Desalination and Water Treatment
www.deswater.com
doi: 10.1080/19443994.2016.1170636

57 (2016) 26916–26931
December

Taylor & Francis
Taylor & Francis Group

Optimization of coagulation–flocculation process for medium density fiberboard (MDF) wastewater through response surface methodology

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Received 2 November 2015; Accepted 20 March 2016

ABSTRACT

This paper compares performance of alum, polyaluminum chloride (PAC), and polelectrolyts (PE) as coagulants to remove suspended solids from wastewater of medium density fiberboard (MDF) manufacture. Response surface methodology was used to optimize coagulation–flocculation (CF) process of MDF wastewater. In the treatments with alum, results revealed that full quadratic model was more adequate for chemical oxygen demand removal and total suspended solids removal, whereas linear squares model was accurate for turbidity removal. In the treatments with PAC and PE, linear squares model had the highest accuracy for all the responses. The linear term of coagulant dosage (X_1) had the largest effect on the responses in the CF process of MDF wastewater with alum and PAC; whereas, the linear term of mixing time (X_3) had the largest effect on the responses in CF process of MDF wastewater with PE. Among the coagulants used in this study, it is concluded that PE was as the best coagulant for CF of MDF wastewater. The optimal conditions of the CF process using alum were 1,500 mg/L dosage, pH 3, and 175 s mixing time. The conditions of 1,500 mg/L dosage, pH 7, and 137 s mixing time were found as the optimum CF conditions of MDF using PAC. For PE coagulant, 20 mg/L dosage, pH 8.8, and 135 s mixing time were found as the optimal conditions for the CF process of MDF with PE.

Keywords: Medium density fiberboard; Wastewater; Coagulation/flocculation; Optimization; Response surface methodology

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1. Introduction

Medium density fiberboard (MDF) is an engineered wood product composed from fine lignocellulosic fibers, combined with a synthetic resin and joined together under heat and pressure to form panels [1]. Nowadays, chip washing is seen as being a compulsory step to remove the bark, soil, sand, and other abrasive contaminates. Washed chips are heated to 40–60˚C at atmospheric pressure in a surge bin. The effluent of MDF manufacture comes from the chips washing and steam pretreated stages. It contains high-suspended solids, chemical oxygen demand (COD), biochemical oxygen demand (BOD), fatty acids, and other soluble substances [2,3]. The wastewater can cause considerable damage to the receiving waters if discharged as untreated. MDF wastewater contains fiber and causes unique solid/liquid separation challenges. Most solid/liquid separation systems have difficulty in operation when the requirements are to produce high-quality water, to remove fine particle, to operate continuously, and to remove high quantities of fiber [4].

Chemical coagulation–flocculation (CF) is a chemical treatment technique followed by sedimentation to enhance the ability of a treatment process to remove suspended particles. Research and practical applications have shown that the CF process will decrease the pollution load and can generate an adequate water recovery [5–7]. CF process could be considered as one of the most typical physicochemical processes used in urban water and wastewater treatments due to its easy operation, relatively simple design, and low-energy consumption [8]. Proper CF process is essential for good clarification and filtration performance and for disinfection byproduct control. Improper CF can cause high aluminum residuals in the treated water and the post-treatment precipitation of particle causing turbidity, deposition, and coating of pipes in water distribution system. CF is mainly done with inorganic metal salts, e.g. aluminum and ferric sulfates and chloride. Polyelectrolyts (PE) of various structures, e.g. polyacryamids, chitosan, polysaccharids, polyvinyl, and many more usually used as coagulant aid to increase the floc density in order to improve the rate of sedimentation. Among them, polyaluminum chloride (PAC) is one of typical kinds and had become most applied [9,10]. In addition to chemical coagulants, natural coagulants such as cactus *Opuntia ficus indica* [11], *Cassia obtusifolia* seed gum [12,13], mustard [14], and rice starch [15,16] were reported to be effective in water and wastewater treatments.

The separation of particulate matter from the liquid phase is one of the important steps in most wastewater treatment processes. CF processes are used to separate the suspended solids portion from the water. The process of CF separation consists of four steps, which is shown in Fig. 1. The initial step is simple: the chemical is added to wastewater. This is followed by the second step, where the solution is mixed rapidly in order to make certain that the chemicals are evenly and homogenously distributed throughout the wastewater. In the third step, the solution is mixed again, but this time is a slow fashion to encourage the formation of insoluble solid precipitates, the process known as "coagulation." The final step is the removal of the coagulated particles by way of filtration or decantation [17].

From the numerous reviews of the fundamental theory and mechanisms of coagulation, various mechanisms for destabilizing contaminants using chemical coagulants have been identified. These mechanisms include double layer compression, adsorption charge neutralization, sweep coagulation, and inter particle bridging. The type of interactions between the chemical coagulant and contaminants determines the mechanism of coagulation. The predominance mechanisms observed during conventional coagulation with metal coagulants are adsorption charge neutralization and sweep coagulation [15,16]. For aluminum salts, the mechanism of coagulation is controlled by hydrolysis

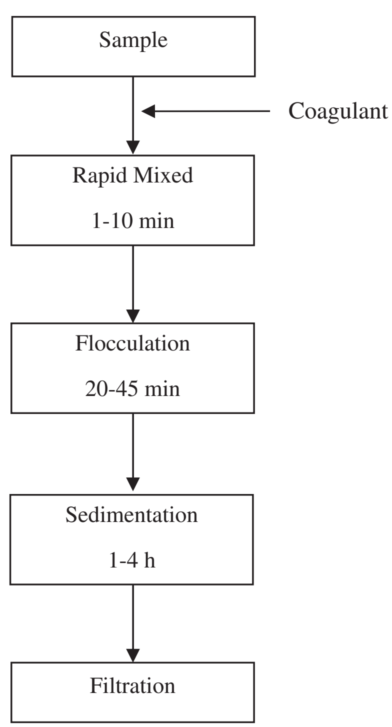

Fig. 1. Steps of coagulation process.

speciation [18]. While PE neutralized or reduces the negative charge of the particle, similar to the effect of alum. PE consists of simple monomers that are polymerized into high-molecular weight substances with molecular weights ranged in 10^4–10^6 D. The intensity of the charge depends upon degree of ionization of the functional groups, the degree of copolymerization, and the amount of substitute d groups within the polymer structure. With respect to charge, organic polymers can be cationic (positive charged), anionic (negative charged), or nonionic (no charge). Polymers in solution generally exhibit low diffusion rates and raised viscosities, thus it is necessary to mechanically disperse the polymer into the water. This is accomplished with short, vigorous mixing (velocity gradients, *G* values of 1,500 1/s, although smaller values have been reported throughout the literature, 300–600 1/s) to maximize dispersion, but not so vigorous as to degrade the polymer or the flocs as they form [19]. PE act in two distinct ways: charge neutralization and bridging between particles. Because MDF wastewater particles are charged negatively, low-molecular weight, cationic PE can act as coagulant that reduces the negative charge of the particles.

The selection of a suitable coagulant is one of the most important decisions for industrial wastewater treatment, but the different coagulants act different quality parameter of water. Several studies have been reported on the examination of CF for the treatment of industrial wastewater treatment, which aims of them were selection of the most appropriate coagulant and optimization of experimental conditions, such as coagulant dosage, pH, and mixing time [20–22]. Unlike conventional approaches, statistical experimental techniques suggest simultaneous, systematic, and efficient variation of all the components economically. Statistical approaches present appropriate ways for process optimization. Nowadays response surface methodology (RSM) is being ordinarily used for optimizing studies in several industrial processes [23–25]. RSM is a useful statistical technique performed for multiple regression analysis using measurable data. The technique solves multivariate data which are found from appropriately designed experiments to solve multivariate equation simultaneously [26]. RSM in several studies has been used to optimize CF process of wastewater such as Yolmeh and Najafzadeh [27] Subramonian et al. [28], Teh et al. [15], Moghaddam et al. [29], and Wang et al. [9]. However, the approach has not been used to optimize CF process of MDF wastewater. Therefore, the objective of this study was to optimize conditions of CF process for MDF using alum, PAC, and PE as coagulants.

2. Materials and methods

2.1. Reagents and stock solutions

Experiments were performed at laboratory scale using a jar test apparatus. Commercial grades of alum (17% w/w AL_2O_3), PAC (30% w/w AL_2O_3), and polyelectorytes (PE) were used as coagulating agents. The wastewater was collected from the equalization tank of wastewater treatment plants of MDF mill in Iran (Kimia Choob Golestan Inc. Company, Gorgan). Table 1 shows the characteristics of MDF wastewater. Jar test procedures were performed using the conventional jar apparatus (Stuart Science Flocculator model, SWI) using 500-mL wastewater samples. The coagulant was added in the range of 20–100 mg L^{-1} to the sample and the pH of the sample solution was adjusted in the range of 6–8 by the addition of the H_2SO_4 (0.1 and 1 N) and NaOH (0.1 and 1 N) solutions. The concentrations of 0.1 and 1 N were used to change pH in low and high amounts, respectively. Then, the coagulant aid of 1 mg L^{-1} was added to the sample. The sample was mixed rapidly at 200 rpm for 2 min followed by mixing at 40 rpm for 15 min. The sample was then allowed to settle for 30 min.

2.2. Optimum coagulant for raw and settled wastewater samples

In order to find the optimum coagulant dose, experiments were carried out in three consecutive steps: (1) control CF for optimization of pH at a fixed coagulant dose; (2) control CF for optimization of alkalinity at a fixed coagulant dose while maintaining optimum pH obtained in the previous step; and (3) control CF for optimization of coagulant dose while maintaining optimum pH and optimum alkalinity obtained in previous steps.

(1) *Control CF for optimization of pH*: the control coagulation and flocculation experiments were carried out at different pH levels, namely 5,6,7,8, and 9 without adjusting alkalinity but using a fixed coagulant dose. The pH that yielded minimum residual turbidity was selected as optimum pH value for the particular raw/settle wastewater sample.
(2) *Control CF for optimization of alkalinity*: the control CF experiments were carried out at the optimum pH as determined in the previous step but varying alkalinity dose while keeping the coagulant dose as fixed. The alkalinity that yielded minimum residual turbidity was selected as optimum alkalinity for the particular raw/settle wastewater sample.

Table 1
The characteristics of MDF wastewater

COD (mg/l)	BOD (mg/l)	TSS (mg/l)	Turbidity (NTU)	pH
4,800	1,700	4,500	3,500	5.5

Note: NTU: Nephelometric turbidity unit.

(3) *Control CF for optimization of coagulant dose*: after optimization both pH and alkalinity levels, the experiments were carried out by varying coagulant dosages from 20 to 100 mg L^{-1}. These experiments were performed by adjusting the pH and alkalinity of the raw/settled wastewater sample at the optimum value as determined in the previous two steps. The coagulant dosage that yielded minimum residual turbidity was selected as the optimum coagulant dose for the particular raw/settled sample.

2.3. Experimental design

A five level, three variables central composite design (CCD) was used to optimize with respect to three important reaction variables the coagulant dosage (dosage), pH, and mixing time (Tables 2 and 3). The CCD was selected in this study because of its efficiency with respect to the number of runs required for fitting a second-order response surface model. In addition, the CCD is ideal for sequential experimentation and allows a reasonable amount of information for testing lack of fit while not involving an unusually large number of design points [28,30]. In order to analyze the obtained results, namely COD removal (%), total suspended solids (TSS) removal (%), and turbidity removal (%), Minitab® version 16.1.1 (Minitab Inc. USA, 2010) was used. The factorial design composed of 3 factors, 1 block, 3 replicates, 20 base run, 60 total run. This design have 8 cube points, 6 center points in cube, 6 axial points, and 0 center points in axial. Regression analysis was performed on the data of variables including the COD removal (%), TSS removal (%), and turbidity removal (%).

2.4. Statistical analysis

The least square multiple regression methodology was used to investigate the relationship between the independent and dependent variables. The experimental design results were fitted by a second-order polynomial equation in order to correlate the response to the independent variables. The general equation to predict the optimal point was explained as follows [31,32]:

Table 2
Coded and uncoded levels of the independent variables of coagulation process for MDF with alum and PAC

Independent variables	Symbol	Coded levels				
		−0.59	−0.35	0	+0.35	+0.59
Coagulant dosage (mg/L)	X_1	500	750	1,000	1,250	1,500
pH	X_2	3	4.5	6	7.5	9
Mixing time (s)	X_3	20	60	100	140	180

Table 3
Coded and uncoded levels of the independent variables of coagulation process for MDF with PE

Independent variables	Symbol	Coded levels				
		−0.59	−0.35	0	+0.35	+0.59
Coagulant dosage (mg/L)	X_1	20	40	60	80	100
pH	X_2	3	4.5	6	7.5	9
Mixing time (s)	X_3	20	60	100	140	180

Table 4
The CCD matrix and the experimental data for the responses

Coagulant	Point type	Run no.	X_1[a]	X_2[b]	X_3[c]	Obs. responses			Pred. responses		
						Y_1 (%)[d]	Y_2 (%)[e]	Y_3 (%)[f]	Y_1 (%)	Y_2 (%)	Y_3 (%)
Alum	Fact	1	−0.35	0.35	−0.35	48.9	37.4	50.9	50.8	38.9	50.6
	Fact	2	0.35	0.35	−0.35	54.6	43.5	57.8	56.9	46.4	62.3
	Center	3	0.00	0.00	0.00	65.4	52.9	70.1	64.9	52.8	70.6
	Fact	4	0.35	−0.35	−0.35	71.8	66.2	75.3	73.3	67.2	76.1
	Fact	5	−0.35	−0.35	0.35	55.8	45.8	62.3	57.1	45.8	63.8
	Fact	6	−0.35	−0.35	−0.35	52.8	39.6	60.4	53.7	41.2	61.5
	Axial	7	−0.59	0.00	0.00	50.5	38.4	49.9	48.5	36.7	49.6
	Axial	8	0.00	0.00	−0.59	61.8	51.9	66.9	59.7	48.7	64.9
	Axial	9	0.00	0.00	0.59	66.5	55.2	70.9	64.3	53.2	69.6
	Axial	10	0.00	0.59	0.00	55.7	45.7	64.0	53.4	43.3	59.4
	Fact	11	0.35	−0.35	0.35	74.6	67.3	81.3	75.8	69.5	79.8
	Fact	12	−0.35	0.35	0.35	51.4	40.1	55.2	53.1	42.5	54.1
	Center	13	0.00	0.00	0.00	65.7	43.2	70.8	64.9	52.8	70.6
	Center	14	0.00	0.00	0.00	64.9	43.0	70.0	64.9	52.8	70.6
	Axial	15	0.00	−0.59	0.00	70.9	62.1	78.2	69.2	60.9	77.5
	Center	16	0.00	0.00	0.00	64.3	55.4	69.6	64.9	52.8	70.6
	Center	17	0.00	0.00	0.00	63.8	54.8	68.7	64.9	52.8	70.6
	Center	18	0.00	0.00	0.00	64.8	55.0	71.5	64.9	52.8	70.6
	Axial	19	0.59	0.00	0.00	72.5	67.1	79.8	69.9	63.8	74.7
	Fact	20	0.35	0.35	0.35	56.8	47.1	60.7	58.8	48.6	65.8
PAC	Fact	1	−0.35	0.35	−0.35	35.2	24.4	37.2	34.3	23.7	36.5
	Fact	2	0.35	0.35	−0.35	45.7	35.8	47.6	44.3	33.5	46.9
	Center	3	0.00	0.00	0.00	38.5	26.9	40.9	38.7	29.1	41.4
	Fact	4	0.35	−0.35	−0.35	35.6	25.8	37.9	37.6	28.7	41.1
	Fact	5	−0.35	−0.35	0.35	31.8	23.0	34.3	31.2	22.7	34.4
	Fact	6	−0.35	−0.35	−0.35	28.1	19.8	30.5	28.2	20.1	30.7
	Axial	7	−0.59	0.00	0.00	30.5	22.5	35.1	31.3	22.6	35.3
	Axial	8	0.00	0.00	−0.59	36.4	25.4	40.0	36.0	25.3	38.7
	Axial	9	0.00	0.00	0.59	39.7	29.1	42.7	40.5	29.4	44.1
	Axial	10	0.00	0.59	0.00	38.3	27.5	39.9	39.6	28.4	41.1
	Fact	11	0.35	−0.35	0.35	42.5	34.1	47.8	41.6	32.3	45.7
	Fact	12	−0.35	0.35	0.35	37.1	25.7	41.0	36.9	26.1	40.4
	Center	13	0.00	0.00	0.00	39.8	30.6	41.2	38.7	29.1	41.4
	Center	14	0.00	0.00	0.00	38.7	30.2	40.5	38.7	29.1	41.4
	Axial	15	0.00	−0.59	0.00	31.2	23.7	33.2	30.4	23.1	32.2
	Center	16	0.00	0.00	0.00	37.8	28.1	40.4	38.7	29.1	41.4
	Center	17	0.00	0.00	0.00	39.2	31.8	44.0	38.7	29.1	41.4
	Center	18	0.00	0.00	0.00	39.0	31.2	43.5	38.7	29.1	41.4
	Axial	19	0.59	0.00	0.00	48.6	37.8	52.5	48.5	37.9	52.4
	Fact	20	0.35	0.35	0.35	47.5	33.9	51.1	47.2	34.9	50.6
PE	Fact	1	−0.35	0.35	−0.35	77.3	71.3	82.7	78.7	71.6	83.4
	Fact	2	0.35	0.35	−0.35	71.2	63.5	76.6	72.5	64.3	78.2
	Center	3	0.00	0.00	0.00	83.4	77.4	89.8	83.9	77.4	92.0
	Fact	4	0.35	−0.35	−0.35	72.5	64.0	78.0	69.5	61.2	76.8
	Fact	5	−0.35	−0.35	0.35	84.6	79.2	90.7	86.6	80.1	92.2
	Fact	6	−0.35	−0.35	−0.35	75.7	68.7	80.9	75.6	67.2	81.2
	Axial	7	−0.59	0.00	0.00	89.8	80.9	93.7	87.9	80.3	92.3
	Axial	8	0.00	0.00	−0.59	64.6	55.3	72.8	65.4	56.6	72.7
	Axial	9	0.00	0.00	0.59	86.7	82.4	95.4	84.3	80.9	92.4

(*Continued*)

Table 4 (*Continued*)

Coagulant	Point type	Run no.	X_1[a]	X_2[b]	X_3[c]	Obs. responses Y_1 (%)[d]	Y_2 (%)[e]	Y_3 (%)[f]	Pred. responses Y_1 (%)	Y_2 (%)	Y_3 (%)
	Axial	10	0.00	0.59	0.00	87.3	82.0	92.6	85.1	80.4	91.6
	Fact	11	0.35	−0.35	0.35	79.0	71.2	83.9	80.4	72.7	87.2
	Fact	12	−0.35	0.35	0.35	88.5	82.3	94.3	89.3	83.6	95.1
	Center	13	0.00	0.00	0.00	82.6	78.5	90.2	83.9	77.4	92.0
	Center	14	0.00	0.00	0.00	82.7	79.0	91.7	83.9	77.4	92.0
	Axial	15	0.00	−0.59	0.00	79.4	70.5	88.3	79.7	71.3	87.1
	Center	16	0.00	0.00	0.00	83.5	75.6	91.7	83.9	77.4	92.0
	Center	17	0.00	0.00	0.00	85.7	77.1	93.3	83.9	77.4	92.0
	Center	18	0.00	0.00	0.00	85.1	77.0	94.9	83.9	77.4	92.0
	Axial	19	0.59	0.00	0.00	77.2	70.2	84.6	77.3	69.7	83.5
	Fact	20	0.35	0.35	0.35	82.5	78.1	90.3	83.1	78.3	90.3

[a]X_1: Coagulant dosage (mg/L).
[b]X_2: pH.
[c]X_3: Mixing time (s).
[d]Y_1: COD removal.
[e]Y_2: TSS removal.
[f]Y_3: Turbidity removal.

$$Y = \beta_{k0} + \sum_{i=1}^{4} \beta_{ki} x_i + \sum_{i=1}^{4} \beta_{kii} x_i^2 + \sum_{i<j=2}^{4} \beta_{kij} x_i x_j \quad (1)$$

where Y is the predicted response (LA); β_{k0}, β_{ki}, β_{kii}, and β_{kij} represent regression coefficients; and $x_i x_j$ are the coded independent factors. The models were compared based on the coefficient of determination (R^2), adjusted coefficient of determination (R^2-adj.), predicted coefficient of determination (R^2-pred.), root mean square error of prediction (RMSEP), and absolute average deviation (AAD). R^2 must be near to 1 and the RMSEP and AAD between the estimated and observed data must be as low as possible [26]. After selecting the most accurate model, the analysis of variance (ANOVA) was used to enquire the statistical significance of the regression coefficients by conducting Fisher's F-test at 95% confidence level. The interactive effects of the factors were observed using surface plots, derived from the chosen model [26]:

$$\text{RMSEP} = \sqrt{\frac{\sum_{i=1}^{N} (y_{\text{pre}} - y_{\text{exp}})^2}{N}} \quad (2)$$

$$\text{ADD} = \left\{ \sum_{i=1}^{N} (|y_{\text{exp}} - y_{\text{pre}}| / y_{\text{exp}}) / N \right\} \times 100 \quad (3)$$

where y_{pre}, y_{exp}, and N are the predicted data, observed data, and number of treatment, respectively.

Finally, the process was optimized. The aim of the optimization was to maximize the COD removal, the TSS removal, and the turbidity removal with the same weight (w = 1) and the credibility of the optimum conditions was diagnosed through the desirability values of the responses which range from 0 to 1. The closer values of desirability to 1 show the more desirable and credible optimal conditions.

3. Results and discussion

3.1. Fitting the response surface models

According to the created designs, 20 experiments were performed for each the coagulants, which the observed results are shown in Table 4.

The values of R^2, R^2-adj., R^2-pred., RMSEP, and ADD for treatments with alum coagulant revealed that the full quadratic models were the more adequate than other models for the COD removal and TSS removal; however, for the turbidity removal, the Linear squares model was observed as the more adequate model (Table 5). Linear squares model was the more adequate than other models for all the responses in treatments with PAC and PE as coagulants (Table 4).

The reduced models (with regardless to insignificant coefficients) for each response in the different coagulant are as follows:

Alum:

$$\text{COD removal}\,(\%) = 64.963 + 6.290\,X_1 - 5.042\,X_2 - 2.130\,X_1^2 - 3.337\,X_1\,X_2 \quad (4)$$

Table 5
The statistics of the four fitted models

			Responses		
Coagulant	Models	Statistics	COD removal (%)	TSS removal (%)	Turbidity removal (%)
Alum	Linear				
		R^2	77.76	75.59	82.00
		R^2-adj.	73.59	71.01	78.62
		R^2-pred.	62.44	62.04	70.36
		RMSEP	10.08	12.67	11.35
		ADD (%)	12.37	17.15	10.83
	Linear-squares				
		R^2	86.31	75.71	92.29
		R^2-adj.	79.98	64.50	88.73
		R^2-pred.	56.86	42.30	72.12
		RMSEP	3.18	6.08	2.38
		ADD (%)	5.74	8.17	2.65
	Linear-interactions				
		R^2	85.37	84.27	84.49
		R^2-adj.	78.62	77.00	77.32
		R^2-pred.	30.09	62.00	36.94
		RMSEP	7.86	9.67	8.75
		ADD (%)	7.86	12.87	7.63
	Full quadratic				
		R^2	93.92	84.39	94.78
		R^2-adj.	88.44	70.33	90.08
		R^2-pred.	55.34	42.34	62.21
		RMSEP	1.62	3.64	4.37
		ADD (%)	2.94	5.62	4.08
PAC	Linear				
		R^2	90.76	79.65	83.46
		R^2-adj.	89.03	75.83	80.35
		R^2-pred.	83.87	67.57	71.55
		RMSEP	8.96	12.76	10.13
		ADD (%)	9.93	15.17	12.64
	Linear-squares				
		R^2	96.06	88.52	92.86
		R^2-adj.	94.24	76.98	89.56
		R^2-pred.	87.60	83.22	79.49
		RMSEP	0.844	1.432	1.336
		ADD (%)	1.82	3.82	2.50
	Linear-interactions				
		R^2	92.23	84.13	84.96
		R^2-adj.	88.65	76.80	78.02
		R^2-pred.	80.97	58.57	62.49
		RMSEP	5.84	9.05	7.96
		ADD (%)	5.87	10.08	9.13
	Full quadratic				
		R^2	97.54	93.00	94.37
		R^2-adj.	95.32	86.70	89.30
		R^2-pred.	83.47	69.24	68.36
		RMSEP	2.97	4.07	3.15
		ADD (%)	3.17	5.87	4.27

(*Continued*)

Table 5 (*Continued*)

Coagulant	Models	Statistics	Responses		
			COD removal (%)	TSS removal (%)	Turbidity removal (%)
PE	Linear				
		R^2	73.91	78.28	78.28
		R^2-adj.	69.02	74.21	74.21
		R^2-pred.	57.52	65.17	65.17
		RMSEP	13.873	11.627	14.643
		ADD (%)	10.06	8.97	9.64
	Linear-squares				
		R^2	93.73	94.04	94.04
		R^2-adj.	90.84	91.29	91.29
		R^2-pred.	77.34	78.31	78.31
		RMSEP	1.468	1.237	1.602
		ADD (%)	1.53	1.43	1.46
	Linear-interactions				
		R^2	74.98	79.07	79.07
		R^2-adj.	63.43	69.40	69.40
		R^2-pred.	25.70	36.40	36.40
		RMSEP	9.643	7.358	8.064
		ADD (%)	7.31	6.48	6.91
	Full quadratic				
		R^2	94.81	94.83	94.83
		R^2-adj.	90.13	90.17	90.17
		R^2-pred.	66.93	64.25	64.25
		RMSEP	4.814	3.983	4.467
		ADD (%)	4.18	3.51	3.85

$$\text{TSS removal}\,(\%) = 50.884 + 8.015\,X_1 - 5.740\,X_2 - 4.375\,X_1\,X_2 \quad (5)$$

$$\text{Turbidity removal}\,(\%) = 70.241 + 7.192\,X_1 - 5.634\,X_2 - 3.028\,X_1^2 \quad (6)$$

PAC:

$$\text{COD removal}\,(\%) = 38.815 + 5.092\,X_1 + 2.888\,X_2 + 1.453\,X_3 - 1.323\,X_2^2 \quad (7)$$

$$\text{TSS removal}\,(\%) = 29.791 + 4.571\,X_1 + 1.720\,X_2 + 1.253\,X_3 - 1.426\,X_2^2 \quad (8)$$

$$\text{Turbidity removal}\,(\%) = 41.736 + 5.174\,X_1 + 2.758\,X_2 + 1.870\,X_3 - 1.615\,X_1^2 - 1.747\,X_2^2 \quad (9)$$

PE:

$$\text{COD removal}\,(\%) = 83.889 - 3.082\,X_1 + 1.536\,X_2 + 5.496\,X_3 - 3.260\,X_3^2 \quad (10)$$

$$\text{TSS removal}\,(\%) = 77.466 - 3.126\,X_1 + 2.302\,X_2 + 6.507\,X_3 - 3.251\,X_3^2 \quad (11)$$

$$\text{Turbidity removal}\,(\%) = 92.021 - 2.564\,X_1 + 5.779\,X_3 - 1.615\,X_1^2 - 3.401\,X_3^2 \quad (12)$$

The ANOVA was used to evaluate the significance of the quadratic polynomial models and validity of the model in addition to R^2 [33]. For each terms in the models, a large F-value and a small p-value would imply a more significant effect on the respective response variable [30]. Therefore, the linear term of coagulant dosage (X_1) had the largest effect on the COD removal, TSS removal, and turbidity removal in the CF process of MDF wastewater with alum (Table 6). Also for CF process of MDF wastewater with PAC, the linear term of X_1 had the largest effect on the responses (Table 7). Whereas, the linear term of X_3 had the largest effect on the responses CF process of MDF wastewater with PE (Table 8).

For CF process of MDF wastewater with alum, the linear terms of X_1 and X_2 showed a significant effect ($p < 0.05$) on the COD removal. The quadric term of X_1^2 and the interactive term of X_1X_2 also had a significant

Table 6
ANOVA of the models for COD removal, TSS removal, and turbidity removal of MDF wastewater coagulated with alum

	Source	Degrees of freedom	Mean of squares	F	p
COD removal (%)	Regression	9	122.431	17.15	0.000
	Linear	3	304.111	42.61	0.000
	Coagulant dosage (X_1)	1	540.294	75.69	0.000
	pH (X_2)	1	347.236	48.65	0.000
	Mixing time (X_3)	1	24.802	3.47	0.092
	Square	3	33.418	4.68	0.027
	X_1^2	1	65.354	9.16	0.013
	X_2^2	1	32.129	4.50	0.060
	X_3^2	1	20.498	2.87	0.121
	Interaction	3	29.765	4.17	0.037
	X_1X_2	1	89.111	12.48	0.005
	X_1X_3	1	0.031	0.00	0.949
	X_2X_3	1	0.151	0.02	0.887
	Residual error	10	7.138	–	–
	Lack-of-fit	5	13.790	28.39	0.091
	Pure error	5	0.486	–	–
	Total	19	–	–	–
TSS removal (%)	Regression	9	167.974	6.01	0.005
	Linear	3	451.385	16.14	0.000
	(X_1)	1	877.444	31.37	0.000
	(X_2)	1	449.858	16.08	0.002
	(X_3)	1	26.852	0.96	0.350
	Square	3	0.720	0.03	0.994
	X_1^2	1	2.036	0.07	0.793
	X_2^2	1	0.014	0.00	0.983
	X_3^2	1	0.125	0.00	0.948
	Interaction	3	51.818	1.85	0.202
	X_1X_2	1	153.125	5.47	0.041
	X_1X_3	1	2.205	0.08	0.785
	X_2X_3	1	0.125	0.00	0.948
	Residual error	10	27.972		
	Lack-of-fit	5	20.390	0.57	0.722
	Pure error	5	35.554	–	–
	Total	19	–	–	–
Turbidity removal (%)	Regression	6	220.994	25.93	0.000
	Linear	3	392.701	46.08	0.000
	(X_1)	1	706.427	82.88	0.000
	(X_2)	1	433.521	50.86	0.000
	(X_3)	1	38.155	4.48	0.054
	Square	3	49.287	5.78	0.010
	X_1^2	1	132.184	15.51	0.002
	X_2^2	1	9.663	1.13	0.306
	X_3^2	1	22.271	2.61	0.130
	Residual error	13	8.523	–	–
	Lack-of-fit	8	13.266	14.21	0.065
	Pure error	5	0.934	–	–
	Total	19	–	–	–

Table 7
ANOVA of the models for COD removal, TSS removal, and turbidity removal of MDF wastewater coagulated with PAC

Response	Source	DF	Mean of squares	F	p
COD removal (%)	Regression	6	87.645	52.86	0.000
	Linear	3	165.618	99.88	0.000
	Coagulant dosage (X_1)	1	354.099	213.55	0.000
	pH (X_2)	1	113.904	68.69	0.000
	Mixing time (X_3)	1	28.851	17.40	0.001
	Square	3	9.673	5.83	0.009
	X_1^2	1	2.015	1.22	0.290
	X_2^2	1	25.228	15.21	0.002
	X_3^2	1	0.352	0.21	0.652
	Residual error	53	1.658	–	–
	Lack-of-fit		2.408	5.25	0.052
	Pure error	45	0.459	–	–
	Total	59	–	–	–
TSS removal (%)	Regression	6	64.330	16.71	0.000
	Linear	3	115.758	30.07	0.000
	X_1	1	285.401	74.14	0.000
	X_2	1	40.406	10.50	0.006
	X_3	1	21.468	5.58	0.034
	Square	3	12.902	3.35	0.052
	X_1^2	1	0.476	0.12	0.731
	X_2^2	1	29.340	7.62	0.016
	X_3^2	1	10.253	2.66	0.127
	Residual error	13	3.850	–	–
	Lack-of-fit	8	3.998	1.11	0.477
	Pure error	5	3.612	–	–
	Total	19	–	–	–
Turbidity removal (%)	Regression	6	95.927	28.17	0.000
	Linear	3	172.429	50.64	0.000
	X_1	1	365.625	107.38	0.000
	X_2	1	103.895	30.51	0.000
	X_3	1	47.766	14.03	0.002
	Square	3	19.425	5.70	0.010
	X_1^2	1	9.585	2.82	0.117
	X_2^2	1	44.019	12.93	0.003
	X_3^2	1	0.037	0.01	0.919
	Residual error	13	3.405	–	–
	Lack-of-fit	8	3.966	1.58	0.318
	Pure error	5	2.507	–	–
	Total	19	–	–	–

effect ($p < 0.05$) on the COD removal. However, an insignificant effect ($p > 0.05$) was observed for other terms. The linear term of X_1 and X_2 and the interactive term of X_1X_2 had the largest effects on the TSS removal ($p < 0.05$). For turbidity removal, significant effect was observed for the linear term of X_1 and X_2 and the quadric term of X_1^2 (Table 6).

For CF process of MDF wastewater with PAC, the all linear terms and the quadric term of X_1^2 had significant effect on each of the three responses. However, the other terms had insignificant effect ($p > 0.05$) on the responses (Table 7).

The all linear terms and the quadric term of X_3^2 had significant effect on each the three responses of CF process for MDF wastewater with PE; whereas, an insignificant effect ($p > 0.05$) was observed for other terms (Table 8).

The fitness of the models was investigated through lack-of-fit test that its values for all responses and coagulants were insignificant ($p > 0.05$), which indicated suitability of models to accurately predict the responses [34].

Table 8
ANOVA of the models for COD removal, TSS removal, and turbidity removal of MDF wastewater coagulated with PE

	Source	DF	Mean of squares	F	p
COD removal (%)	Regression	6	121.456	32.41	0.000
	Linear	3	191.532	51.12	0.000
	Coagulant dosage (X_1)	1	129.724	34.62	0.000
	pH (X_2)	1	32.249	8.61	0.012
	Mixing time (X_3)	1	412.624	110.12	0.000
	Square	3	51.379	13.71	0.000
	X_1^2	1	3.387	0.90	0.359
	X_2^2	1	4.169	1.11	0.311
	X_3^2	1	153.175	40.88	0.000
	Residual error	13	3.747	–	–
	Lack-of-fit	8	5.065	3.09	0.115
	Pure error	5	1.639	–	–
	Total	19	–	–	–
TSS removal (%)	Regression	6	157.028	34.20	0.000
	Linear	3	261.418	56.93	0.000
	X_1	1	133.477	29.07	0.000
	X_2	1	72.384	15.76	0.002
	X_3	1	578.394	125.97	0.000
	Square	3	52.638	11.46	0.001
	X_1^2	1	11.225	2.44	0.142
	X_2^2	1	5.812	1.27	0.281
	X_3^2	1	152.348	33.18	0.000
	Residual error	13	4.592	–	–
	Lack-of-fit	8	6.555	4.52	0.057
	Pure error	5	1.451	–	–
	Total	19	–	–	
Turbidity removal (%)	Regression	6	127.693	23.35	0.000
	Linear	3	189.559	34.66	0.000
	X_1	1	89.802	16.42	0.001
	X_2	1	22.763	4.16	0.062
	X_3	1	456.113	83.39	0.000
	Square	3	65.827	12.04	0.000
	X_1^2	1	37.599	6.87	0.021
	X_2^2	1	18.953	3.47	0.085
	X_3^2	1	166.662	30.47	0.000
	Residual error	13	5.470	–	–
	Lack-of-fit	8	6.613	1.82	0.265
	Pure error	5	3.640	–	–
	Total	19	–	–	–

3.2. Effects of the CF conditions on responses

3.2.1. CF process with alum

Fig. 2(a) presents the interaction between the mixing time and dosage on the COD removal when pH was kept at the central point. Giving the figure, the COD removal was gently increased by increasing mixing time. On other hand, the COD removal was increased by increasing dosage of the coagulant, although the intensity was reduced at the high dosages. This is due to there are further negatively charged complexes that have caused larger and more stable clots. As well as, further accelerate and increasing particle collisions lead to accumulation of colloidal particles and they convert to the settle-able particles [35].

According to Fig. 2(b), the COD removal was initially increased by increasing mixing time; however, it was almost constant at the high mixing time. This fact is most likely due to saturation of the CF process at long mixing time, in which case the process is not

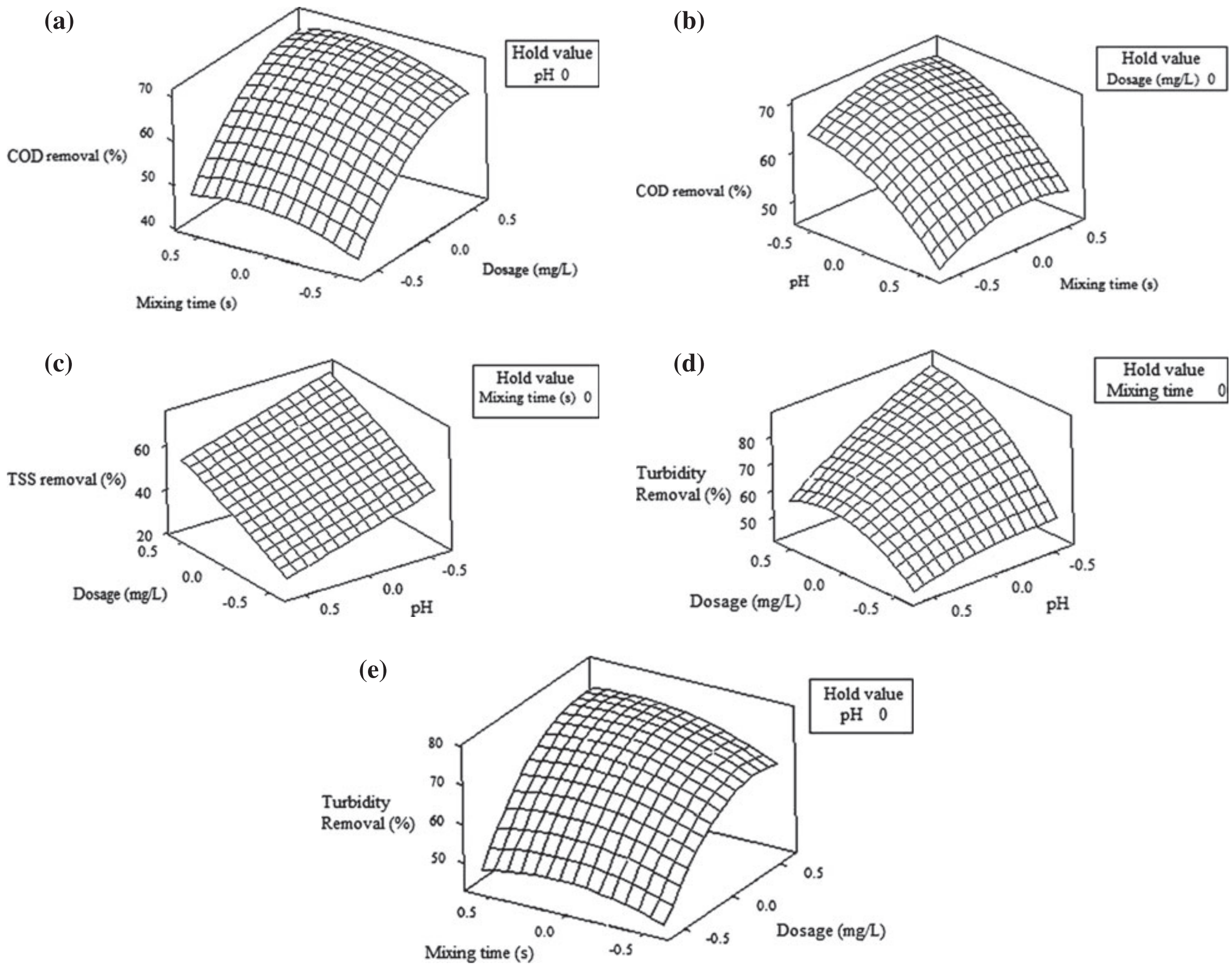

Fig. 2. (a) The interactive effect of mixing time and dosage on the COD removal as using alum as coagulant, (b) the interactive effect of pH and mixing time on the COD removal as using alum as coagulant, (c) the interactive effect of dosage and pH on the TSS removal as using alum as coagulant, (d) the interactive effect of dosage and pH on the turbidity removal as using alum as coagulant, and (e) the interactive effect of mixing time and dosage on the turbidity removal as using alum as coagulant.

done more. Since amount of positive ion (H^+) is increased by reducing pH and thereby neutralize the negatively charged anionic compounds, COD removal was remarkably increased by decreasing pH [36].

Fig. 2(c) shows the interaction between dosage and pH on the TSS removal when mixing time was kept at the central point. TSS removal was increased by increasing dosage and decreasing pH. As shown in Fig. 2(c), it can be concluded that the TSS removal was maximum when pH and dosage were approximately 3 and 1,500 mg/L, respectively.

As is obvious in Fig. 2(d), turbidity removal was initially increased by increasing dosage up to 1,250 mg/L, but subsequently decreased at high pH. This phenomenon probably due to there are too alum complexes. Since these complexes have similarly electric charge that caused the repulsive force and eventually the turbidity is returned by movement and breakdown of larger particles [36]. However, at low pH, the decreasing trend of turbidity removal was not observed. Turbidity removal was increased by reducing pH especially at high coagulant dosage (Fig. 2(d)). Our finding is in agreement with results of Ghafari et al. [36] that used polyaluminum chloride and alum for the treatment of partially stabilized leachate.

The interactive effect of mixing time and dosage on the turbidity removal is shown in Fig. 2(e). At low coagulant dosage, turbidity removal was slightly increased and then decreased by increasing mixing time. However, at high coagulant dosage, turbidity removal was increased by increasing mixing time. This is due to at low coagulant dosage providing the

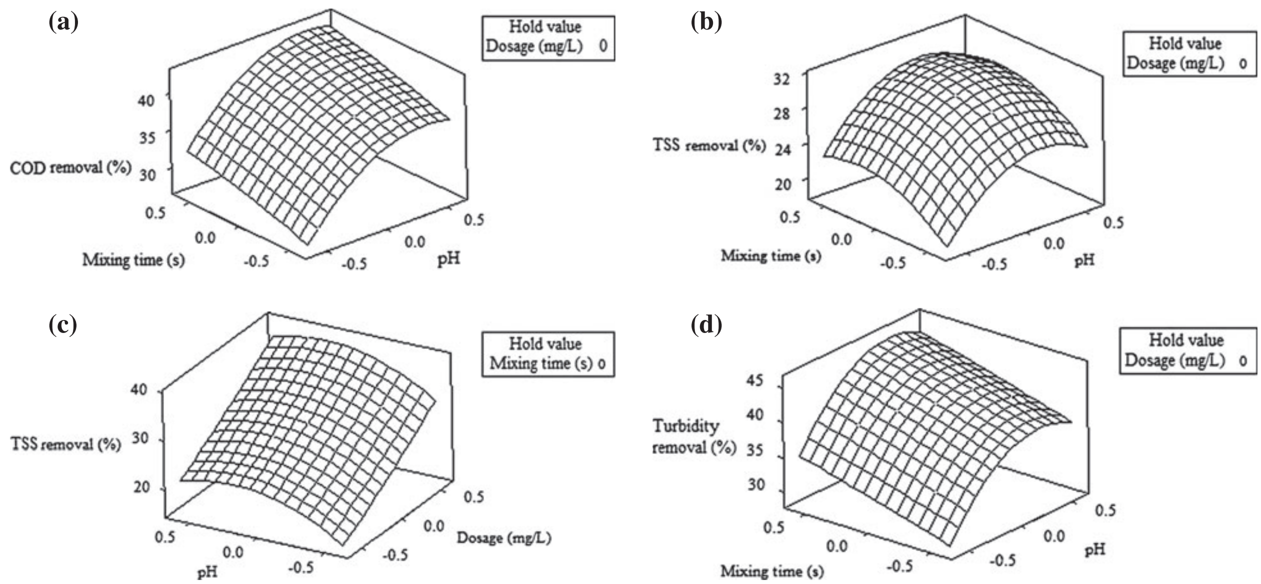

Fig. 3. (a) The interactive effect of mixing time and pH on the COD removal as using PAC as coagulant, (b) the interactive effect of mixing time and pH on the TSS removal as using PAC as coagulant, (c) the interactive effect of pH and dosage on the TSS removal as using PAC as coagulant, and (d) the interactive effect of mixing time and pH on the turbidity removal as using PAC as coagulant.

mixing time is not important and the process is performed in a short time and even at times too long, process remains incomplete due to low dose and thereby in again turbidity. However, at high coagulant dosage providing the mixing time is very important. Kumar et al. [37] studied decolorization and reduction in COD of dyeing wastewater from a cotton textile mill. Their result revealed that the time needed to complete CF process is increased by increasing coagulant dosage.

3.2.2. CF process with PAC

Fig. 3(a) shows the interaction between dosage and pH on the COD removal when mixing time was kept at the central point. The COD removal was increased by increasing pH and dosage. By comparison Figs. 2(b) and 3(a), it can be concluded that PAC and alum had considerable difference in optimum pH for COD removal operating, which this is due to differences in nature of the two coagulant (optimum pH of alum and PAC were low and high, respectively).

The interactive effect of mixing time and pH on the TSS removal is shown in Fig. 3(b). Giving the figure, TSS removal was initially increased by increasing mixing time and pH, but it subsequently decreased, so that the maximum TSS removal was measured at mixing time of 100 s and pH 7. Irfan et al. [22] studied the removal of COD, TSS, and color of black liquor by CF process at optimized pH, settling, and dosing rate. They reported that the maximum TSS removal is observed at about neutral pH [22]. It is noteworthy that the maximum TSS removal with PAC was measured at neutral pH, however, this value with alum was observed at acidic pH.

TSS removal was increased by increasing pH and coagulant dosage at constant mixing time (Fig. 3(c)). Irfan et al. [22] also observed that the TSS removal is increased by increasing dosage.

Giving Fig. 3(d), turbidity removal was increased by increasing mixing time. On other hand, by increasing pH, turbidity removal was initially increased but subsequently decreased. Giving the Fig. 3(d), the maximum TSS removal was observed at mixing time of 180 s and pH about 8. As mentioned above and in Fig. 3(d) is also clear, pHs above the around neutral increasing in turbidity removal were not observed and the value remains almost constant.

3.2.3. CF process with PE

Fig. 4(a) describes the interaction between dosage and mixing time on COD removal when pH was kept at the central point. Giving the figure, COD removal was initially increased with increasing mixing time; however, it remains almost constant by over increasing mixing time, which this is due to breakdown clots that had grown. As is obvious by comparison in

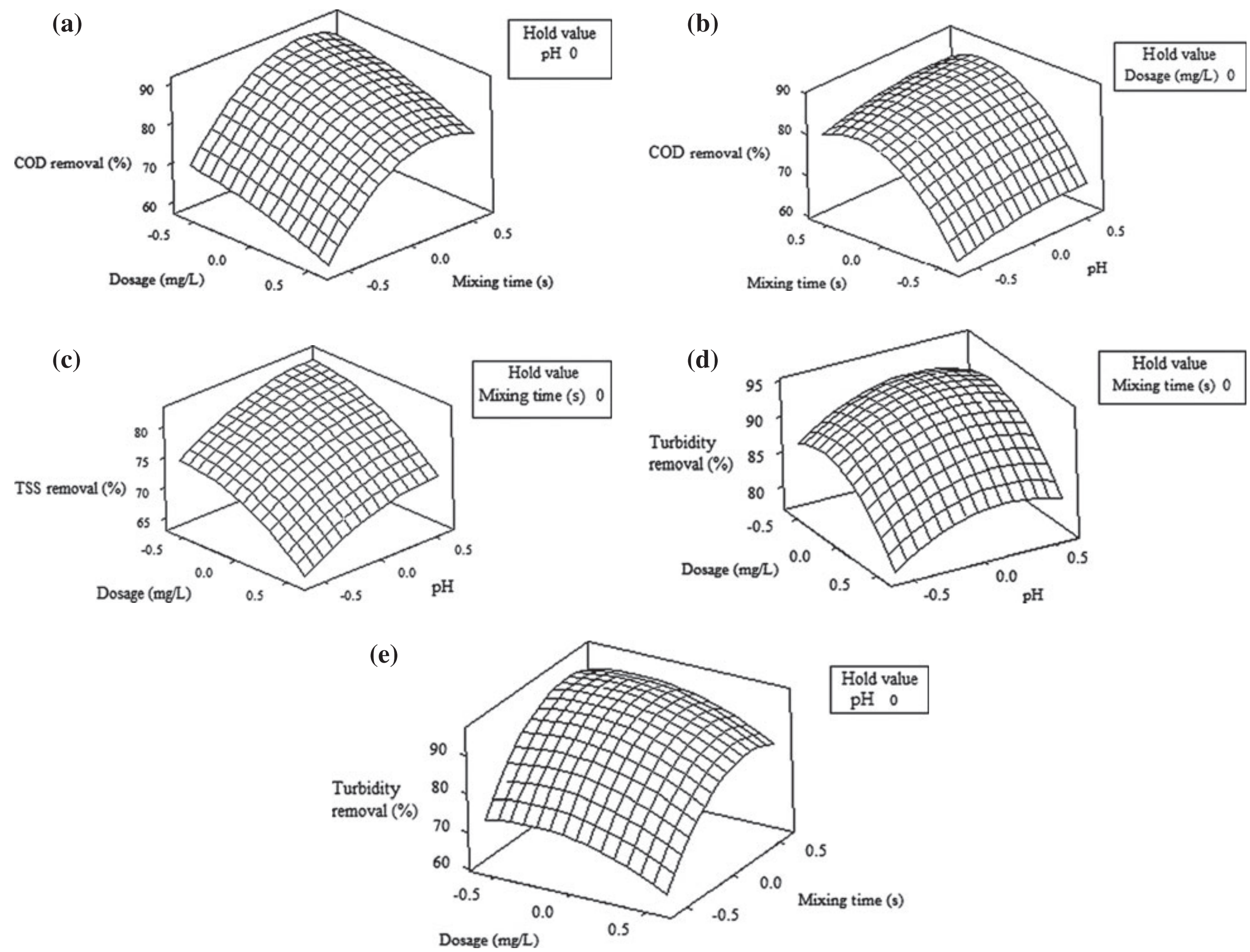

Fig. 4. The interactive effect of dosage and mixing time on the COD removal as using PE as coagulant, (b) the interactive effect of mixing time and pH on the COD removal as using PE as coagulant, (c) the interactive effect of dosage and pH on the TSS removal as using PE as coagulant, (d) the interactive effect of dosage and pH on the turbidity removal as using PE as coagulant, and (e) the interactive effect of dosage and mixing time on the TSS removal as using PE as coagulant.

Figs. 2(a), 3(a), and 4(a), PE coagulant acted contrast to alum and PAC, so that the COD removal was increased by decreasing coagulant dosage, which this is due to difference between the nature of the coagulants.

COD removal was gently increased by increasing pH; on other hand, it was remarkably increased by increasing mixing time; however, COD removal decreased by too increasing mixing time (Fig. 4(b)). Freitas et al. [38] studied optimization of CF process for the treatment of industrial textile wastewater using okra (*A. esculentus*) mucilage as natural coagulant, which they observed similar result and reported that the COD removal was decreased at too long time.

Giving Fig. 4(c), the TSS removal was increased by decreasing the coagulant dosage and increasing pH, so that the maximum TSS removal was observed at the coagulant dosage of 20 mg/L and pH 9.

Fig. 4(d) shows the interactive effect of dosage and pH on the turbidity removal as using PE as coagulant. Turbidity removal was initially increased by decreasing the coagulant dosage and increasing pH, but subsequently decreased, so that the maximum turbidity removal was observed almost at the central point (dosage of 60 mg/L, pH of 6, and mixing time of 100 s) (Fig. 4(d)). Leiviska and Ramo [39] evaluated coagulation of wood extractives in chemical pulp bleaching filtrate by cationic PEs. Their result showed that the turbidity removal was decreased at too much amounts of the coagulant dosage and pH.

According to Fig. 4(e), initially decreasing dosage and increasing pH had positive effect on turbidity removal. However, further increasing these changes decreased turbidity removal.

3.3. Optimization of CF process

The numerical optimization technique was used to optimize the CF process for each coagulant, when weight and importance value for the three responses were considered equal (Weight = 1, importance = 1) [34].

The optimal conditions of the CF process with alum were 1,500 mg/L dosage, pH 3, and 175 s mixing time. The conditions of 1,500 mg/L dosage, pH 7, and 137 s mixing time were found as the optimum CF conditions of MDF with PAC. For PE coagulant, 20 mg/L dosage, pH 8.8, and 135 s mixing time were found as the optimal conditions for the CF process of MDF. The COD removal, TSS removal, and turbidity removal were acquired 73, 74.98, and 87.48%, respectively, for alum with composite desirability of 0.999; 51.02, 38.93, and 55.52%, respectively, for PAC with composite desirability of 0.998; and 91.09, 85.50, and 93.38%, respectively, for PE with composite desirability of 1.

4. Conclusions

RSM was successfully used to optimize CF process of MDF wastewater with alum, PAC, and PE. PE had a more performance for CF process of MDF wastewater compared to two other coagulants. This is could be attributed to the particles of MDF wastewater that are charged negatively, PE neutralized and reduced the negative charge; in addition, PE is attached itself to another particles by forming a bridge. The highest COD removal, TSS removal, and turbidity removal were observed at coagulant dosage of 20 mg/L, pH 8.8, and mixing time of 135 s as the optimum condition of the CF process with PE.

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