



REMOVAL OF ACID RED 398 DYE FROM AQUEOUS SOLUTIONS BY COAGULATION/FLOCCULATION PROCESS

Maryam Hasani Zonoozi¹, Mohammad Reza Alavi Moghaddam^{1*}, Mokhtar Arami²

¹ Civil and Environmental Engineering Department, Amirkabir University of Technology (AUT), Hafez st., Tehran, Iran;

² Textile Engineering Department, Amirkabir University of Technology (AUT), Hafez Ave., Tehran, Iran

Abstract

The removal of Acid Red 398 (AR398) dye from dye-containing solution using coagulation/flocculation process with polyaluminum chloride (PAC) and Alum was investigated. The effect of different parameters involving pH, dosage of coagulant, initial dye concentration, and bentonite as a natural coagulant aid was examined. According to the obtained results, the optimum pH, at which the maximum removal occurred, was about 4 and 5 for PAC and Alum, respectively. However, PAC performed efficient in a broader pH range. In the case of PAC, the best removal efficiency was about 80% for the dosage of 100-120 mg/l, while, it was about 60% for 140-160 mg/l of Alum. With the increase of initial dye concentration in the range of 25-250 mg/l, the removal efficiency for both coagulants increased at first (from 25-100 mg/L) and then declined. Bentonite, as a coagulant aid, slightly enhanced the removal efficiency. By adding 20 mg/l of bentonite, the efficiency increased by 15% and 9% for Alum and PAC, respectively.

Keywords: Acid Red 398, Coagulation/Flocculation, Dye Removal

1. Introduction

Dyes are widely used in many industries such as textile, rubber, paper, plastic, cosmetic etc. Among them, textile ranks first in usage of dyes (Saiful Azhar et al., 2005). Presently, more than 10,000 of different commercial dyes and pigments are available (Eren and Acar, 2006; Ozer et al., 2006), and more than 7×10^5 tons per year are produced world wide (Crini, 2006; Saiful Azhar et al., 2005). Two percent of dyes that are produced are discharged directly in aqueous effluents (Crini, 2006). These colored compounds are not only aesthetically displeasing, but they also impede light penetration, retard photosynthetic activity and inhibit the growth of biota. Some dyes are also toxic and carcinogenic (Eren and Acar, 2006). Presence of very small amounts of dyes in water (less than 1 ppm for some dyes) is highly visible and undesirable (Crini, 2006). Therefore it is necessary to eliminate dyes from wastewater before it is discharged.

Since dye compounds are specifically designed to be recalcitrant with poor biodegradability, they are very stable and difficult to degrade by conventional aerobic biological treatments, such as activated sludge process. Thus, they are usually treated by physico-chemical processes (Ozer et al., 2006; Shi et al., 2007).

Coagulation/flocculation is one of the most popular unit operations in water and wastewater treatment trains. Also it is one of the most effective chemical treatment methods for dye removal from industrial wastewaters (Gao et al., 2007; Golob et al., 2005).

In this work, the removal efficiency of Acid Red 398 dye was investigated using PAC and Alum as coagulants.

The study focused on the effect of different parameters such as pH, coagulant dosage, initial dye concentration and bentonite (as a coagulant aid) on the dye removal efficiency.

* Author to whom all correspondence should be addressed: phone: 0098-912-2334600, fax: 0098-21-66414213, e-mail: alavim@yahoo.com, alavi@aut.ac.ir

2. Materials and methods

In this study, commercial grades of PAC (30% w/w Al₂O₃) and Alum (17% w/w Al₂O₃) were used. All experiments were performed at laboratory scale. Synthetic wastewater was prepared by dissolving Acid red 398 (AR398, trade name: Erionyl Red BL), which was provided by Ciba Company. This dye is widely used in textile industries in Iran. Stock dye solutions of 1000 mg/l were prepared and then diluted using deionized water to obtain desired concentrations. pH of the synthetic wastewater was adjusted with H₂SO₄ and NaOH solutions. Bentonite was sieved by 75 µm sieve. The particles under 75 µm were used in further experiments.

The effects of various parameters were determined by jar test procedure. A period of 2 min was allowed for the rapid mixing of the dye containing solutions at 200 rpm followed by a 10 min period of slow mixing at 30 rpm. Then, the solutions were allowed to settle for 45 min. After settling, samples for measurement of dye concentration were withdrawn using a pipette from a height of 2-3 cm below the surface in each jar. The maximum absorbance (λ_{max}) of the dye with the background of deionized water was 504 nm, which was determined according to scanning pattern performed on HACH spectrophotometer DR/4000. Percentage of dye removal was calculated by Eq. (1):

$$\text{dye removal (\%)} = \frac{C_r - C_t}{C_r} \times 100 \quad (1)$$

where, C_r and C_t are the dye concentration in raw and treated solutions, respectively.

3. Results and discussion

3.1. Effect of pH on the removal of AR398 dye

pH plays an important role in coagulation/flocculation process using inorganic coagulants. Charge on hydrolysis products and precipitation of metal hydroxides are both controlled by pH variations. Thus, pH must be controlled to establish optimum conditions for coagulation (Li and Gregory, 1991). To study the effect of pH on AR398 dye removal efficiency, dosages of PAC and Alum were kept constant at 100 mg/L, while varying pH of the samples using H₂SO₄ and NaOH. Dye concentration was 100 mg/l for all solutions during the experiment.

As shown in Fig. 1, removal of AR398 dye was absolutely dependent on the pH variations. The optimum pH, at which the maximum removal occurred, was about 4 and 5 for PAC and Alum, respectively. However, PAC was effective in a broader pH range relative to Alum. In the other words, the performance of Alum was more sensitive to the pH variations. This is in consistent with the findings reported by some other researchers. (Jiang, 2001; Ye et al., 2007).

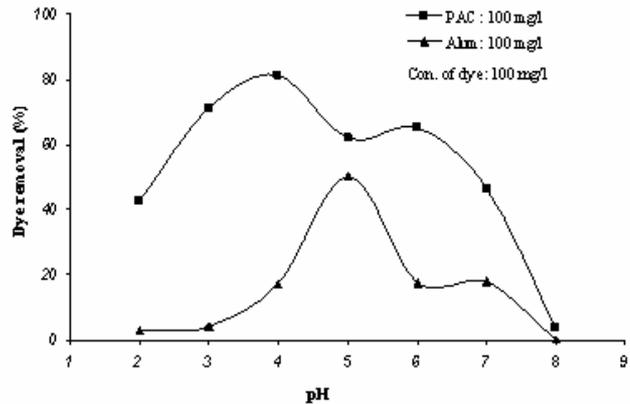


Fig. 1. Effect of pH on the removal of AR398 dye with PAC and Alum

3.2. Effect of the coagulants concentration

In this step, to study the effect of coagulant dosage on dye removal efficiency, different amounts of PAC and Alum were dosed into the dye-containing solutions. Dye concentration was kept constant at 100 mg/l and pH was adjusted to 4 and 5 (optimum pH) for PAC and Alum, respectively. The variations of the dye removal with coagulants dosage are shown in Figure 2.

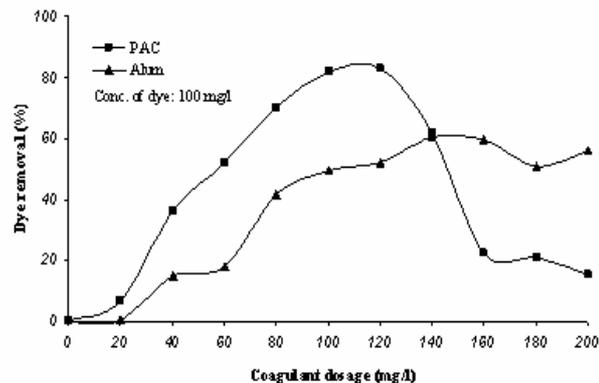


Fig. 2. Effect of coagulant dosage on the removal efficiency of AR398 dye, pH: 4 for PAC; 5 for Alum

According to the results, with the increase of the coagulants dosage, the removal efficiencies increased at first (from 0 to 120 mg/L) for both of the coagulants. However, the efficiencies were much higher for PAC. For the dosages more than 120 mg/l, the curve relatively approached plateau for Alum, while, for PAC, the efficiency decreased rapidly which was probably due to re-stabilization phenomenon.

3.3. Effect of initial dye concentration

This step was performed to determine the influence of initial dye concentration on dye removal efficiency, using a constant coagulant dosage (100

mg/L for PAC, and 140 mg/L for Alum, the dosages which led to the maximum dye removal).

In addition, the variations of the amount of the removed dye per unit mass of coagulant (Q), versus the initial dye concentration are presented. The results are illustrated in Fig. 3.

For PAC, with the increase of initial dye concentration from 25 mg/L to 100 mg/L, the removal efficiency increased dramatically from 14% to about 81%. While, for dye concentrations more than 100 mg/L, the efficiency decreased and reached to about 37% for the dye concentration of 250 mg/L. The highest value of Q was 1.3 mg dye/ mg PAC for the dye concentration of 200 mg/L. (Fig. 3(a)).

In the case of Alum, the curves relatively showed similar changing trends. However the values of the removal efficiency and the Q were superior for PAC. The largest value of the Q (0.7 mg dye/ mg Alum) was observed for initial dye concentration of 200 mg/l, while the highest removal efficiency was 60% for dye concentration of 100 mg/L (Fig. 3b).

Little data was found in literature to compare the influence of dye concentration on coagulation efficiency. According to the data of Klimiuk group, at the optimum coagulant (polyaluminum chloride)

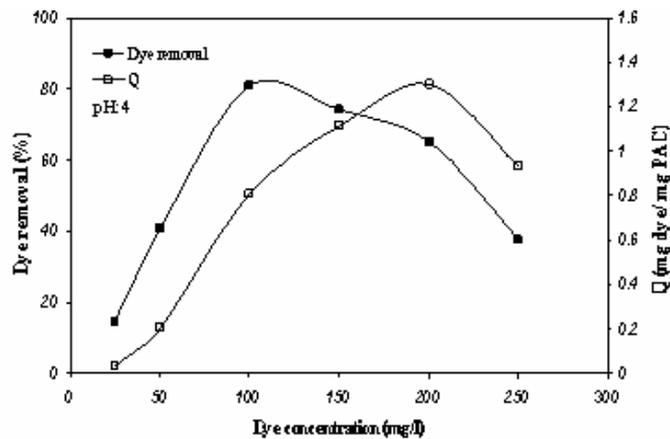
dosage, the removal degree (the amount of the removed dye per unit mass of Al) was associated with the initial concentration of the selected dyes, and the smallest removal degree was obtained for smallest concentrations of the dyes (Klimiuk et al., 1999).

This result is in consistent with the results obtained for PAC and Alum in this study, which showed the smallest values of Q for dye concentrations of 25 and 50 mg/L.

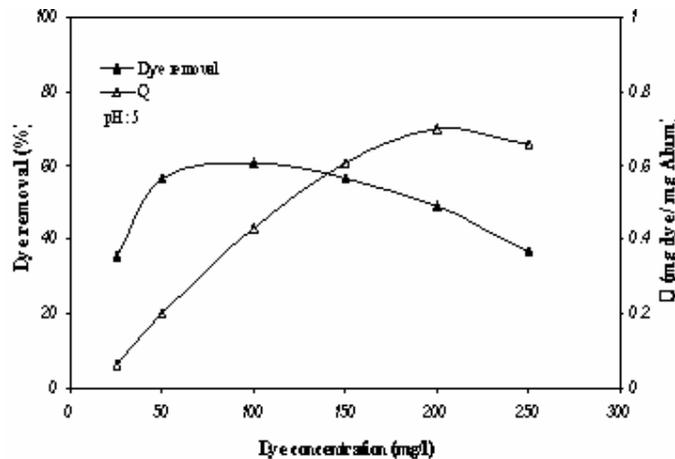
3.4. Effect of addition of bentonite as a coagulant aid

Coagulant aids such as activated silica, clay and polyelectrolytes are used in coagulation/flocculation process, usually to obtain higher efficiency, to reduce the amount of required coagulant, and to form stronger and more settleable flocs (AWWA, 2003).

In this step, the effect of bentonite as a natural coagulant aid on AR398 dye removal efficiency was investigated. The experiments were conducted under two selected dosage levels for both coagulants (60 mg/L and 80 mg/L), with different concentrations of the coagulant aid. The results are shown in Fig. 4.



a)



b)

Fig. 3. Effect of initial dye concentration on dye removal efficiency and Q for: (a) PAC, (b) Alum

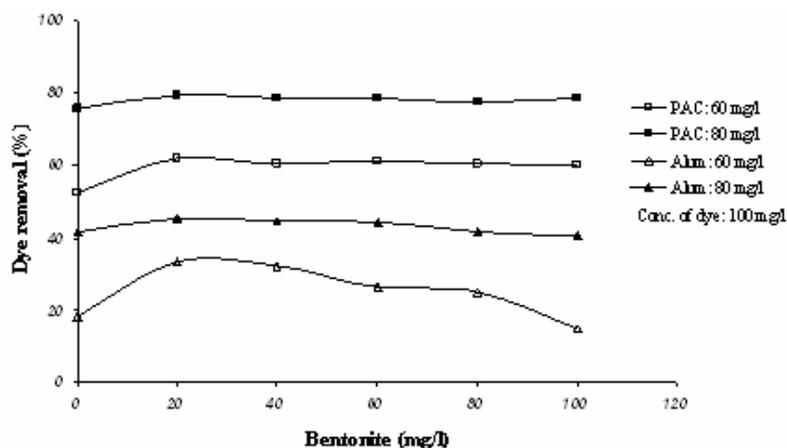


Fig. 4. Effect of bentonite on AR398 dye removal efficiency, pH: 4 for PAC; 5 for Alum

By adding bentonite to the coagulation/flocculation process, the dye removal efficiencies increased slightly. When the coagulants concentration was 60 mg/L, with the aid of 20 mg/l of bentonite, the removal efficiency increased from 18% to about 33% for Alum, and from 52% to 61% for PAC. No significant change was observed when the coagulants concentration was 80 mg/L.

According to the reports of other researchers, in some cases coagulant aids positively affected on dye removal and in some others, they acted reversely and reduced the removal efficiency. For example, Nabi Bidhendi group reported that Polyelectrolyte did not have any positive effect on dye removal with Alum and $MgCl_2$ (Nabi Bidhendi et al., 2007). Also, Joo group found that with Alum alone, dye removal efficiency was 20% or less, while it increased to 99% with the aim of 150 mg/l of a synthetic polymer (Joo et al, 2007).

4. Conclusions

In this study, the following conclusions were drawn:

- Removal of AR398 dye was absolutely pH dependent. The maximum removal efficiency occurred when the pH was about 4 and 5, for PAC and Alum, respectively.
- PAC performed more efficient than Alum and removed the dye with higher efficiencies. However, for the dosages more than 120 mg/l, a removal reduction or re-stabilization phenomenon was happened for PAC.
- With the increase of initial dye concentration (in the range of 25 to 250 mg/L), both coagulants showed similar changing trends, however, the values of the removal efficiency and the Q were higher for PAC.
- By adding bentonite as a coagulant aid, the removal efficiency of AR398 dye increased slightly. For the coagulants concentration of 60 mg/l, by adding 20 mg/l bentonite, the removal efficiency

increased by 15% and 9% for Alum and PAC, respectively.

Acknowledgements

The authors are grateful to the Research and Technology Affairs of Amirkabir University of Technology (AUT) for their financial support of this study. In addition, the authors wish to express their thanks to Ms. Maryam Akbari and Ms. Elham Paseh for their assistance during the analyses of the samples.

References

- AWWA, (2003), *Principles and Practices of Water Supply Operations: Water Treatment*, 3rd Edition, American Water Works Association.
- Crini G., (2006), Non-conventional low-cost adsorbents for dye removal: A review, *Bioresource Technology*, **97**, 1061–1085.
- Eren Z., Acar F. N., (2006). Adsorption of Reactive Black 5 from an aqueous solution: equilibrium and kinetic studies. *Desalination*, **194**, 1–10.
- Gao B.Y., Yue Q.Y., Wang Y., Zhou W.Z., (2007), Color removal from dye-containing wastewater by magnesium chloride. *Journal of Environmental Management*, **82**, 167–172.
- Golob V., Vinder A., Simonic M., (2005), Efficiency of the coagulation/flocculation method for the treatment of dye bath effluents, *Dyes and Pigments*, **67**, 93-97.
- Jiang J.Q., (2001), Development of coagulation theory and pre-polymerized coagulants for water treatment, *Separation and Purification Methods*, **30**, 127-141.
- Joo D.J., Shin W.S., Choi J.H., Choi S.J., Kim M.C., Han M.H., Ha T.W., Kim Y.H., (2007), Decolorization of reactive dyes using inorganic coagulants and synthetic polymer, *Dyes and Pigments*, **73**, 59-64.
- Klimiuk E., Filipkowska U., Korzeniowska A., (1999), Effects of pH and coagulant dosage on effectiveness of coagulation of reactive dyes from model wastewater by polyaluminium chloride, *Polish Journal of Environmental Studies*, **8**, 73-79.
- Li G., Gregory J., (1991), Flocculation and sedimentation of high turbidity waters. *Water Research*, **25**, 1137–1143.

- Nabi Bidhendi G.R., Torabian A., Ehsani H., Razmkhah N., (2007), Evaluation of Industrial Dyeing Wastewater Treatment with Coagulation and Polyelectrolyte as a Coagulant aid. *Iranian Journal of Environmental Health Science and Engineering*, **4**, 29-36.
- Ozer A., Akkaya G., Turabik M., (2006), The removal of Acid Red 274 from wastewater: Combined biosorption and biocoagulation with *Spirogyra rhizopus*, *Dyes and Pigments*, **71**, 83-89.
- Saiful Azhar S., Ghaniey Liew A., Suhardy D., Farizul Hafiz K., Irfan Hatim M.D., (2005), Dye Removal from Aqueous Solution by using Adsorption on Treated Sugarcane Bagasse, *American Journal of Applied Sciences*, **2**, 1499-1503.
- Shi B., Li G., Wang D., Feng C., Tang H., (2007), Removal of direct dyes by coagulation: The performance of preformed polymeric aluminum species. *Journal of Hazardous Materials*, **143**, 567-574.
- Ye C., Wang D., Shi B., Yu J., Qu J., Edwards M., Tang, H., (2007), Alkalinity effect of coagulation with polyaluminum chlorides: Role of electrostatic patch. *Colloids and Surfaces A: Physicochem. and Engin. Aspects*, **294**, 163-173.