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Multi-response optimization of coagulation–flocculation process for stabilized landfill leachate treatment using a coagulant based on an industrial effluent

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ABSTRACT

The aim of this study is to assess the performance of an industrial effluent steel industrial wastewater (SIWW) highly loaded with ferric salts from Maghreb Steel Company as coagulant for stabilized landfill leachate treatment. Response surface methodology and central composite design were applied to optimize the coagulation–flocculation process and to model the relationships between independent variables (coagulant dose and effluent pH) and responses (chemical oxygen demand (COD) and turbidity removal, and sludge production). Quadratic polynomial models developed for these responses indicated that the optimum conditions were 8 mL/L of SIWW at pH 2.75. These results showed good agreement between experimental and model predictions. 55.43% and 87.55% of COD and turbidity removal, respectively and with 19 mL/L of sludge production from treatment of this leachate by coagulation–flocculation using a SIWW-based coagulant were demonstrated. Accordingly, it is concluded that SIWW can be used as an effective and alternative coagulant for the pre-treatment of stabilized landfill leachate by a coagulation–flocculation process.

Keywords: Stabilized landfill leachate; Coagulation–flocculation; Response surface methodology; Steel industrial wastewater; Multi-response optimization

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1. Introduction

According to Morocco's Ministry of Environment, the current production of municipal solid waste in Morocco is estimated at more than 6.31 million tonnes per year [1]. During storage and under the combined action of rainwater and natural fermentation, these wastes produce over 800.000 m^3 of landfill leachates [2]. Indeed, leachate is the residual liquid generated by the percolation of water and liquids through a storage area for waste, chemicals or simply soil contaminated with pollutants [3]. This product of the dissolution of organic matter and trace elements (heavy metals, organic and chemical pollutants, radionuclides, etc.) is a source of air, soil and water pollution, and more particularly the groundwater [4–7].

In Morocco, since the publication of Solid Waste Management Law 28-00 in November 2006, the direct discharge of leachate has become prohibited [8]. Subsequently, these discharges must be carefully collected and treated before being discharged into the natural environment.

Unlike wastewater, there is no standard treatment for landfill leachates. Landfill leachate treatment solutions are often assessed on a case-by-case basis. Even if the techniques used derive for the most part from wastewater treatment, leachates are very complex effluents whose treatment requires special expertise. The size of the site, the landfill conditions, the nature and age of the waste, as well as the weather conditions all influence the composition and quantity of leachate produced [9].

In order to define the most suitable landfill leachate treatment plants, studies carried out upstream are therefore essential to know the concentrations and volumes of leachate to be treated [10]. Indeed, even after years of landfill closure, landfill leachate continues to form due to the slow natural processes of municipal solid waste biodegradation in landfills. Thus, we can classify the leachate into three categories according to its age, namely, young (<5 y), medium or intermediate (5–10 y) and old or stabilized (>10 y). Young municipal landfill leachates are characterised by low pH levels and high concentrations of easily degradable organic matter such as volatile fatty acids [11].

In mature municipal landfills, their leachates are characterised by high pH and hardly biodegradable organic matters, mainly humic and fulvic fractions [12]. The biochemical oxygen demand (BOD)/chemical oxygen demand (COD) ratio in the mature landfill leachate decreases over time because it is the non-biodegradable part of the COD that remains largely. Therefore, biological treatments are only possible for young and intermediate landfill leachates [13]. Whereas, stabilized leachates are difficult to treat biologically due to a low BOD/COD ratio.

Indeed, physico-chemical processes seem better adapted for stabilized landfill leachate treatment such as adsorption [14], ion-exchange [15], advanced oxidation processes [16], membrane technology [17], ammonia stripping [18], ozonation [19], nanofiltration [20], reverse osmosis [21], microwave oxidation [22], struvite precipitation [23], electrochemical [24] and coagulation–flocculation [25].

Generally, coagulation–flocculation is an excellent process for both pre-treatment and post-treatment of stabilized landfill leachate, which can effectively remove refractory pollutants, such as heavy metals and non-biodegradable dissolved organic matter, and subsequently improve the biodegradability of this leachate [26]. This process implicates the destabilization of colloidal particles brought about by the addition of coagulants, leading to the agglomeration of destabilized particles into micro floc and after into bulky floccules easily separable. Among the most widely used coagulants are mineral-based coagulants, such as alum [27–28], polyaluminium chloride [29–30], lime [31–32], ferric [33–34] and polyferric salts [35–36]. In addition, many factors can influence on the performance of the coagulation–flocculation process such as type and dose of coagulant/flocculant, effluent pH and mixing speed/time [37]. In most studies, optimization of these performances was achieved via conventional method by varying one factor while keeping all other factors unchanged. Unlike classical methodology, the response surface methodology allows to highlight all independent variables and their interactions at the same time [38].

Recently, we have shown that steel industrial wastewater (SIWW) as excellent coagulant for treatment of domestic and industrial wastewater effluents [39–41]. Following on from our ongoing program to develop wastewater treatment processes, we describe in this paper the use of SIWW as a new coagulant for stabilized landfill leachate treatment. The optimization of key operating parameters for the coagulation–flocculation process was also investigated using the response surface methodology.

2. Materials and methods

2.1. Leachate sampling and characterization

Leachate samples were collected from Mesbahait closed landfill site (MCL) [42]. This landfill site is located in Mohammedia City, approximately 24 km to the northeast of Casablanca City, Morocco at latitude 33°42′00″N and longitude 007°23′00″W. Moreover, Mohammedia City enjoys a Mediterranean climate characterized by mild, wet winters and hot, dry summers. Its proximity to the Atlantic Ocean tends to cool the city in summer and warm it in winter. It benefits greatly from the sun all year round. The average precipitation in Mohammedia is approximately 432 mm/y.

The MCL has a total area of 6.5 ha. It is an old quarry of limestone characterized by schists and representing fissures. It also has been operating for 25 y since 1987. The site was fully closed in 2012. During operation, MCL received approximately 175 tons of solid waste per day. In total, over 1.5 million tons of these solid wastes are landfilled by simply piling them up at the bottom of the landfill site and form a front of about 3 m. In the lower part of MCL, the leachate flows by gravity with a high flow rate, thus generating a lake of about 30 m in diameter. After the closure of MCL since 2012, these landfill leachates were reserved in stabilization ponds for natural treatment.

Leachate samples were manually collected from the ponds of MCL and placed in 50 L polyethylene containers. The samples were immediately transported to the laboratory and stored at 4°C. These samples were placed at room temperature for 2 h before analysed. The pH, COD, five-day biochemical oxygen demand (BOD_5), turbidity, conductivity and ammoniacal nitrogen, were determined for each sample. All the analytical procedures were performed according to the standard methods for the determination of water and wastewater [43].

The main physico-chemical characteristics of the MCL landfill leachate compared to the Moroccan Wastewater Discharge Standards are presented in Table 1.

From this table, we see that this landfill leachate is a dark colour, highly turbid liquid, with pH higher than 7, with also high concentration of ammoniacal nitrogen in the range of 632–951 mg/L, BOD_5 of 228–411 mg/L and COD of 2,153–2,707 mg/L. These concentrations are not tolerated by Moroccan Wastewater Discharge Standards.

Indeed, the brown color of this landfill leachate was due to the presence of humic substances which are generally non-biodegradable. Moreover, the BOD_5/COD ratio is less than 0.1, which indicates that this leachate is not easily biodegradable. However, the high concentration of ammonia causes the dysfunction of natural biological leachate treatment processes.

According to its characteristics, the effluent of this closed landfill was classified as a stabilized leachate. This correlates with what has been reported in the literature [44–45].

In our case, biological methods are not recommended for the treatment of this landfill leachate. Therefore, they should be more likely to be treated by physico-chemical techniques, such as coagulation–flocculation.

2.2. Experimental procedure

Coagulation–flocculation experiments were performed using a conventional jar-test apparatus equipped with six beakers and six rectangular blade propellers to mix leachate sample and coagulant.

In this study, the variables investigated were coagulant dose and effluent pH. Hydrochloric acid (HCl, 37%) and sodium hydrate (NaOH, 1 M) were used to adjust the pH of the leachate samples.

The experimental process consisted of three successive stages. The sample was mixed rapidly at 200 rpm for 2 min followed by slow mixing at 40 rpm for 15 min. The time and speed for rapid and slow mixing were set with an automatic controller. After mixing process, the flocs were left untouched to allow settling for 30 min.

Afterwards, the sludge volume was measured and then the supernatant was withdrawn from the beaker to analyze the leachate pollutants, namely COD and turbidity.

The removal of the studied parameters from leachate was calculated based on the following equation:

$$\text{Removal}\left(\%\right)=\frac{C_i-C_f}{C_i}\times 100 \tag{1}$$

where C_i and C_f are the initial and final concentrations of leachate pollutants, respectively.

2.3. SIWW-based coagulant

The SIWW was collected from Maghreb Steel Company, located in Casablanca, Morocco. Indeed, Maghreb Steel is a leading Moroccan company in the steel industry with a total production capacity exceeding 3 million tonnes of steel per year, which also resulted in high flow rate of SIWW. The physicochemical characterization of SIWW is reported in Table 2.

From this table, we notice that SIWW is an industrial effluent highly loaded with ferric salts. Consequently, our research group has shown that SIWW as excellent coagulant for treatment of domestic and industrial wastewater effluents [46]. In this study, SIWW will also be used as a coagulant in stabilized landfill leachate treatment.

2.4. Response surface methodology

Generally, response surface methodology (RSM) was used for analysis, modelling and process optimization [47–48]. Indeed, the central composite design (CCD) is the most widely used model for fitting RSM [49]. A central composite rotatable design for k independent variables was employed to design the experiments in which the variance of the predicted response, Y, at some points of independent variables, X, is only a function of the distance from the point to the design centre. The design of experiment is intended to reduce the number of experiments and to arrange the experiments with various combinations of independent variables. In the rotatable design, the standard error, which depends on the coordinates of the point on the response surface at which Y is evaluated and, on the coefficients, is the same for all points that are the same distance from the central point. These designs consist of a $2k$ factorial then augmented by 2^*k axial points and 2 centre points. The method of least squares for multiple regression was used to investigate the relationship between the independent and dependent variables. The results of this experimental design were fitted using a second-order polynomial equation in order to correlate the response to the independent variables.

The general model for predicting optimal conditions was expressed as follows:

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

where the predicted response is represented by Y; the coefficients of the intercept, linear, quadratic and interaction terms among independent factors (X) are indicated respectively by β_0, β_i, β_{ii} and β_{ij}; and the model error is denoted by e.

In this study, the results of this experimental design were analyzed and interpreted by Statgraphics-Plus statistical software [50]. The desirability function approach has been used to simultaneously optimize multiple response processes.

3. Results and discussion

3.1. Statistical analysis

The central composite design was used in this study to optimize the most influential operating variables of the coagulation–flocculation process, namely coagulant dose (X_1) and effluent pH (X_2). While the COD removal (Y_1), turbidity

Table 1
Characteristics of landfill leachate and Moroccan Wastewater Discharge Standards

Parameters	Unit	Range	Mean	Discharge Standards
pH	–	7.8–8.9	8.3	5.5–9.5
COD	mg/L	2,153–2,707	2,478	500
BOD_5	mg/L	228–411	236	100
NH_3–N	mg/L	632–951	723	–
Turbidity	NTU	62.9–140	117.9	–
Conductivity	mS/cm	25.6–35.9	31.13	2.7

Table 2
Characteristics of SIWW

Parameters	Unit	Value
pH	–	<1
Fe^{3+}	g/L	101.3
Cu^{2+}	mg/L	1.145
Conductivity	mS/cm	20.2

Table 4
Experimental design and results

Runs	Independent variables		Responses		
	X_1	X_2	Y_1	Y_2	Y_3
1	+1	–1	36.13	77.68	29
2	+1	+1	39.32	72.94	82
3	–1	–1	35.68	63.82	46
4	–1	+1	34.88	61.15	59
5	+1.4142	0	31.74	87.22	61
6	–1.4142	0	28.29	69.07	57
7	0	+1.4142	43.85	56.96	72
8	0	–1.4142	42.16	62.28	30
9	0	0	56.04	88.12	22
10	0	0	55.96	87.87	22

removal (Y_2) and sludge production (Y_3) values were chosen as the response variables. The input and output variables as well as the range and levels of the independent variables are reported in Table 3.

The range and levels of independent variables were determined from preliminary tests and by various investigations of stabilized landfill leachate treatment by coagulation–flocculation. The design matrix with ten experiments for 2 independent variables that were carried out and their 3 responses notified, shown in Table 4.

Five levels for each factor were selected according to this design matrix data to investigate the effects of factors and to locate the optimal conditions. The relationship between independent variables and response was drawn by second-order polynomial equation. The regression equation coefficients were calculated by least squares method. The second-order models for the decolourization efficiency and COD removal of dye from textile effluent in terms of coded variables are shown in Eqs. (3)–(5), respectively.

$$Y_1 = 56 + 1.207X_1 + 0.604X_2 - 13X_1^2 - 6.5X_2^2 + X_1X_2 \quad (3)$$

$$Y_2 = 88 + 6.432X_1 + 1.884X_2 - 4.938X_1^2 - 14.188X_2^2 - 0.5X_1X_2 \quad (4)$$

$$Y_3 = 22 + 1.457X_1 + 15.675X_2 + 18.25X_1^2 + 14.25X_2^2 + 10X_1X_2 \quad (5)$$

Generally, positive sign in front of the terms indicates synergistic effect, whereas negative sign indicates antagonistic effect. The quality of the model developed was evaluated based on the correlation coefficient value.

The *R* value for Eqs. (2)–(4) was 0.9997, 0.9999 and 0.9991, respectively. All these *R* values obtained were relatively high, indicating that there was a good agreement between the experimental and the predicted values from

Table 3
Input and output variables

	Independent variables				
	Natural variables (X_i)	Unit	Levels of coded variables X_i		
Input variables			Low (–1)	Center (0)	High (+1)
	X_1 = Coagulant dose	mL/L	6	8	10
	X_2 = Effluent pH	–	2	3	4
	Responses				
Output variable	Y_1 = Chemical oxygen demand removal (%) Y_2 = Turbidity removal (%) Y_3 = Sludge production (mL/L)				

the models. The R^2 values for these equations were 0.9995, 0.9998 and 0.9982, respectively. This indicated that 99.95%, 99.98% and 99.82% of the total variation in the removal of COD, turbidity and sludge production, respectively, was attributed to the experimental variables studied.

Indeed, the adequacy of these models was further justified through analysis of variance (ANOVA), as shown in Table 5.

From this table, we notice that the model F-value was greater than the critical value [$F_{0.01}(5,4) = 15.52$] at 1% level of significance, which implies that the regression is globally significant at a confidence level of 99% for all these second-order polynomial equations. In addition, linear terms (X_1 and X_2), squared terms (X_1X_1 and X_2X_2), and interaction terms (X_1X_2) were significant model terms greater than or equal to 95% at a confidence level for each response.

According to these statistical results obtained, it was shown that the above models were adequate to predict these responses by coagulation–flocculation process in stabilized landfill leachate using a SIWW-based coagulant within the range of variables studied.

3.2. Process analysis

3.2.1. COD removal

The influence of each independent variable on the COD removal efficiency by coagulation–flocculation using a SIWW-based coagulant for the stabilized treatment of landfill leachate is shown in Fig. 1.

From this figure, it can be seen that the dose of coagulant (X_1) and the pH of the effluent (X_2) have a positive effect on the efficiency of removing COD from the stabilized landfill leachate. However, looking at this figure we see that they have a negative effect in the positive area, which can be explained by the negative effect of the interaction of the squared term. Whereas, the positivity of the two independent variables was well reflected on the positive interaction between coagulant dose and effluent pH.

Contour plots and response surface plots were also prepared for the purpose of evaluated the independent variables on the COD removal, as shown in Fig. 2.

The geometric plots are approximately symmetrical in shape with circular contours showing clear peaks, involving that the optimal conditions for maximum response values are assigned to pH and dosage in the design space. The two-dimensional geometric representation of the responses on the dosage-pH level (contour plot) shows concentrically closed curves whose centers represent the optimal conditions.

From these response surface plots, we can conclude that when the coagulant dose and the effluent pH increase individually or simultaneously, the COD removal efficiency increases to an optimum at about 56% and then decreases in the other side of the experimental study area, which agrees with findings of Ghafari et al. for an old leachate [51].

3.2.2. Turbidity removal

Practically, the coagulation–flocculation process is a very suitable method for wastewater turbidity removal. Fig. 3 illustrates the effect of coagulant dose and effluent pH on the turbidity removal efficiency by coagulation–flocculation using a SIWW-based coagulant for treatment of stabilized landfill leachate.

This figure shows that in general the coagulant dose has a positive effect on the efficiency of turbidity removal. The effluent pH also has a positive effect on this response and then it becomes negative from the center of the study area, which can be explained by the negative effect of the

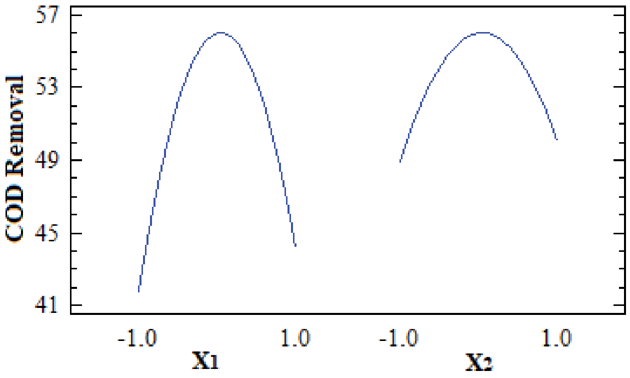

Fig. 1. Main effects plot for COD removal.

Table 5
Analysis of variance

Response	Terms	Sum of squares	Df	Mean square	F_{exp}	Significance test
	Regression	795.971044	5	159.1942088	1,484.48	***
Y_1	Residue	0.428956	4	0.107239	–	
	Total	796.4	9	–	–	
	Regression	1,287.829806	5	257.5659612	3,813.05	***
Y_2	Residue	0.270194	4	0.0675486	–	
	Total	1,288.1	9	–	–	
	Regression	4,136.53378	5	827.306756	443.23	***
Y_3	Residue	7.46622	4	1.86655	–	
	Total	4,144.0	9	–	–	

***$p \leq 0.01$; **$p \leq 0.025$; *$p \leq 0.05$; NS: No significant.

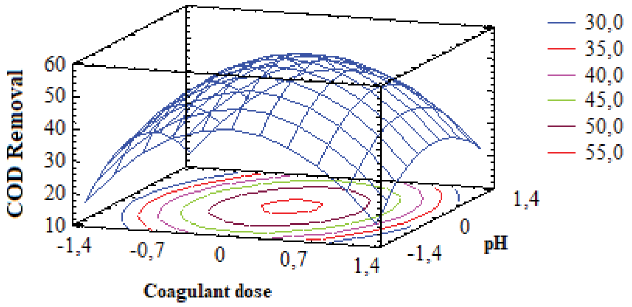

Fig. 2. Response surfaces for COD removal.

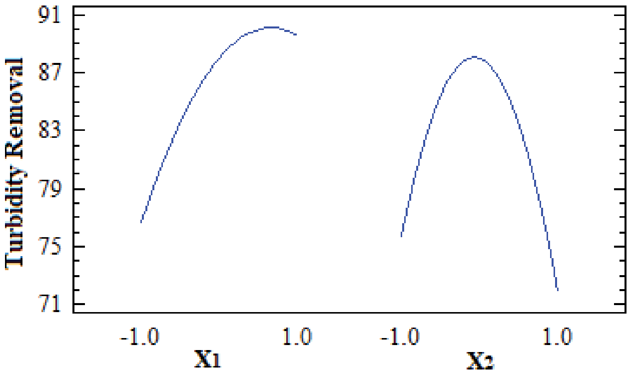

Fig. 3. Main effects plot for turbidity removal.

interaction of the squared term. While, the opposite signs of the both independent variables were well reflected on the negative interaction between the dose of coagulant and the pH of the effluent.

Interaction between independent variables on the turbidity removal from old landfill leachate by coagulation–flocculation treatment is shown in Fig. 4.

From these response surface plots, we can conclude that when the coagulant dose increases, the turbidity removal efficiency increases until an optimum is obtained at 90% which remains almost unchanged in the experimental study area. On the contrary, for interactions involving the pH of the effluent, obtaining this optimum is followed by a remarkable decrease in the turbidity removal efficiency from stabilized landfill leachate. A similar effect of these independent variables on turbidity removal was observed in findings of Lessoued et al. for an old leachate [52].

3.2.3. Sludge production

To choose of a suitable coagulant, it is necessary to take into account, on the one hand, the efficiency of pollution removal, and on the other hand, the cost of treatment, which should be as low as possible. However, the sludge production may affect the economic feasibility of the proposed method.

The contribution of independent variables and their interaction on sludge production during the treatment of stabilized leachate by coagulation–flocculation using a SIWW-based coagulant are illustrated in Figs. 5 and 6.

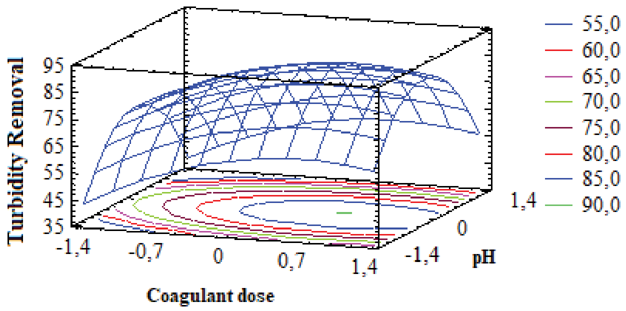

Fig. 4. Response surfaces for turbidity removal.

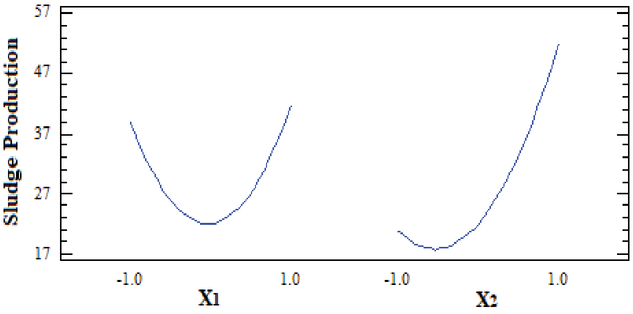

Fig. 5. Main effects plot for sludge production.

Fig. 5 indicates that the coagulant dose and the effluent pH have a positive effect on the sludge production in the positive domain while the reverse is reported in the negative zone of the experimental study area.

It can also be concluded from Fig. 6 that when the dose of coagulant and the pH of the effluent increase individually or simultaneously, the sludge production decreases to a minimum towards 18 mL/L and then increases on the other side of the experimental range.

According to these results, SIWW has the advantage of generating low sludge production compared to conventional coagulants as reported by Assou et al. [53].

3.3. Process optimization

In order to optimize the stabilized landfill leachate treatment by coagulation–flocculation processes using a SIWW-based coagulant, we established by CCD three models, that is, three responses with different objectives but with the same independent variables.

For this, it is obligatory to adapt an acceptable compromise between these three models by using the desirability function as shown in Fig. 7.

As shown in Fig. 7, the optimum conditions of COD and turbidity responses can be identified by superimposing their contours in an overlay plot in which the optimum area is easily localized. The optimization criteria for chosen responses as reported in Table 6.

These constraints were chosen relatively close to the maximum removal efficiencies of leachate pollutants and the minimum production of sludge in order to obtain a moderately precise optimum zone.

Table 6
Optimum conditions

Responses		Limits			Optimum		
		Y_1	Y_2	Y_3	Y_1	Y_2	Y_3
		>55	>85	<20	55.43	87.55	19
Independent variables	X_1	[–0.16; 0.23]			0		
	X_2	[–0.35; -0.16]			–0.25		

Table 7
Comparison with other coagulants

Coagulant	Coagulant dose	Effluent pH	% COD removal	Reference
$FeCl_3$	0.53 g/L Fe	6	45	[54]
Alum	2.5 g/L	6	27	[54]
PAC	2 g/L	7	43	[55]
SIWW	8 mL/L	2.75	56	This study

According to Fig. 7 and Table 6, the optimal conditions for the treatment of stabilized landfill leachate by coagulation–flocculation processes using a SIWW-based coagulant were obtained at pH = 2.75 and coagulant dose = 8 mL/L of SIWW. The predicted response values are 55.43%, 87.55% and 19 mL/L for COD removal, turbidity reduction and sludge production, respectively. The experimental checking in these optimal conditions, confirms that these results have a good reproducibility of the proposed models in the experimental range of this study.

The COD removal from stabilized landfill leachate obtained with SIWW-based coagulant were compared with those of other coagulants cited in the literature (Table 7).

In fact, the SIWW-based coagulant can remove up to 56% of COD from stabilized landfill leachate. Thus, with 0.53 g/L Fe of $FeCl_3$ and with 2.5 g/L of alum, 45% and 27% COD removal from stabilized landfill leachate, respectively, were found by Aziz et al. [54]. Ghafari et al. [55] demonstrates that polyaluminium chloride (PAC) can remove up to 43% of COD from stabilized landfill leachate.

These results are agreed with findings reported by Amokrane et al. [56] using conventional coagulants for pre-treatment of stabilized landfill leachate by a coagulation–flocculation process.

4. Conclusions

In summary, an industrial effluent (SIWW) highly loaded with ferric chloride from Maghreb Steel Company was used as coagulant for stabilized landfill leachate treatment. Response surface methodology (RSM) was used for the analysis, modelling and optimization of independent variables influencing the coagulation–flocculation process using a central composite design with multiple responses.

The optimum conditions obtained for stabilized landfill leachate treatment by coagulation–flocculation were at pH 2.75 and 8 mL/L of SIWW. The model predictions are in good agreement with experimental data. Under these optimal conditions, 56% and 88% of COD and turbidity removal, respectively and with 20 mL/L of sludge production from treatment of this leachate by coagulation–flocculation were archived. The advantages of the proposed coagulation in addition to pollution removal, process using SIWW were mainly, simplicity, no cost, and easy onsite implementation.

Therefore, this present study reveals that SIWW can be used as an efficient and alternative coagulant for the pre-treatment of stabilized landfill leachate by a coagulation–flocculation process.

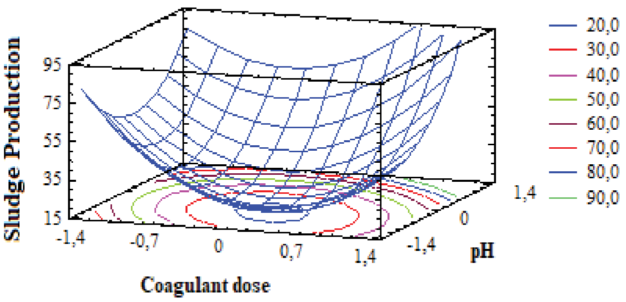

Fig. 6. Response surfaces for sludge production.

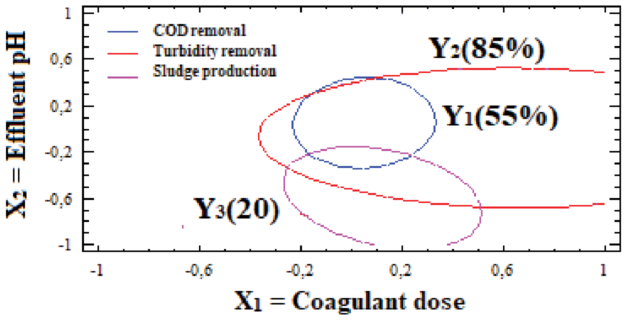

Fig. 7. Overlay multiple response plots for optimal region.

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