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Statistical modeling and process optimization of coagulation–flocculation for treatment of municipal wastewater

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ABSTRACT

In this study coagulation–flocculation method was evaluated to treat municipal wastewater using aluminum sulphate and calcium hydroxide as coagulants, and polyacrylamide as flocculant by varying pH from 5 to 7 and coagulant dosage from 100 to 300 mg L^{-1} at constant flocculant dosage (10 mg L^{-1}), time (1 min for rapid mixing followed by 30 min for slow mixing), temperature (30°C ± 2°C), agitation speed (150 rpm for rapid mixing followed by 50 rpm for slow mixing) and settling time (2 h) by jar test method. The responses were percentages reduction in turbidity, chemical oxygen demand (COD) and biochemical oxygen demand (BOD). The sample was real-time wastewater from sewage treatment plant of Salalah Sanitary Drainage Services (SSDS) Company, Salalah, Sultanate of Oman. The trials were performed with single and mixed or combined coagulants. Response surface methodology (RSM) based central composite design (CCD) was used for the optimization of pH and coagulant dosage to maximize percentage reduction in turbidity, COD and BOD. The maximum of 99.7%, 98%, and 95.5% were achieved for turbidity removal using aluminum sulphate, calcium hydroxide and combined coagulants respectively. However, maximum of 60%, 58.1%, and 42.5% were achieved for COD removal using calcium hydroxide, combined and aluminum sulphate respectively. Moreover, maximum of 79.5%, 78.5%, and 54.5% were achieved for BOD removal using aluminum sulphate, calcium hydroxide and combined coagulants respectively. The obtained results indicate that treatment using combined coagulant is effective for removal of turbidity and COD, and could be used for the treatment of municipal wastewater.

Keywords: Coagulation; Flocculation; Municipal wastewater; Turbidity; COD; BOD

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1. Introduction

A municipal wastewater is mainly comprised of water (99.9%) together with a small amount of concentration of suspended and dissolved organic and inorganic solids [1]. It causes risk to human health due to exposure of micro-organisms, and high load of organic pollutants in soil and aquatic life from untreated wastewater in industrial and municipal worldwide [2]. Municipal sewage treatment plant of SSDS collected 3,500–4,000 m^3 d^{-1} of influent from domestic and commercial activities [3]. In the Sultanate of Oman, like in many other Mediterranean countries, demands on water resources for households, commercial, industrial, and agricultural use are on the increase due to rapid population growth and demographic shifts [4].

The increasing population, and extensive industrialization and urbanization in the region of Salalah produces a large quantity of liquid effluent. The main goal of most global sewage treatment plant is to produce a disposable effluent without causing harm to the environment and prevent pollution [5]. Many countries are suffering from water shortages and forced to use non-conventional resources such as cost effective harvesting technology and wastewater treatment [6]. In Salalah city, sewage treatment plant processed effluent for agriculture, industrial and municipal

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purposes, and feeding the aquifer by natural recharge to maintain the composition of the groundwater in the future. The more rapid withdrawal of large quantity of groundwater from the aquifer leads to the intrusion of seawater [7].

The physical, physico-chemical and biological methods for treatment of water and wastewater include coagulation–flocculation, advanced oxidation process, activated carbon adsorption, chlorination and ozonation disinfection have been commonly used on large scale [8–10]. The process of coagulation–flocculation is mostly widespread technique for yielding high pollutant removal efficiency such as small particles of color, turbidity, and bacteria in water and wastewater treatment prior to sedimentation and filtration [11]. Many coagulants and flocculants are widely used in water and wastewater treatment processes and are classified as inorganic such as aluminum and ferric salts due to its low cost and ease to use [12]. Similarly, organic polymeric flocculants such as polyacrylamide derivatives has its remarkable ability to flocculate efficiently with low dosage. Many coagulants are widely used in water and wastewater treatment processes and are classified as inorganics such as aluminum and ferric salts, and organics like polyacryl amide derivatives. The effectiveness of aluminum and ferric coagulants are well recognized from their ability to form multi-charge polynuclear complexes with enhanced adsorption characteristics and its complexity formed has been controlled by the pH system [13].

The cheaper and widely used metal coagulants in water and wastewater treatments around the world are aluminum salts due to its low cost and ease of use [14]. Other chemicals used as coagulants include calcium hydroxide and magnesium carbonate to increase the pH of raw water before the water is treated with alum or ferric sulphates [12]. The advantages of using calcium hydroxide as coagulant are its low cost, safety in handling and sludge captures more metals on the treatment with fewer tendencies to leach them out to produce high fecal coliform reduction [15,16]. Furthermore, high quality of calcium hydroxide is available abundantly in the governorate of Dhofar, Sultanate of Oman.

Full and fractional factorial designs are commonly used methods for process optimization [17]. But, the former is expensive, laborious and leads to unpredictable optimal region [18]. The latter helps to achieve optimal region with minimum experiments, error, and cost [19]. RSM is a fractional factorial design and useful for the modeling and analysis of problems in which dependent variables are influenced by independent variables [20]. Box-Wilson and Box-Behnken are the designs of RSM. Box-Wilson design or CCD is used for process with two or more variables whereas Box-Behnken design (BBD) is used for three or more variables [21].

The present study aimed to investigate the potential of coagulation–flocculation process using single and combined coagulants of aluminum sulphate and calcium hydroxide, and polyacrylamide as flocculant for the treatment of municipal wastewater for compliance of Sultanate of Oman discharge standards in terms of turbidity, COD, and BOD.

2. Materials and methods

2.1. Materials

The municipal wastewater was collected from SSDS water reclamation plant. The sample was filled in air-tight plastic bottle and transported to the laboratory, and characterized. The coagulants and flocculants used were of analytical grade and used without further purification.

2.2. Batch experimental studies using jar test

Coagulant dosage (100–300 mg L^{-1}) and pH (5–7) were independent variables at constant flocculant dosage (10 mg L^{-1}), time (1 min for rapid mixing followed by 30 min for slow mixing), temperature (30°C ± 2°C), agitation speed (150 rpm for rapid mixing followed by 50 rpm for slow mixing) and settling time (2 h) and turbidity, COD and BOD were dependent variables for coagulation–flocculation process by jar test apparatus. The apparatus is equipped with six beakers of 500 mL volume each at room temperature. The wastewater samples were thoroughly shaken before transferring to the corresponding jar test beakers for the re-suspension of possible settling solids.

The sample was adjusted initial pH from 5.0 to 7 by adding 1 N HCl or NaOH solution. Jar tests were dose with mixed coagulant–flocculant dosage from 110 to 310 mg L^{-1}. After performing the jar test method, the supernatant was withdrawn from the beaker for chemical analysis and sludge (dense floc) was discarded. The experiments were performed in triplicate the mean values were taken as response. The coefficient of variance was found to be within 5%. The characteristics of supernatant separated after settling was performed according to the standard operating procedures reported elsewhere [22].

2.3. Statistical modeling and process optimization

The main purpose of coagulation–flocculation is to remove total solids (both suspended and dissolved) from wastewater [1]. The factors affecting the performance of coagulation–flocculation process are pH of feed solution, the dosage of coagulant and flocculant, time for fast and slow mixing, temperature, agitation speed, and settling time [23]. The present study focused on the variation of pH and coagulant dosage while all other factors fixed at a constant value. The main advantage of RSM is that it is used to study the interaction effect between the factors [20]. Face-centered CCD is used in the present study since it can be used for two or more variables.

The number of experiments performed in CCD for k factors is $2^k + 2k + c$ where c is number of center points [24]. 11 trials were performed with 3 center points for 2 factors as per the CCD matrix as shown in Table 1. All the trials were performed in triplicate and the average value was taken as a response. The experimental data were fitted to the quadratic model as shown in Eq. (1) and model coefficients were evaluated using Eq. (2). The model coefficients were analyzed for comparing experimental and predicted values. Analysis of variance (ANOVA) was used to test the significance of the model. The best of fit between experimental and predicted values were analyzed by regression coefficients R^2, adjusted R^2 and predicted R^2. The value of R^2 between 0.9 and 1 reveals the best fit of data between experimental and predicted values.

$$Y = \beta_0 + \sum_{i=1}^{n}\beta_i X_i + \sum_{i=1}^{n}\beta_{ii} X_i^2 + \sum_{i=1}^{n-1}\sum_{j=i+1}^{n}\beta_{ij} X_i X_j + \varepsilon \tag{1}$$

Table 1
CCD matrix for reduction in turbidity, COD and BOD by coagulation–flocculation using aluminium sulphate, calcium hydroxide and combined aluminium sulphate-calcium hydroxide as coagulants and polyacrylamide as flocculant

pH	Coagulant-flocculant dosage (mg L^{-1})	Aluminium sulphate-polyacrylamide			Calcium hydroxide-polyacrylamide			Aluminium sulphate–calcium hydroxide–polyacrylamide		
		% Turbidity reduction	% COD reduction	% BOD reduction	% Turbidity reduction	% COD reduction	% BOD reduction	% Turbidity reduction	% COD reduction	% BOD reduction
6	110	22	15.85	68	58.3	9	96.3	87.6	78.5	30.1
7	210	99.1	50	53.5	88.9	64	21.4	82.6	67.5	26.3
5	310	88.4	41.4	42.1	98.9	76	70.5	40.3	43	21.1
6	210	99.7	42.5	79.5	97.3	76	28.5	76	93.6	8
5	210	99.6	75.6	52.6	97.5	65	27.3	97	62.8	1.2
6	210	99.7	42	80	97.8	76.4	31.5	75.8	93.2	76
5	110	24.5	20.5	66.1	56.5	9.76	44.9	85.5	54	27.3
6	210	99.7	42	80	98.3	75.8	31	76.4	94.2	8.4
6	310	84.6	26	90.5	98.3	51	80	47.6	85.1	32.6
7	110	36.5	16	45.8	27.3	12	89.8	95.5	58.1	54.5
7	310	84.7	30	63.3	98.3	53.5	81	51.4	78.3	69

$$\beta = \left(X^T X\right)^{-1}\left(X^T Y\right) \tag{2}$$

where Y is the response, X_i and X_j are independent factors, b_0 is an intercept, b_i, b_{ii}, b_{ij} are linear, squared and interaction coefficients, respectively and ε is error. The statistical analysis of CCD was investigated using Design Expert® software version 11.1.0.1 from Statease Inc., USA.

3. Results and discussion

3.1. Characteristics of municipal wastewater

The wastewater was characterized, and turbidity, electrical conductivity, pH, BOD, COD, and TSS were found to be 225 NTU, 2,000 mS cm^{-1}, 7.25, 50, 500, and 400 mg L^{-1} respectively.

3.2. Statistical modeling and process optimization

3.2.1. Effect of pH and coagulant-flocculant dosage on turbidity, COD, and BOD removal using aluminum sulphate as coagulant and polyacrylamide as flocculant

Table 1 shows the CCD matrix designed for independent variables with experimental percentage reduction in turbidity, COD, and BOD using aluminum sulphate as coagulant and polyacrylamide as flocculant. pH 5–7 and coagulant flocculant dosage (110–310 mg L^{-1}) were the independent variables (inputs) and percentage reduction in turbidity, COD and BOD were the dependent variables (responses). All the factors were simultaneously varied to perform 11 experiments for optimization (Table 1). The quadratic equations based on actual factors were obtained to evaluate the effect of each factor on the response:

Table 2 presents an ANOVA for the quadratic model of percentage turbidity reduction. It is noted that the all terms involving coagulant-flocculant dosage were significant with p-value less than 0.05 and the terms involving pH show p-value greater than 0.05. This shows that dosage is more significant than pH for the removal of turbidity [25]. The positive and negative signs of coefficients indicate positive and negative effects, respectively [26]. ANOVA of regression model demonstrates that the model with high Fisher's F-test (247.11) and low p-value (<0.05) is significant. Therefore, the developed model was statistically significant within ±5%.

The ANOVA for the quadratic model of percentage COD reduction is presented in Table 2. It is noted that the

Table 2
Optimal values for reduction in turbidity, COD and BOD by coagulation–flocculation using aluminium sulphate, calcium hydroxide, and combined aluminium sulphate-calcium hydroxide as coagulants and polyacrylamide as flocculant

Coagulant-flocculant	pH	Coagulant-flocculant dosage	% Turbidity reduction	% COD reduction	% BOD reduction
Aluminium sulphate-Polyacrylamide	6	210	99.7 (Experimental) 96.1 (Predicted)	42.5 (Experimental) 45.8 (Predicted)	79.5 (Experimental) 79.3 (Predicted)
Calcium hydroxide-Polyacrylamide	5.5	310	98 (Experimental) 99.8 (Predicted)	60 (Experimental) 64.2 (Predicted)	78.5 (Experimental) 79.9 (Predicted)
Aluminium sulphate-Calcium hydroxide-Polyacrylamide	7	110	95.5 (Experimental) 93.12 (Predicted)	58.1 (Experimental) 54.7 (Predicted)	54.5 (Experimental) 51.3 (Predicted)

Table 3
ANOVA for reduction in turbidity, COD and BOD by coagulation–flocculation using aluminium sulphate as coagulant and polyacrylamide as flocculant

Source	df	% Turbidity reduction				% COD reduction				% BOD reduction			
		SS	MS	*F*-value	*p*-value	SS	MS	*F*-value	*p*-value	SS	MS	*F*-value	*p*-value
Model	5	10166	2033.2	247.11	<0.001	2846.5	569.29	11.347	0.0093	2250.7	450.14	10.092	0.0120
A-pH	1	10.14	10.14	1.2324	0.3174	287.04	287.04	5.7211	0.062242	0.5400	0.5400	0.0121	0.9167
B-Coagulant-flocculant dosage	1	5086.6	5086.6	618.24	<0.001	338.25	338.25	6.7418	0.048455	42.667	42.667	0.9565	0.3729
AB	1	61.622	61.622	7.4896	0.0409	11.902	11.903	0.2372	0.646813	430.56	430.56	9.6526	0.0267
A^2	1	16.918	16.918	2.0562	0.2110	416.09	416.09	8.2931	0.034597	1668.5	1668.5	37.404	0.0017
B^2	1	4786.1	4786.1	581.71	<0.001	2139.2	2139.2	42.638	0.00126	0.7301	0.7301	0.0164	0.9032
Residual	5	41.138	8.2276			250.86	50.172			223.03	44.605		
Lack of Fit	3	41.138	13.712			250.46	83.485	410.58	0.002431	222.62	74.207	364.95	0.0027
Pure Error	2	0	0			0.4067	0.2033			0.4067	0.20333		
Cor Total	10	10207				3097.3				2473.7			

squared term involving coagulant-flocculant dosage was significant with *p*-value less than 0.05 and the other terms show *p*-value greater than 0.05. This shows that coagulant-flocculant dosage is more significant at high concentration than pH for the removal of COD [27]. Finally, the developed model with *p*-value <0.05 shows that the model was statistically significant.

Table 2 presents an ANOVA for the quadratic model of percentage BOD reduction. Like COD, the interactive term, and squared term involving pH was significant with *p*-value less than 0.05 and the other terms show *p*-value greater than 0.05. This shows that pH is more significant than coagulant-flocculant dosage for the removal of BOD [28]. Therefore, the developed model was statistically significant. R^2 value greater than 0.95 confirmed that experimental values fit well with the predicted data (Table 3).

The three-dimensional response surface plots of the regression equations are presented in Fig. 1. The effect of pH and coagulant–flocculant dosage and their interaction on percentage reduction in turbidity, COD and BOD are shown in Figs. 1a–c respectively. Coagulant–flocculant dosage should be optimized for maximum removal of turbidity, COD, and BOD. Turbidity, COD, and BOD removal of 24.5%, 20.5%, and 66.1% were observed at pH of 5 and coagulant-flocculant dosage of 110 mg L^{-1} respectively. As the pH increased to 6, the percentage reduced to 22%, 15.85%, and 68% for turbidity, COD and BOD respectively. The increment of pH from 6 to 7 leads to an increase in removal of turbidity, COD and BOD [29]. The maximum removal of 99.7%, 42.5%, and 79.5% were achieved for turbidity, COD and BOD respectively at optimal values of pH 6 and coagulant-flocculant dosage 210 mg L^{-1}.

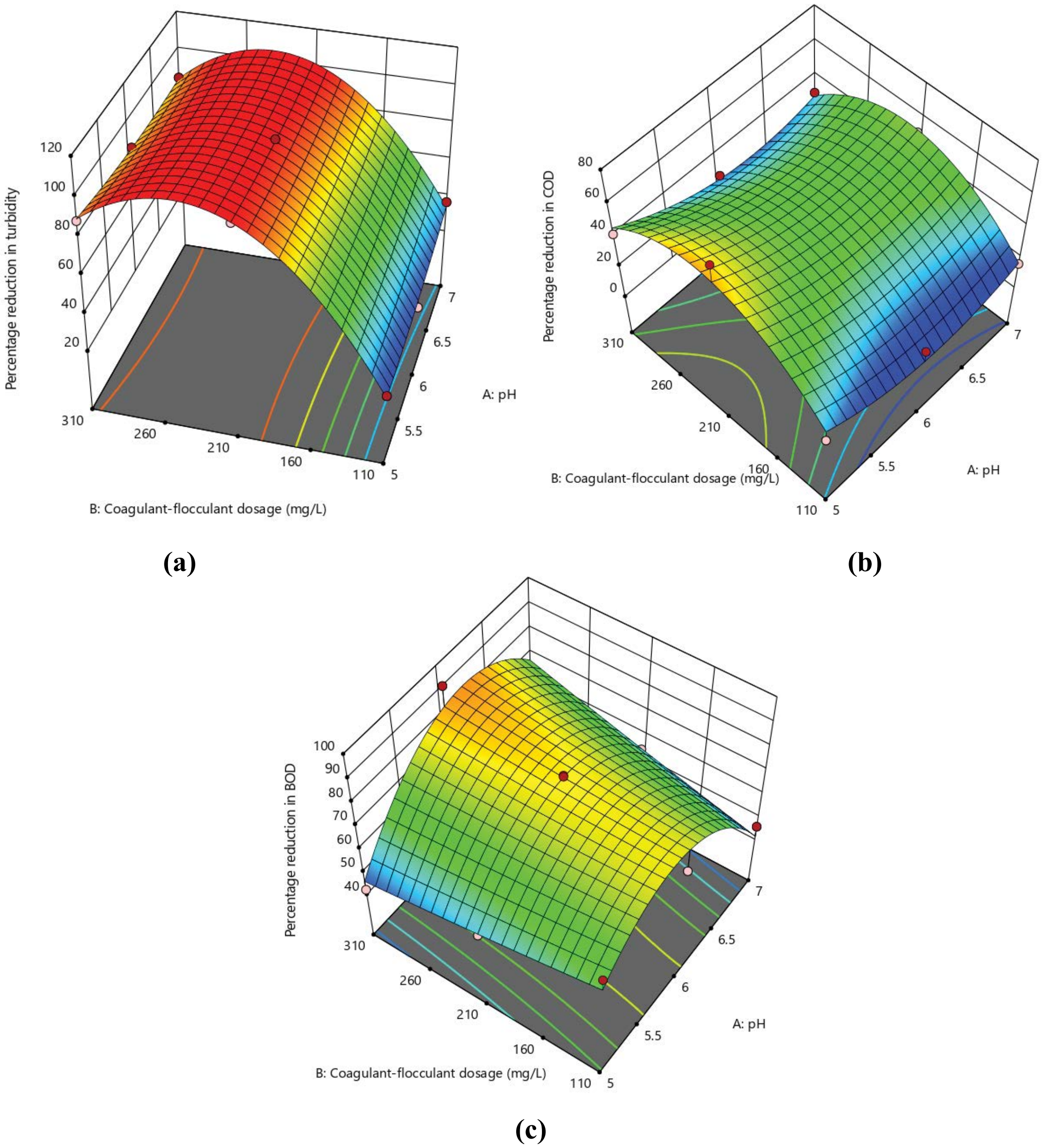
3.2.2. Effect of pH and coagulant-flocculant dosage on turbidity, COD and BOD removal using calcium hydroxide as coagulant and polyacrylamide as flocculant

Table 1 shows the CCD matrix designed for independent variables with experimental percentage reduction in turbidity, COD, and BOD using calcium hydroxide as coagulant and polyacrylamide as flocculant. pH (5–7) and coagulant flocculant dosage (110–310 mg L^{-1}) were the independent variables (inputs) and percentage reduction in turbidity, COD and BOD were the dependent variables (responses). All the factors were simultaneously varied to perform 11 experiments for optimization (Table 1). The quadratic equations based on actual factors were obtained to evaluate the effect of each factor on the response (Table 3).

Table 4 presents an ANOVA for the quadratic model of percentage turbidity reduction. It is noted that the all terms except squared term of pH were significant with *p*-value less than 0.05. This shows that dosage is more significant than pH for the removal of turbidity [30]. Therefore, ANOVA of regression model demonstrates that the model with low *p*-value (<0.05) is significant. The ANOVA for the quadratic model of percentage COD reduction is presented in Table 4. It is noted that the all terms except squared term involving pH were significant with *p*-value less than 0.05. Finally, the developed model with *p*-value <0.05 shows that the model was statistically significant.

Table 4 presents an ANOVA for the quadratic model of percentage BOD reduction. Unlike turbidity and COD, all the terms except linear term of coagulant–flocculant dosage were significant with p-value less than 0.05. This shows that pH is more significant than coagulant–flocculant dosage for the removal of BOD [31]. Therefore, ANOVA of regression model demonstrates that the model is significant. R^2 value greater than 0.98 confirmed that experimental values fit well with the predicted data (Table 3).

The three-dimensional response surface plots of the regression equations are presented in Fig. 2. The effect of pH and coagulant–flocculant dosage and their interaction on percentage reduction in turbidity, COD, and BOD are shown in Figs. 2a–c respectively. Coagulant–flocculant dosage should be optimized for maximum removal of turbidity, COD and BOD. Turbidity, COD and BOD removal of 56.5, 9.76, and 44.9% were observed at pH of 5 and coagulant–flocculant dosage of 110 mg L^{-1} respectively. As the pH increased to 6, the percentage increased to 58.3%, 9% and 96.3% for turbidity, COD and BOD respectively. The increment of pH from 6 to 7 leads to an increase in removal of COD and BOD and decrease in removal of turbidity [32]. The maximum removal of 98%, 60%, and 78.5% were achieved for turbidity, COD and BOD respectively at optimal values of pH 5.5 and coagulant–flocculant dosage 310 mg L^{-1}.



(a) **(b)**

(c)

Fig. 1. Three dimensional surface plots showing the effect of pH and coagulant-flocculant dosage on reduction in (a) turbidity, (b) COD, and (c) BOD by coagulation–flocculation using aluminium sulphate as coagulant and polyacrylamide as flocculant.

Table 4
Regression analysis and model coefficients for reduction in turbidity, COD and BOD by coagulation–flocculation using aluminium sulphate, calcium hydroxide, and combined aluminium sulphate-calcium hydroxide as coagulants and polyacrylamide as flocculant

Coagulant-flocculant	Response	Regression analysis			Model coefficients					
		R^2	Adj. R^2	Pred. R^2	Intercept	A	B	AB	A^2	B^2
Aluminium sulphate-	% turbidity reduction	0.9960	0.9919	0.9647	–118.53	–21.468	2.3522	–0.0393	2.5842	–0.0043
Polyacrylamide	% COD reduction	0.9528	0.9056	0.6589	382.69	–157.08	1.3990	–0.0173	12.816	–0.0029
	% BOD reduction	0.9817	0.9636	0.8505	–718.98	286.47	–0.6184	0.1038	–25.663	$5.37\text{x}10^{-5}$
Calcium hydroxide-	% turbidity reduction	0.9812	0.9623	0.8050	–151.64	55.575	0.7219	0.0715	–6.4158	–0.0021
Polyacrylamide	% COD reduction	0.9987	0.9973	0.9864	–244.43	25.835	2.1270	–0.0619	–1.3658	–0.0036
	% BOD reduction	0.9927	0.9854	0.9240	–311.69	165.16	–1.6768	–0.0860	–11.571	0.0052
Aluminium sulphate-	% turbidity reduction	0.9997	0.9994	0.9979	298.06	–81.156	0.4145	0.0028	6.8079	–0.0015
Calcium hydroxide-	% COD reduction	0.9804	0.9609	0.7991	–829.92	301.17	–0.0553	0.0780	–25.850	–0.0009
Polyacrylamide	% BOD reduction	0.9699	0.9398	0.6950	402.91	–100.27	–1.4031	0.0518	8.8421	0.0026

Table 5
ANOVA for reduction in turbidity, COD and BOD by coagulation–flocculation using calcium hydroxide as coagulant and polyacrylamide as flocculant

Source	df	% Turbidity reduction				% COD reduction				% BOD reduction			
		SS	MS	*F*-value	*p*-value	SS	MS	*F*-value	*p*-value	SS	MS	*F*-value	*p*-value
Model	5	5922.5	1184.5	51.856	<0.001	7549.9	1510.0	24.184	0.0016	7630.0	1526.0	11.630	0.0088
A-pH	1	245.76	245.76	10.759	0.0220	75.331	75.331	1.2065	0.3221	408.38	408.38	3.1124	0.1380
B-Coagulant-flocculant dosage	1	3921.9	3921.9	171.70	<0.001	3737.0	3737.0	59.852	<0.001	0.0417	0.0417	<0.001	0.9865
AB	1	204.49	204.49	8.9523	0.0304	153.02	153.02	2.4507	0.1783	295.84	295.84	2.2547	0.1935
A^2	1	104.28	104.28	4.5652	0.0857	4.7256	4.7256	0.0757	0.7942	339.19	339.19	2.5851	0.1688
B^2	1	1151.1	1151.1	50.392	<0.001	3258.8	3258.8	52.192	<0.001	6910.6	6910.6	52.669	<0.001
Residual	5	114.21	22.842			312.19	62.438			656.04	131.21		
Lack of Fit	3	113.71	37.904	151.61	0.0066	312.00	104.00	1114.3	<0.001	650.87	216.96	83.983	0.0118
Pure Error	2	0.5000	0.2500			0.1867	0.0933			5.1667	2.5833		
Cor Total	10	6036.7				7862.1				8286.0			

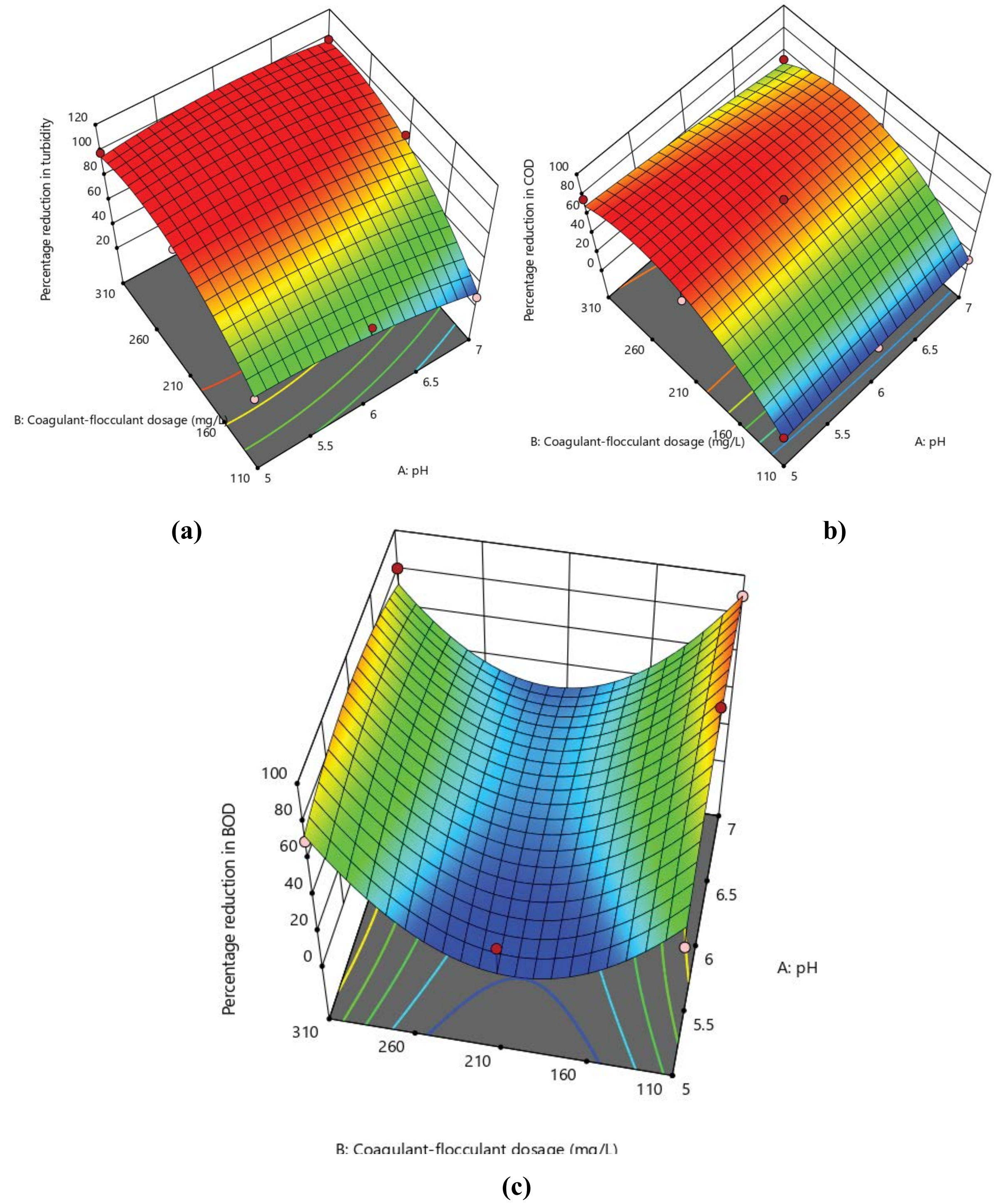


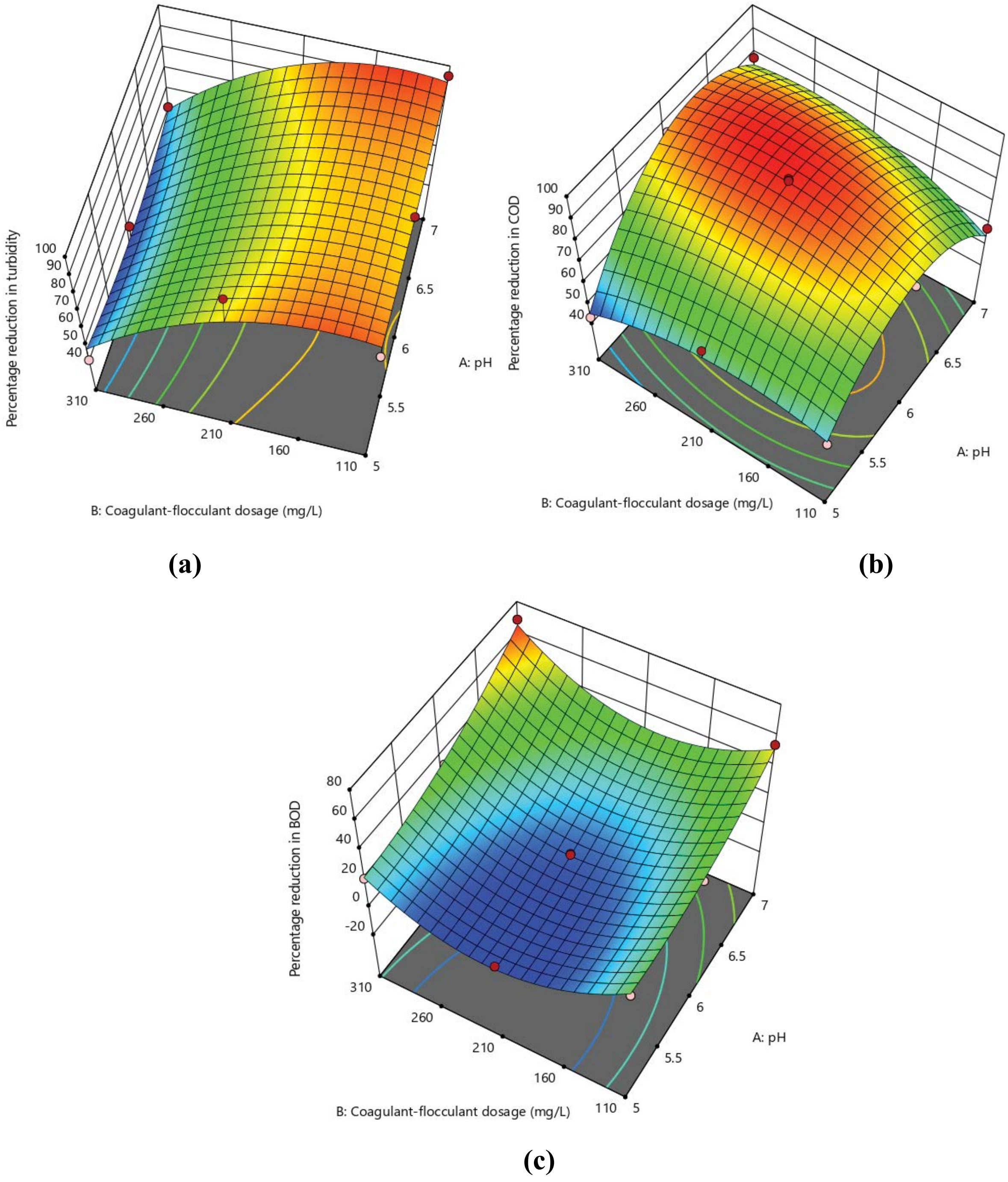
Fig. 2. Three dimensional surface plots showing the effect of pH and coagulant-flocculant dosage on reduction in (a) turbidity, (b) COD, and (c) BOD by coagulation–flocculation using calcium hydroxide as coagulant and polyacrylamide as flocculant.

3.2.3. Effect of pH and coagulant–flocculant dosage on turbidity, COD and BOD removal using combined aluminum sulphate-calcium hydroxide as coagulant and polyacrylamide as flocculant

Table 1 shows the CCD matrix designed for independent variables with experimental percentage reduction in turbidity, COD and BOD using combined aluminum sulphate-calcium hydroxide as coagulant and polyacrylamide as flocculant. pH (5–7) and coagulant flocculant dosage (110–310 mg L^{-1}) were the independent variables (inputs) and percentage reduction in turbidity, COD and BOD were the dependent variables (responses). All the factors were simultaneously varied to perform 11 experiments for optimization

(Table 1). The quadratic equations based on actual factors were obtained to evaluate the effect of each factor on the response:

Table 5 presents an ANOVA for the quadratic model of percentage turbidity reduction. It is noted that the all terms except squared term of pH were significant with p-value less than 0.05. This shows that dosage is more significant than pH for the removal of turbidity [30]. Therefore, ANOVA of regression model demonstrates that the model with low p-value (<0.05) is significant. The ANOVA for the quadratic model of percentage COD reduction is presented in Table 5. It is noted that the all terms were significant with p-value less than 0.05. This shows that both dosage and pH were equally significant for the removal of COD [33–35]. Finally, the developed model with p-value <0.05 shows that the model was statistically significant.



(a)

(b)

(c)

Fig. 3. Three dimensional surface plots showing the effect of pH and coagulant-flocculant dosage on reduction in (a) turbidity, (b) COD, and (c) BOD by coagulation–flocculation using combined aluminium sulphate-calcium hydroxide as coagulant and polyacrylamide as flocculant.

Table 6
ANOVA for reduction in turbidity, COD and BOD by coagulation–flocculation using combined aluminium sulphate-calcium hydroxide as coagulant and polyacrylamide as flocculant

Source	df	% Turbidity reduction				% COD reduction				% BOD reduction			
		SS	MS	*F*-value	*p*-value	SS	MS	*F*-value	*p*-value	SS	MS	*F*-value	*p*-value
Model	5	3414.3	682.86	10.530	0.0109	3008.3	601.66	32.329	<0.001	4260.1	852.02	57.033	<0.001
A-pH	1	7.4817	7.4817	0.1154	0.7479	324.14	324.14	17.417	0.0087	1673.3	1673.3	112.01	<0.001
B-Coagulant-flocculant dosage	1	2786.4	2786.4	42.967	0.0012	41.607	41.607	2.2356	0.1951	19.440	19.440	1.3013	0.3057
AB	1	0.3025	0.3025	0.0047	0.9482	243.36	243.36	13.076	0.0153	107.12	107.12	7.1707	0.0439
A^2	1	117.41	117.41	1.8106	0.2362	1692.8	1692.8	90.960	<0.001	198.06	198.06	13.258	0.0149
B^2	1	600.19	600.19	9.2551	0.0287	214.42	214.42	11.521	0.0193	1771.3	1771.3	118.57	<0.001
Residual	5	324.25	64.850			93.053	18.611			74.694	14.939		
Lack of Fit	3	324.06	108.02	1157.4	<0.001	92.547	30.849	121.77	<0.001	74.374	24.792	154.95	0.0064
Pure Error	2	0.1867	0.0933			0.5067	0.2533			0.3200	0.1600		
Cor Total	10	3738.6				3101.3				4334.8			

Table 5 presents an ANOVA for the quadratic model of percentage BOD reduction. Unlike turbidity and COD, all the terms except linear term of pH were significant with *p*-value less than 0.05. This shows that dosage is more significant than pH for the removal of turbidity [30]. Therefore, ANOVA of regression model demonstrates that the model is significant. R^2 value greater than 0.96 confirmed that experimental values fit well with the predicted data (Table 3).

The three-dimensional response surface plots of the regression equations are presented in Fig. 3. The effect of pH and coagulant–flocculant dosage and their interaction on percentage reduction in turbidity, COD, and BOD are shown in Figs. 3a–c respectively. Coagulant-flocculant dosage should be optimized for maximum removal of turbidity, COD, and BOD. Turbidity, COD, and BOD removal of 85.5%, 54%, and 27.3% were observed at pH of 5 and coagulant-flocculant dosage of 110 mg L^{-1} respectively. As the pH increased to 6, the percentage changed to 87.6%, 78.5%, and 30.1% for turbidity, COD and BOD respectively. The increment of pH from 6 to 7 leads to an increase in removal of COD and BOD and decrease in removal of turbidity [32]. The maximum removal of 95.5%, 58.1%, and 54.5% were achieved for turbidity, COD and BOD respectively at optimal values of pH 7 and coagulant-flocculant dosage 110 mg L^{-1} (Table 6).

4. Conclusion

The pH and coagulant-flocculant dosage were varied at constant flocculant dosage (10 mg L^{-1}), time (1 min for rapid mixing followed by 30 min for slow mixing), temperature (30°C ± 2°C), agitation speed (150 rpm for rapid mixing followed by 50 rpm for slow mixing) and settling time (2 h) to evaluate the percentage removal of turbidity, COD, and BOD by coagulation–flocculation in jar test method. The obtained results indicate that treatment using combined coagulant is effective for removal of turbidity and COD, and could be used for treatment of municipal wastewater.

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