Desalination and Water Treatment
www.deswater.com

doi: 10.1080/19443994.2015.1067837

57 (2016) 14488–14495
July

Taylor & Francis
Taylor & Francis Group

Coagulation–flocculation process for landfill leachate pretreatment and optimization with response surface methodology

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Received 12 January 2015; Accepted 22 June 2015

ABSTRACT

The present paper aimed to characterize and treat a landfill leachate using a coagulation–flocculation process. The leachate was obtained from a landfill in the city of Sousse, Tunisia. Its physicochemical characterization showed high levels of chemical oxygen demand (COD), ammonium, and heavy metal contents. The coagulation–flocculation process was applied as pretreatment effluent to reduce these pollutants. The key operating parameters (pH, coagulant dose, flocculant dose, and mixing speed in the flocculation step) on the coagulation–flocculation process were optimized using response surface methodology to investigate COD removal in landfill leachate. In this regard, a hybrid design was carried out to seek optimal conditions which were as follows: pH: 3.36; concentration of coagulant: 0.87 g/l; concentration of flocculant: 26 mg/l; and mixing speed: 48 rpm. These conditions have been proved experimentally.

Keywords: Landfill leachate; Pollutants; COD removal; Coagulation–flocculation; Response surface methodology; Hybrid design

1. Introduction

The physicochemical and biological decomposition of wastes and percolation of rainwater through compacted waste layers in municipal landfills generate a contaminated liquid called leachate. It usually contains high concentrations of dissolved and finally suspended organic matter and inorganic compounds such as ammonia [1]. The main indicators of organic pollutants are the chemical oxygen demand (COD) and the biochemical oxygen demand (BOD). In addition, the ammonia content, usually present in concentrations higher than 2,000 mg/l, does not decrease and often constitutes a major long-term pollutant in leachate [2]. Heavy metals are also present in leachate even in low concentrations. Therefore, leachate can pose a serious threat to its surrounding environments. Thus, to protect the environment, it is essential that leachate be treated before being discharging into the natural environment. The leachate characteristics are very variable and complex. They depend on the waste type,

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hydrology site, seasonal weather variations, landfill age, and the decomposition stage in the landfill [2–4]. Due to this complexity, treatment of landfill leachate is very difficult. The biological treatments, including aerobic and anaerobic processes, are the most economically efficient method for the removal of biodegradable organic compounds, but it is dependent on the BOD/COD ratio. Physicochemical treatments are also applied in the organic pollution treatment. Coagulation–flocculation process is a relatively simple technique that can be employed successfully in treating old landfill leachate [5]. Moreover, it could be a simple technique in the pretreatment of fresh leachate prior to other physicochemical or biological treatments that reduce pollutant loads, remove recalcitrant compounds and therefore, improve leachate biodegradability [6]. By the addition of chemical reagents (coagulants), the coagulation process involves the destabilization of colloidal suspension disorders that cannot settle naturally. Iron-based coagulants are more efficient than aluminum ones [7–10] and pose less health risks than the latter in the event of an overdose. In the present study, ferric chloride 6-hydrate ($FeCl_3 \cdot 6H_2O$) was investigated for the landfill leachate pretreatment. The flocculation process promotes the aggregation and binding of destabilized particles microflocs into larger flocks that can be removed by filtration or sedimentation. The efficiency of this process is governed by a great number of factors. Usually to achieve optimization of a treatment, one should begin by screening the whole retained factors to select the effective ones and then look for optimal levels of the fewer retained influent factors. Preliminary experiments allowed us to select four factors among eight potentially influent factors (pH, coagulant dosage, coagulation time, mixing speed in the coagulation step, dosage of flocculant, time flocculation, mixing speed in the flocculation step and decantation time). To optimize the levels of the selected variables, one can use the conventional experimentation method that involves changing one variable at a time, and maintaining the others fixed [5,10,11]. However, this experimentation method requires many experimental runs, that are time-consuming, and ignores the interaction effects between the parameters and leads to a low optimization efficiency [12]. Unlike the conventional method, statistical experimental designs can avoid these limitations and allow estimation of the effects of independent parameters and their interactions. In particular, response surface methodology (RSM) is an efficient way to design, build models, and analyze the effects of several independent variables in order to optimize processes for different research types. It has been applied successfully in various scientific and technical fields such as chemical engineering, biochemistry, biotechnology, and environment [10,12,13]. Few studies were reported on the RSM application to optimize the operation conditions of the coagulation–flocculation process applied for landfill leachate treatment. The main objective of this study was the examination of coagulation–flocculation process efficiency for the raw leachate treatment, especially in terms of organic matter trying to reach the legal limits of release into natural environment. Among the most applied experimental designs for optimization (Box Behnken, Central composite, Doehlert ... [14,15]), the Roquemore design may be regarded as an efficient tool requiring fewer experiments than the cited designs while keeping a relatively high precision [16,17]. Its matrix design is created via an imagination idea that involves the use of a central composite design for $k-1$ variables and the variable levels are supplied in such a way as to obtain certain symmetries in the matrix.

2. Materials and methods

2.1. Leachate

Leachate samples were collected from the sanitary landfill of Sousse, Tunisia. Samples were immediately transferred to the laboratory and stored at 4°C. Their main physicochemical characteristics are presented in Table 1.

Table 1
Landfill leachate characteristics and legal limits for discharges into urbanized stream

Parameter	Values	LLD[a]
pH	8–8.2	6.5–9
Turbidity (NTU)	800–1,000	nd[b]
TSS (g/l)	0.7–1	0.4
TDS (g/l)	47–48	nd[b]
COD (g O_2/l)	19–21	1
BOD_5 (g O_2/l)	5–6	0.4
BOD/COD	0.26–0.28	
NTK (g/l)	1.71–1.8	0.1
NH_4^+ (g/l)	1.46–1.48	nd[b]
Pb (mg/l)	<0.005	1
Cd (mg/l)	<0.005	0.1
Fe (mg/l)	10.2–12.8	5
Cr (mg/l)	1.7–1.9	1
Ni (mg/l)	0.05–0.07	2

[a]LLD: legal limits for discharge into urbanized streams (NT. 106.002);
[b]Nd: not determined.

2.2. Analytical techniques

The physicochemical characteristics of leachate samples were validated according to the French standard NFXPT 90-210 [18]. The pH was measured using pH meter (INOLAB WTW 720). Turbidity was measured using 2020 Turbidimeter (LaMotte). BOD_5 was determined by the manometric method with a respirometer (BSB-Controlled Model OxiTop (WTW)) and the COD was estimated using the method described by Knechtel [19]. Total nitrogen contents (TN) were measured by the Kjeldahl method using an automated apparatus (Buchi, Switzerland). The total suspending solid content (TSS) was assessed by drying at 105˚C for 12 h [20]. The total concentrations of Fe, Ni, Cr, and heavy metals were determined using atomic absorption flame emission spectroscopy AAS (Thermo scientific).

2.3. Jar test

Coagulation–flocculation experiments were performed using a conventional jar-test apparatus (Prolabo), equipped with six 1,000-ml beakers at room temperature (around 20˚C). Leachate samples were removed from the refrigerator and kept for about 2 h at room temperature before performing any test. pH adjustment was made using solutions of HCl or NaOH. The coagulant solution (20 g/l ferric chloride $FeCl_3$, $6H_2O$) and the flocculant solution (cationic polyacrylamide) were prepared before each experiment. After the coagulation–flocculation tests, samples were taken from the liquid level about 3 cm underneath the surface.

2.4. COD removal

The COD removal was determined by measuring the COD before and after the treatment. The following equation was used to calculate the removal efficiency:

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

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

2.5. Toxicity test

2.5.1. Phytotoxicity

Leachate phytotoxicity was assessed by measuring the germination index (GI) of tomato seeds (Lycopersicon esculentum). The phytotoxicity was determined according to the method of Zucconi et al. [21], based on the determination of the GI. Ten seeds of the plant species were placed in a 90-mm-diameter Petri dish, already containing a sterile filter paper and a 5-ml test solution. A control in distilled water was used systematically for each species and for each test medium concentration. Four different dilutions in distilled water were performed in order to widen the response field to the toxicity of each sample. The Petri dishes were then covered and incubated at 25 ± 2˚C for 72 h.

2.5.2. Microtoxicity

The test principle consisted in measuring the inhibition rate of bioluminescence of the bacterium *Vibrio fischeri*, strain DSM 2167. A LUMIStox equipment (GmbH, Düsseldorf, Germany), in accordance with the ISO/DIS 11348-2 (1998) standard, was selected for this measurement. The percentage inhibition of bioluminescence (IB %) was measured by mixing 0.5 ml of landfill leachate and 0.5 ml of luminescent bacterial suspension. After 15-min incubation at 15˚C, the decrease in bioluminescence was determined. The sample toxicity was expressed as the percentage of inhibition of the bioluminescence (IB %) relative to a control (distilled water).

3. Results and discussion

3.1. Physicochemical characteristics of the leachate

The characteristics of the leachate samples collected during the research period are summarized in Table 1. The leachate samples were alkaline (pH > 7) with a brownish color due probably to the presence of humic substances [22], a high organic load expressed in COD (20 g/l), and low BOD_5 (6 g/l) with a BOD_5/COD ratio of 0.27 indicating the leachate low biodegradability. The COD measured level was much higher than the authorized levels (Tunisian norms). Nitrogen is present mainly as ammonium. This is correlated with what was reported in the literature. Indeed, several studies have shown that almost all of the nitrogen present in the leachate is in ammonia [23–25]. The presence of NH_4 in the solution is toxic for bacteria [4]. We also noted the presence of some micropollutant minerals such as iron and chrome. These data suggest that most of the mineral pollutants are insoluble at this high pH value [26]. These characteristics are typical of a young leachate.

3.2. Leachate toxicity

Microtoxicity tests were carried out using the luminescent bacterium *V. fischeri*. The test principle consists in measuring the bioluminescence inhibition (IB %). The test performed showed that the leachate samples exhibited high microtoxicity since they inhibited the bioluminescence of *Vibrio fischeri* at a rate of 100%. Leachate toxicity was also evaluated on plants in order to assess inhibition risk of seed germination contacted with different concentrations of the test product. Seed germination is completely inhibited (growth inhibition (%) = 0), when the leachate applied is raw or diluted 10 times suggesting a high phytotoxicity. Indeed, Zucconi et al. [21] reported that the effluent is phytotoxic if the GI is below 50%. Although the leachate 50-fold diluted decrease the toxicity, an inhibitory effect was still observed (Fig. 1).

3.3. Optimization of coagulation–flocculation process: modeling of COD removal

3.3.1. Optimization method

The statistical experimental design employed (Roquemore design [27]) was selected to optimize four operating variables influencing the coagulation–flocculation process, namely the pH (X1), the coagulant dose (X2), the flocculant dose (X3), and the mixing speed in the flocculation step (MSFS) (X4). Five levels for each factor were selected according to the design matrix data (Table 3) to investigate the effects of factors and to locate the optimal conditions. The range and levels of the variables (X1, X2, X3, and X4) are provided in Table 2. The response variable (COD removal) was fitted to a second-order model that describes the relationship between independent variables X_J and the dependent output variables (y). The better model was developed by a second-order mathematical model (Eq. (2)):

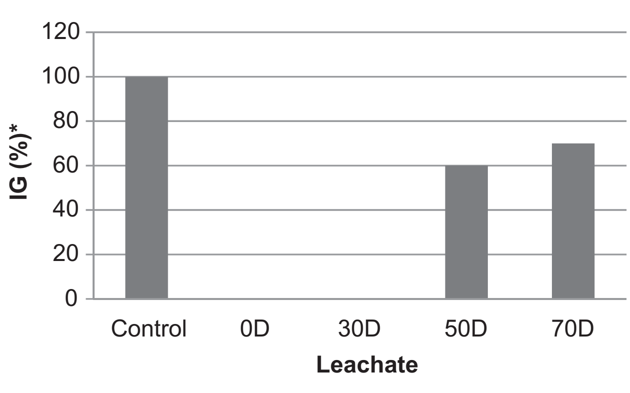

Fig. 1. Leachate phytotoxicity. *IG: index germination; D: dilution factor.

Table 2
Level variables range of the experimental design

Parameter	Center	Step variation
X1: pH	5	2.00
X2: Coagulant dosage (g/l)	0.6	0.5
X3: Flocculant dosage (mg/l)	30	20
X4: MSFS (rpm)	50	20

$$\hat{Y} = b_0 + \sum_{J=1}^{4} b_J X_J + \sum_{J=1}^{4} b_{JJ} X_J^2 + \sum_{\substack{k=1 \\ J\neq k}}^{4} b_{Jk} X_J X_k \tag{2}$$

where $\hat{Y}$ is the predicted response, b_0 is the model constant, b_J is the linear effect, b_{JJ} is the squared effect, b_{Jk} is the interaction effect and X_J are independent variables. The levels of the four variables (Table 2) are chosen according to Table 3. The levels of the other parameters were fixed as follows: coagulation time: 4.5 min, coagulation speed: 150 rpm, flocculation time: 60 min, and decantation time: 30 min.

3.3.2. Model equation

The postulated model coefficients were calculated by least squares method on the basis of the experimental responses (Table 3) using the NEMROD.W software. The fitted model expressed in coded variables was represented by the following equation:

$$\begin{aligned} Y = {} & 57.1 - 27.1X_1 + 2.1X_2 - 2.5X_3 + 2X_4 - 7X_1^2 - 2.4X_2^2 \\ & - 6.1X_3^2 - 26.3X_4^2 - 20.8X_1X_2 + 16.3X_1X_3 + 2.1X_2X_3 \\ & + 9.7X_1X_4 - 16.3X_2X_4 - 11X_3X_4 \end{aligned} \tag{3}$$

3.3.3. Variance analysis and the model validation

To attest the good quality of the fitting, it is important to analyze the variance as demonstrated in Table 4. Indeed, this table shows that the regression sum of squares is statistically significant at 99%. The value of F (critical value of fisher) for significance of regression (the ration of mean square due to the regression to mean square of real error) was 8.74 greater than $F_{0.01}(14.9)$. The coefficient of determination R^2 equal to 0.931 means that 93.1% of the observed variations of the response are attributed to the variable effects. These results confirm the good fitting of the model. The model obtained was used to predict response values in the studied domain and to draw isoresponse contour plots and response surfaces.

Table 3
Matrix hybrid design, real experimental conditions, and measured responses

N°Exp	pH[a]	Coagulant dose (g/l)	Flocculant dose (mg/l)	MSFS (rpm)	COD (%)
1	0.000 (5.00)	0.000 (0.60)	0.000 (30.00)	1.000 (70.00)	(33.55)
2	−0.540 (3.92)	−0.540 (0.33)	−0.542 (19.16)	0.327 (56.55)	(62.18)
3	0.540 (6.08)	−0.540 (0.33)	−0.542 (19.16)	0.327 (56.55)	(50.12)
4	−0.540 (3.92)	0.540 (0.87)	−0.542 (19.16)	0.327 (56.55)	(80.40)
5	0.540 (6.08)	0.540 (0.87)	−0.542 (19.16)	0.327 (56.55)	(22.35)
6	0.540 (3.92)	−0.540 (0.33)	0.542 (40.84)	0.327 (56.55)	(55.00)
7	0.540 (6.08)	−0.540 (0.33)	0.542 (40.84)	0.327 (56.55)	(40.45)
8	0.540 (3.92)	0.540 (0.87)	0.542 (40.84)	0.327 (56.55)	(54.42)
9	0.540 (6.08)	0.540 (0.87)	0.542 (40.84)	0.327 (56.55)	(36.58)
10	0.820 (3.36)	0.000 (0.60)	0.000 (30.00)	−0.569 (38.62)	(69.00)
11	−0.820 (6.64)	0.000 (0.60)	0.000 (30.00)	−0.569 (38.62)	(16.00)
12	0.000 (5.00)	−0.820 (0.19)	0.000 (30.00)	−0.569 (38.62)	(36.32)
13	0.000 (5.00)	0.820 (1.01)	0.000 (30.00)	−0.569 (38.62)	(55.00)
14	0.000 (5.00)	0.000 (0.60)	−0.822 (13.55)	−0.569 (38.62)	(40.00)
15	0.000 (5.00)	0.000 (0.60)	0.822 (46.45)	−0.569 (38.62)	(45.00)
16	0.000 (5.00)	0.000 (0.60)	0.000 (30.00)	0.000 (50.00)	(53.14)
17	0.000 (5.00)	0.000 (0.60)	0.000 (30.00)	0.000 (50.00)	(51.50)
18	0.000 (5.00)	0.000 (0.60)	0.000 (30.00)	0.000 (50.00)	(55.00)
19	0.000 (5.00)	0.000 (0.60)	0.000 (30.00)	0.000 (50.00)	(60.00)
20	0. 000 (5.00)	0.000 (0.60)	0.000 (30.00)	0.000 (50.00)	(57.00)
21	−0.395 (4.21)	−0.220 (0.49)	−0.161 (26.77)	−0.125 (47.50)	(67.90)
22	−0.395 (5.79)	0.220 (0.49)	−0.161 (26.77)	−0.125 (47.50)	(45.00)
23	0.000 (5.00)	0.460 (0.83)	−0.161 (26.77)	−0.125 (47.50)	(60.00)
24	0.000 (5.00)	0.000 (0.60)	0.484 (39.68)	−0.125 (47.50)	(60.50)

[a]( ) real experimental conditions.

Table 4
Variance analysis for hybrid design

Source of variation	Sum of squares	Degrees of freedom	Mean squares	F_0	Significance
Regression	4673.86	14	333.847	8.7387	0.129 [a]
Residual	343.831	9	38.2034		
Total	5017.69	23			

[a]indicates significant at the level 99%.

3.3.4. Interpretation of the response surface model

3.3.4.1. Ridge analysis. This method (response surfaces, design, and analyses [28]) is used to plot the curve (optimal path) corresponding to the maximum values of the response on concentric spheres of varying radii (distance to the domain center) as well as the location of the point corresponding to the maximum value obtained. The representation of the optimum path for the removal of COD analysis by the response surface is shown in Fig. 2. The left and right parts of both plots indicate the minimization and the maximization of the response, respectively. Fig. 2(a) shows that the optimum response reached is dependent on the distance. The percentage of COD removal increases from the center of the domain to the boundary where it reaches 85%. Fig. 2(b) displays the coordinates for each factor, in codified variables. It can be seen that the maximum of COD reduction is more related to the variation of the pH (X_1) and the concentration of coagulant (X_2) than the other parameters. To reach the maximum, the first factor (X1) tends to a very low value, the second factor (X2) must tend to a high level, and the third (X3) and the fourth (X4) factors must tend somewhat to low values.

3.3.4.2. Isoresponse contour plots. The isoresponse curves and the three-dimensional response surface curves are plotted as a function of two factors, while the other

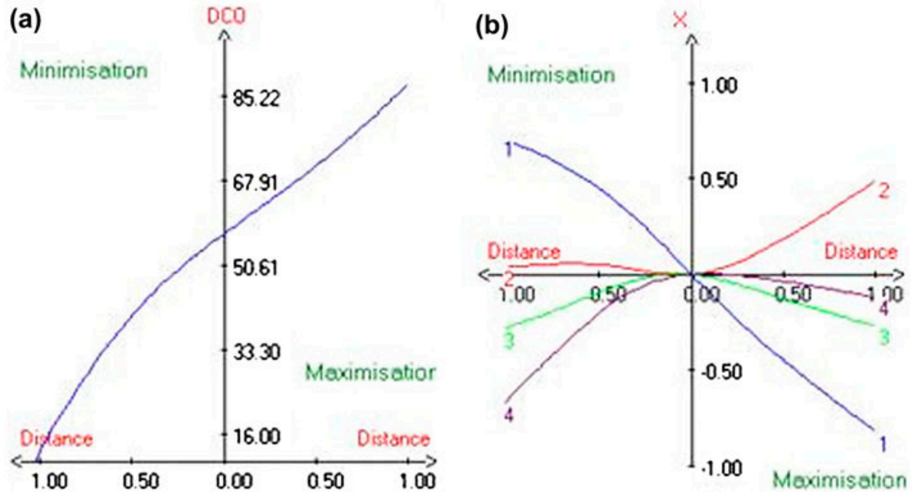

Fig. 2. Ridge analysis: optimal response plot (a) and optimal coordinate plot (b).

factors are kept constant at mean levels (Fig. 3). The contour plots are one the most revealing ways of illustrating and interpreting the response surface system. The contour plots are merely two-dimensional graphs that show contours of constant response with the axis system being a specific pair of the design variables [29]. The examination of these figures shows the high interactive effect between the pH and the dose of coagulant. Fig. 3 shows that the coagulant dose has no significant effect when the pH is high (above 5). However, the coagulant concentration has a significant positive effect when the pH is below 5. Guo et al. [4] also showed that the COD removal efficiency increased with increasing pH up to 5. For pH > 5, the COD removal decreased. The response (COD removal) was highly enhanced by raising the coagulant dose and lowering the pH. The most important reduction is observed at pH 3. Indeed, an increasing of one unit of the pH leads to a decrease of 20% in COD. These results are in agreement with those described by other reports. Indeed, Maranon et al. [30] showed that the optimum pH should be of 3.8 with a ferric chloride dose of 0.5 g/l for treatment of young leachates. Similarly, Verdenne et al. [8] obtained 18% COD removal at pH 3 by applying the coagulation–flocculation for the treatment of old leachates. Moreover, Turki et al. [11] showed also that the pH played a vital role in COD removal that reached the highest value (45%) with a pH of 5.5. In general, chemical coagulation is a process which is highly pH dependent. The pH influences the nature of produced polymeric metal species that will be formed as soon as the metal coagulants

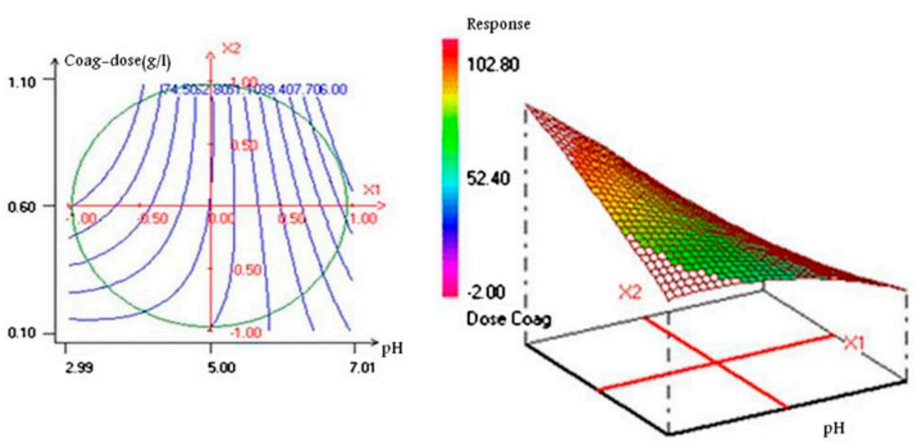

Fig. 3. Three-dimensional response surface and contour plots: the effect of pH (X1) and the dose of coagulant (X2) and their mutual interaction on the COD removal at a constant dose of flocculant (26 mg/l) and MSFP (48 rpm).

are dissolved in water. The influence of pH on chemical coagulation may be considered as a balance of two competitive forces: (1) between H^+ ions and metal hydrolysis products for interaction with organic ligands and (2) between OH^- and organic anions for interaction with metal hydrolysis products [31]. So the results found can be explained by the presence of iron in the form Fe^{3+} in an acid medium. Therefore, the iron added interacts with the negative colloids present in the leachate and neutralizes their charges. When the pH increases the iron salt can react with hydroxyl (OH^-) and form Fe $(OH)_3$ and Fe $(OH)^-$ leading to the COD removal and turbidity decrease [5]. Also, at pH 3, the condition is favorable for the flocculation of organic colloids [32]. In contrast, some research works have showed that the maximum COD removal is obtained at very high pH. For example, Tasti et al. [33] investigated the coagulation–precipitation process efficiency for the treatment of partially stabilized (by re-circulation) leachates, especially in terms of organic matter and solids removal. These authors showed that the highest COD removal value reached about 80% for an iron dosage of 2 g/l and a pH value adjusted to 10. Samadi et al. [34] showed that relatively high removals of suspended solids, and COD were observed at low and high pH values (pH = 4 and 12). The extent of pH range is affected by the types of coagulant used and by the chemical composition of wastewater as well as by the concentration of coagulant. It can be concluded from Fig. 3 that the response (COD removal) was highly enhanced by raising the coagulant dose. As reported by other studies, the COD removal increased with increasing coagulant dosages up to the optimum dosage. Then, it was decreased. This is mainly due to the fact that the optimum coagulant dosage produced flocs having a good structure and consistency. But in doses lower than the optimum one, the produced flocs are small and influence the settling velocity of the sludge.

According to this analysis, the optimal experimental conditions are fixed at pH 3.36, coagulant dose 0.87 g/l, flocculant dose 26 mg/l, and speed of flocculation 48 rpm. To confirm this prediction, an additional experiment was conducted using the optimal conditions. The results obtained show that the COD removal observed (80%) was somewhat lower than the predicted value (85%).

Summarizing the aforementioned results, the pretreatment of landfill leachate by the application of coagulation/flocculation process resulted in the efficient removal of organic and polluting load (Table 5). The fresh leachates did not show any concentrations of heavy metals, superior to standards except for Cr and Fe. The obtained results showed that this process allowed also a decrease in Cr concentration at a level suitable for the legal limits for discharge into urbanized streams. However, the use of ferric chloride during the coagulation–flocculation increased the Fe content in the treated leachates to high level (47 mg/l) that needs subsequent treatment.

Table 5
Organic load removal of landfill leachate after pretreatment by coagulation/flocculation

Parameters	Removal rate (%)
Turbidity	90
COD (mg O_2/l)	80
BOD_5 (mg O_2/l)	40
TKN (mg/l)	35

4. Conclusion

A hybrid design was used to optimize the key operating parameters (pH, coagulant dose, flocculant dose, and mixing speed in the flocculation step) on the coagulation–flocculation process applied for the landfill leachate treatment. The optimal conditions obtained for the desirable response, COD removal, were as follows: pH 3.36, coagulant dosage of 0.87 g/l, flocculant dosage of 26 mg/l, mixing speed in the flocculation step of 48 rpm, and setting time of 30 min. These conditions have been confirmed experimentally and allowed the removal of 80% COD. Despite the significant reduction of the organic load present in the leachate, this treatment still did not fulfill the legal requirements of Tunisian discharge standards in the sewer regarding COD (1,000 mg l^{-1}). Therefore, reduction of COD to the imposed legislative limits can be accomplished by the application of supplementary treatment like adsorption process in an additional step.

Acknowledgments

The authors express their gratitude to the Laboratory, Water Energy and Environment, National Schools of Engineering (Sfax, Tunisia) for providing us with facilities and support during the present study.

We wish to extend our thanks to Mr Jamil JAOUA, Founder and former Mead of the English Department, Sfax Faculty of Science, Tunisia for having edited our paper.

We wish to thank also Dr Annelise HAENNI, from Institut Jacques MONOD (Paris-France) for having edited our paper.

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