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Response Surface Methodology for Treatment of Paper Mill Wastewater by Using Inorganic Polymeric Coagulant

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ABSTRACT

Inorganic polymeric ferric chloride (POFC) coagulant with proposed structure of $Fe_n\,(Cl_{2.2}OH_{0.8})_n$ is synthesized using waste materials and characterized by XRD. In the current work scrutinized efficiency of POFC for paper mill wastewater (PMW) treatment using response surface methodology (RSM) with central Composite Design (CCD) modeling. Different factors; dose, rapid mixing speed, and rapid mixing time are used for optimize the coagulation process using POFC for treating PMW. The turbidity and chemical oxygen demand (COD) removals are the indicators for assessing POFC efficiency. The obtained result for XRD confirms the production of new material of inorganic polymeric coagulants. Based on RSM modelling, there is a high correlation between the experimental and predicated removals of turbidity and COD. Subsequently, the model is significantly applied for predicating COD and turbidity removals at different operation condition. Conclusively, the obtained results proposed for practical application of POFC coagulant for treatment of paper mill wastewater for COD and turbidity elimination. Furthermore, the applied RSM with CCD is talented model for optimizing treatment of PMW.

Keywords: paper mill wastewater, RSM**,** CCD, POFC, inorganic polymeric coagulant.

INTRODUCTION

The paper industry is considered the third industry for water consumption subsequent to the metal and chemicals industry and discharged large amounts of wastewater (Jaria etal., 2017; Soucy et al., 2014; Adhikari et al., 2015). It produces wastewater quantity 1.5 and 60 m^3 per ton for produced paper (Szolosi, 2003; Thompson et al., 2001). It is recognized that PMW is vastly contaminated by diverse categories of refractory organic contaminants such as adsorbing organic halides (AOXs), as well as residual chemicals generally used during the manufacturing of paper (Kamali and Khodaparast, 2015; Chandra et al., 2018; Kumar et al., 2018). However, the quantity of contaminants PMW effluents is greatly reliant on the paper production technology and the kind of the raw materials used for the production of pulp (Hubbe et al., 2016). The challenges that facing paper industry are protection of human health and environment through efficiently treatment for discharged wastewater or reprocessing of treated wastewater into the production (Pellegrin et al., 1999; Pokhrel and Viraraghavan, 2004; Emiliano et al., 2018). Coagulation/flocculation technique is a coupled effective process used widely as a critical part of the overall treatment of wastewater to decrease turbidity due to the presence of suspended particles (Tatsi et al., 2003; Zhong et al., 2003; Ahmad et al., 2005; Nasser et al., 2006; Sharma et al., 2006; Wong et al., 2006; Yue et al., 2008). From long time, the most common used coagulants are the multivalent inorganic metals salts as alum, ferric chloride and calcium chloride that is due to their low cost and simple application method (Joo et al., 2007). On the contrary, they have some drawbacks, for instance pH adjustment before or after treatment, over dosages use, and their sensitivity to wastewater composition. Moreover, the utilization results have two worse ecological outputs; high sludge production and metal residues in the treated water which may have harmful implication on environment and public health (Flaten, 2001, Lombi et al., 2010).

In the conventional multi-factorial trials, factors optimizing for wastewater treatment conducted via changing one variable with keeping the further factors constant, that was considered time consuming and incompetent of effective optimization (Kim etal., 2003; Wang et al., 2007; Ayodele et al., 2014). Recently, response surface methodology (RSM) with CCD modeling had been employed to optimize the individual factors as well as their interacting effects and understand the performance of complex systems (Shaykhi and Zinatizadeh 2014; Suárez-Escobar et al., 2016). RSM is a procedure for manipulating attempts and assisting investigators to construct models, estimate the influence of numerous variables and optimize the response with lower quantity of trials (Ali et al., 2018; Adel Alaeddini et al., 2013; Mohammed et al., 2015). ANOVA affords the statistical outcomes and investigative confirmatory checks that assess satisfactoriness of the models. Herein, in the current work, new inorganic polymeric ferric chloride (POFC) coagulants was employed for remediation of PMW as cost-effective material, and the central composite design (CCD) incorporated with RSM was used for optimizing the operating parameter of treatment processes; coagulant dose, rapid mixing speed and mixing time.

MATERIALS AND EXPERIMENTAL DESIGN

Preparation and characterization of polymeric coagulant for laboratory experiments

The polymeric coagulant; POFC was prepared via neutralizing 100 ml of iron-containing waste with 100 ml of sodium carbonate according to submitted Egyptian Patent No.-1096/2018 with proposed structure $(Fe_n(OH)_{0.8}\,Cl_{2.2})_n)$ whose XRD is analyzed using PANalytical X-Pert Pro-diffractometer equipped with a diffracted beam monochromator Cu Ka source.

Coagulation/flocculation experiments

The coagulation experiments were conducted using jar test apparatus (JLT6-6, Jar Test, VELP Scientifica Co., Italy) at room temperature. Composite wastewater samples were collected from paper mill factory, Alexandria, Egypt. The factory discharged about 23000 m^3/day and the criteria of wastewater are displayed in Table 1. Five hundred milliliter of paper mill wastewater was used for jar test trials. Under rapid mixing ranged from (1-5) with the set agitation speed ranged from 100 to 300 round per min (rpm), predetermined amount of coagulant of POFC was introduced. Then, the speed was switched to speed of 40 rpm for 10 min. Finally, after quiescent settling of 30 min, the clarified treated wastewater was withdrawn from 2 cm below the surface for analyzing the residual turbidity and COD. The turbidity of the supernatant was determined with a turbidity-meter (Lovibond TB 210 IR, Lovibond Company, Germany) and chemical oxygen demand (COD) was analyzed using APHA (Rice, 2017). The trials are repeated for three times. As well, the analysis is preformed triplicate.

Experimental design

The required number of experimental trial (N_c) to construct the CCD is given by the following equation:

$$N_c = 2^k \text{ factorial point} + 2K \text{ avia; ponts} + 6 \text{ ventral point} \quad (1)$$

where: N_c – trials number;
k – the number of changeable factors which is $K = 3$ in the present study.
Thus, $N = 20$ – the number of runs required for CCD with three changeable factors (Box and Hunter, 1957).

Table 1. Characteristics of industrial papermill wastewater

Parameter	Unit	Value
pH	-	7.3
Turbidity	NTU	2130
Total Suspended Solid (TSS)	mg/L	1588
Biological Oxygen Demand (BOD)	mg/L	1073
Chemical oxygen demand (COD)	mg/L	2094
Oil and grease	mg/L	32

Note: * nephelometric turbidity units (NTU)

Table 2 shows the range of values of the independent natural variables (X_i) and there levels for the CCD. The relation between the code values or the levels ($-\alpha$, −1, 0, 1, α) and the natural values X for the CCD can be obtained from the following set of equations (Napier-Munn, 2000):

$$
\begin{aligned}
-\alpha &= X_{min} \\
-1 &= \left[\frac{X_{max}+X_{min}}{2}\right] - \left[\frac{X_{max}-X_{min}}{2\beta}\right] \\
0 &= \left[\frac{X_{max}+X_{min}}{2}\right] \\
1 &= \left[\frac{X_{max}+X_{min}}{2}\right] + \left[\frac{X_{max}-X_{min}}{2\beta}\right] \\
\alpha &= X_{max}
\end{aligned}
\tag{2}
$$

where: $\beta = 2^{k/4}$, X_{min} and X_{max} are the minimum and maximum natural values, respectively.

Three changeable factors are nominated for investigating their effect on the efficiency, according to Table 2 the changeable factors are: the dose (X_1), rapid mixing speed (rpm) (X_2), and rapid mixing time (X_3).

A second order polynomial is then used for fitting the efficiency y_1 (percentage of turbidity removal) and y_2 (percentage of COD removal) achieved from the trials. The quadratic polynomial is given by Eq. 3:

$$
y_m = \beta_0 + \sum_{i}^{k} \beta_i X_i + \sum_{i}^{k} \beta_i X_i^2 + + \sum_{i=1}^{k} \sum_{j=i+1}^{k} \beta_{ij} X_{ij} + \varepsilon
\tag{3}
$$

where: y_m – the response variable (y_1 or y_2),
X_i – the changeable factors,
$\beta's$ – the unknown coefficients.

The experimental CCD is depicted in Tables 2, where for each set of combination among the changeable factors and the analogous observed response. The experimental detected efficiency values in addition to the values of the changeable factors are then substituted in equation 3 to obtain a set of equations which in matrix form can be written as Eq. 4:

$$
Y = X\beta + \varepsilon
\tag{4}
$$

where: Y is the matrix for the measured efficiency values;
X is the matrix of the input factors;
β is the regression coefficients matrix.

The solution of equation 4 can be obtained by using the method of least square (Ali et al., 2018). The Matlab software is used to perform the multilinear regression of the responses in Y on the input factors in X in order to obtain coefficients of the quadratic equations in the β matrix.

RESULTS AND DISCUSSION

Characteristic for the patent coagulant/flocculent polymer

XRD analysis for inorganic polymeric coagulant POFC

Figure 1 displays the XRD diffraction configuration for different prepared polymeric ferric chloride (POFC) coagulants. XRD of POFC showed diffraction pattern of prepared polymeric coagulant from iron waste. It is noticeable that characteristic diffractive peaks of crystalline material are observed at certain 2 ϴ of 27.9°, 32.2° and 46.02° that are revealed the presence of new other crystals. As shown, the XRD spectra reveals that no existence of $FeCl_3 \cdot H_2O$ and $Na_2(CO_3) \cdot (H_2O)$. While diffraction peaks are related to $FeCl_3 \cdot H_2O$ at 2 ϴ of 17.5° (Cao et al., 2013, Louvain et al., 2013) and $Na_2 (CO_3) \cdot (H_2O)$ at 18.2° and 30° (Ma et al., 2010; Zhao et al., 2018). The existence of XRD diffraction at 2 ϴ of 27.9°, 32.2° and 46.02° in the polymeric coagulant reveals the presence of new other crystals with high intensity indicating non-standard crystalline structure in POFC.

Table 2. Input variables and their levels employed in the 2^4 central composite design

Variables	Range of values and levels for CCD				
	-1.6818	-1	0	1	1.6818
X_1, Dose (g/l)	0.21	0.6	1.2	1.8	2.2
X_2, Rapid mixing speed (rpm)	116	150	200	250	284
X_3, Rapid mixing time (min)	1.32	2	3	4	4.68

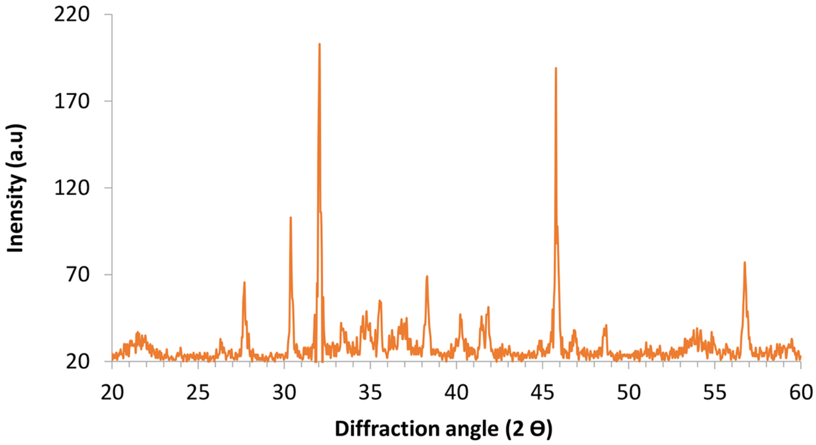

Figure 1. XRD pattern for prepared polymeric coagulant POFC

Meanwhile, the intensities of other peaks are weak. Finally, a new combination has been formed from inorganic polymer coagulants (POFC).

The response surface methodology for optimization turbidity and COD removal from wastewater

Central composite design

Response surface method and the central composite design are employed to obtain the optimal conditions for the input parameters; coagulant dose, rapid mixing speed, and rapid mixing time that maximize the turbidity and COD removal. The experimental and the predicted results are shown in Table 3. Multiple regression analysis of these results gives the following equations (Eq. 5–6) that embody the relationship between the efficiency y_1 (turbidity), and y_2 (COD), and the input factors; x_1 (coagulant dose), x_2 (rapid mixing speed) and x_3 (mixing time):

$$y_1 = 108.342 + 4.843x_1 - 0.1281x_2 - 2.9732x_3 + \\ +0.0119x_1x_2 + 1.2609x_1x_3 - 0.0047x_2x_3 - \\ -3.6473x_1^2 + 0.0003x_2^2 + 0.4467x_3^2 \quad (5)$$

$$y_2 = 65.4054 + 14.8151x_1 - 0.3046x_2 + 25.2404x_3 + \\ +0.0241x_1x_2 - 1.1529x_1x_3 - 0.0487x_2x_3 - \\ -5.9258x_1^2 + 0.0011x_2^2 - 2.6689x_3^2 \quad (6)$$

According to quadric equation for turbidity and COD response, coagulant dose and mixing time has a positive impact on the COD removal %, meanwhile mixing speed has a negative impact one. But only dose has a positive impact on turbidity removal. The statistical significance of the models was performed with ANOVA. The probability (i.e. P value) of the model terms was calculated at 95% confidence level. The ANOVA data are given in Table 4. It was noted that CCD model has high significance with low P-value (P-value of 0.0001) for predicting the turbidity and COD removal rates after coagulation treatment using. Previously, Momeni et al. (2018), investigated the removal of color and turbidity via coagulation treatment process by using RSM model, they found that lack of fit value is not significant and not effectual; this means the used variable in CCD model is adequate for optimization of the coagulation treatment process. Currently, ANOVA analysis for COD removal showed that lack of fit value is highly insignificant; this means the used variables are enough to obtain the maximum COD removal via coagulation treatment of wastewater. Meanwhile, it was found the corresponding value for turbidity is significant. Figure 2 represents a relationship between the observed and the predicted efficiencies for turbidity and COD. It noteworthy well consistence among the observed and the predicted efficiencies with regression coefficients of 0.94 and 0.96 for turbidity and COD removal efficiency, respectively. Moreover, the regression of COD removals indicates that 96% of the variations in the response are due to the model independent variables and only 4% of the variations are not explained by the model. Meanwhile, the regression of turbidity removals indicates only 6% of the variations are not explicated by the model.

Influence of dose and rapid mixing speed on turbidity and COD removals

The variation of the turbidity and COD removals rate from wastewater with POFC dose

Table 3. CCD of the experiment with related the observed and predicted responses

Run	Dose (g/L/10)	Rapid mixing speed (rpm)	Rapid mixing time (min)	Turbidity removal (%)		COD removal (%)	
				Observed	Predicted	Observed	Predicted
1	0.6	150	2	95.01	94.91	77.66	77.95
3	0.6	150	4	94.80	94.43	81.28	80.42
9	0.6	250	2	95.09	94.61	84.13	84.52
10	0.6	250	4	94.19	93.19	78.23	77.26
11	1.8	150	2	94.80	95.39	80.21	80.24
5	1.8	150	4	97.86	97.93	81.28	79.94
6	1.8	250	2	96.56	96.52	89.79	89.70
7	1.8	250	4	98.43	98.13	80.91	79.67
8	0.21	200	3	88.90	89.90	75.20	75.44
2	2.2	200	3	94.70	94.32	78.09	79.22
4	1.2	116	3	98.30	98.00	87.87	88.53
12	1.2	284	3	97.02	97.9	93.14	93.82
13	1.2	200	1.32	96.67	96.50	79.64	78.82
14	1.2	200	4.68	96.68	97.44	70.32	72.48
15	1.2	200	3	95.45	95.71	85.11	83.18
16	1.2	200	3	95.74	95.71	81.91	83.18
17	1.2	200	3	95.81	95.71	83.40	83.18
18	1.2	200	3	95.79	95.71	82.77	83.18
19	1.2	200	3	95.81	95.71	82.98	83.18
20	1.2	200	3	95.78	95.71	83.19	83.18

Table 4. The ANOVA analysis for turbidity and COD removal % of CCD

Parameter	Source of variance	Degree of freedom	Sum of squares	Mean square	F-value	P-value
Turbidity Removal(%)	Model	9.00	70.803	7.867	15.038	0.0001
	Residual	10.00	4.566	0.457	-	-
	Lack of fit	5	4.469	0.894	45.883	0.0004
	Pure error	5	0.097	0.019	-	-
	Total	19.00	75.370	-	-	-
COD Removal(%)	Model	9.00	461.612	51.290	27.938	0.0001
	Residual	10.00	18.358	1.836	-	-
	Lack of fit	5	12.798	2.560	2.302	0.1908
	Pure error	5	5.561	1.112	-	-
	Total	19.00	479.971	-	-	-

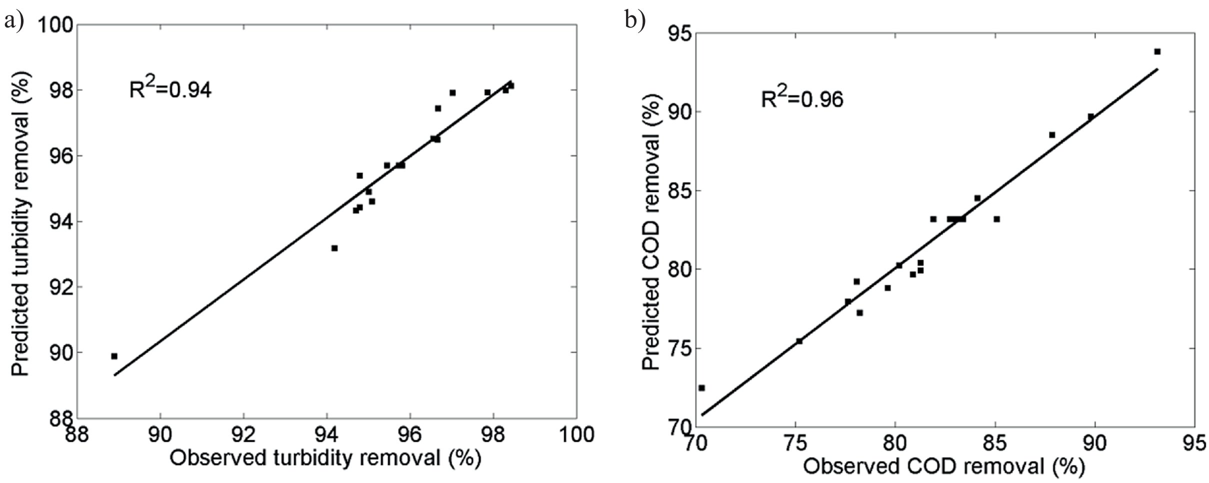

Figure 2. The relation between the experimental and the predicted values of the percentage of turbidity and COD removals

and rapid mixing speed is shown in Figure 3a-d by keeping mixing time at 3 min. The obtained results reveal that turbidity and COD removals increased with increasing rapid mixing speed and POFC dose. The turbidity removal rate recorded a maximum value of 99% at dose of 0.16 g/L and rapid mixing of 300 rpm. The corresponding recorded removal value of COD is 94%. Figures 5b and 5d shows a considerable interaction between the dose and the rapid mixing on turbidity removal and COD removal which helps for optimization coagulation treatment of wastewater. Also, adding lower coagulant dose to wastewater led to minimum COD removal at rapid mixing range (150-250 rpm). It was noted that POFC dose and the fast speed are the detrimental factors for optimizing coagulation treatment of wastewater, where POFC dose enhances sorption and flocs development and also fast mixing of highly turbid wastewater disperses the coagulant and improves collision of particle velocity (Kan et al., 2002; Sheng et al., 2006). Kan et al., 2002 investigated the fast mixing speed influence on turbidity removal with initial 380 NTU using polyaluminum chlorides (PACl). The obtained high turbidity removal of 98% at speed of 350 rpm which is higher than that used in current study with turbidity elimination efficiency of 99% (i.e. the current mean turbidity value in PMW is 2130 NTU).

Influence of dose and rapid mixing time on the removals

The turbidity removal from wastewater against POFC dose and mixing time illustrates in Figure 4 (a-d). The turbidity removal is inversely proportional to mixing time, where and maximum turbidity removal was recorded at the lower mixing time. Meanwhile, the increasing the POFC dose leads to a significant increase in the turbidity removal and maximum turbidity removal ~98% from PMW was achieved at 0.18

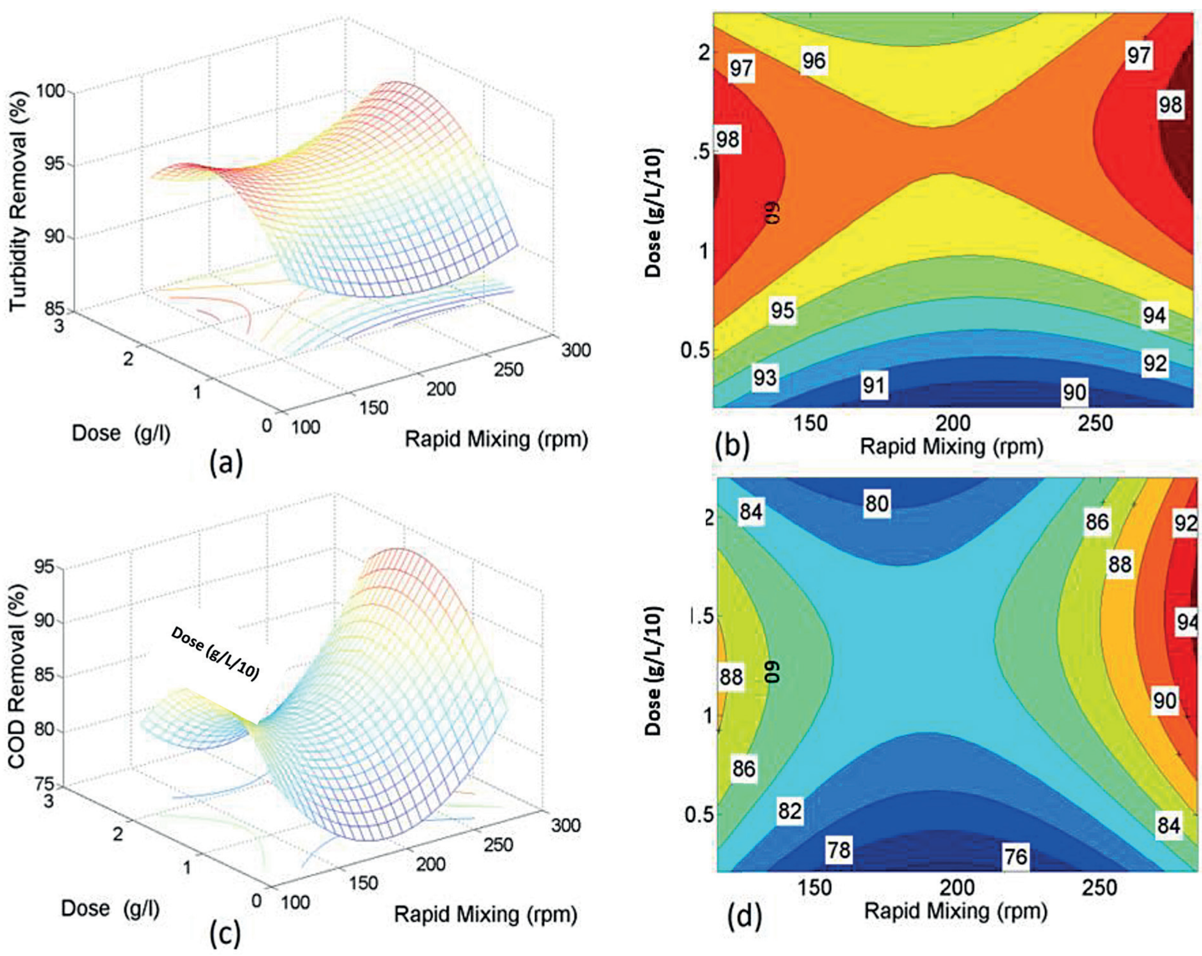

Figure 3. Effect of dose and rapid mixing on the turbidity removal (a, b) and the COD removal (c, d), the rapid mixing time of 3 min

g/L. Lower doses of POFC are not adequate to neutralize the surface charge of suspended particulates. Therefore, the coagulant dose has the highest influence on the turbidity removal. For COD data, removal rate increased with increasing time to 3 min and dose to 0.16 g/L. Figure 4c-d shows well interactive effect between dose and mixing time on COD removal value. The maximum observed COD removal is achieved at mixing time of 1.4 min and POFC dose of 0.14 g/L. The excess of POFC dose declines the coagulation treatment efficiency, owe to restabilize suspended and colloidal particle in solution as well destroying flocs at higher dose of coagulants (Zhang et al., 2004). Also, the optimization of POFC dose will achieve low sludge formation and lower treatment cost.

Influence of mixing speed and mixing time on the removals

The turbidity removal from wastewater as a function of mixing speed and mixing time illustrated in Figure 5a-d. It is observed that, there is lower interaction among rapid mixing time and speed on turbidity removing efficiency. Where, it is inversely proportional to turbidity removal, the maximum value was achieved at high rapid mixing speed with lower mixing time. Meanwhile, the COD removal was increased with increasing rapid mixing speed and lowering mixing time (Figure 5c). There is no clear impact on COD removal due to interaction between mixing speed and mixing time. The optimal COD removal is 95% at rapid mixing of 280 rpm and time of 2 min. as shown in Figure 5d. This finding can be explicated as following; the lowering of mixing time assists the formation of bridging between POFC particle and suspended particulates in wastewater and establishing of flocs. Meanwhile, the similar charged flocs will be repelled with more mixing time that leads to destabilizing flocs and re-stabilizing suspended and colloidal particles in wastewater.

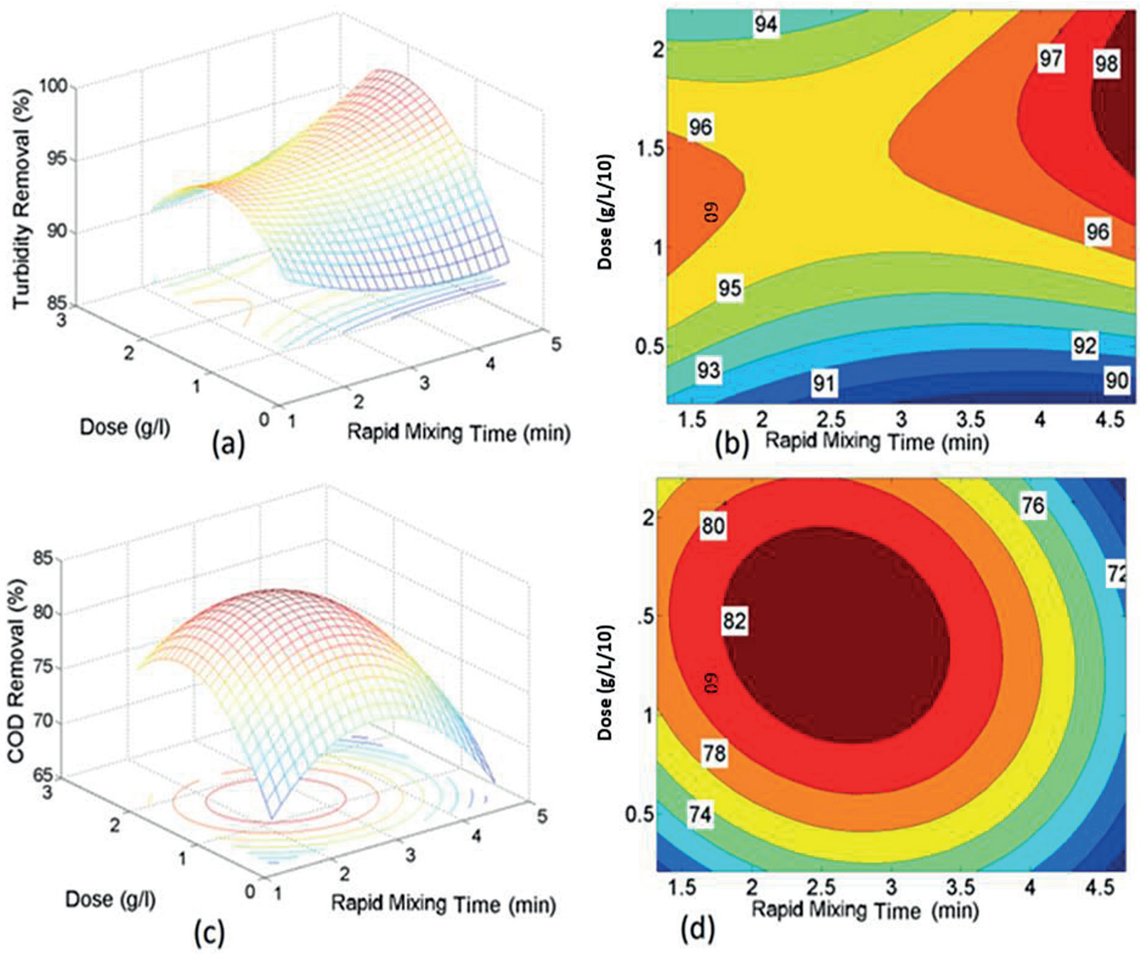

Figure 4. Effect of POFC dose and mixing time on turbidity and COD removal from paper mill wastewater at rapid mixing of 200 rpm

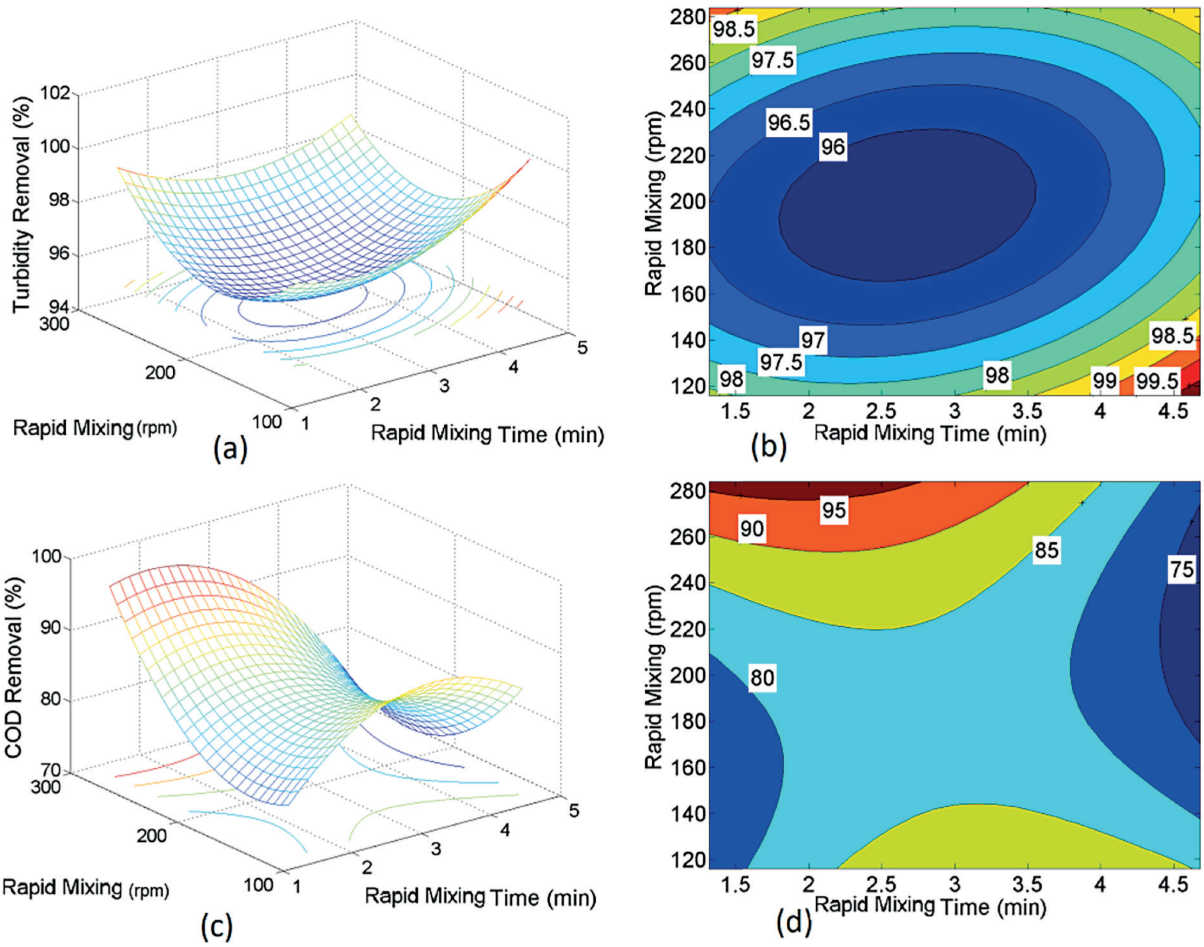

Figure 5. Effect of rapid mixing speed and mixing time on turbidity and COD removal from paper mill wastewater at dose of 0.12 g/l

Global effect of all variables on the turbidity and COD removal

Matlab graphical user interface (GUI) is used for optimization to makes this visualization more perceptive which is shown in Figure 6. While, the optimal value of the individual removal within the range of experimental results can be estimated. As clearly seen, the removal percentage for turbidity or COD is not directly proportional to the input parameters and a careful estimation for the values of the inputs is required to maximize the removals. The optimal predicted values of turbidity and COD removal from PMW after coagulation with POFC are recorded to be 97.5% and 85%, respectively at POFC dose of 0.16 g/L, rapid mixing speed of 280 rpm, and mixing time of 2.2 min. Upon applying the optimal parameters for treatment process the experimental removals of COD and turbidity 86% and 97%, respectively, that are too close model results confirming application of RSM model for optimizing paper mill wastewater treatment using POFC.

Treatment efficiency of paper mill wastewater at optimal condition

The efficiency of chemical coagulation treatment using POFC for paper mill wastewater was estimated at optimal condition of dose of 0.16 g/L, rapid mixing speed of 280 rpm, and mixing time of 2.2 min and depicted in Table 5. The obtained results revealed high removal efficiency for turbidity, TSS, BOD, COD and oil & grease. The removal efficiencies for TSS, and COD from paper mill wastewater recorded 98.4% and 86%, respectively which is much higher than that previously achieved for paper mill treatment using 3.7 g/L of PAC and 35 mg/L of PAM (Kim, 2016).

CONCLUSION

An inorganic polymeric ferric chloride coagulant was prepared from waste material and employed for treatment of PMW. RSM with central composite design was effectively developed to

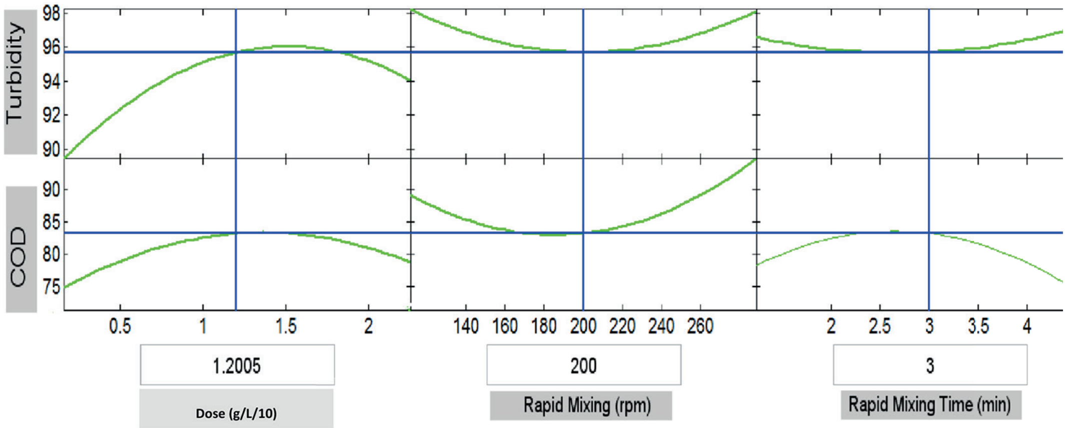

Figure 6. Graphical user interface for optimizing investigated factors

Table 5. Characteristic of treated effluent after chemical coagulation using POFC

Parameter	Unit	Treated effluent	R %
pH	-	7.2	-
Turbidity	mg/L	19	99
Total suspended solid (TSS)	mg/L	25	98.4
Biological oxygen demand (BOD)	mg/L	152	85.8
Chemical oxygen demand (COD)	mg/L	290	86
Oil and grease	mg/L	< 2	100
Residual Iron	mg/L	0.42	-

attain the optimal circumstances for coagulation treatment process of PMW; POFC dose, mixing time, and rapid mixing speed. The results revealed that the studied factors have a significant impact on turbidity and COD elimination efficiency. The ANOVA analysis proved that the applied model is highly noteworthy for prediction of turbidity and COD elimination from wastewater after coagulation treatment using POFC. At optimum treatment conditions; POFC dose of 0.12 g/L, rapid mixing of 280 rpm, and mixing time of 2.2 min, the highest removal percentages with POFC coagulant are 85% for COD removal and 97.5% for turbidity removal, which are close with experimental data; 86% for COD and 99% for turbidity. Also, coagulation and flocculation method was powerfully optimized by RSM with CCD model. Consequently, POFC is promising proper coagulant for COD, TSS and turbidity removal from industrial PMW.

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