (g/mol) | 696.663 ± 0.04 g/mol |
| pKa | >4 |
| $\lambda_{max}$ | 500 nm |
| $T$° fusion | >360 |
| Water | High solubility |
| Utility | Textile |

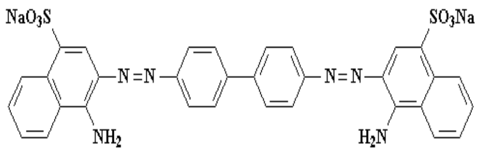


Fig. 1. Chemical structure of CR [35].

*2.1.4. Dosage of the CR dye*

The reduction percentage of the CR dye tested is evaluated by measuring the absorbance (A). These measurements are carried out on a spectrophotometer of the "UV/Vis OPTIZEN 2120 UV" type, at a wavelength $\lambda_{max}$ = 500 nm. To plot the CR calibration curves, we determined that, absorbance values corresponding to different concentrations of the standard solutions.

*2.2. Description of jar-test*

The jar-test tests were carried out on a flocculator with 6 agitators (Flocculator Fisher 1198) with an individual rotation speed varying between 0 and 200 rpm. This apparatus allows the solutions contained in a series of 500 mL beakers to be agitated simultaneously. The solutions enriched in CR and in reagents (coagulant and coagulants aid) are subjected for 3 min to rapid stirring of 150 rpm. The speed has then reduced to 45 rpm for a duration of 17 min.

For the tests with the coagulant alone, even in combination with the coagulants aid, and after decantation for 30 min, the supernatant is recovered to be filtered under vacuum on an OSMONICS INC membrane with a porosity of 0.45 µm. The filtrate has then been assayed by analysis with a spectrophotometer. The evaluation of the percentage of CR dye removal was calculated by the following equation (Eq. (1)):

$$\text{CR Removal}(\%) = \left(\frac{\left(C_0 - C_f\right)}{C_0}\right) \times 100 \quad (1)$$

where $C_0$ and $C_f$, respectively, represent the initial and final concentrations of CR expressed in mg/L.

Different reaction parameters will also be studied, such as:

- Effect of the variation in the dose of the coagulant, we considered a range going from 5 to 100 mg/L of AS.
- Effect of varying the initial CR content of 1–50 mg/L.
- Effect of pH variation between 1 and 10.

The last trial concerns the hybrid effect of coagulant AS and three coagulants aid, namely: PAC, polymers, and lime in the form of ($Ca(OH)_2$).

## 3. Results and discussion

*3.1. Effect of the coagulant dose*

Before starting the tests concerning the effect of the reaction parameters (variation of the initial concentration of the dye, pH, and the effect of coagulants aid concentration), the synthetic solutions of the CR dye are prepared at a concentration of 30 mg/L, in distilled water with an unadjusted pH (pH = 4.50 of synthetic solution). These solutions are subjected to the jar-test under the variation of the doses of coagulant in the form of AS. The range of coagulant varies between 5 and 100 mg/L. This range was chosen in order to determine the optimal dose of AS. The results obtained have been presented in (Fig. 2).

The results clearly show that the elimination yield of the CR dye increases progressively with the increase in the dose of AS. However, yields have reduced beyond an optimal dose. These results indicate (Fig. 2) that, the optimal dose of the coagulant is 40 mg/L with a rate of elimination of CR in the order of 97.24%.

It is important to note there is a remarkable discoloration of the CR accompanied by good yields at very low doses of coagulant. This could be due to the existence of a proportion between the negative charges of CR and the positive charges of the coagulant, this strong attraction of charges, which improves the appearance of flocs. The flocs formation were observed from the first doses.

Likewise, the decrease in yields has been explained by the destabilization of colloidal particles, caused by the overdose of the coagulant. The latter, being in excess, plays an opposite role, neutralizing all the particles, which are found to be positively charged and repulsive forces are again exerted between them. We will therefore have very charged water by coagulant with poor coagulation or clarification. The phenomena is schematized in the picture of Fig. 3.

All our results can be confirmed by similar works [22,23] whose tested water treatment residuals (WTR) as a coagulant on an acid dye. They have shown that the elimination of color increases with an increasing dose of coagulant up to a yield of 94.2%. Further, previous studies on disperse dyes employing aluminum as coagulant reported color removal in the range of 80%–90% at doses of 40–200 mg/L [36,37], whereas with ferric salt as coagulant gave 75% color removal at a dose of 60 mg/L [19,26]. Puchana-Rosero et al. [27] showed thatthe elimination of the dye from tannery wastewater increases rapidly and reaches 100% at 71 mg/L of coagulant. Hoong and Ismail [38], also raised the same observations during their study on removal of dye in wastewater by adsorption–coagulation combined system with hibiscus sabdariffa as the coagulant.

However, it should be noted that the final pH decreases with the variation in the concentration of the doses of the

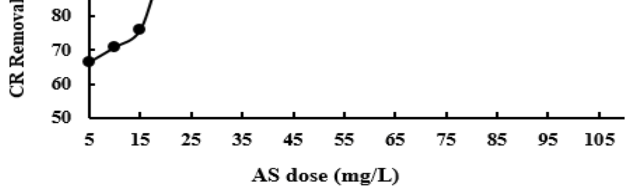


Fig. 2. Effect of the AS coagulant doses.

coagulant injected (Fig. 4). Then a slight increase appeared. The results reveal that the optimal conditions for removing the dye were at pH 3.39 (Fig. 4). This drop-in pH is explained by the fact that, in aqueous solution, $Al^{3+}$ is a strongly hydrated ion and is found surrounded by six coordinated water molecules in an octahedral configuration. The highly positive charge on the central metal ion causes some polarization of the O–H bonds and there is a tendency for protons to dissociate giving one or more hydroxylated species [39]. The chemical reaction represents this process by the following equation (Eq. (2)), where the complexes of $Al^{3+}$ ions in water act as weak acids.

$$Al(H_2O)_6^{3+} \leftrightarrow Al(H_2O)_5(OH)^{2+} + H^+ \quad (2)$$

### 3.2. Effect of initial dye content on CR removal

During this step, several experiments with different initial concentrations of CR dye were carried out. Keeping the other parameters constant, such as the pH of the medium (4.50), rapid stirring conditions (stirring speed (150 rpm for 3 min), and slow stirring (stirring speed 60 rpm for 17 min). The coagulation of these solutions has been carried out with a dose of aluminum sulfate of 40 mg/L (optimal dose). The variation in the initial content of the dye ranges between 1 and 50 mg/L. The results in Fig. 5. show that as the dye concentration increases, the dye removal efficiency increases to an optimum then a slight decrease appeared. In our case, the optimum corresponds to the content of 40 mg/L of CR with 98.26% removal efficiency.

In fact, the elimination efficiency, which is more important for CR on the one hand, can be attributed to its higher molecular weight (696.663 g/mol, Table 1), and on the other hand to its chemical structure aromatic, which contains amino and phenyl functions. These functional categories, in general, can affect the efficiency of the coagulation process by facilitating their removal by: (1) charge neutralization [40] and (2) floc formation, extension, and sedimentation [41,42].

In similar tendencies, some authors have shown that there is an optimum in the elimination of dyes and then there is a relaxation in the yields. Among these works, we can cite the coagulation–flocculation process for dye removal using WTR: modeling through artificial neural networks. Where the authors have suggested that, the maximum color removal has obtained at a lower pH of 3.0 and at this pH reduced color removal has obtained at higher initial dye concentrations, the increase in dye concentration [26].

In the literature, these authors have recorded that results of reduced color removal with the increase in initial dye concentration at optimum pH have been reported for acid dyes with polyaluminum chloride (PACl), and also with WTR as coagulants [22,23,26]. This phenomenon

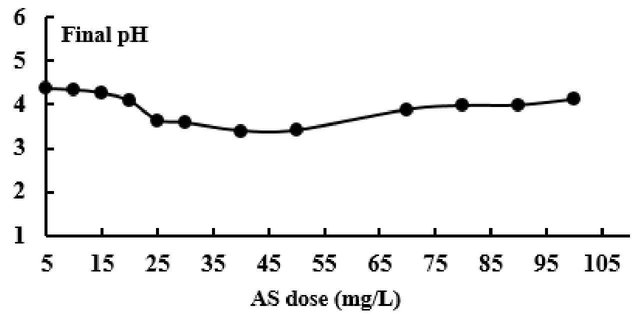


Fig. 4. Effect of final pH after variation of AS doses.

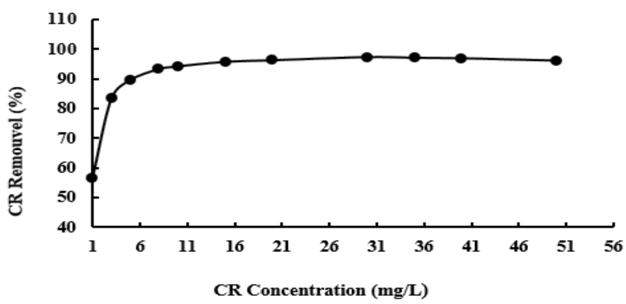


Fig. 5. Effect of initial CR dye concentration (AS = 40 mg/L, pH = 4.50).

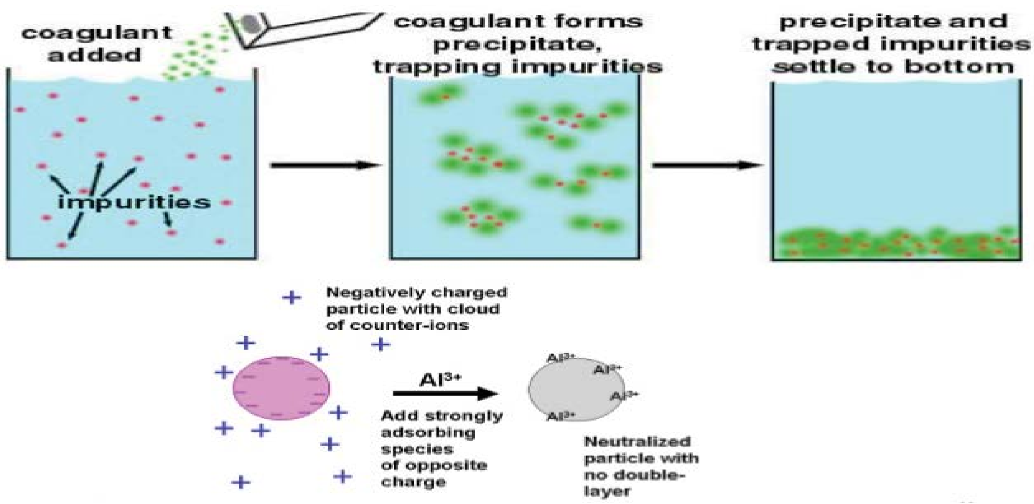


Fig. 3. Picture explains the mechanisms of coagulation–floculation process.

could be due to increased dye aggregation and/or depletion of hydrolysis products of the coagulant [22]. At higher pH contrary effects of dye concentration may be due to the bridging of dye particles on to WTR in the absence of ionization of dye or destabilized WTR. Identical results were also obtained with Ghernaout et al. [42], in the aim to remove two dyes BF cibacete blue (CB) and red solophenyl 3BL (RS) by using aluminum sulfate and ferric chloride.

Other works [10]on the application of bipolar electrocoagulation (EC) with iron electrode indicates the effect of the initial dye concentration (IDC) on color removal and shows by varying the IDC from 30 to 140 mg/L, the removal efficiency significantly drops. When IDCs were less than 50 mg/L, the efficiencies were above 40%, but with its increase to 140 mg/L, the efficiencies fell down immediately around 24%. After this point, the IDC 50 mg/L corresponding to 41.9% dye removal has been chosen for further studies.

In return, contradictory results were also obtained during decolorization of reactive dyeing wastewater such as the dyes, Blue 19, Black 5, and Red 195 by poly aluminum chloride, the authors have recorded there is an immediate decrease with the increase in the initial content of the three dyes tested [30].

### *3.3. Effect of pH on color removal CR*

It has been established that pH plays an important role in the performance of the dyes coagulation process Because, it could affect the specialization of coagulant and the hydrolysis behavior of dyes [43,44]. A series of experiments have been carried out by varying the pH between 1 and 10. We adjusted the pH of the solutions using HCl and/or NaOH with 0.1 N for both middle. In this essay, we considered 30 mg/L the concentration of CR and 40 mg/L of AS. The results obtained are presented in the histograms of the Fig. 6.

The histograms of Fig. 6 indicated that pH is strongly influenced by the removal of reactive dyes such as CR. The coagulation effect has effectively depended on the pH of the dye solution. Our results are schowing.

At pH in the range between 4 and 6, the best performance in terms of color removal was achieved. This range of pH corresponds to the region where positively charged $Al(OH)_2^+$ and $Al2(OH)^{2+}$, and insoluble $Al(OH)_3$ species prevailed (the hydrolysis diagram of aluminum as a function of pH has also been proposed in Fig. 7). In this condition, the coagulation middle is acidic, it further enhances the positively-charged environment by increasing the density of positive charges ($H^+$) around the coagulants hydrolysates. Charge neutralization and complex reaction between the coagulant AS and dissociated organic compounds of CR dominate the coagulation mechanism [21,27] This mechanism confirms the precipitation of the insoluble Al-CR complexes will be produced.

At pH = 6 is the optimum pH at which 98.20% dye removal has observed.

At pH > 6, the removal efficiency of dye has decreased, which could be due to the formation of soluble compounds of aluminum ions. At this stage, the alkalization of the medium is obviously unfavorable in the case of aluminum treatment. This is probably due to the presence of negatively charged species, such as Al $(OH)_4^-$ (Fig. 7), which reduces their attraction to anionic organic compounds.

All our results can be confirmed by the works of certain researchers:

Mahesh et al. [26] mentioned, the maximum color removals of 68%, 67%, and 64% were obtained at pH 3. Similar results have been reported in the literature, the optimum pH, at which the maximum removal of Acid Red 398 occurred, was about 4 and 5 for polyaluminum chloride (PACl) and aluminum, respectively [21]. Moghaddam et al. [22,23] and Mahesh et al. [26] have used PACl- and ferric chloride-based WTR as coagulants, the maximum removals of dyes were 94.2%–96.5% were obtained at pH values 3.42–3.50, respectively, using PACl [23,26] and ferric chloride [22]-based WTR.

Mukhlish et al. [24] during the removal of reactive dye from aqueous solution using coagulation–flocculation coupled with adsorption on papaya leaf. They noticed that the best performance in terms of color removal was achieved in the range of pH 4–8. The pH 5.5 is taken to be the optimum, pH at which 83.64% dye removal was observed. They were indicated that the performance of dye removal efficiency at very low and high pH was very poor, only 3.6% and 29.5% at pH 2 and 10, respectively.

In return, other works have obtained contradictory results Puchana-Rosero et al. [27], during the coagulation–flocculation process combined with adsorption using activated carbon obtained from sludge for dye removal from tannery wastewater. They were shown in the acid pH range between 1 and 3, only 15.31% dye removal was achieved.

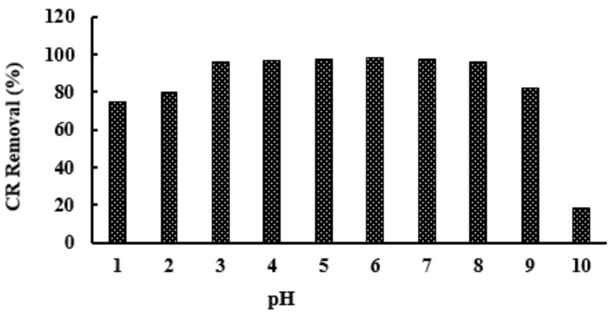


Fig. 6. Effect of pH on the removal of CR dye by coagulation with AS.

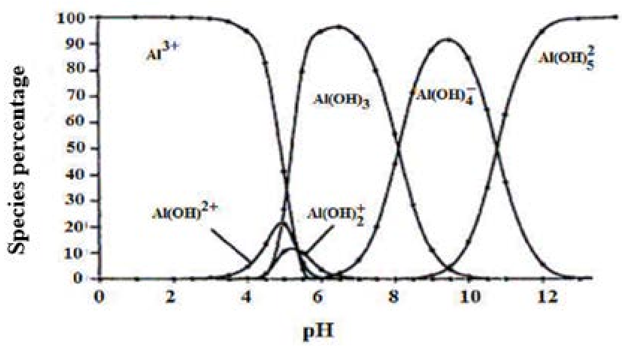


Fig. 7. Different aluminum species percentage as a function of pH of the medium.

But, at pH 4 the dye removal percentage increases quickly reaching 62.80% and was most efficient at basic pH with 93.2% removal at pH 10. According to Papic et al. [16], these could be explained by the fact that, the auxochrome group of the dye molecule has a high degree of dissociation and thus the solubility of alum is high. In the same context, a work carried out by Yuan-Shing and Ha-Manh [30], on the decolorization of reactive Red 195 solution by electrocoagulation process. They indicated that the lowest color removal was recorded between pH 3 and 6, with removal efficiencies below 18%. Nevertheless, when the pH of the dye solutions increased from 6 to 11 the decolorization raised considerably and reached a peak at pH 11, then significantly dropped in the strong basic zone (pH 12).

However, it is important to note that other researchers Hoong and Ismail [38] and Ghernaout et al. [42], whose show in the acidic condition the coagulation is favorable and possesses many advantages.

### *3.4. Effect of coagulants aid on color removal CR*

During our study, we tested three types of coagulants aid depending on the role each of them can play. These are PAC, an agriculture food polymer used at drinking water treatment plants, and the lime ($Ca(OH)_2$). To achieve our objective, our tests were carried out according to the jar-test protocol at optimal dose of coagulant (AS 40 mg/L and 30 mg/L CR). The variable doses of coagulants aid range from 0.0 to 0.5 g/L for PAC and 0.0–2 mg/L for lime ($Ca(OH)_2$) but, for the polymer the concentrations are between 0.0 and 0.20 g/L. The results that we obtained have presented on the curves of Figs. 8–10.

From the results obtained (Figs. 8–10), we note that, the elimination yields increase with increasing in the concentration of PAC and polymers until an optimum, then a remarkable decrease has been recorded. In the range when there is the removal of the CR, the flocs formed are large and settle quickly. In the case of lime, the phenomenon appeared contradictory; there is a decrease in the elimination yields of the CR, then an improvement in the yields is exhibited.

For PAC, 0.05 g/L improves the elimination of CR at 97.68%, the PAC plays the role of an adsorbent. While the 0.02 g/L of polymers record an almost total elimination, it is from 99.06%.

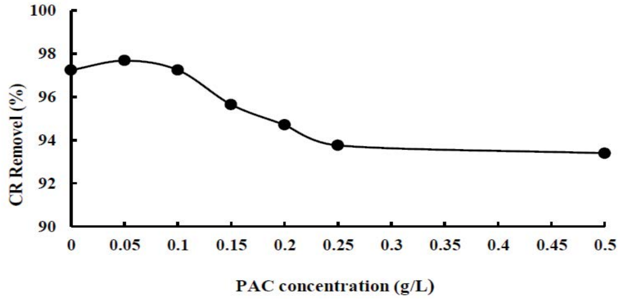


Fig. 8. Effect of the variation of PAC on CR removal (CR = 30 mg/L, AS = 40 mg/L).

For lime, ($Ca(OH)_2$), the improvement in the elimination yields of CR appeared gradually from $Ca(OH)_2$ concentration of 0.4 g/L, the maximum yield is 94.63%. In this test, there is a precipitation phenomenon, which has appeared accompanied by an increase in pH values (Table 2). Indeed, the higher pH, the higher negative charges had by the solution and the aluminum coagulants have needed to neutralize and precipitate the charged species.

We compare the three coagulants aids each with other, we find that the use of the polymer improves the elimination of CR or the decoloration is very well, as well as PAC. According to the coagulation theory [15,21,24,45], the good elimination could be explained by the fact, the PAC and the polymer neutralize the charge of the dye molecule and it binds to the PAC and the polymer surfaces by weak forces like van der Waals forces. These flocs are combines with other flocs to form a high-density flocs, which are easily removed. In contrast, the dye removal percentage

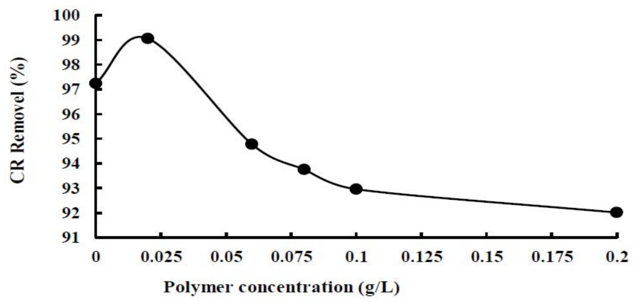


Fig. 9. Effect of the variation of agriculture food polymer on CR removal (CR = 30 mg/L, AS = 40 mg/L).

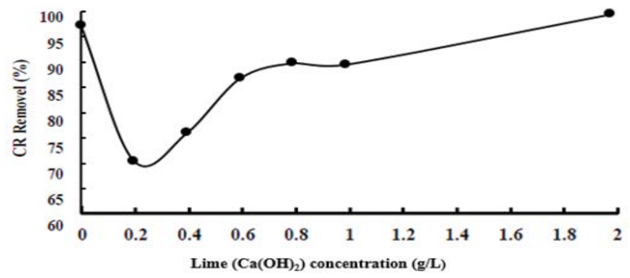


Fig. 10. Effect of the variation of lime ($Ca(OH)_2$) on CR removal (CR = 30 mg/L, AS = 40 mg/L).

Table 2
Final pH avec combinaison coagulant and lime ($Ca(OH)_2$

| $Ca(OH)_2$ (g/L) | pH final |
|---|---|
| 0 | 4.25 |
| 0.2 | 6.75 |
| 0.4 | 9.85 |
| 0.6 | 10.59 |
| 0.8 | 11.8 |
| 1 | 11.91 |
| 2 | 12.19 |

undergoes a remarkable decrease with further increase in the amount of the coagulant aids is the reason can be best explained by the producing and the restabilized of the colloids on excessive PAC and polymer dosages.

However, all our results have confirmed by the works of Mahesh et al. [26], Puchana-Rosero et al. [27], Bisht and Lal [15], and Riera-Torres et al. [46] who have proven the excellent discoloration of dyes by the presence of coagulants aids.

## 4. Conclusions

Among the different technologies available to treat dyes such as physical, chemical, biological, and advanced methods, chemical coagulation–flocculation, which has been carried out by adding chemical reagents, called coagulants (sulfate alumina, ferric sulfate, etc., is still a suitable technology because of its cost-effectiveness and simplicity. In this context, our objective is articulated.

In this study, AS is used as a widely used coagulant to treat textile wastewater containing an anionic dye (CR). AS recorded 97.24% removal for dye concentration 30 mg/L under an optimum condition.

The CR dye removal was greatly affected by the pH of the system, with a range of pH 4–6 showing higher removal. This corresponds to the region where positively charged $Al(OH)^{2+}$ and $Al(OH)_2^+$, and insoluble $Al(OH)_3$ species prevailed. With increasing initial dye concentration (in the range of 1–50 mg/L), the results have shown, an increase in the elimination yields of CR with the increase in the initial content of the dye to an optimum then a slight decrease appeared. The combination process of coagulation–flocculation with coagulants aid such as PAC, agrifood polymer, and the lime showed effective results in the removal of CR dye. But, the agrifood polymer appeared the most effective adjuvant.

Overall, the results of this research indicated that the coagulation–flocculation treatment method, AS combined with PAC or polymer as the operating agent, is an effective and rapid method for removing direct dye from wastewater colored, the aim of which is to protect the environment and living beings.

On the whole, the findings of this work can be applied on the one hand, to treating industrial wastewater although further research should increase our understanding of various aspects of the problem. On the other hand, we will improve our work by using other coagulants (ferric chloride, chitosan, etc.), a technical-economic study is very necessary between the different coagulants. Likewise, in our perspectives, we have come to study the effect of coagulation–flocculation process in the binary elimination of dyes.

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