



Response surface optimization of acid red 119 dye from simulated wastewater using Al based waterworks sludge and polyaluminium chloride as coagulant

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ABSTRACT

In this research, the performance of Polyaluminium Chloride (PAC) and Polyaluminium Chloride sludge (PACS) as coagulants for acid red 119 (AR119) dye removal from aqueous solutions were compared. The sample of PACS was collected from “Baba Sheikh Ali” water treatment plant (Isfahan, Iran) where PAC is used as a coagulant in the coagulation/flocculation process. A response surface methodology was applied to evaluate the simple and combined effects of the operating variables including initial pH, coagulant dosage and initial dye concentration and to optimize the operating conditions of the treatment process. Results reveal that the optimal conditions for dye removal were initial pH 3.42, coagulant dosage of 4.55 g dried PACS/L and initial dye concentration of 140 mg/L for PACS, while the optimal initial pH, coagulant dosage and initial dye concentration for PAC were 3.8, 57 mg/L and 140 mg/L, respectively. Under these optimal values of process parameters, the dye removal efficiency of 94.1% and 95.25% was observed for PACS and PAC, respectively. Although lower amount of PAC in comparison with PACS was needed for specific dye removal, the reuse of PACS as a low-cost material can offer some advantages such as high efficiency for AR119 dye removal and economic savings on overall water and wastewater treatment plant operation costs.

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1. Introduction

Industries such as textile, leather, paper, plastics, etc., are some of the sources for dye effluents (Weber and Morris, 1964). The discharge of highly colored effluent is currently one of the world's major environmental problems as some of the dyes and their metabolites are either toxic or mutagenic and carcinogenic and pose a potential health hazard to all forms of life (Reife and Freeman, 1994). Various methodologies have been investigated for treating dye bearing effluents that can be classified in four categories: (i) physical (ii) chemical (iii) biological and (iv) acoustical, radiation, and electrical processes (Gupta and Suhas, 2009). Coagulation/flocculation is one of the most effective chemical treatment methods for dye removal from industrial wastewaters (Gao et al., 2007). Typical chemicals (coagulation reagents) used for water and wastewater treatment are predominantly inorganic salts of Al or Fe (Rizzo et al., 2005). Moreover, during the last decades, alternative coagulation reagents have started to be used in treatment facilities such as the

inorganic polymeric coagulants or flocculants, byproduct of industrial process, etc.

Polyaluminium chloride (PAC), an inorganic polymer flocculants (IPFs), has been developed rapidly and become applied widely for treatment of water and wastewater (Wang et al., 2004; Ye et al., 2007). Several research groups reported that PAC shows excellent properties in coagulation/flocculation processes for removal of various dyestuffs (Shi et al., 2007; Zonoozi et al., 2008, 2009). It should be noted that such chemical treatment method (Coagulation/flocculation process) imposes a high operational cost incurring from the use of coagulants and the treatment and disposal of a large amount of chemical sludge (Guan et al., 2005). While current sludge disposal methods may still suffice for the time being, the need for environmental sustainability and fiscal responsibility coupled with population increases will continually provide the drive toward beneficial reuse (Babatunde and Zhao, 2007). Under these circumstances, the idea of using sludge generated from water treatment plants may be favorable (Chu, 2001). More than 11 possible ways for reusing the waterworks sludge were reported by Babatunde and Zhao (2007). Among these possible routes, the reuse of waterworks sludge as a coagulant in wastewater treatment can be considered as an attractive method that was studied by several research groups (Guan et al., 2005; Chu, 2001).

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On the other hand, the appropriate implementation of coagulation/flocculation process is important and depends upon how precisely effective factors are chosen. Therefore, trial and error has been conventionally practiced to optimize the variables (Ghafari et al., 2009). This method is not only time consuming, but also usually incapable of reaching the true optimum due to ignoring the interactions among variables (Wang et al., 2004). Response surface methodology (RSM) is an effective tool for optimizing the process when a combination of several independent variables and their interactions affect desired responses (Ravikumar et al., 2007). With RSM, several variables are tested simultaneously with a minimum number of trials according to special experimental designs based on factorial designs. This methodology has the advantage of being less expensive and time consuming than the classical methods (Silva et al., 2004).

The main objective of this study is to compare the performance of Polyaluminium chloride (PACS) and PAC as coagulating agents to remove an anionic dye (acid red 119) from dye-containing solutions. The appropriate implementation of coagulation/flocculation depends upon how precisely effective factors are chosen. In this study, initial pH, coagulant dosage and initial dye concentration were selected as operating variables that were studied by some other research groups (Sadri Moghaddam et al., 2010; Anouzla et al., 2009; Zonoozi et al., 2009). This study focused on the optimization of operational conditions for both coagulating agents (PACS and PAC) using RSM to develop a mathematical correlation between the initial pH, coagulant dosage and initial dye concentration for acid red 119 dye removal.

2. Materials and methods

2.1. Preparation of synthetic wastewater

Synthetic wastewater was prepared by dissolving acid red 119 (AR119, trade name: Polar red brown V), which is widely used in the textile industries in Iran. A stock dye solution of 1000 mg/L was prepared in distilled water and then was diluted according to the working concentrations. The pH was adjusted by (0.1 N and 1 N) H_2SO_4 or (0.1 N and 1 N) NaOH and measured using a 340i/SET pH meter (WTW-Germany).

2.2. Coagulants

Commercial grade of PAC (purity: 30% w/w Al_2O_3) and a sample of PACS were used as coagulating agents in this study. The PACS was collected from coagulation/flocculation unit of a water treatment plant in Isfahan (Iran), where PAC is used as a coagulant. The collected sample was stored at room temperature ($25 \pm 1^\circ C$) in the form of suspension and taken for use in the experiments. Chemical analyses of the dried PACS were carried out by X-ray fluorescence (XRF, X"Unique II-philips). Detailed chemical compositions of the dried PACS are presented in Table 1. As shown, the amount of aluminum and iron, the effective components on the coagulation/flocculation process, are 19.2% (as Al_2O_3) and 3.02% (as Fe_2O_3), respectively. It should be noted that the trace elements analyzed were not reported due to the very small amount.

A scanning electron microscope (SEM) (Seron technology AIS-2100/korea) was also used to characterize the PACS sample for morphological information. For this purpose, the PACS sample was dried and covered with a thin layer of gold and its surface was observed by SEM, operated at 20 kV accelerating voltage.

2.3. Coagulation/flocculation studies

Experiments were performed at laboratory scale. Jar-test apparatus from Zag-Chemi Co. (Iran) was used for the experiments.

Table 1
The chemical compositions of PACS.

Chemical composition	Amount (Wt %)
Al_2O_3	19.2
Fe_2O_3	3.02
CaO	15.5
MgO	1.64
SiO_2	29.5
SO_3	4.5
Na_2O	0.65
K_2O	0.9
TiO_2	0.41
MnO	0.23
P_2O_5	0.2
L.O.I. ^a	24.14

^a L.O.I = Lost on Ignition.

Different amounts of PAC and PACS were added to 250 mL of AR119 dye wastewater. The coagulation/flocculation procedure involved 2 min of rapid mixing at 100 rpm, followed by slow mixing at 40 rpm for 30 min and 30 min of settling. After settling, the additional centrifuging (5000 rpm for 5 min) was performed to obtain clear liquid. A UV–Vis HACH spectrophotometer DR/4000 (USA) was used to quantify the remaining color in the supernatant layer after coagulation at a wavelength corresponding to the maximum absorbance of 526 nm (λ_{max}) for AR119 dye.

2.4. Preliminary study: determination of the effective ranges for pH and dosage of coagulants

A batch preliminary experiment was conducted to determine a narrower range of initial pH and coagulant dosage for both coagulants prior to designing the experimental runs. Experiments were carried out by varying a single factor while keeping all other factors fixed at a specific set of conditions. For this purpose, the behavior of dye removal after coagulation was investigated under various initial pH ranging between 3 and 12, while the dosages of coagulants (5.5 g/L for PACS and 50 mg/L for PAC) were kept constant. After that, different amounts of each coagulant (ranging between 0.5 and 9 g/L for PACS and 15–120 mg/L for PAC) were dosed into the solutions, to determine the optimum dosage of coagulants for AR119 dye removal at optimum pH. Initial dye concentration was 80 mg/L for all solutions. Results reveal that the both coagulants presented the best performance at lower pH (acidic medium) and by increasing coagulant dosage beyond a specific value (5.4 g dried PACS/L for PACS and 70 mg/L for PAC); the increase in dye removal was dramatically attenuated.

2.5. Experimental design and data analysis

RSM is a collection of statistical and mathematical techniques useful for developing, improving and optimizing process (Myers and Montgomery, 2002; Montgomery, 1996). In recent years, RSM is used in different fields of science that are highlighted by many research groups for its feasibility and efficiency. Sadri Moghaddam et al. (2010), Anouzla et al. (2009) and Ravikumar et al. (2007), have effectively applied RSM to optimize different processes variables for dye removal.

In the present study, a central composite design (CCD) was employed for determining the optimum condition for the dye removal by PACS and PAC. The statistical software "MiniTab", version 15.1.1.0 was used for CCD and the obtained data analyzing. Initial pH, coagulant dosage and initial dye concentration were chosen as three independent variables in the coagulation/flocculation process. As mentioned previously, experiments were initiated as a preliminary

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