



## Coagulation/flocculation process for dye removal using sludge from water treatment plant: Optimization through response surface methodology

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### ABSTRACT

In this study, performance of a waterworks sludge (FCS: ferric chloride sludge) for the removal of acid red 119 (AR119) dye from aqueous solutions were investigated. For this purpose, response surface methodology (RSM) was applied to optimize three operating variables of coagulation/flocculation process including initial pH, coagulant dosage and initial dye concentration. The results showed that the decrease of initial pH was always beneficial for enhancing dye removal and no re-stabilization phenomenon was occurred even at the used maximum FCS dosage. It seems that iron hydroxides of the FCS could neutralize the negative charges on dye molecules or cause to the trapping of the dye ones. Therefore, the sweep flocculation and/or the charge neutralization might play key roles in the enhancement of dye removal. The optimum initial pH, FCS dosage and initial dye concentration were found to be 3.5, 236.68 mg dried FCS/L and 65.91 mg/L, respectively. Dye removal of 96.53% is observed which confirms close to RSM results. Therefore, it can be concluded that reusing the FCS as a low-cost material into the coagulation/flocculation process in wastewater treatment plants can offer some advantages such as high efficiency for AR119 dye removal and economic savings on overall treatment plant operation costs.

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### 1. Introduction

Textile industry is considered as one of the most polluting sectors in terms of effluent composition and volume of discharge. Dyes are gradually emerging as a class of anthropogenic organic substances that pose serious threat to environment [1]. Disposal of this colored water into receiving water can be toxic to aquatic life. In addition, they pose a problem because they may be mutagenic and carcinogenic and can cause severe damage to human beings, such as dysfunction of organs like kidney [2,3]. 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 [4].

Coagulation/flocculation process is one of the most efficient methods that are widely employed for dye removal from industrial wastewater as it is efficient and simple to operate [5–7]. The removal of pollutants by the aid of coagulation is a promising process, but the use of conventional coagulants may not be so admirable because of the chemical costs. Under these circum-

stances, the idea of using sludge generated from water treatment plants (WTPs) may be favorable [8,9].

Waterworks sludge due to its high content of metal hydroxide, largely and easily available nature coupled with the fact that it is free of charge can be considered as a valuable raw material for treatment of various pollutants in wastewater [10,11]. Therefore, with a continual increase in the production of waterworks sludge and in line with the prevailing legislative and economic drives pointing toward waste avoidance and beneficial reuse of waste streams, a number of research efforts have been made particularly in recent years to reuse waterworks sludge in many beneficial ways [11]. Such beneficial reuses include the use of waterworks sludge as a coagulant [9,12,13] or adsorbents [14–20] in wastewater treatment.

In this research, the potential and effectiveness of a waterwork sludge (ferric chloride sludge (FCS)) was studied as an alternative coagulant for the removal of acid red 119 (AR119). For this purpose, the response surface methodology (RSM) is used to develop a mathematical correlation between the initial pH, coagulant dosage and initial dye concentration for the dye removal. The main objective of using RSM is to determine the optimum operational conditions for the system or to determine a domain that satisfies the operating specifications [21]. Several research groups have also applied RSM to optimize coagulation/flocculation process for dye removal [22,23].

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**Table 1**  
Experimental range and levels of the independent test variables.

Variables	Factor	Unit	Range and level				
			$-\alpha$	-1	0	1	$\alpha$
Initial pH	$X_1$	-	2.31	3	4	5	5.68
Coagulant dosage	$X_2$	mg dried FCS/L	35.68	80	145	210	254.3
Initial dye concentration	$X_3$	mg/L	65.91	100	150	200	234.08

## 2. Materials and methods

### 2.1. Chemicals and materials

Synthetic wastewater was prepared by dissolving Acid red 119 dye (AR119, commercial name: Polar red brown V) which was provided by Ciba Company (Iran) in distilled water. This dye is a commercial dye and is used widely in textile industry in Iran. First, a stock dye solution of 1000 mg/L was prepared in deionized water and then was diluted according to the working concentrations. The required pH was adjusted by (0.1N and 1N)  $H_2SO_4$  or (0.1N and 1N) NaOH. pH measurement was carried out using a 340i/SET pH meter (WTW-Germany).

Sample of the FCS was collected from Jalaliyeh WTP in Tehran, Iran where ferric chloride is being used as coagulating agent in coagulation/flocculation process. The collected sample (FCS) was stored and used at room temperature in the form of suspension. A scanning electron microscope (SEM) equipped with energy dispersive X-ray spectroscopy (EDS) (Seron technology AIS-2100/Korea) was used to characterize the FCS sample for its basic constituents and morphological information.

### 2.2. Experimental procedure

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. The coagulation/flocculation procedure involved 2 min of rapid mixing at 100 rpm, followed by 30 min of slow mixing at 40 rpm, and 30 min of settling. The additional centrifuging (5000 rpm for 5 min) was performed to obtain clear liquid for all samples before analysis. Dye concentration was measured using UV-vis HACH spectrophotometer DR/4000 at a wavelength corresponding to the maximum absorbance 526 nm ( $\lambda_{max}$ ) for AR119 dye. Percentage of dye removal was calculated by the following equation:

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

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

### 2.3. Experimental design and data analysis

The most popular class of second-order designs called central composite design (CCD) was used for the RSM in the experimental design. The CCD was first introduced by Box and Wilson in 1951, and is well suited for fitting a quadratic surface, which usually works well for the process optimization [24–26].

In this research, the rotatable experimental plan was implemented as a CCD. The effect of three variables in the coagulation/flocculation process including initial pH of the solution, coagulant (FCS) dosage and initial dye concentration was investigated. A total of 20 experiments according to a  $2^3$  full factorial CCD, consisting of eight factorial points (coded to the usual  $\pm 1$  notation), six axial points ( $\pm\alpha, 0, 0$ ),  $(0, \pm\alpha, 0)$ ,  $(0, 0, \pm\alpha)$ , and six replicates at the center points  $(0, 0, 0)$  were conducted. The value of  $\alpha$  for rotatability depends on the number of points in the factorial portion of

the design, which is given in Eq. (2):

$$\alpha = (N_F)^{1/4} \quad (2)$$

where  $N_F$  is the number of points in the cube portion of the design ( $N_F = 2^k$ ,  $k$  is the number of factors). Therefore,  $\alpha$  is equal to  $(2^3)^{1/4} = 1.682$  according to Eq. (2).

At first, preliminary experiments were conducted to determine a narrower range of pH and coagulant dosage prior to designing the experimental runs. For this purpose, experiments were carried out by varying a single factor while keeping all other factors fixed at a specific set of conditions. The experimental results showed that FCS presented high performance at lower initial pH values. In addition, by increasing coagulant dosage beyond a specific value, the increase in dye removal was dramatically attenuated and finally the curve approached plateau. According to the obtained experimental data, levels of three main parameters investigated in this study are presented in Table 1. For statistical calculations, the variables  $X_i$  (the real value of an independent variable) were coded as  $x_i$  (dimensionless value of an independent variable) according to the following equation:

$$x_i = \frac{(X_i - X_0)}{\delta X} \quad (3)$$

where  $X_0$  is the value of  $X_i$  at the center point and  $\delta X$  represents the step change.

Experimental data was analyzed using MiniTab v15.1.1.0 and fitted to a second-order polynomial model to optimize the variables in the coagulation/flocculation process. The quadratic equation model for predicting the optimal conditions can be expressed as Eq. (4):

$$Y = b_0 + \sum_{i=1}^n b_i x_i + \sum_{i=1}^n b_{ii} x_i^2 + \sum_{i=1}^{n-1} \sum_{j=i+1}^n b_{ij} x_i x_j \quad (4)$$

where  $Y$  is the predicted response (dye removal efficiency),  $b_0$  the constant coefficient,  $b_i$  the linear coefficients,  $b_{ii}$  the quadratic coefficients,  $b_{ij}$  the interaction coefficients and  $x_i, x_j$  are the coded values of the variables. Dedicated RSM program and “Matlab” (version 5) software were used for response surface and counter plotting, respectively. Adequacy of the proposed model is then revealed using the diagnostic checking tests provided by analysis of variance (ANOVA). The quality of the fit polynomial model was expressed by the coefficient of determination  $R^2$ . The  $R^2$  values provide a measure of how much variability in the observed response values can be explained by the experimental factors and their interactions. These analyses are done by means of Fisher’s ‘F’ test and  $P$ -value (probability). Model terms were evaluated by the  $P$ -value with 95% confidence level. Finally, the optimal values of the critical parameters were obtained by analyzing the surface and counter plots and also by solving the regression equation using LINGO 9.0 software.

## 3. Results and discussions

### 3.1. Characterization of the FCS

As determined by EDS, detailed chemical compositions of the dried FCS (by weight) are presented in Table 2. The results of EDS analysis from an average of scanned points showed that the major

**Table 2**  
Chemical composition of FCS.

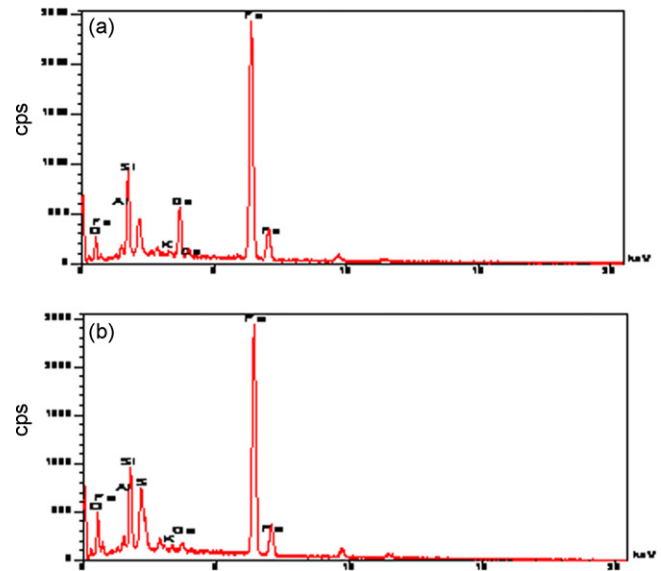
Composition	Fe	O	C	Si	Ca	Al	K
Amount (wt%)	60.19	13.83	10.47	9.3	4.63	1.45	0.13

elements of FCS particles are: Fe, O, C, Si, Ca, Al, K (Fig. 1(a)) and after the coagulation/flocculation process, a small increase in C (First peak), S and O elements was appeared due to the trapped dye molecules which contain aromatic molecules and sulphonic groups (Fig. 1(b)).

The SEM images of the used dried FCS sample (in 10  $\mu\text{m}$ -scale) before and after the experiment are presented in Fig. 2(a) and (b), respectively. As shown in Fig. 2(a), the dried FCS was regular in shape. Following the coagulation/flocculation process for AR119 dye removal, the FCS particles agglomerated with the dye molecules and resulted in the larger size particles of dye-loaded sludge (Fig. 2(b)).

### 3.2. Development of regression model equation and validation of the model

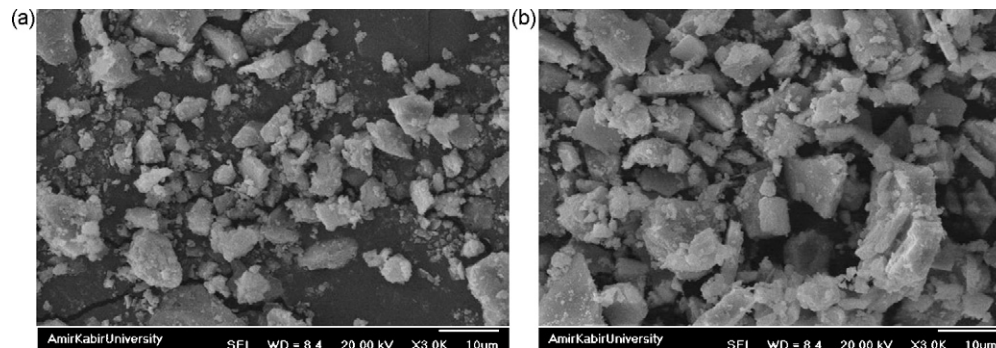
In order to study the combined effect of the factors, experiments were performed for different combinations of the parameters using statistically designed experiments. The experimental design matrix together with the maximum observed and predicted decolorization efficiencies are listed in Table 3.



**Fig. 1.** EDS of the FCS sample (a) before process; (b) after process.

The coefficients of the response function (Eq. (4)), the  $t$  and  $P$ -values for AR119 dye removal efficiencies were obtained using experimental data and are presented in Table 4.

Using the experimental results, the following second-order polynomial equation was fitted to the decolorization results and



**Fig. 2.** SEM of the used FCS sample.

**Table 3**  
Full factorial CCD matrix for AR119 dye removal.

Run order	Initial pH ( $x_1$ )	FCS dosage ( $x_2$ , mg dried FCS/L)	Initial dye concentration ( $x_3$ , mg/L)	Dye Removal (%)	
				Experimental	Predicted
1	0	0	0	51.3	51.26
2	-1	-1	-1	73	71.06
3	1	1	1	36.65	38.32
4	0	0	0	51.3	51.26
5	0	0	0	51.2	51.26
6	0	0	0	51.3	51.26
7	0	0	0	51.25	51.26
8	0	0	0	51.3	51.26
9	1	1	-1	43	44.68
10	-1.68	0	0	94	93.01
11	-1	-1	1	46.75	44.79
12	1	-1	-1	20.7	21.35
13	0	1.68	0	78.3	73
14	-1	1	-1	96.3	102.5
15	0	0	1.68	39	43.47
16	0	-1.68	0	13.1	18.78
17	1.68	0	0	9.8	11.17
18	-1	1	1	86.85	85.93
19	0	0	-1.68	75	70.91
20	1	-1	1	11.9	5.36

**Table 4**  
Estimated regression coefficients for dye removal efficiency (%) in coded units.

Term	Coefficient	SE coefficient	t	P
Constant	51.2639	1.808	28.347	0.000
$x_1$	-24.329	1.2	-20.276	0.000
$x_2$	16.1167	1.2	13.432	0.000
$x_3$	-8.1567	1.2	-6.798	0.000
$x_1^2$	0.2937	1.168	0.251	0.807
$x_2^2$	-1.8983	1.168	-1.625	0.135
$x_3^2$	2.0968	1.168	1.795	0.103
$x_1 x_2$	-2.0437	1.568	-1.304	0.222
$x_1 x_3$	2.5688	1.568	1.639	0.132
$x_2 x_3$	2.4062	1.568	1.535	0.156

obtained in terms of coded factors:

$$Y = 51.2693 - 24.329x_1 + 16.1167x_2 - 8.1567x_3 + 0.2937x_1^2 - 1.8983x_2^2 + 2.0986x_3^2 - 2.0437x_1x_2 + 2.5688x_1x_3 + 2.4062x_2x_3 \quad (5)$$

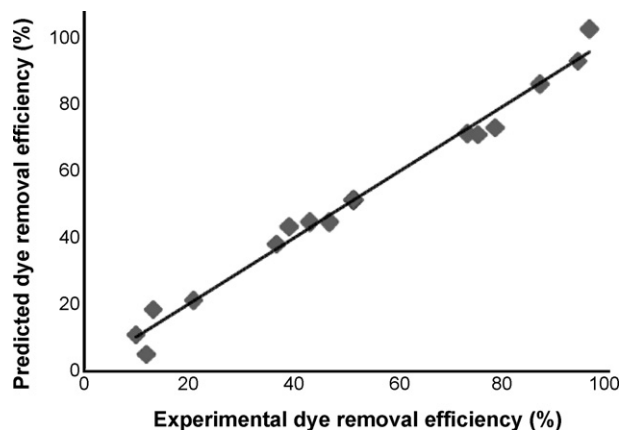
Positive sign in front of the terms indicates synergistic effect, whereas negative sign indicates antagonistic effect. The dye removal efficiency results predicted by the Eq. (5), at each experimental point, are presented in Table 3.

It was observed from Table 4 that the coefficients for the initial pH of dye solution ( $x_1$ ), coagulant dosage ( $x_2$ ) and dye concentration ( $x_3$ ) ( $P < 0.000$  for all) were highly significant whereas the square terms ( $x_{12}$ ,  $x_{22}$  and  $x_{32}$ ) and the interaction terms ( $x_1x_2$ ,  $x_1x_3$  and  $x_2x_3$ ) were insignificant to the response.

For a model to be reliable, the response should be predicted with a reasonable accuracy by the model when compared with the experimental data. Fig. 3 compares experimental dye removal efficiency (%) with the predicted values obtained from the model. The figure indicated good agreements between the experimental and predicted values of dye removal efficiency.

The adequacy of the model was further justified through ANOVA. The results of the ANOVA for AR119 dye removal are shown in Table 5. In this case, the  $P$ -value of 0.000 ( $P < 0.05$ ) for regression model equation implies that the second-order polynomial model fitted to the experimental results well. The lack-of-fit was also calculated from the experimental error (pure error) and residuals. "Lack-of-fit  $F$ -value" of 22469.34 implies the significance of model correlation between the variables and process response for dye removal.

A high  $R^2$  value, close to 1, is desirable and ensures a satisfactory adjustment of the quadratic model to the experimental data. Also a reasonable agreement with adjusted  $R^2$  is necessary [27].



**Fig. 3.** Parity plot for the experimental and predicted value of dye removal (%).

**Table 5**  
Analysis of variance (ANOVA) for dye removal efficiency (%).

Source	DF	Seq SS	Adj SS	Adj MS	F-value	P
Regression	9	12801.0	12801.0	1422.33	72.34	0.000
Linear	3	12539.4	12539.4	4179.80	212.59	0.000
Square	3	129.1	129.1	43.02	2.19	0.153
Interaction	3	132.5	132.5	44.17	2.25	0.145
Residual error	10	196.6	196.6	19.66		
Lack-of-fit	5	196.6	196.6	39.32	22469.34	0.000
Pure error	5	0.00	0.00	0.00		
Total	19	12997.6				

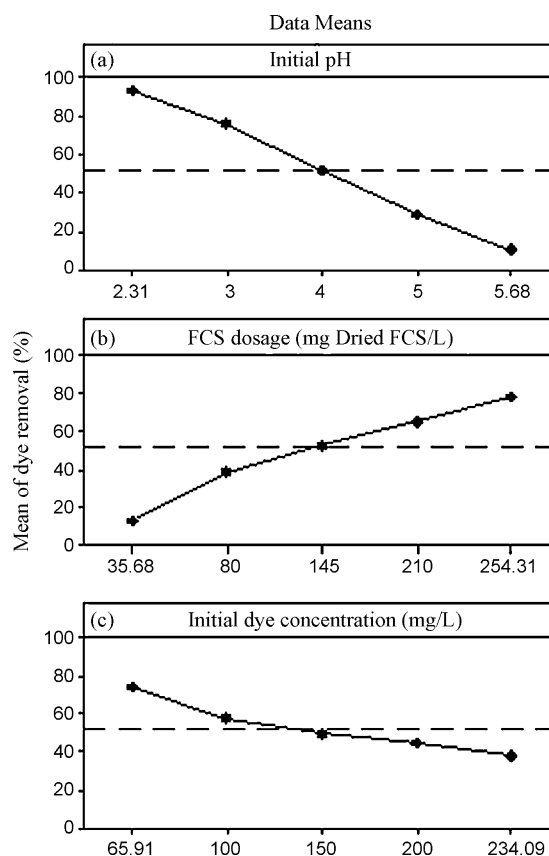
Note:  $R^2 = 98.49\%$ ;  $R^2(\text{adj}) = 97.13\%$ .

According to the ANOVA results (Table 5), the value of  $R^2 = 98.49\%$  and  $R^2(\text{adj}) = 97.13\%$  confirm the accuracy of the model.

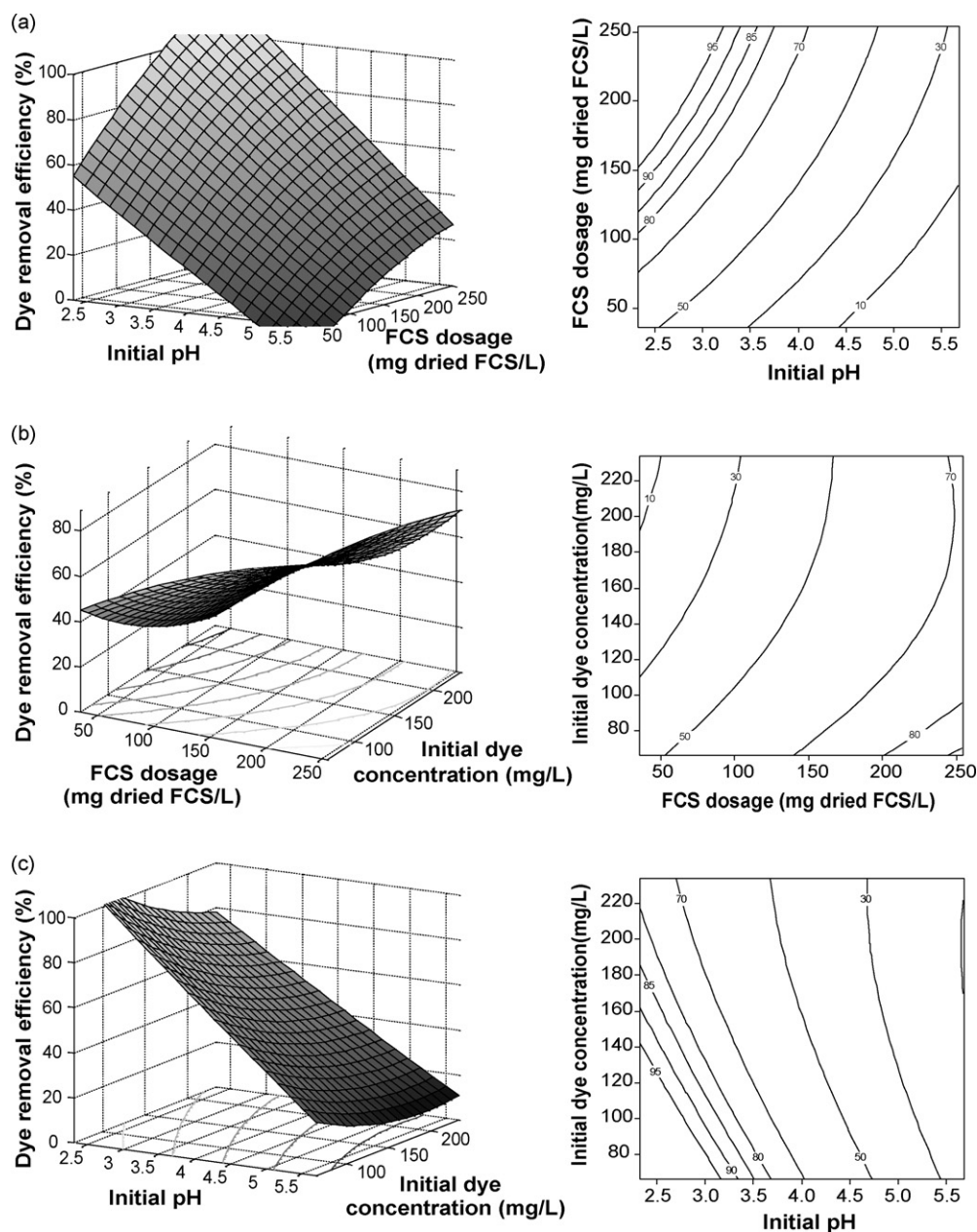
### 3.3. Response surface and counter plotting for evaluation of operational parameters

The most important parameters, which affect the efficiency of dye removal using coagulation/flocculation process, are initial pH, coagulant (FCS) dosage and initial dye concentration. Main effects plot of each parameter for AR119 dye removal using FCS are shown in Fig. 4.

As shown in Fig. 4(a), the effectiveness of FCS in removing the AR119 dye is highly dependent on initial pH. It is seen that FCS showed higher dye removals at low initial pH values and the maximum removal was obtained when the initial pH of solutions was 2.31. In other words, the dye removal efficiency decreased with increasing the initial pH of dye-containing solutions and reached



**Fig. 4.** Main effects plot of (a) initial pH, (b) FCS dosage and (c) initial dye concentration on dye removal efficiency using FCS.



**Fig. 5.** Surface plots and its corresponding contour plots for AR119 dye removal as a function of: (a) initial pH and FCS dosage at initial dye concentration of 150 mg/L; (b) FCS dosage and initial dye concentration at pH 4; (c) initial pH and initial dye concentration at 145 mg dried FCS/L.

to 11.17% at pH 5.68. Thus, pH must be controlled to establish optimum conditions for coagulation.

The results illustrated in Fig. 4(b) indicate that with the increase of FCS dosage, the removal efficiency increased and the maximum dye removal efficiency was achieved at maximum coagulant dosage (254.31 mg dried FCS/L). Furthermore, it was also observed from Fig. 4(c) that as initial dye concentration increased, dye removal efficiency decreased.

For a better explanation of the independent variables and their interactive effects on the decolorization of AR119 dye solutions, 3D plots and its corresponding contour plots are represented in Fig. 5.

pH plays an important role in the coagulation/flocculation process. Charge on hydrolysis products and precipitation of metal hydroxides are both controlled by pH variations [28]. As it can be seen from Fig. 5(a) and (c), the dye removal efficiency was sensitive even to small alterations of the initial pH. By decreasing initial pH, the dye removal efficiency increased. Charge neutralization

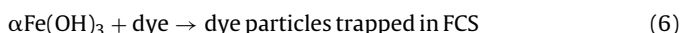
is considered to be a prerequisite condition for most coagulation processes to occur [29]. As the functional groups of acid dyes are anionic, hydrolysis products of the FCS (substantially iron hydroxides) can neutralize the negative charges on dye molecules. Therefore, the most likely mechanism dealing with removal of AR119 dye seems to be charge neutralization. On the other hand, with the decrease of pH, dye protonation processes could lead to reduction of charge density and cause self-aggregation of anionic dye molecules [29]. Therefore, less coagulant would be required to destabilize them. According to the observed results (Fig. 5(a)), at lower initial pH values, less FCS dosage was required to obtain a certain dye removal efficiency which is in agreement with this theory.

Fig. 5(a) and (b) shows that with the increase of FCS dosage, the removal efficiency steadily increased and no “re-stabilization zones” with negative dye removals were found even at the applied maximum dosage (254.3 mg dried FCS/L). The higher removal

**Table 6**  
Optimum values for decolorization of AR119 dye solutions.

Variables	Unit	Optimum Values ( $X_i$ )
Initial pH of dye solution	–	3.5
Ferric chloride sludge dosage	mg dried FCS/L	236.68
Initial dye concentration	mg/L	65.91
Dye removal efficiency (predicted)	%	100
Dye removal efficiency (experimental)	%	96.53

might be due to the sweep flocculation mechanism, which is inclined to occur at high FCS dosage. Therefore, it can be concluded that charge neutralization was not the only mechanism by which removal of the dye particles occurred. The FCS apparently served as condensation nuclei and the dye particles were enmeshed as the precipitate was settled. Theoretically, FCS could contribute to the trapping of dye particles as shown in Eq. (6).



This theory could be proved by the larger size particles of dye-loaded FCS after the experiment (Fig. 2(b)).

Some other research groups similarly reported that no re-stabilization phenomenon (removal reduction with increase of dosage) was observed even at the used maximum coagulant dosage [29,30].

At constant value of the initial dye concentration (150 mg/L), when the FCS dosage increases and the initial pH of dye solution decreases, the dye removal efficiency increases and finally reaches to 100% (Fig. 5(a)). It seems that charge neutralization and enmeshment in the precipitate both contributed to coagulation of AR119 dye in this situation.

As it shown in Fig. 5(b) and (c), the initial dye concentration slightly influenced the process efficiency. However, at a fixed coagulant dosage, the percentage of dye removal decreased with increasing the dye concentration (Fig. 5(b)). In other words, the residual concentration of dye molecules will be higher for higher initial dye concentrations. This could be ascribed to the accompanying increase in dye aggregation and/or depletion of accessible hydrolysis products of the coagulant.

### 3.4. Process optimization

In order to maximize the dye removal efficiency, regressions equation (Eq. (5)) was optimized using LINGO 9.0 software. In this study, a cost driven approach was preferred to determine the maximum dye removal with minimum chemicals consumption such as acid and base in comparison with FCS that is freely available. Therefore, several scenarios were examined in order to find the optimal conditions and the optimization results are shown in Table 6. The optimum values of the process parameters were calculated in coded units ( $x_i$ ) and then converted into uncoded units ( $X_i$ ) using Eq. (3).

Finally, the optimum values were further validated by actually carrying out the experiment at the optimal condition. The experimental checking in this optimal condition confirms good agreements with RSM results. The optimum values for AR119 dye removal were also confirmed with surface and counter plots (Fig. 5).

### 3.5. Cost evaluation

Cost of coagulation/flocculation process is comprised of two main items namely, chemicals and sludge handling and disposal. Therefore, economic interest to use hydroxide sludge for decolorization of dyeing wastewaters was considered from two points of view: (1) significant reductions in present and future sludge disposal costs at WTPs; (2) fresh coagulant savings in wastewater treatment plants (WWTPs).

Often the costs of disposal and handling the enormous quantities of waterworks sludge can account for a significant part of the overall operating costs of WTPs, and they are likely to increase due to increasingly stringent regulations. In addition, the limited land available for waterworks sludge disposal makes this a considerable worry for water purification authorities [31–33]. Hence, reusing the FCS in the WWTPs for dye removal can offer great advantages such as cost reduction in sludge disposal at WTPs and economic drives pointing toward beneficial reuse of waste streams in accordance with the concept of sustainable development.

On the other hand, hydroxide sludge generated from WTPs cost less than those conventional coagulants (inorganic salts of Al or Fe) used in WWTPs under the optimum operating conditions. Therefore, reusing the FCS as a low-cost material (almost free of charge) can provide a considerable cost benefit with fresh coagulant savings in WWTPs.

Fundamentally, such approaches at beneficial reuses offer two distinct advantages, in terms of economic savings on overall treatment plant operation costs and environmental sustainability. Therefore, it is reasonable to conclude that FCS can be fruitfully used as low-cost coagulant for the removal of AR119 dye from dye-containing solutions.

## 4. Conclusions

In this research, a coagulation/flocculation process was studied to remove AR119 dye from solutions by reusing the Ferric chloride sludge (sludge of a WTP in Tehran, Iran). Statistical optimization method (a central composite design coupled with response surface methodology (RSM)) overcomes the limitations of classical methods and was successfully employed to obtain the optimum process conditions while the interactions between process variables were demonstrated. The results clearly showed that the dye removal efficiency of FCS was severely influenced by initial pH variations and with the increase of FCS dosage, no removal reduction was observed. It was found that apart from the charge neutralization mechanism, the FCS apparently served as condensation nuclei and the dye particles were enmeshed as the precipitate was settled (sweep flocculation mechanism). From the optimization, the maximum dye removal efficiency was obtained at initial pH of dye solution, FCS dosage and initial dye concentration of 3.5, 236.68 mg dried FCS/L and 65.91 mg/L, respectively. ANOVA showed a high  $R^2$  value of regressions model equation ( $R^2 = 0.9849$ ), thus ensuring a satisfactory adjustment of the second-order regression model with the experimental data.

Therefore, RSM has been proved to be a powerful tool for optimizing the coagulation/flocculation process for the AR119 dye removal from synthetic wastewater and the FCS can be considered as an appropriate alternative for conventional costly coagulants that are widely used in WWTPs.

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