

EFFECT OF PRAESTOL AS A COAGULANT-AID TO IMPROVE COAGULATION-FLOCCULATION IN DYE CONTAINING WASTEWATERS

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ABSTRACT

This study was conducted to investigate the effect of praestol, as a coagulant-aid, to improve coagulation-flocculation process in the removal of disperse red 60 from aqueous solutions. The effect of various parameters including coagulants dose (10-1000 mg l⁻¹), praestol dose (0-1000 mg l⁻¹), solution pH (3-11), initial dye concentration (100-500 mg l⁻¹), flocculation speed (30-60 rpm), flocculation time (15-30 min), settling time (5-60 min) and ionic strength (0-6 mg l⁻¹) was evaluated on the dye removal. The dye removal efficiency was substantially increased by using praestol in the concentration of 80 mg l⁻¹ and 400 mg l⁻¹ for coagulation with alum and polyaluminum chloride (PACl), respectively. The maximum dye removal by alum coupled with praestol (AI-P) and PACl coupled with praestol (PA-P) was found to be 97.8% and 98.7%, respectively that were occurred at pH 7. The results showed that the application of PA-P or AI-P can be effectively used to remove disperse red 60 (DR 60) in aqueous solutions.

Keywords: Wastewater, Dye, Coagulation, Praestol, Alum, PACl

1. Introduction

In recent years, aquatic ecosystems pollution due to the irregular disposal of dye containing wastewaters has been substantially increased, especially in developing countries (Noorimotlagh *et al.*, 2014). It has been approximated that more than 700,000 tons of various dye materials are annually produced in the world (Riera-Torres *et al.*, 2010). Many industries including textile, dyeing, leather, rubber, plastic, carpet, food processing, pulp and paper are considered as the major sources of dye-containing wastewaters production (Soltani *et al.*, 2014; Nourmoradi *et al.*, 2015a). Discharge of the color wastewaters into the water bodies can reduce light penetration and then can disrupt the photosynthesis process in the aqueous environments (Anouzla *et al.*, 2009). Moreover, most of dyes materials can cause adverse effects including allergy, dermatitis, skin irritation and cancer on human health (Anouzla *et al.*, 2009). The presence of very low concentration of dyes (less than 1 mg l⁻¹) in an effluent, because of the mentioned effects, is objectionable and it should be removed prior to discharging the wastewater into the environment (Sadri Moghaddam *et al.*, 2010). Different treatment methods like adsorption, coagulation-flocculation, chemical oxidation, photo-degradation and biological conversion are applied to remove dyes from wastewaters. However, the dyes are difficult to remove by conventional treatment processes (Merzouk

et al., 2011). Among the above procedures, coagulation-flocculation process is extensively used in removing dyes from industrial wastewaters (Huang *et al.*, 2014b). Dyes can be removed through coagulation-flocculation without decomposition, thus no potentially detrimental and poisonous intermediate substances are created. Furthermore, the operation and maintenance (O&M) of this process is relatively uncomplicated and cost effectiveness (Shi *et al.*, 2007; Huang *et al.*, 2014a). Many coagulants such as aluminum sulphate, ferric chloride and polyaluminum chloride (PACl) are extensively used in the conventional wastewater treatment process (Anouzla *et al.*, 2009).

Table 1. Dye removal from textile wastewaters by coagulation-flocculation process.

Type of Coagulant	Type of Dye	Color Removal (%)	Reference
Ferric based	Real textile wastewater	50	(Zahrim <i>et al.</i> , 2011)
Ferric chloride	Reactive dye	12	(Zahrim <i>et al.</i> , 2011)
Ferrous sulphate	Real textile wastewater	50	(Zahrim <i>et al.</i> , 2011)
Aluminium sulphate	Cotton-textile wastewater	60	(Selcuk, 2005)
Ferric sulphate	Real textile wastewater	58	(Verma <i>et al.</i> , 2012)
Aluminium sulphate	Direct dye	50	(Zahrim <i>et al.</i> , 2011)
Ferrous sulphate	Real textile wastewater	50	(Baraoidan <i>et al.</i> , 2007)

Application of these inorganic coagulants without coagulant-aid in some dye containing wastewaters has been resulted in low dye removal, Table 1. But, using coagulant-aid, especially polymers, in the coagulation process enhances the dye removal from these wastewaters (Joo *et al.*, 2007; Zahrim *et al.*, 2011). Polymers have been used in water and wastewater treatment for several decades. Polymers including polyacrylamides, chitosan, polysaccharides, polyvinyl are usually employed as coagulant-aid to improve the larger flocs formation and to improve the rate of sedimentation. Polyacrylamide is a crystalline poly-functional molecule that contains a vinylic carbon-carbon double bond and an amide group. Using polyacrylamide, as a relatively low cost coagulant-aid, along with common coagulants leads to a significant increase in the solids settling velocity (Ahmad *et al.*, 2008). Praestol is a high molecular weight and white color grainy polymer that is obtained through organic synthesis on the basis of polyacrylamids (Wira and Wira, 2006). Although PACl and alum have extensively been applied in dye removal, but, to the best of our knowledge, there was no study by common coagulants and praestol, as a coagulant aid, for the removal of dye in wastewaters. The aim of this work was to investigate the influence of praestol, as a coagulant-aid, to improve coagulation-flocculation process in the removal of a dye, disperse red 60 (DR 60), from aqueous solutions.

2. Materials and methods

2.1. Materials

Disperse red 60 (DR 60) was purchased from Alvan Sabet Co (Iran). The chemical structure and general characteristics of the dye are shown in Fig 1 and Table 2, respectively.

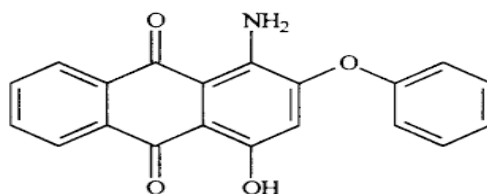


Figure 1. The chemical structure of disperse red 60

Commercial grade of polyaluminum chloride (PACl) and aluminum sulfate (alum), coagulant agent, and praestol as a coagulant-aid were provided from Chemkimia Co (Malaysia). Other analytical grade chemicals, NaOH (98%), H₂SO₄ (95-97%), and NaCl were supplied by Merck Co (Germany).

Table 2. The characteristics of disperse red 60.

Characteristic	DR 60
Generic name	Disperse Red 60
Chemical formula	C ₂₀ H ₁₃ NO ₄
Molecular weight (g mol ⁻¹)	331.326
λ _{max} (nm)	586
Appearance	Red powder

2.2. Preparation of synthetic wastewater

A stock dye solution of 1000 mg l⁻¹ was prepared in distilled water and then was diluted according to the working concentrations. The solution pH was adjusted by (0.1 N and 1 N) H₂SO₄ and (0.1 N and 1 N) NaOH. The solution pH was quantified using a pH meter (Metrohm Herisau Digital E 532).

2.3. Coagulation-flocculation experiments

Coagulation-flocculation experiments were performed in batch mode by a six beaker jar-test apparatus (Zag-Chemi, Iran). The coagulation-flocculation procedure involved rapid mixing at 200 rpm for 2 min followed by slow mixing and then settling (Ahmad *et al.*, 2008; Wu *et al.*, 2012). One liter of the dye solution was transferred into each Plexiglas beaker. The effects of various coagulants dose (10-1000 mg l⁻¹), praestol dose (10-1000 mg l⁻¹), solution pH (3-11), initial dye concentration (100-500 mg l⁻¹), flocculation stirring speed (30-60 rpm), flocculation mixing time (15-30 min), settling time (5-60 min) and ionic strength (0-6 g l⁻¹) were tested in the removal of DR 60 at room temperature (24±1 °C). After conducting the treatment process, the supernatant was taken, at 5 cm below the effluent surface, to determine the dye removal efficiency. All the experiments were carried out in triplicates and the average values were applied to analyze the treatment method. The removal efficiency was calculated by Eq. (1).

$$R (\%) = \frac{(C_0 - C_e)}{C_0} \times 100 \quad (1)$$

Where; R (%) is the removal efficiency of DR 60. C₀ (mg l⁻¹) and C_e (mg l⁻¹) are the initial and equilibrium dye concentrations, respectively (Faghihian *et al.*, 2014; Nourmoradi *et al.*, 2015b). An UV-Vis spectrophotometer (HACH, DR5000, USA) was used to determine DR 60 concentration at the maximum absorbance wavelength (586 nm).

3. Results and discussion

3.1. Effect of praestol dose

PACl and alum were used as coagulants coupled with praestol, as coagulant-aid (flocculant). The effect of the flocculant dosages (0-1000 mg l⁻¹) were separately measured on the dye removal with a fixed dosage of alum (100 mg l⁻¹) and PACl (100 mg l⁻¹) and initial dye concentration of 100 mg l⁻¹ at initial solution pH of 6. Also, during this step, the coagulation mixing speed was 200 rpm for 2 min followed by slow mixing (40 rpm for 15 min) and then settling (30 min). The results of dye removal by alum coupled with praestol (Al-P) and PACl coupled with praestol (PA-P) are shown in Fig 2. As seen, the maximum dye removal by alum and PACl (without praestol) was 65.8% and 0.7%, respectively. The removal efficiency of dye was substantially increased by adding praestol as flocculant in the coagulation process. The dye removal was increased from 65.8% to over 98% by increasing praestol dose from 0 to 80 mg l⁻¹ in coagulation process by alum (100 mg l⁻¹). The removal efficiency was nearly constant by increasing flocculant dose more than 80 mg l⁻¹ in alum coagulation. The dye removal was also improved from 0.7% to 99% with enhancing praestol from 0 to 400 mg l⁻¹ in PACl (100 mg l⁻¹) coagulation process. The removal efficiency was not changed with increasing flocculant dose (more than 400 mg l⁻¹) in PACl coagulation. The previous studies also showed that better dye removal could be achieved when polymer was applied in combination with

common coagulants (Wu *et al.*, 2012; Oladoja and Aliu, 2009). In this study, praestol doses of 80 mg l⁻¹ and 400 mg l⁻¹ were selected for Al-P and PA-P treatments, respectively for the rest of experiments.

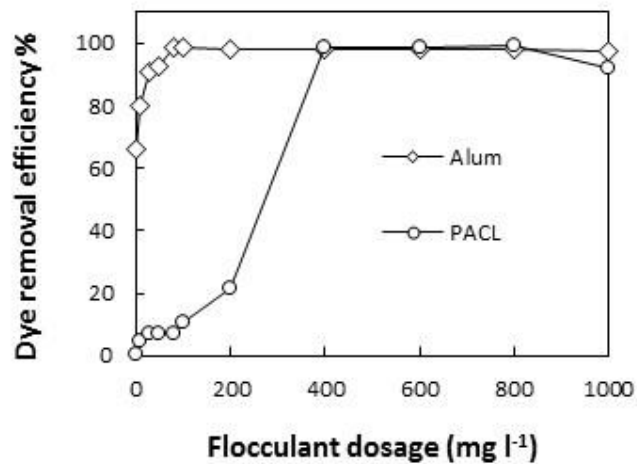


Figure 2. The effect of praestol dose on DR 60 removal

3.2. Effect of coagulant dose

The effect of coagulant dosage is one of the most important parameters that should be considered to determine the optimum condition in coagulation-flocculation process. Insufficient dosage or overdosing of coagulant can be resulted in the poor removal performance in the process. Therefore, it is significant to determine the most favorable dosage in order to minimize the chemical cost, sludge formation and also to acquire the optimum removal efficiency in the treatment process (Patel and Vashi, 2012). The coagulation experiments were carried out with various doses of coagulants from 10 to 1000 mg l⁻¹, fixed amount of praestol dosage of 80 mg l⁻¹ (for coagulation with alum) and 400 mg l⁻¹ (for coagulation with PACl), initial dye concentration of 100 mg l⁻¹ and flocculation and settling time of 15 and 30 min, respectively. The dye removal as a function of coagulant concentration is shown in Fig 3.

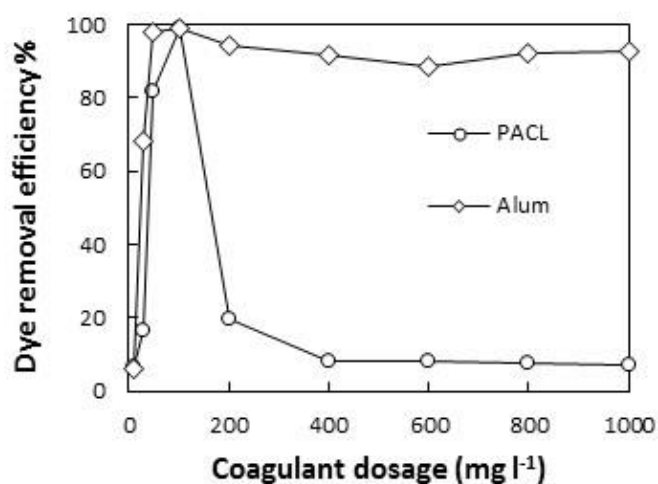


Figure 3. The effect of coagulant dose on DR 60 removal

The removal efficiency of DR 60 was increased with increasing the coagulant dose up to 50 mg l⁻¹ and 100 mg l⁻¹ for alum and PACl, respectively. The dye removal efficiency was decreased with increasing the

coagulants doses greater than 50 mg l^{-1} and 100 mg l^{-1} for alum and PACl, respectively. This decline in the dye removal efficiency may be associated to charge turnaround and destabilization of colloidal dye flocs because of overdosing of coagulant (Saritha *et al.*, 2015).

3.3. Effect of solution pH

The solution pH not only influences on the surface charge of coagulants, but also alters the stabilization of the dye suspension (Saritha *et al.*, 2015). Fig. 4 depicts the effect of the solution pH (3 to 11) on the dye removal by Al-P and PA-P. The dosage of polymer as a coagulant-aid for coagulation of DR 60 by PACl and alum was 400 mg l^{-1} and 80 mg l^{-1} , respectively. The coagulants doses were also 50 mg l^{-1} and 100 mg l^{-1} for alum and PACl, respectively. As can be seen, the dye removal efficiency was increased by Al-P from 6.8% to 97.8% as the pH increased from 3 to 7, and the removal efficiency was dramatically dropped to 5.8% at pH 11.

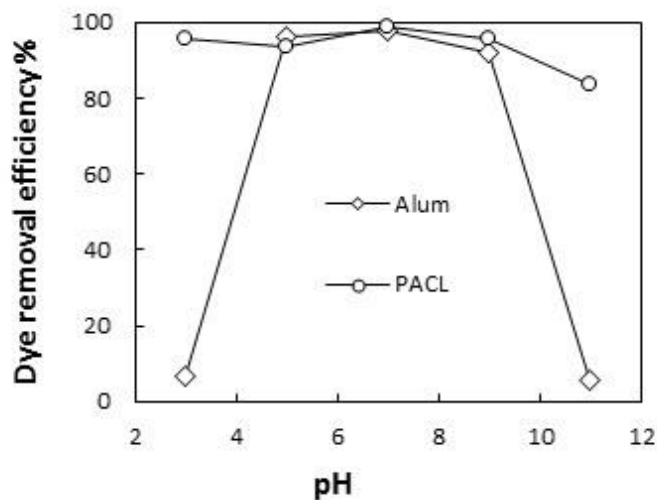


Figure 4. The effect of pH on DR 60 removal

Similar trend was observed for PA-P in the solution pH of 3 to 7, so, the removal efficiency was slowly increased from 95.5% to 98.7% as the pH increased from 3 to 7 and it was decreased to 83.7% at pH 11. As depicted in Fig 4, the small change in removal efficiency of DR 60 dye was observed for PA-P in comparison with Al-P in all pH ranges. It can be concluded that PA-P compared to Al-P could operate well in wide range of pH for the removal of DR 60. Several researchers reported that polyacrylamide-based polymers such as PACl in comparison with other common coagulants gave better dye removal efficiency in broader range of pH (Yeap *et al.*, 2014; Sanghi *et al.*, 2006; Baraoidan *et al.*, 2007). The maximum dye removal by coagulation with Al-P (97.8%) and PA-P (98.7%) was obtained at pH 7. The most predominated species of Aluminum-based coagulants at this pH value (pH=7) are positively charged $\text{Al}(\text{OH})_2^+$, $\text{Al}_2(\text{OH})_2^+$ and also insoluble $\text{Al}(\text{OH})_3$ that are very effective to remove dye via sweep precipitation (Merzouk *et al.*, 2011). In higher and lower sides of the optimum pH, the removal efficiency of DR 60 was reduced maybe due to the formation of more soluble forms of Aluminum (Merzouk *et al.*, 2011). Therefore, the solution pH of 7 was used as the optimum pH to remove DR 60 for the rest of the experiments.

3.4. Effect of flocculation speed and time

Fig. 5 illustrates the effect of flocculation mixing speed and time (20–60 rpm for 15–30 min) on the dye removal efficiency. The coagulation test was conducted in alum and PACl dosages of 50 mg l^{-1} and 100 mg l^{-1} , respectively, praestol dosage of 80 mg l^{-1} (for coagulation with alum) and 400 mg l^{-1} (for coagulation with PACl) and initial dye concentration of 100 mg l^{-1} at pH 7. As seen in Fig. 5, by increasing the flocculation speed from 20 rpm to 40 rpm at flocculation time of 15 min, the dye removal efficiency was decreased. But, the dye removal at mixing time of 15 min was increased as the flocculation speed

was increased from 40 rpm to 60 rpm for both the coagulants. Otherwise, the dye removal by increasing the flocculation speed from 20 rpm to 60 rpm was firstly increased (up to 40 rpm) and it was then decreased (from 40 rpm up to 60 rpm) for both the coagulants at mixing time of 30 min. The maximum dye removal of 98.1% and 99.4% was achieved for Al-P and PA-P at the flocculation speed and time of 20 rpm and 15 min, respectively.

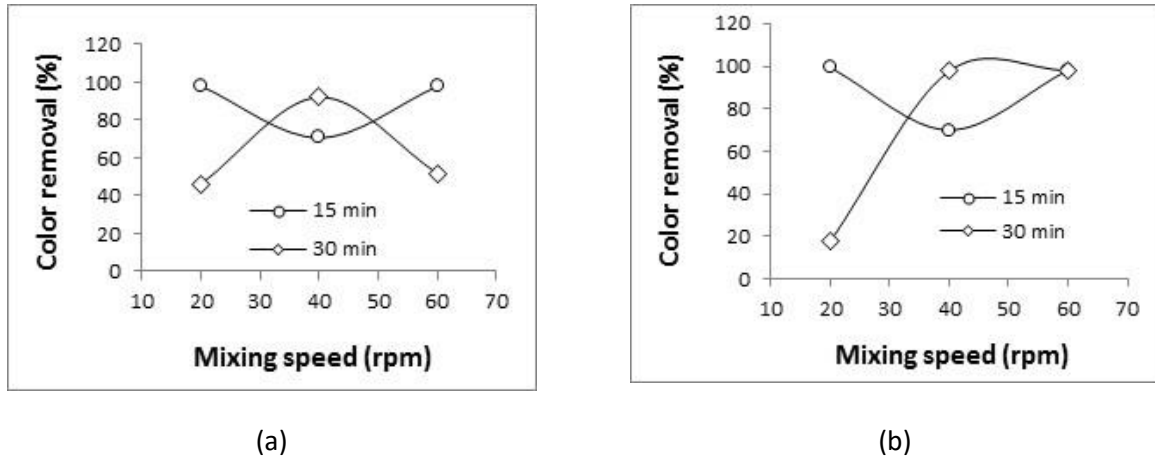


Figure 5. The effect of flocculation speed and time for DR 60 removal by a) Al-P and b) PA-P

3.5. Effects of settling time

In the coagulation–flocculation process, the settling time of the flocs is an important parameter because of the influence on the overall cost as well as efficiency of the process. The results of the effect of settling time on the dye removal efficiency are presented in Fig. 6.

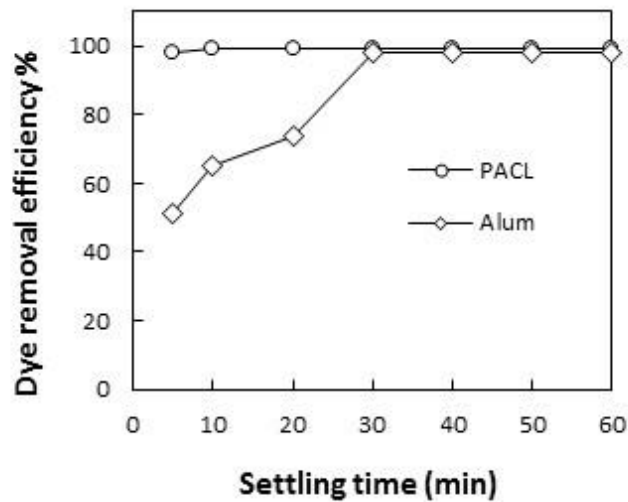


Figure 6. The optimum settling time for DR 60 removal

The addition of flocculant (praestol) had a significant effect on the settling time. This may be due to the bridging mechanism of the high molecular weight of the flocculant, praestol, which can produce more compact, high strength and larger flocs (Glover *et al.*, 2000; Negro *et al.*, 2006). The increase of flocs size improves the flocs settling speed and therefore reduces the settling time of the flocs. The flocs generated by PA-P settled faster than the flocs generated by Al-P. The increment in settling time more than 5 min had no effect on the dye removal efficiency by PA-P. But, in the coagulation process with Al-P, the dye removal efficiency was increased by increasing the settling time up to 30 min and after that it was nearly

constant. The maximum dye removal of 98.1% over 30 min settling time and 98.2% over 5 min settling time was obtained by alum+ praestol and PACI+ praestol, respectively.

3.6. Effects of solution ion strength

Various types of impurities including dissolved compounds can be found in the wastewaters from textile and dyeing industries. Ions like calcium, sodium, potassium, chloride, sulphate, carbonate, nitrate, etc are the major components of textile and dyeing wastewaters (Szyguła *et al.*, 2008). The ionic strength can diminish the zeta potential of the solution because of its affect on the electrical charges, which causes coagulation easier. Therefore, sodium chloride may be poured in wastewater prior to coagulation in order to enhance the ionic strength of the suspension to be treated, particularly when the wastewater is polluted by non-ionic pollutants (Merzouk *et al.*, 2011). Raising wastewater ionic strength by sodium chloride also has other advantages such as chloride anions can substantially decline the unfavorable influences of other anions on the coagulation, such as SO_4^{2-} and HCO_3^- (Merzouk *et al.*, 2011). In this study, the effect of solution ionic strength by NaCl (0 to 6 g l^{-1}) was investigated on the coagulation process under the optimal conditions (dye concentration of 100 mg l^{-1} , optimum content of praestol, alum and PACI (see Sec 3.1 to 3.3) at pH 7). The results are shown in Fig 7.

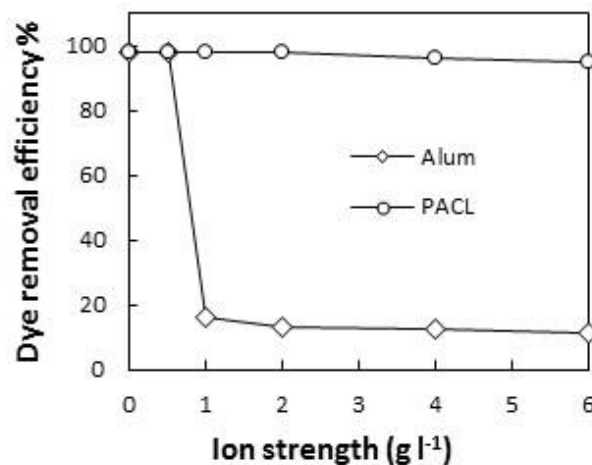


Figure 7. The optimum ion strength for DR 60 removal

As seen, increasing the solution ion strength from 0 to 6 g l^{-1} NaCl had slightly negative effect on the performance of dye removal by PA-P. The performance of dye removal by Al-P was nearly constant with increasing the solution ion strength up to 1 g l^{-1} NaCl. But, the removal efficiency of Al-P was substantially decreased from 98.05% to 11.11% as the solution ion strength increased from 1 to 6 g l^{-1} , respectively. The decrease in the dye removal efficiency by increasing ion strength may be due to the absence of HCO_3^- in the artificial wastewater. Therefore, it was concluded that NaCl addition into the dye solution resulted in decrease of DR 60 removal efficiency.

4. Conclusions

The effect of praestol, as a coagulant aid, was evaluated on the improvement of coagulation process with alum and PACI for the removal of a common disperse dye, DR 60. When praestol was used in combination with alum and PACI, the color removal was increased at lower doses of the coagulants. The maximum dye removal by alum coupled with praestol (Al-P) and PACI coupled with praestol (PA-P) was found to be 97.8% and 98.7%, respectively that were occurred at pH 7. The flocs generated by PA-P settled faster than the flocs generated by Al-P. Using these coagulants, especially PACI, along with praestol as a coagulant-aid can effectively remove DR 60 from aqueous solutions.

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