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Removal of phenol and colour of leachate of municipal solid waste by physico-chemical treatment using a liquid waste as a coagulant

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Abstract

A coagulation flocculation process combined with a Steel Industry Wastewater rich in ferric chloride (SIWW) and polymer were used to treat fresh leachate from municipal solid waste of the city Mohammedia. A central composite experimental design and response surface methodology were employed to evaluate and optimize the reagents dosage and to achieve a balance between efficiency and operational costs. The influence of pH was also evaluated to determine the most suitable pH condition. The best regression coefficients (R^2) were obtained for phenol removal, colour removal and decanted sludge, with values of 0.89, 0.93 and 0.92 respectively. The most significant factors in the analysis of variance (ANOVA) in this study were pH and SIWW dosage. However, flocculant dosage was not most significant factor. Multiple response optimizations fits the optimum values of the factors and the responses as 40 $mL.L^{-1}$ of coagulant, 12 $mL.L^{-1}$ of polymer and 91% of phenol removal, 70% of colour removal and 38 $mL.L^{-1}$ of decanted sludge at pH=6.

Key words: Municipal Solid Waste; Wastewater valorization; physicochemical process; Fresh leachate; Response surface methodology; central composite experimental design.

1. Introduction

Landfilling is one of the least expensive methods for disposal of solid waste. It is reported that about 90% of municipal solid waste (MSW) is disposed in open dumps and landfills unscientifically, creating problems to public health and the environment [1]. Leachates may contain large amounts of organic matter (biodegradable, but also resistant to biodegradation), where humic-type constituents consist an important group [2], as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts [3]. Landfill leachates have been identified as potential sources of ground and surface waters contamination, as they may percolate through soils and subsoils. Leachates present considerable variations chemical composition [4-6].

If landfills are not properly managed, these can cause uncontrolled gaseous and liquid emissions. The landfill can be classified into three categories based on age: young, middle and old. A landfill which is within 5 years is termed as young age landfill. It consists of large amount of biodegradable matters and a higher COD value of 20000 mg/l. [7]

The leachate characteristics vary with time and from site to site because it depend on type of wastes disposed, rainfall, age of the landfill and design of the landfill etc [8]. The characteristics of the landfill leachates can usually be represented in terms of the parameters such as COD, BOD_5, ratio of BOD_5/COD, colour, pH, temperature, alkalinity, oxidation reduction potential and heavy metals [9].

The treatment processes used for leachate of municipal solid waste often involve a combination of appropriate techniques. Coagulation/flocculation is an essential process in water and in industrial wastewater treatment. Several studies have been reported on the examination of coagulation–flocculation for the treatment of leachate, aiming at performance optimization, i.e. selection of the most appropriate coagulants and flocculants determination of experimental conditions, assessment of pH effect and investigation of flocculant addition [10]. Coagulation and flocculation is a relatively simple technique that may be employed successfully in treating old landfill leachates [11-13].

The advantages of the proposed physico-chemical method for the treatment of leachates are mainly simplicity, low cost, good removal efficiencies and easy onsite implementation. This method could be used for pre- or post-leachate treatment in combination with biological treatment process. As a result of the apparent inability of the method for sufficient pollutant removal, the cost of the high chemical dosages that are required, and the associated problems of the chemical sludge that is generated, it could be suggested that no single leachate treatment method, biological or physicochemical, is able to produce an effluent with acceptable quality, and that both approaches should be appropriately combined.

The main objective of coagulation and flocculation process is to remove turbidity of organic compounds and heavy metals from the leachate. Several authors have used response surface methodology and optimization to improve the coagulation–flocculation processes of wastewater of different origins [14-20]. These authors agree that the type and dosage of coagulant and flocculant reactants are decisive to the success of the coagulation–flocculation process.

The aim of this study is to evaluate and optimize variables for using of Steel Industry Wastewater for a physico-chemical water treatment process of leachate resultant from compacting of solid waste (Mohammedia, Marocco). A statistically analyze experimental data is must enable a compromise between efficiency and operational costs of a real industrial process.

2. Materials and methods

2.1. Sample collection

The leachate studied is a fresh leachate resulting from compaction of municipal solid waste in Mohammedia city (Marocco); it is recovered from a reservoir which is in the dump trucks.

To obtain a collection of leachate, a Garbage Compactor Truck was chosen at random, which contained approximately 5.5 tons of solid waste. Garbage Compactor Trucks are trucked specially adapted for the collection of MSW. They come equipped with an automated garbage recovery system and also a compacting system.

2.2. Chemicals and Materials

The SIWW taken from Maghreb steel (Morocco society) discharges is rich in $FeCl_3$ (29.5 %) and was used as a coagulant in this study. This liquid waste was valorized as coagulant in the treatment of leachate.

Maghreb steel uses hydrochloric acid in various cleaning process. Iron oxide and hydrochloric acid react to form iron chloride (ferrous and ferric), which is soluble in water. Rust is literally dissolved. After soaking in acid, the workpiece is covered immediately with a thin film of rust when exposed to air. It is necessary to neutralize with a base. The sodium hydroxide solution, commercially available as used for unblocking sinks is well suited. Washing soda (used as a mild detergent) is also suitable. After neutralization, dry with compressed air and paint if necessary.

The liquid waste of rinsing with hydrochloric acid is the SIWW waste liquid rich in ferric chloride. The characteristics of this coagulant are given in Table 1.

Table 1: Main characteristics of SIWW

Parameter	Value
pH	<1
Conductivity (ms/cm)	20
Fe^{3+} (g/l)	101.3
$FeCl_3$ (g/l)	295

The flocculant used is an anionic polymer 35 %; its trade name is Hymoloc DR3000. The characteristics of this flocculant are shown in table 2.

The experimental set-up used for the coagulation–flocculation experiments at laboratory scale consisted of a Jar-test device (Jar Test Flocculator FC-6S Velp Scientifica) in which six stirring blades were connected to a motor that operated under adjustable conditions. The system permitted the experiments to be performed with ease and the different variables affecting the removal of suspended fat and organic matter to be interpreted such as pH, stirring time and speed, retention time or reactant concentrations.

Coagulant dosages (SIWW) varied in the range of 26–54 mL.L^{-1} (equivalent to 7.8 – 16.2 g $FeCl_3$/L), while flocculant dosages (Hymoloc DR3000 0.3%) ranged from 5 to 19 mL.L^{-1}.

Table 2: Characteristics of polymer Hymoloc DR3000

Appearance Milky	Value
Parameter	Weight High
Density	35%
Viscosity	3.0-4.1
pH	<600cp
Cationicity	1.2 g/cm3
Molecular	White Liq.

Sixteen experiments were carried out for coagulant SIWW. After the addition of coagulant, the leachate was stirred at 160–180 rpm for 10 min. The flocculant was then added and the medium stirred at 40–50 rpm for 20 min. Samples were taken from the supernatant and analyzed after leaving the medium to settle for two hours.

The pH is one of the most restrictive parameters in the coagulation step and affects the hydrolysis equilibrium produced by the presence of the coagulant agent. The pH of leachate was adjusted by addition of NaOH (40 %) or H_2SO_4.

2.3. Analytical Experiments

The coloration was determined using European Standard Method (ISO 7887:1994) [21] with UV/Visible spectrophotometer (Model 9200 UV/VIS).

Samples of leachates, treated with Folin Ciocalteu, develop a blue color; the absorption was then measured at 725 nm after being left in the dark for an hou. Temperature and reaction time influence the development of the coloration.

The volume of decanted sludge was estimated by the volumetric method using the Imhoff cones, after 2 hours of settling, the sludge production is determined by direct reading as mL.L^{-1} of sludge of leachate treated.

Removal efficiency of phenol and colour were obtained according to the formula given below:

$$\text{Removal}\,\% = (1 - \frac{C}{C0}) \times 100 \qquad (1)$$

Were C_0 and C are the initial and final concentrations of phenol and colour of leachate after treatment by coagulation flocculation with SIWW.

2.4. Experimental design

A central composite rotatable design for *k* independent variables was employed to design the experiments [22] in which the variance of the predicted response, $\hat{Y}$, at some points of independent variables, *X*, is only a function of the distance from the point to the design centre.

These designs consist of a 2^k factorial (coded to the usual ±1 notation) augmented by $2{*}k$ axial points ($\pm \alpha$, 0, 0), (0, $\pm \alpha$, 0), (0, 0, $\pm \alpha$), and 2 centre points (0, 0, 0) [23]. The value of α for rotatability depends on the number of points in the factorial portion of the design, which is given in Eq. (2):

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

Where N_F is the number of points in the cube portion of the design ($N_F = 2^k$, *k* is the number of factors). Since there are three factors, the N_F number is equal to 2^3 (=8) points, while α is equal to $(8)^{1/4}$ (=1.682) according to Eq. (2).

In this study, the responses were phenol removal (Y_{phenol}), colour removal (Y_{colour}) and volume decanted sludge (Y_{vds}) of leachate of municipal solid waste. Each response was used to develop an empirical model that correlated the response to the coagulation processes activated variables using a second-degree polynomial equation as given by Eq. (3) [24]:

$$\hat{Y} = \beta_o + \beta_1 X_1 + \beta_2 X_2 + \beta_3 X_3 + \beta_{12} X_1 X_2 + \beta_{13} X_1 X_3 + \beta_{23} X_2 X_3 + \beta_{11} X_1^2 + \beta_{22} X_2^2 + \beta_{33} X_3^2 \quad (3)$$

Where β_o the constant coefficient, β_i the linear coefficients, β_{ij} the cross-product coefficients and β_{ii} the quadratic coefficients.

The software JMP® 10 was used for the experimental design, data analysis, model building, and graph plotting.

3. Results and discussion

3.1. Physico-chemical characterization of raw leachates

The physico-chemical characterization of raw leachate produced by compaction of solid waste is determined in the following table:

Table 3: Physico-chemical characterization of raw leachate

Parameter	Value
pH	4.45
Conductivity (ms/cm)	11.57
Turbidity (NTU)	4000
Colour	>3
COD (mg.L^{-1})	57 600
BOD$_5$ (mg. L^{-1})	6 800
BOD$_5$/COD	0.12
Phenol (mg. L^{-1})	182
Surfactant (mg. L^{-1})	35.6
Settled volume (mL.L^{-1})	16
Suspended matter (mg. L^{-1})	6530
Orthophosphate (mg. L^{-1})	0.32
Total phosphorus (mg. L^{-1})	1879
Ammonia nitrogen (mg.L^{-1})	2.23
NTK (mg.L^{-1})	1290
SO_4^{2-} (µg.L^{-1})	156
Cu (mg.L^{-1})	1
Zn (mg.L^{-1})	1.45
Cr (mg.L^{-1})	2.5
Ni (mg.L^{-1})	0.27
Pb (mg.L^{-1})	0.53
Sb (mg.L^{-1})	0.9
Sn (mg.L^{-1})	0.6

The characteristics of leachate of municipal solid waste can usually be represented in terms of the basic parameters such as COD, BOD$_5$, ratio of BOD$_5$/COD, colour and pH. The investigated leachate was characterized by high levels of organic matter, in terms of COD values, reached 57 600 mg.L^{-1} in leachate (Table 3).

The BOD$_5$/COD ratio was between 0.12; it shows that the samples collected are biodegradable. The variation of leachate characteristics was attributed to a number of causes, such as variations in the composition, age and moisture of the solid waste...Variations in the composition of municipal solid waste are one of the main factors affecting leachate characteristics. In terms of BOD$_5$, the value reported reached 6 800 mg.L^{-1}. It should be mentioned that fresh leachate presented relatively low pH values (around 5) rather low BOD$_5$/COD ratio, high COD levels and very high phenol and surfactant content.

Another parameter was investigated in this study, which is color of leachate. The results show that the leachate is dark color, which is explained by their high organic matter content.

3.2. Development of the regression model equation

Preliminary experiments were carried out to screen the appropriate parameters and to determine the experimental domain. From these experiments, the effects of initial pH of leachate (X1), coagulant dosage in mL.L^{-1} (X2) and flocculent dosage in mL.L^{-1} (X3) are investigated on three responses: phenol removal, colour removal and volume sludge decanted. The parameter levels and coded values were given in Table 4.

The coefficient of determination R^2 in this study were relatively high, indicating a good agreement between the model predicted and the experimental values. Meanwhile, adjusted R^2 permitting for the degrees of freedom associated with the sums of the squares is also taken into account in the lack-of-fit test, which should be an approximate value of R^2. When R^2 and adjusted R^2 differ dramatically, there is a good chance that insignificant terms have been included in the model [25]. As shown in Table 5, the two R^2 values were not significantly different.

Table 4: Study field and coded factors.

Natural variables (X_j)	Unit	Coded variables X_1, X_2, X_3^*				
		A	-	0	+	B
X_1= initial pH	-	4.3	5	6	7	7.7
X_2= Coagulant dosage	$mL.L^{-1}$	26.5	32	40	48	53.4
X_3= Flocculent dosage	$mL.L^{-1}$	5.3	8	12	16	18.7

* The coded values $X_j = \pm 1$ are obtained by equation: $X_j = (x_j - \bar{x}_j)/\Delta$

Table 5: Regression coefficient R^2 and adjusted R^2

	Phenol removal (%)	Colour removal (%)	Decanted sludge ($mL. .L^{-1}$)
R^2	0.89	0.93	0.92
R^2_{adj}	0.72	0.82	0.81

The experimental design matrix, the corresponding experimental parameters and response value were shown in Table 6.

Table 6: Experimental design and results for phenol and colour removal and decanted sludge.

Configuration	X_1	X_2	X_3	Phenol removal (%)	Colour removal (%)	Decanted sludge ($mL.L^{-1}$)
− − −	5	32	8	85	85	88
− − +	5	32	16	86	85	68
− + −	5	48	8	92	67	82
− + +	5	48	16	94	69.5	78
+ − −	7	32	8	89.5	84.5	160
+ − +	7	32	16	94.5	87	126
+ + −	7	48	8	94	94	148
+ + +	7	48	16	90	91	120
a00	4.32	40	12	88	75	80
A00	7.68	40	12	96	84	190
0a0	6	26.5	12	87	82	40
0A0	6	53.4	12	93.5	72	54
00a	6	40	5.3	91	83	75
00A	6	40	18.7	91.5	86.5	36
0	6	40	12	91	73	40
0	6	40	12	91	68	41

3.3. Process optimization

The regression coefficient (R^2) is a number that indicates the proportion of the variance in the dependent variable that is predictable from the independent variable [22]. It is a statistic used in the context of statistical models whose main purpose is either the prediction of future outcomes or the testing of hypotheses, on the basis of other related information. It provides a measure of how well observed outcomes are replicated by the model, based on the proportion of total variation of outcomes explained by the model [23-25]. The plots of the experimental value versus the predicted value for phenol removal, colour removal and sludge decanted are shown in Fig. 1. The experimental values are distributed relatively near to the straight line.

The analysis of variance test (ANOVA) for the response surface model is provided in Table 7. Since the p-value for the model was lowers than 0.05, there was a statistical relationship between phenol removal, colour removal and sludge decanted and the selected variables at a 95% confidence level. As can be observed in the ANOVA table (Table 7), the significant terms in the model were X_1, X_2 and X_1X_2 of phenol and colour removal, while, $X_1$and X_2 where the most significant factors of sludge decanted. Other model terms (X_3, X_1X_3, X_2X_3 and X_2^2) were not significant.

The goodness of fit of the model for phenol removal (for example) was evaluated by the regression coefficient R^2=0.89. The 89 % sample variation observed for phenol removal was attributed to the variables selected (pH, coagulant and flocculant dosages), while the model did not explain 11 % of the variations.

However, phenol removal, colour removal and sludge decanted response were found at a pH=6 and a high level of coagulant dosage 40 $mL.L^{-1}$ equivalent to 12 g $FeCl_3$/L. The interaction between this factors causes the most significant variation as can be observed at Fig. 2, for example, if we set the coagulant dosage and flocculant dosage at 40 and 12 $mL.L^{-1}$ respectively, phenol removal, colour removal and sludge decanted can be achieve 91%, 70 % and 38 $mL.L^{-1}$ respectively at pH equal at 6.

These results show that the coagulation–flocculation mechanism differs depending on the pH value and dosage of coagulant. Several studies have reported the examination of this process for the treatment of industrial wastewater, especially with respect to performance optimization of coagulant/flocculant, determination of experimental conditions, assessment of pH and investigation of flocculant addition [26].

Fig. 1 shows the three-dimensional response a surface which was constructed to show the effects of the coagulant dosage, flocculent dosage and the pH on the phenol and colour reduction and sludge decanted of fresh leachate by coagulation processes with SIWW. The optimum removal points by three-dimensional (3D) surface (93, 85 % and 115 $mL.L^{-1}$) obtained at around dose 48 $mL.L^{-1}$ correspond to 14.4 g $FeCl_3$/L and initial pH=7 as can be observed by the saddle shape at dosage flocculant fixed at 12 $mL.L^{-1}$.

The three-dimensional (3D) surface plot is approximately symmetrical in shape with circular contours. The responses of phenol removal, colour removal and sludge decanted plot show clear peak, which indicate that the optimum conditions for maximum value of the response are determined by dose coagulant and initial pH inside the design boundary. The decline in these responses efficiencies is observed when moving away from this point, implying that neither increase nor decrease in any of the tested variables is desired.

The responses permitted the development of mathematical equations where each response (Y) was estimated as a function of X_1, X_2 and X_3, and computed as the sum of a constant, three linear effects (terms in X_1, X_2 and X_3), three quadratic effects (X_1^2, X_2^2 and X_3^2), and three interactions effect (X_1X_2, X_1X_3, X_2X_3) according to three equations:

$$Y_1=91.05 +1.79X_1 +1.89X_2 +0.35X_3 -1.87X_1X_2 -0.25X_1X_3 -X_2X_3 +0.22X_1^2 -0.39X_2^2 -0.04X_3^2 \quad (4)$$
$$Y_2=70.32+4.77X_1-2.69X_2+0.58X_3+5.87X_1X_2-0.37X_1X_3-0.37X_2X_3+3.61X_1^2+2.73X_2^2+5.47X_3^2 \quad (5)$$
$$Y_3=37.87+30.97X_1+0.7X_2-11.1X_3-2.75X_1X_2-4.75X_1X_3+2.75X_2X_3+39.75X_1^2+8.64X_2^2+11.75X_3^2 \quad (6)$$

Where Y_1, Y_2 and Y_3 were responses of phenol removal, colour removal and sludge decanted respectively.

Table 7: ANOVA for phenol removal, colour removal and decanted sludge response surface models.

	Source	Degrees of freedom[b]	Sum of squares[c]	F-value[d]	Rapport t	p-value[e]
Phenol removal	X1(5-7)	1	43.79	15.25	3.91	0.0079[a]
	X2(32-48)	1	49.24	17.15	4.14	0.0061[a]
	X3(8-16)	1	1.72	0.60	0.77	0.4688
	X1*X2	1	28.12	9.80	-3.13	0.0203[a]
Colour removal	X1(5-7)	1	310.66	24.33	4.93	0.0026[a]
	X2(32-48)	1	99.26	7.77	-2.79	0.0317[a]
	X3(8-16)	1	4.55	0.36	0.60	0.5722
	X1*X2	1	276.12	21.62	4.65	0.0035[a]
Decanted sludge	X1(5-7)	1	13101.60	31.19	5.58	0.0014[a]
	X2(32-48)	1	6.67	0.02	0.13	0.9038
	X3(8-16)	1	1682.63	4.00	-2.00	0.0922
	X1*X2	1	60.50	0.14	-0.38	0.7174

[a] Significant at the 95% confidence level.

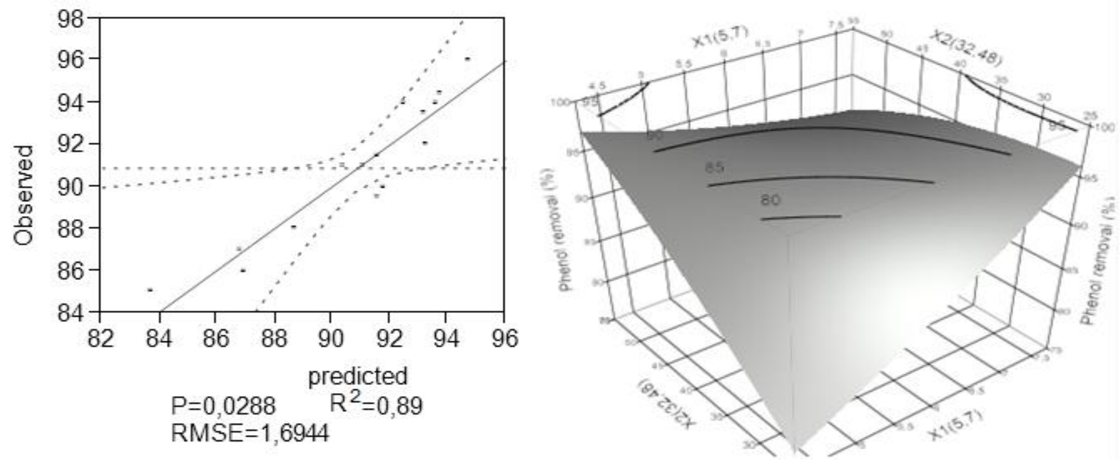
[b] Degrees of freedom: an estimate of the number of independent categories in a particular statistical test or experiment.

[c] Sum of squares: the sum of squares is a mathematical approach to determining the dispersion of data points. The sum of squares is used as a mathematical way to find the function which best fits (varies least) from the data.

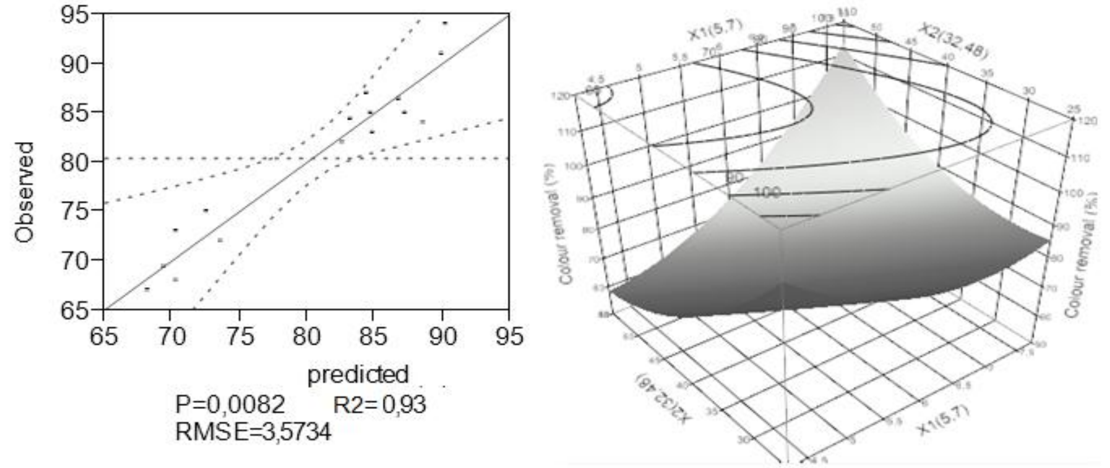
[d] F-value: value calculated by the ratio of two sample variances. The F statistic can test the null hypothesis: (1) that the two sample variances are from normal populations with a common variance; (2) that two population means are equal; (3) that no connection exists between the dependent variable and all or some of the independent variables.

[e] p-Value: p value is associated with a test statistic. It is the probability, if the test statistic really were distributed as it would be under the null hypothesis, of observing a test statistic [as extreme as, or more extreme than] the one actually observed.

(a) Phenol removal (%)



(b) Colour removal (%)



(c) Decanted sludge (mL.L^{-1})

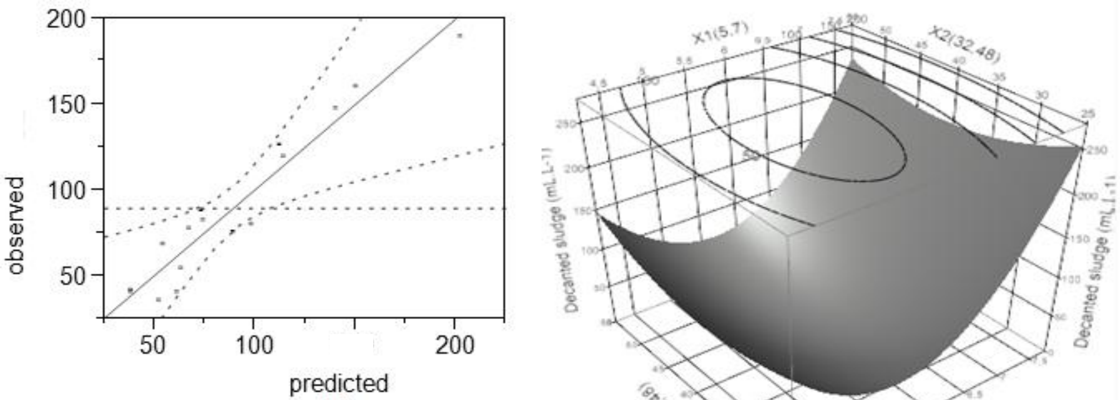


Figure 1: Experimental values versus predicted values and Response surface plots as a function of pH (X1) and coagulant dosage (X2) at flocculant dosage equal at 12 mL.L^{-1}: (a) Phenol removal (%); (b) colour removal (%); (c) decanted sludge (mL.L^{-1}).

3.4. Model validation

To further validate the models under even higher reactant dosages, an additional experiment (6 of pH, 40 mL.L^{-1} of coagulant, equivalent to 12 g $FeCl_3$/L, and 12 mL.L^{-1} of flocculant) was performed. The sample used to optimize coagulation process is the same one used to validate the model. The responses are listed in Table 8 along with the predicted measured results.

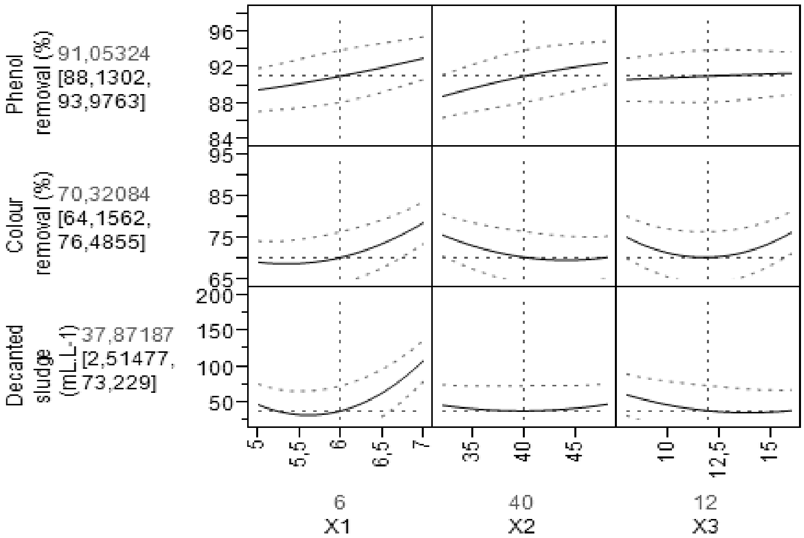


Figure 2: Profiler forecast of phenol removal, colour removal and sludge decanted.

As can be seen in the table, the three responses were close to the responses that were estimated using response surface methodology. Besides, an evaluation of the model's overall performance known as the regression coefficient and denoted by R^2 must be considered. The R^2 values in this study were relatively high, indicating a good agreement between the model predicted and the experimental values. Therefore, it can be concluded that the models accurately represent phenol removal, colour removal and decanted sludge over the experimental range studied. Table 8 shows the optimum values for the responses and the factors. The values were calculated by means of the desirability function and the models obtained using response surface methodology. The validation of this model show the optimum values of the factors and the responses were 6 of the pH, 40 $mL.L^{-1}$ of coagulant (equivalent to 4 g Fe^{3+}/L and 12 g $FeCl_3$/L), 12 $mL.L^{-1}$ of flocculant and 91 %, 70 % and 38 $mL.L^{-1}$ of phenol removal, colour removal and decanted sludge, respectively. The optimum dose of a coagulant or flocculant is defined as the value above which there is no significant difference in the increase in removal efficiency with a further addition of coagulant or flocculant.

Table 8: Validation of the models

	Phenol removal (%)	Colour removal (%)	Decanted sludge ($mL.L^{-1}$)
Validation of the models at pH= 6, at 40 $mL.L^{-1}$ of coagulant and 12 $mL.L^{-1}$ of flocculant			
Optimum response predicted (%)	91	70	38
Optimum response experimental (%)	80	68	42

Conclusion

1- A central composite experimental design and response surface methodology were used to optimize the coagulation–flocculation process of leachate resultant from compacting solid waste of city Mohammedia for reducing the number and cost of experiments and improving the process at industrial scale.
2- The best regression coefficients (R2) were obtained for phenol removal, colour removal and decanted sludge at variable pH (4.4-7.7): 0.89, 0.93 and 0.92, respectively.
3- Coagulant dosage and pH seems to be the most significant factors in the soluble removal of phenol and colour.
4- For flocculant dosage, however, the trend is not given that this factor influences in treatment of fresh leachate (for a low concentration of flocculant).
5- Multiple response optimization allowed the coagulant and flocculant dosages to be minimized, while maximizing the phenol and colour removal percentages, and to decrease the volume of decanted sludge.

6- The pH was also evaluated to determine the most suitable condition for the coagulation–flocculation process of operational, economic and post-treatment factors.

7- The validation of this model show the optimum values of the factors and the responses were 6 of the pH, 40 mL.L-1 of coagulant (equivalent to 12 g FeCl3/L), 12 mL.L-1 of flocculant and 91 %, 70 % and 38 mL.L-1 of phenol removal, colour removal and decanted sludge, respectively.

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References

1. Kang K.H., Shin H. S., Park H., *Wat. Res.* 36 (16): (2002) 4023-4032.
2. Wang Q., Matsufuji Y., Dong L., Huang Q., Hirano F., Tanaka A., *Waste Manag.* 26 (2006) 815-824.
3. Tatsi A. A., Zouboulis A. I., Matis K. A., Samaras P., *Chemosphere* 53 (2003) 737-744.
4. Bu L., Wang K., Zhao Q. L, Wei L. L., Zhang J., Yang J. C., *J. Hazard. Mater.* 179 (2010) 1096-1105.
5. Niua J., Zhangb T., Heb Y., Zhouc H., Zhaoc A., Zhaoa Y., *J. Hazard. Mater.* 252-253 (2013) 250-257.
6. Afshin M., Zazouli M. A., Izanloo H., Rezaee R., *American-Eurasian J. Agric. & Environ. Sci.* 5 (5) (2009) 638-643.
7. Silva A. C., Dezotti M., Sant'Anna Jr G. L., *Chemosphere* 55 (2004) 207-214.
8. Aziz S. Q, Aziz H. A., Yusoff M. S., Bashir M. J. K., *J. Hazard. Mater.* 189 (2011) 404–413.
9. Amuda O. S., Amoo I. A., *J. Hazard. Mater.* 141 (2007) 778-783.
10. Mufeed S., Kafeel A., Gauhar M., Trivedi R. C., *Waste Manag.* 28 (2008) 459-467.
11. Dikshit A. K., *IPCBEE* 6 (2011) *IACSIT Press, Singapore.*
12. Camba A., González-García S., Bala A., Fullana-i-Palmer P., Teresa Moreira M., Feijoo G., *J. Clean. Product.* 67, 15 (2014) 98-106.
13. Wiszniowski J., Surmacz-Górska J., Robert D., Weber J. V., *J. Environ. Manag.* 85 (2007) 59-68.
14. Ahmad A. L., Ismail S., Bhatia S., *Environ. Sci. Technol.* 39 (2005) 2828–2834.
15. Carvalho G., Delée W., Novais J. M., Pinheiro H. M., *Color. Technol.* 118 (2002) 215–219.
16. Bathia S., Othman Z., Ahmad A. L., *Chem. Eng. J.* 133 (2007) 205–212.
17. Wang J. P., Chen Y. Z., Ge X. W., Yu H. Q., *Colloids Surf.* 302 (2007) 204–210.
18. Omar F. M., Rahman N. N. N. A., Ahmad A., *Water Air Soil Pollut.* 195 (2008) 345–352.
19. Anouzla A., Abrouki Y., Souabi S., Safi M., Rhbal H., *J. Hazard. Mater.* 166 (2–3) (2009)1302–1306.
20. Martin M. A, Gonzalez I., Berrios M., Siles J. A, Martin A., *Chem. Eng.* 172 (2011) 771-782.
21. AFNOR, French Association for Standardization, *Collection of French standards* (1999), *Standard Test Methods for Water. Paris, France.*
22. Istadi I., Amin N. A. S., *J. Chem. Eng.* 106 (2005) 213–227.
23. Clarke G. M., Kempson R. E., *Arnold, London* (1997).
24. Ravikumar K., Pakshirajan K., Swaminathan T., Balu K., *J. Chem. Eng.* 105 (2005) 131–138.
25. Montgomery D. C., *Fifth ed., John Wiley & Sons, New York* (2001).
26. Renou S., Givaudan J. G., Poulain S., Dirassouyan F., Moulin P., *J. Hazard. Mater.* 150 (2008) 468–493.

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