DECOLORIZATION OF AN ACIDIC DYE FROM SYNTHETIC WASTEWATER BY SLUDGE OF WATER TREATMENT PLANT

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ABSTRACT
In this research, sludge of Jalaliyeh water treatment plant in Tehran was used as an alternative coagulant for the removal of a specific type of acid dye (acid red 119 (AR119)). For this purpose, the effect of initial pH, coagulant dose and initial dye concentration on dye removal efficiency were investigated. Results showed that the dye removal rates were largely dependent on pH. When the solution pH was increased from 3 to 8, the dye removal rates decreased from 96.3% to 2.3%. The removal efficiencies of the dye using 130-350 mg dried sludge/L were more than 90% at initial pH =3. With the increase of initial dye concentration in the range of 10-200 mg/L, the removal efficiency increased at first (from 10-40 mg/L) and then declined. So, sludge of Jalaliyeh water treatment plant may be considered as an appropriate coagulant for the removal of AR119 dye.

Key words: Acid dye; Coagulation; Removal efficiency; Water treatment plant; Sludge

INTRODUCTION
Effluent from industries, such as dyeing and textile industries, contains many coloring substances, which are toxic (Benefield et al., 1982; Chu, 2001). Even the presence of very low concentrations of dyes (less than 1 mg/L) in the effluent is considered undesirable and needs to be removed before the wastewater can be discharged into the environment (Vandevivere et al., 1998).

Various treatment technologies for dye removal has been investigated extensively such as the photochemical oxidation (Rezaee et al., 2008), membrane (Yu et al., 2010), chemical coagulation (Nabi Bidhendi et al., 2007; Szyguła et al., 2009), adsorption (Sreedhar Reddy and Kothaiah, 2006; Qiu et al., 2009; Nagda and Ghole, 2009) and biological processes (Vaigan et al., 2009; Mehrali et al., 2010, Naimabadi et al., 2009).

The coagulation/flocculation process is one of the most effective physicochemical treatment methods that is widely employed for treating dye bearing effluents of industries (Gao et al., 2007; Tan et al., 2000; Jiang et al., 1996). Coagulation effectiveness and cost depend on coagulant type and concentration, solution pH, ionic strength, as well as both concentration and nature of the organic residues in effluent (Robert et al., 1996; Duan et al., 2003). Typical chemicals (coagulation reagents) used for water and wastewater treatment are predominantly inorganic salts of Al or Fe (Rizzo et al., 2005). The use of such conventional coagulants for dye removal is applicable, but it may not be so desirable because of their considerable costs. Therefore, the idea of using cheaper substitutes may be so favorable.

On the other hand, sludge is an unavoidable by-product of the processing of drinking water in water treatment works. In recent years, the management of sludge derived from water treatment plant has become the most critical environmental issues due to the very fast increase in sludge production and the associated disposal costs and limitations (Yang et al., 2006). Under these circumstances,
the reuse of sludge as beneficial material has been concerned to mitigate these effects. Such beneficial reuses include the use of waterworks sludge as a coagulant or adsorbents in wastewater treatment that was studied by several research groups (Sujana et al., 1998; Guan et al., 2005; Yang et al., 2006).

The main objectives of this research are to investigate the acid red 119 dye removal from aqueous solution by sludge of a water treatment plant as an alternative coagulant and study the effect of different operating variables such as initial pH, coagulant dose and initial dye concentration on the performance of coagulation/flocculation process.

MATERIALS AND METHODS

The sludge sample used in this study was collected from Jalaliyeh water treatment plant in Tehran, where ferric chloride is being used as coagulating agent in coagulation/flocculation process. The collected sample was stored and used at room temperature in the form of suspension.

Synthetic wastewater was prepared by dissolving Polar red brown V (CI: Acid red 119 (AR119)) which was provided by Ciba Company. This dye is a commercial dye and is widely used widely in textile industry in Iran. At first, a stock dye solution of 1000 mg/L was prepared by adding 1 gram of AR119 dye to 1 L of deionized water. Then, it was diluted according to the working concentrations. The required pH was adjusted by H\(_2\)SO\(_4\) or NaOH. pH measurement was carried out using a 340i/SET pH meter (WTW-Germany).

A six beaker jar-test apparatus from Zag-Chemi Co. (Iran) was used to simulate the coagulation/flocculation process. Each beaker contained 250 mL of the dye solution. A period of 2 min was allowed for the rapid mixing at 100 rpm followed by a 30 min period of slow mixing at 40 rpm. Then the solutions were allowed to settle for 30 min. After settling, the additional centrifuging (5000 rpm for 5 min) was performed for all samples before analysis.

The remaining color in the supernatant layer of treated solutions was measured using HACH spectrophotometer (DR/4000) at a wavelength corresponding to the maximum absorbance for AR119 dye. The maximum absorbance (\(\lambda_{\text{max}}\)) of the dye with the background of deionized water was 526 nm, which was used for all the absorbance readings. Percentage of dye removal was calculated by the following equation:

\[\eta(\%) = \frac{(C_r - C_t)}{C_r} \times 100\]  

Where: \(\eta\) is dye removal efficiency; \(C_r\) and \(C_t\) are dye concentrations in raw and treated solutions, respectively.

The variables of this study were as follows:

- \(\text{pH}=3-8\)
- Coagulant dosage: 25-350 mg dried sludge/L
- Initial dye concentration: 10-200 mg/L

RESULTS

Effect of initial pH

The coagulation/flocculation performance was investigated under various initial pH ranging between 3 to 8, while the dosages of coagulant and dye concentration were kept constant at 150 mg dried sludge/L and 80 mg/L, respectively.

The results are presented in Fig. 1. As it is shown, the dye removal yield by sludge increased with decreasing initial pH in the solution. It is seen that the maximum efficiency is observed at pH=3 with 96.3% dye removal efficiency. Results indicate that the effectiveness of coagulant in removing dye is highly dependent on initial pH. The removal efficiency decreased sharply when the pH value exceeded 3 and the dye removal reduced to only about 2.3% at pH=8.

![Fig. 1: Effect of initial pH on the removal of AR119 dye with sludge; Coagulant dosage: 150 mg dried sludge/L; Initial dye concentration: 80 mg/L](image)
**Effect of coagulant dosage**
The effect of coagulant dosage on dye removal efficiency was investigated. For this purpose, different amounts of sludge were dosed into the dye-containing solutions at constant dye concentration (80 mg/L) and pH = 3. Analytical data for the coagulated samples are presented in Fig. 2.

The results illustrated in Fig. 2 indicate that with the increase of coagulant dosage, the removal efficiency increased. The highest decrease in dye occurred in experiments carried out with 150 mg dried sludge/L. Beyond this value, regarded as being optimal, this increase was dramatically attenuated and finally the curve approached plateau. With the increase of dosage, no re-stabilization phenomenon or removal reduction was observed. From these results, it became clear that coagulant dosage of 150 mg dried sludge/L presented the best performance. Thus, this concentration of sludge was applied for the further step.

**Effect of initial dye concentration**
Effect of initial dye concentration on process performance was studied at optimum conditions (pH=3 and 150 mg dried sludge/L). The results are shown in Fig. 3. With the increase of initial dye concentration (10 mg/L to 200 mg/L), the rate of the efficiency reduction was not significantly changed. The removal efficiency increased slightly with the increase of initial dye concentration from 10 mg/L to 40 mg/L and then declined. In addition, the variations of the amount of the removed dye per unit mass of coagulant (Q), versus the initial dye concentration are presented in Fig. 3. It is shown that Q increased steadily with the increase of dye concentration and reached to 0.96 mg removed dye/mg dried sludge.

![Fig. 2: Effect of coagulant dosage on the removal efficiency of AR119; Initial pH=3; Initial dye concentration: 80 mg/L.](image)

![Fig. 3: Effect of initial dye concentration on dye removal efficiency and the amount of the removed dye per unit mass of coagulant (Q); Initial pH=3; Coagulant dosage = 150 mg dried sludge/L.](image)
DISCUSSION

The results of the study on coagulation/flocculation of solutions containing an acidic dye (AR119 dye) proved that reusing Jalaliyeh water treatment plant sludge as a coagulating agent in acid red 119 dye removal may be considered so efficient. However, the conducted tests showed that the efficiency of the process varied considerably and was associated with the initial pH, coagulant dosage and initial concentration of dye solutions.

The decrease of initial pH was beneficial for enhancing dye removal and by adding coagulant to the coagulation/flocculation process, the efficiencies tended to increase. As the functional groups of acid dyes are anionic, they release negative charges when dissolved in water. It seems that the positively charged species of coagulant can cause destabilization of negatively charged molecules by adsorption onto the surface of colloids and Charge neutralization (Benefield et al., 1982).

Furthermore, sludge apparently served as condensation nuclei, and the dye particles were enmeshed as the precipitate was settled. Hence, it can be found that charge neutralization was not the only mechanism by which removal of the dye particles occurred. Similar result was reported by Chu (2001) for the removal of hydrophobic dye with recycled alum sludge, who has mentioned that with the overdose of recycled alum sludge, no restabilization phenomenon or reduction removal was observed and therefore, the concentration of recycled alum sludge used in the process needs no precise control (Chu, 2001).

In our study, as shown in Fig. 3, the amount of the dye removed by the used coagulant increased steadily with an increase in the initial concentration of dye solution and no reduction of Q occurred. This trend is also similarly reported by Zonoozi et al. (2009), concerning the application of polyaluminum chloride in Acid Blue 292 dye removal. They found that with the increase of Acid Blue 292 dye concentration, Q increased at first and reached to its highest value (2.1 mg dye/mg coagulant), when the dye concentration was 150 mg/L, and then it decreased rapidly to 0.9 mg dye/mg coagulant.

Several researchers have been used conventional coagulants such as ferric chloride for dye removal from aqueous solutions. Kim research group reported that ferric chloride shows satisfactory properties in coagulation/flocculation processes for decolorization of disperse dye solutions (Kim et al., 2004). They found that the maximum removal efficiencies of 97.7% and 99.6% for the disperse blue 106 and disperse yellow 54 dyes were obtained at the optimum coagulant dosage of 0.93 mM and 0.74 mM ferric chloride, respectively (Kim et al., 2004). In addition, Karadag research group determined the optimum ferric chloride dosage as a coagulant for the removal of Reactive Blue 4 (RB4, anthraquinone type of reactive dye) and Reactive Yellow (RY, an azo type of reactive dye) from aqueous solution (Karadag et al., 2006). They reported that optimum coagulant (FeCl₃) dosages were 0.4 g/L for RY and 0.3 g/L for RB4, while the maximum removal efficiencies of 99.67% and 99.74% were obtained (Karadag et al., 2006).

As it can be seen, optimal ferric chloride (fresh coagulant) dosage for removal of reactive dyes was higher than the sludge dosage needed for AR119 dye removal whereas optimum ferric chloride dosage for disperse dyes removal was little lower. These differences in removal efficiency might result from the different chemical structures and solubility behaviors of the dyes.

According to our results, the main results of this study can be summarized as follows. Initial pH was found to be the most important parameter in coagulation/flocculation process for acid red 119 dye removal using Jalaliyeh water treatment plant sludge. The maximum removal efficiency (more than 96.3%) occurred when the initial pH was about 3. The highest dye removal (96.3%) occurred in experiment carried out with 150 mg dried sludge/L at constant pH=3. No restabilization of dye materials or removal reduction was observed in the process at different sludge concentrations. Furthermore, it was found that initial dye concentration (in the range of 10-200 mg/L) had not significantly effect on dye removal efficiency. On the other hand, Q increased steadily with the increase of dye concentration and reached to 0.96 mg removed dye/mg dried sludge.
It can be concluded that the reuse of such kind of sludge as a low-cost material instead of conventional costly coagulants can offer great advantages such as high efficiency for AR119 dye removal from dye-containing solutions, fresh coagulant savings in wastewater treatment plants and significant reductions in present and future sludge disposal costs at wastewater treatment plant.

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