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Reducing Pollution of Stabilized Landfill Leachate by Mixing of Coagulants and Flocculants: A Comparative Study

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Abstract

The physico-chemical process of coagulation-flocculation is very efficient and economical for the treatment of leachate. The latter can have considerable impacts on the environment. The leachate from the landfill of the city of Mohammedia is characterized by a high COD content which varies between 2200 and 2700 mg/l, a total Kjeldahl nitrogen concentration varying from 1080 to 1405 mg/l while the ammonium content has a concentration varying between 587 and 1410 mg/l. Organic matter is not readily biodegradable (BOD₅/COD: 0.2 to 0.13). Metal concentrations ranged from 0.1 to 4.2 mg/l for Cr, 40 to 5 mg/l for Cd, and 0.3 to 0.8 mg/l for lead. For monitoring the leachate treatment, several coagulants and flocculants were used (FeCl₃, Al₂(SO₄)₃, Alginate, cationic flocculants, anionic flocculants). In parallel with the monitoring of the physicochemical parameters we followed the production of the volume of the settled sludge over time. Treatment with all coagulants and flocculants used is pH dependent. Ferric Chloride has been shown to be effective at a pH of 6.5 while for Aluminum Sulfate the optimum pH is 5.3. The results showed that coagulation-flocculation by Ferric Chloride and Aluminum Sulfate is very effective in reducing turbidity. This reduction reaches 95 and 98% respectively for FeCl₃ and Al₂(SO₄)₃, while the reduction in COD for the two coagulants is around 60%. Organic flocculants alone do not lead to a significant reduction in turbidity and COD, while their combination with coagulants marks a good reduction in pollution. Hydrated iron hydroxides precipitate more easily than flocs formed by aluminum, resulting in more efficient removal of pollutants than that obtained at lower pH values. The order of introduction strongly influences the coagulation flocculation. The optimal doses of the various coagulants and flocculants chosen for the study vary from one reagent to another. FeCl₃ remains the most suitable coagulant to further eliminate organic and metal pollution. The cost associated with the treatment using flocculants remains much higher when the flocculant is used in admixture with a coagulant.

Keywords: leachate, coagulation and flocculation, test jar, optimization

1. Introduction

Leachate is the juice produced during the fermentation of household waste stored in public landfills, which can pose many problems during the design and maintenance of a landfill. Leachate from landfills has been identified as potential sources of contamination of soil, groundwater and surface water, as it can percolate through soils, causing pollution of waterways and groundwater, s’ they are not properly collected, processed and safely disposed of [1–4].

The climate and landfill are the main factors influencing the production and composition of the leachate. Where the climate is prone to higher levels of precipitation, there will be no more water penetration into the landfill, and therefore no more leachate generated. Another factor is the topography of the site, which influences the runoff regimes and the water balance of the site.

The treatment of landfill leachate is a complicated process due to the type of contaminants it contains and the variation in volume. The percolation of rainwater through municipal landfill waste from leachate produced by the biological and chemical processes that occur in the waste increases the volume of leachate juice.

Figure 1. Diagram of landfill leachate treatment techniques, based on Abbas et al. [6]; Renou et al. [7], Shuokr et al. [8].

The combination of the above factors could generate an effluent whose properties also largely depend on the age of the landfill [2–5].

Solid waste from municipal landfills is considered a very important source of pollution since it contains enormous amounts of organic and mineral matter, some types of organic matter of which are biodegradable, where humic and fluvic acids represent a significant amount in old leachate. Optimizing the factors controlling the treatment of this liquid effluent can greatly increase the efficiency of the process. The physico-chemical treatment process can be applied to landfill leachate without being affected by the toxicity of the leachate and could provide a simple, selective and economically acceptable alternative to traditional methods. The different treatment techniques often used to reduce pollution from leachate discharges are illustrated in **Figure 1** [6, 8].

The flocculation coagulation methods which are the most widely applied for the fight against pollution from leachate discharges [9–12]. State-of-the-art physico-chemical treatment processes have been developed [13, 14] and applied for the decontamination of leachate discharges. These different techniques depend on several parameters: leachate age, quality, cost and waste to be treated (household and industrial waste). Biological treatment processes such as activated sludge is problematic due to the low kinetics of degradation and foam production [15]. In the treatment of leachate many factors can influence the effectiveness of treatment by coagulation flocculation), such as the type and the optimal concentration of the coagulant/flocculant and the optimal pH [16–19]. The sanitary landfill method for final solid waste disposal continues to be widely accepted and used because of its economic benefits in developing countries [20].

The main objective of this study is the optimization of the conditions of the coagulation flocculation process in the treatment of landfill leachate, and the determination of the most appropriate dose for different coagulants and flocculants such as ferric chloride, sulphate aluminum, Alginate, ... at optimum pH which considerably reduces organic matter, suspended solids and metals present in leachate.

2. Materials and methods

2.1 Sampling techniques

The leachate samples were collected from the Mohammedia town landfill in 50 L plastic bins, transported to the laboratory and stored at 4° C. The leachate samples were placed for 2 hours at room temperature before performing the flocculation coagulation tests. Then, the samples were thoroughly shaken to resuspend the deposited solids before further tests were performed.

2.2 Coagulants/flocculant dosage

Several coagulants and flocculants have been tested for the treatment of leachate discharges such as ferric chloride ($\text{FeCl}_3 \cdot 6\text{H}_2\text{O}$), aluminum sulphate ($\text{Al}_2(\text{SO}_4)_3 \cdot 18\text{H}_2\text{O}$), alginate, ... for destabilize suspended solids and colloids and eliminate pollution afterwards.

In addition, two flocculants such as the Astral flocculant and the cationic polyelectrolyte Superfloc supplied by companies in Casablanca, the flocculants Chemic1, Chemic2, Chemic3 supplied by an Italian company.

The experimental process consists of three steps: a rapid mixture of leachate containing coagulation reagents of the flocculation reagents at 160 rpm for 10 min, followed by slow agitation for 20 minutes and 30 rpm, then a step of final settling for 1 hour. The coagulation-flocculation was carried out with the optimized and previously determined operating parameters.

Furthermore, the treatment tests were carried out by the Jar test technique using six polyethylene beakers of volume equal to 1 liter to examine the different doses of coagulant with a well-defined initial pH (optimal pH). The tested samples were mixed thoroughly to resuspend the deposited solids, and the appropriate volume of the sample was transferred to the corresponding beakers. First, the optimum pH for FeCl_3 activity was determined. A known volume of the solution of ferric chloride, aluminum sulfate or other coagulants and flocculants was added to 1 L of landfill leachate at various pH values adjusted with H_2SO_4 and NaOH . To study the optimum dose of coagulant, the pH of the leachate solution is maintained at the optimum value and variable doses of FeCl_3 or $\text{Al}_2(\text{SO}_4)_3$ were then added. After 60 minutes of decantation, the supernatant was taken for analysis. To evaluate the effectiveness of ferric chloride, aluminum sulphate or other coagulants and flocculants for the treatment of leachate, the following parameters were determined: turbidity, chemical oxygen demand (COD)... settled sludge and metallic elements not eliminated.

2.3 Analytical techniques

The turbidity was determined by a HI 93703 microprocessor turbidimeter. The Chemical Oxygen Demand (COD) and the other physicochemical parameters (NTK, total phosphate, etc.) for the characterization of the leachate were determined according to standard methods [21].

The pH of the solutions was measured using an “Accumet Basic AB15 pH-meter” from the Fisher Scientific brand with a combined Ag/AgCl glass electrode according to standard NF T 90-008 February 2001 (T90-008). The conductivity was measured using a conductivity meter brand “YK-2001PH intelligent conductivity pH-meter” according to standard NF EN 27888 January 1994 (T90-031). The COD was carried out according to the AFNOR standard in force [NF T90-101 February 2001 (T90-101)]. The measurement of the biological oxygen demand after five days (BOD5) was facilitated by the use of the gauge method [(EN 1899 May 1998) (T90-103)] using the BOD5 meter of the VELP brand. The absorbance measurements were carried out using a UV-visible spectrophotometer.

Spectrophotometer 9,200 RAYLEIGH 2-beam 1 nm bandwidth. Turbidity was measured using a turbidimeter according to NF EN ISO 7027 March 2000 (T 90-033). The determination of the materials in suspension was carried out by the centrifugation method of the standard [NF T 90-105 January 1997 (T 90-105)]. The phenolic compounds were determined by the colorimetric method using the Folin–Ciocalteu reagent [22]. The nitrate assay was carried out by the spectrometric method in the presence of sulfosalicylic acid according to standard EN ISO 78-90 January 1997 (T 90-045). The determination of the total phosphorus was carried out by spectrometric method according to NF T 90-023 January 1997. The determination of ammoniacal nitrogen NH_4 was carried out by the spectrophotometric method with indophenol blue according to AFNOR NF T 90-015 January 1997. The method of heavy metal analysis is atomic absorption spectrophotometry with a graphite furnace (VARIAN AA 20 model).

3. Results and discussions

3.1 Leachate characteristics

To assess the impacts of a landfill on the environment, it is necessary to characterize the effluents it generates. Indeed, whatever the mode of operation of a landfill, the leachate constitutes, if it is not treated before its discharge, a source of

Parameters	Values	Average	Moroccan Standards
pH	7.7–8.92	8.3	6.5–8.5
Conductivitéms/cm	25.6–35.9	31.2	2.7
Turbidity NTU	63–140	102.2	—
HPO ₄ ²⁻ mg/l	592.4–2128	1693.3	—
Sulphate mg/l	77.47–218.7	156.04	—
Tot phosphate mg/l	1226.6–2217	1879.5	10
TKN mg/l	1080–1405	1289.75	30
COD mg/l	2153–2707	2473.9	500
BOD5 mg/l	526–290	399	—
BOD5/COD	0.2–0.135	0.16	—
O ₂ mg/l	0–0.2	0.03	—
NO ₃ ⁻ mg/l	36.3–453.9	173.2	—

Table 1.
Physico-chemical parameters of leachate (mg/l) from Mohammedia landfill.

nuisance which is added to the numerous problems of contamination of the surrounding environment. These liquids loaded with mineral and organic substances resulting from the decomposition of waste can be entrained by runoff and reach surface water, or infiltrate through the bedrock of the landfill and contaminate the water of the water table which is not deep.

The different physicochemical parameters analyzed in the landfill leachate are presented in **Table 1**. In addition, the results of the analysis of metals (Cu, Zn, Cr, Ni, Pb, Sb and Sn) in the three sampling companies were carried out (**Table 2**).

The characteristics of a leachate can generally be represented in terms of basic parameters such as COD, BOD, BOD5/COD ratio, color, pH, content of metallic elements [4]. These results showed that the leachate is characterized by high concentrations of organic matter and high concentrations of ammonium and nitrogen compound (**Table 1**). The organic matter, in terms of COD varies between 2153 and 2707 mg/l (stabilized leachate), while high concentrations of ammoniacal nitrogen (587 to 1410 mg/l) and NTK (1080 to 1405 mg/l) have been detected. The main concentrations of heavy metals are between 0.1 to 4.2 mg/l for Cr, 0.04–0.005 mg/l for cadmium and 0.8 to 0.3 mg/l for Pb. The characterization of the average leachate showed that the BOD5/COD ratio varies between 0.2 to 0.13, which shows that the leachate is rich in organic matter and not biodegradable (humic and fulvic substances).

In addition, turbidity and conductivity are very high values, far exceeding the standards for treated wastewater. This shows that decision-makers in Morocco must make an effort to save the current situation of public landfills in order to protect the

Sampling date	Cu	Zn	Cr	Cd	Ni	Pb	Sb	Sn
25/03/2000	0.7	0.2	0.1	0.035	0.3	0.5	1.3	—
10/8/2002	1.2	2.3	4.2	0.018	0.4	0.3	0.9	—
5/10/2007	1.2	1.9	3.2	0.005	0.2	0.8	0.5	0.6

Table 2.
Analysis of the metallic elements of leachate from the Mohammedia landfill.

health of the population. The low values of the BOD5/COD ratio (0.2–0.13) thus show that the leachate is rich in non-biodegradable organic matter, which can cause several impacts on surface water and groundwater [2]. Souabi et al. [1], showed that for leachate discharges from Mohammedia (main collector), the BOD5/COD ratio varies between 0.2 and 0.13, showing that the leachate is not easily biodegradable and can therefore cause several impacts on surface water. (Oued El Maleh). The same authors have shown that the COD values obtained vary between 2301 and 2750 mg/l and remain much lower than the content detected by [23] discharged into the sea. The variation in the characteristics of the leachate has been attributed to many factors, such as variations in the composition of the solid waste, the age of the landfill, the hydrogeology of the landfill, precipitation and specific weather conditions. and waste moisture [7, 24, 25]. From 5 to 10 years, the landfill is known as “middle age” and its leachate has a COD of between about 3000 to 15000 mg/l [20]. After 10 years, a landfill contains less biodegradable material and the leachate has a COD value of less than 2000 mg/l [4, 11].

For a given leachate, fluctuations in pH, flow, COD and BOD5 contents, etc. have been observed over time [4, 11]. This can influence the efficiency of the elimination by different treatment techniques and disturb the receiving environment if the leachate is rejected without treatment, which justifies the installation of a homogenization basin. The age of the landfill is one of the main factors that also influences the characteristics of the leachate.

Landfill leachate contains chemicals, including metal ions such as iron. Indeed, efficient and cost-effective leachate treatment techniques are difficult to implement [26]. Rivas and Gimento [27] pointed out that old landfills produce stabilized leachate with a relatively low COD content ranging from 500 to 5000 ppm, a slightly basic pH of 7.5 to 8.5, low biodegradability (BOD5/COD less than 0.1) and a significant amount of heavy metals and high molecular weight compounds (humic and fulvic substances). Leachate exhibits considerable variation in composition [11]. The concentration of contaminants are mainly influenced by the age of the landfill, as well as by the type of waste deposited at the landfill and other hydrogeological factors [11, 17].

The characterization of the average leachate has shown that the BOD5/COD ratio varies between 0.2 to 0.13, which shows that the leachate is rich in organic matter and not biodegradable (humic and fulvic substances) which can cause several impacts on the water surface and groundwater [28].

3.2 Leachate settling

The study of the elimination of leachate pollution by sedimentation is presented in **Table 3**. The settling time varies around 120 minutes. The supernatant was collected to analyze the COD after a stabilization time of 120 minutes.

Raw leachate (COD mg/l)	Decanted leachate (COD mg/l)	Abatement %
2707	1800	34.5
2301	1632	29
2153	1536	29.5
2688	2544	5
2240	1728	23
2540	1866	26.5

Table 3.
Study of leachate sedimentation for six sampling campaigns settling time 120 min.

The COD removal rate varies between 5 and 34.5%, which is essentially a function of the quality of the leachate collected. Indeed, sedimentation remains essential to reduce leachate pollution at the lowest cost.

Settling is a process to reduce turbidity by allowing the water to stand for 2–24 hours for the particles to settle to the bottom of the basin. The advantage of settling is that it does not require any equipment other than settling tanks. However, the drawbacks of this method require several settling tanks when the polluting load and the flow rate are high.

The removal of suspended particles by sedimentation depends on the size and density of these particles. Settleable solids are measured as the visible volume accumulated at the bottom of an Imhoff cone after settling for one hour.

Unhindered settling is a process that removes discrete particles at a very low concentration without interference from nearby particles. The re-stabilization of the system by adding excess coagulant is not effective, since increasing the settling time promotes clarification of the supernatant beyond the critical coagulation concentration and the charge reversal point.

3.2.1 Effects of Fe^{3+} and Al^{3+} on the removal of leachate pollution

The coagulation results for the $FeCl_3$ and $Al_2(SO_4)_3$ leachate are shown in **Figures 2 and 3**.

The comparative study of coagulation-flocculation by Fe^{3+} and Al^{3+} is given in the **Table 4**.

3.2.2 Effect of pH on $FeCl_3$ leachate coagulation

In the coagulation-flocculation process [29, 30]. It is very important to adjust the pH since coagulation occurs within a specific pH range for each coagulant and depending on the type and characteristic of the raw effluent to be treated.

The COD removal efficiency tests, using $FeCl_3$ were performed for pH values between 2 and 12 (**Figure 4**). The optimum pH of the raw leachate, before the addition of the coagulant is 6.5. At this pH, the COD and the turbidity dropped by 84% and 96% respectively. This conclusion is in agreement with that of [27].

Figure 2.
Effect of $FeCl_3$ on the elimination of leachate pollution. Experimental conditions: initial COD of the leachate = 3175 mg/l and initial turbidity = 128 NTU, pH = 7.8.

Figure 3. Effect of $\text{Al}_2(\text{SO}_4)_3$ on COD removal, turbidity and settled sludge. Experimental conditions: initial COD of leachate = 2400 mg/l, initial turbidity = 140 NTU, pH = 7.8.

	Optimal concentration mmol/l	%Turbidity removal	%COD removal	Sludges ml/l	optimal pH
Fe^{3+} mmol/l	18.5	95	67	800	6.5
Al^{3+} mmol/l	5.82	98	60	230	5.3

Table 4. Comparative study of coagulation-flocculation by Fe^{3+} and Al^{3+} .

Figure 4. Effect of pH on leachate coagulation using FeCl_3 . Experimental conditions: (optimal FeCl_3 concentration: 3500 mg/l), initial leachate COD = 3168 mg/l, initial turbidity = 110 NTU and pH 7.8.

3.2.3 Effect of pH on leachate coagulation using $\text{Al}_2(\text{SO}_4)_3$

Figure 5 shows the effect of pH on leachate coagulation using aluminum sulfate. The results show a removal of turbidity which varies around 84% at pH 5.3. The maximum sludge volume, generated at optimum pH, is 240 ml/l. Partial dissolution of aluminum oxide and the appearance of a net positive charge on the surface should be considered.

Figure 5.
Effect of pH on leachate coagulation using $\text{Al}_2(\text{SO}_4)_3$. Experimental conditions: (optimal concentration of $\text{Al}_2(\text{SO}_4)_3$; 1000 mg/l), initial leachate COD = 2.400 mg/l, initial turbidity = 140 NTU and pH 7.8.

Flocs generated under strongly acidic or basic conditions were significant, but in very small numbers, especially in an acidic environment. The reduction in pH is due to the acidic character of Al^{3+} from $\text{Al}_2(\text{SO}_4)_3$. For the other coagulant doses tested, the flocs were microscopic and similar in size.

4. The effect of coagulants and flocculants on the removal of COD and turbidity

4.1 Effect of the mixture: Astral Flocculant + FeCl_3

To improve the elimination of leachate pollution, we tested a flocculant supplied by the company. This flocculant is currently used at the wastewater treatment plant which treats the wastewater of company by coagulation flocculation. The effect of Astral flocculant on turbidity and COD removal is shown in **Figure 6**.

Figure 6.
Reduction of turbidity and COD by the Astral Flocculant alone.

The results show that the COD and turbidity parameters decrease with the increase in the concentration of Astral flocculant. The optimum concentration of the flocculant varies around 200 mg/l. From this value optimal, the concentration of COD and turbidity increases with the increase in the concentration of flocculant. The COD goes from 3300 mg/l to 1600 mg/l and the turbidity goes from 140 to 40 NTU with a removal efficiency of 53% and 71% respectively for the COD and the turbidity.

Indeed, the mode of action of polymers is however very different and depends mainly on their electrical charge properties and the concentration of flocculant. The stabilized leachate may contain too much organic matter (biodegradable, but also refractory to biodegradation) consisting mainly of humic substances [31, 32] as well as ammoniacal nitrogen, heavy metals, organochlorines and inorganic salts [33].

The pH is an essential parameter to be taken into account for the physicochemical treatment of liquid discharges by coagulation flocculation. Indeed, we have studied the effect of pH on coagulation flocculation by the flocculant Astral. The results are shown in **Figure 7**. The effect of pH on the elimination of pollution from leachate discharges by coagulation flocculation has shown that there is no variation in turbidity and COD in the field. With a pH of between 2.5 and 12. The quantity of sludge formed during treatment remains lower. In addition, the results showed that the turbidity of raw leachate increases from 115 NTU to 40 NTU starting from acidic pH 2.5 while the COD goes from 3300 mg/l to 1550 mg/l with a yield of 65% and 53% respectively for turbidity and COD.

The study of coagulation flocculation by FeCl_3 (2500 mg/l) in the presence of variable concentrations of Astral flocculant is illustrated in **Figure 8** while monitoring the performance by measuring the parameters of COD, Turbidity and sludge production. These results showed that the increase in the concentration of Astral Flocculant made it possible to considerably reduce the COD and turbidity parameters while increasing the production of sludge. The turbidity value stabilizes around 15 NTU while it appears that the COD is around 1150 mg/l for a flocculant concentration of 600 mg/l with a yield of 62% and 89% respectively for the COD and the turbidity. The volume of sludge produced is around 800 ml/l with complete elimination of the coloring. at pH 6,5.

Furthermore, the study of the reduction in the pollution of leachate discharges by FeCl_3 in the presence of a constant concentration of Astral flocculant (200 mg/l) is given in **Figure 9**. The results have shown that the increase in

Figure 7.
Effect of pH on the reduction of turbidity and COD by the Astral Flocculant alone (200 mg/l).

Figure 8.
Study of the mixture: FeCl_3 (2500 mg/l FeCl_3) + Astral flocculant variable flocculant COD = 3056 mg/l, pH = 6.5.

Figure 9.
Study of the mixture: FeCl_3 + ASTRAL flocculant (200 mg/l) variable FeCl_3 . COD = 3100 mg/l, pH = 6.5.

addition of FeCl_3 increases the removal of COD and turbidity while increasing the production of sludge that requires treatment. It appears that the optimum concentration varies around 5000 mg/l of FeCl_3 for a pH of 6.5. Indeed, in our study, the elimination of leachate pollutants was improved by the addition of cationic polyelectrolytes (Astral and Lesieur Flocculant). It should be noted that at this stage hydrolysis, precipitation and adsorption reactions of leachate pollutants are greatly affected by the presence of humic and fulvic substances. Indeed, specific interactions can appear between humic substances, the surface of flocculates and dissolved ferric species, influencing the efficiency of the coagulation-flocculation process. The effect of a coagulant and flocculant mixture addition on the removal of COD and partially stabilized leachate turbidity could improve leachate remediation efficiency.

4.2 Effect of the mixture: Flocculant Lesieur + FeCl₃

To get an idea of the effectiveness of the leachate treatment by another flocculant used by the company Lesieur in Casablanca for the depollution of wastewater by SBR (sequential batch reactor), we carried out coagulation flocculation tests by the flocculant alone or mixed with FeCl₃. The effect of the Lesieur flocculant on the removal of turbidity and COD is illustrated in **Figure 10**. The results showed that the COD and turbidity parameters decrease with increasing concentration of Lesieur flocculant. The optimum concentration of the flocculant varies around 200 mg/l (**Figure 10**). From this value the COD concentration increases with the increase in the flocculant concentration. The results showed that the COD goes from 3300 to 1300 mg/l and the turbidity goes from 140 to 35 NTU (optimal value) with a removal efficiency of 39% and 75% respectively for the COD and the turbidity.

To compare the effect of pH on Astral flocculant versus Lesieur flocculant, we studied the effect of pH on coagulation flocculation by Lesieur flocculant. The results are shown in **Figure 11**. The effect of pH on the elimination of pollution from leachate discharges by coagulation flocculation has shown that the optimum

Figure 10.
Reduction of turbidity and COD by Lesieur Flocculant alone COD = 3300 mg/l, Turb = 130 NTU.

Figure 11.
Effect of pH on the reduction of turbidity and COD by Lesieur Flocculant (200 mg/l) COD = 3300 mg/l, Turb = 115 NTU.

Figure 12.
Study of the mixture: FeCl_3 + Flocculant Lesieur (200 mg/l flocculant) variable FeCl_3 . COD = 3200 mg/l, Turb = 130 NTU to be reviewed.

Figure 13.
Study of the mixture: FeCl_3 (2500 mg/l) + Lesieur Flocculant. Turb = 125 NTU.

pH varies around 6. In addition, the turbidity of raw leachate increases from 115 NTU to 30 NTU while the COD drops from 3300 mg/l to 1550 mg/l for the optimal concentration of 200 mg/l of the flocculant with a yield of 74% and 54% respectively for turbidity and COD.

FeCl_3 as a coagulant alone or as a mixture with other flocculants remains the most appreciated for better elimination of pollution from leachate discharges [34] in the form of COD and turbidity. We therefore studied the effect of FeCl_3 (200 mg/l of the flocculant, pH = 6) on the Lesieur flocculant.

Figure 12 shows that the mixture (FeCl_3 + Flocculant Lesieur) made it possible to reduce pollution, with a reduction in COD from 3200 mg/l (crude) to 800 mg/l for an optimal concentration of 200 mg/l of flocculant of Lesieur and 3000 mg/l of FeCl_3 while the turbidity has a minimum value for a concentration of 100 mg/l of FeCl_3 . The volume of the settled sludge increases to reach a maximum of 200 ml/l for the concentration of 3000 mg/l of FeCl_3 as the optimum value (**Figure 12**).

To get an idea of the efficiency of the leachate treatment with the cationic flocculant used at the Lesieur company wastewater treatment plant in Casablanca, we studied the flocculation coagulation of leachate discharges by fixing the FeCl_3 concentration while at the same time varying that of the lesieur cationic flocculant. The results are illustrated in **Figure 13**. These results showed that the mixture made it possible to reduce the pollution, with a decrease in the COD from 3200 mg/l (crude) to 1500 mg/l (200 mg/l of Lesieur flocculant). The volume of the settled sludge increases to reach a maximum of 220 ml/l. Certainly, it can be concluded that the optimal dose of the cationic flocculant in the mixture (FeCl_3 + Lesieur flocculant) (FeCl_3 2500 mg/pH 6) is 20 mg/l for the lesieur flocculant.

4.3 Effect of the mixture: Alginate + FeCl_3

4.3.1 Alginate alone

The study of the elimination of the pollution of leachate by coagulation flocculation by the alginate alone is given in **Figure 14**. The results obtained showed that the optimal dose of alginate for the elimination of COD and the turbidity are respectively 60 and 75% with an optimal dose of 120 mg/l of alginate, while producing a small quantity of sludge (20 ml/l).

The effect of pH on alginate flocculation coagulation (**Figure 15**), showed that the latter has little effect on the removal of COD and turbidity. For a pH between

Figure 14.
Effect of alginate alone on COD removal and turbidity Turb = 125 NTU.

Figure 15.
Effect of pH on the reduction of turbidity and COD by alginate (120 mg/l).

1 and 10, no sludge production was observed for Alginate concentrations below 800 mg/l, while the volume of sludge increases from pH 10. Furthermore, the study of the elimination of pollution from leachate discharges by the mixture of FeCl_3 and alginate is given in **Figure 16** (fixed alginate: 120 mg/l and variable FeCl_3).

The results obtained showed that the mixture of FeCl_3 and Alginate has an optimal concentration of around 120 mg/l of Alginate and a variable concentration of FeCl_3 increases the COD from 2900 to 1100 mg/l with a yield 60% COD 65% turbidity. The COD value decreases with the increase in the FeCl_3 concentration. As for the variation in turbidity, this remains negligible (30 NTU) from a concentration of 120 mg/l of alginate. The amount of sludge formed increases with an increase in alginate. The optimal concentrations for the removal of COD and turbidity by Alginate vary around 20 mg/l with a FeCl_3 concentration of 2500 mg/l at pH 6.5.

Figure 16.
Study of the mixture: Alginate (120 mg/l) + variable FeCl_3 .

Figure 17.
Study of the mixture FeCl_3 (2500 mg/l) + alginate.

Figure 18.
Variation of turbidity (a) and COD (b) during coagulation-flocculation by flocculants supplied by an Italian company.

Furthermore, **Figure 17** illustrates the effect of Alginate on flocculation coagulation while keeping a fixed concentration of FeCl_3 (2500 mg/l).

The results showed that the Alginate and FeCl_3 mixture has an optimal concentration of around 2500 mg/l, which resulted in very high COD removal with a 50% removal efficiency. The COD value increases with that of the Alginate. The turbidity which varies around 30 NTU and remains constant from 50 mg/l of Alginate, while the volume of sludge produced increases with the increase of the Alginate stabilizes around 125 ml/l. The optimum concentration of Alginate is 120 mg/l. COD and turbidity values for optimum FeCl_3 and Alginate values show values of 1360 mg/l and 30 NTU, respectively.

4.4 Effect of flocculants chemec 1, 2 and 3 supplied by an Italian company

We carried out a comparative study on the elimination of pollution from leachate using three flocculant (Chimec 1, 2 and 3) supplied by an Italian company. The results (**Figure 18a**) showed that the turbidity decreases from 190 to 40 NTU for Chimec 1,2, for an optimal concentration of 4 mg/l then increases to 100 NTU for 30 mg/l of Chimec1 and 2. As regards the flocculant Chemic 3, the results showed a final elimination of the turbidity which is around 60 NTU for the same optimal concentration (4 mg/l) while the turbidity remains stable for Chimec 3 for a concentration of 10 mg/l.

As for the COD content (**Figure 18b**) during the treatment with the three flocculants, the results showed a decrease up to 1500 mg/l for 4 mg/l of flocculant, with an elimination efficiency of 40%, then increased for Chimec 1 and 2 arriving at around 2000 mg/l for 30 mg/l of flocculant. The COD remains almost stable for Chimec3 from 4 mg/l. In fact, no significant difference was observed as regards the elimination of COD by the three flocculants while producing a small amount of sludge in the case of the three flocculants.

5. Comparative study of the elimination of leachate pollution by different coagulants and flocculants

The comparative study on the elimination of turbidity and COD by different coagulants and flocculants is shown in **Figure 19**.

These results show that FeCl_3 gives a significant removal of COD and Turbidity with a high yield compared to other coagulants and flocculants. Aluminum sulphate allows elimination comparable to that obtained with FeCl_3 . With regard to the flocculants, the results showed that the Astral flocculant alone allowed a very interesting elimination of 53% and 75% respectively for the COD and the turbidity. No significant difference was observed for the removal efficiency of COD and turbidity by the flocculants Chimec supplied by the Italian company.

The effect of optimal concentrations of coagulant FeCl_3 and flocculants (Astral, Supefloc, Alginate) alone or mixed on the production of sludge produced during the coagulation/flocculation process Coagulant type and dose volume of sludge is given in **Table 5**.

The addition of organic flocculants has been examined to improve the removal of organic matter.

FeCl_3 + Superfloc showed a removal efficiency of 30 and 66% respectively of COD and turbidity (**Figure 20**). These values are lower than the results obtained for the FeCl_3 + Astral or FeCl_3 + Alginate mixture.

Figure 19.
Comparative study of the elimination of COD and turbidity by different coagulants and flocculants.

	Optimal concentrations alone mg/l	Optimal concentrations in mixture mg/l FeCl ₃ + Flocculants	Sludge in mixture ml/l FeCl ₃ + flocculants
FeCl ₃	2500	—	—
Astral	192	500	800
Superfloc	200	140	230
Alginate	120	20	80

Table 5.
Effect of optimal concentrations of coagulant and flocculants alone or mixed on the production of sludge.

Figure 20.
Effect of FeCl₃ mixture, Superfloc flocculant, Astral flocculant using optimal doses on reduction of COD and turbidity.

The combined action of ferric chloride-polyelectrolyte mixtures was investigated. Addition of flocculant at a coagulant: flocculant ratio (Astral 2.5: 0.5, Superfloc 2.5: 0.14 Alginate 2.5: 0.02) (g), gave removal capacities of COD and variable turbidity (**Figure 6**).

Similar results were observed, during the addition of polyelectrolyte K1370 and A321 to stabilized leachate samples without pH correction [26]. Addition of polyelectrolyte superfloc did not substantially affect organic matter removal, which did not exceed 30%, as compared with the results FeCl₃ + Astral (62%), and FeCl₃ + Alginate (50%). However, the removal of pollutants was enhanced during the addition of polyelectrolytes in the samples. It should be mentioned at this point that hydrolysis, precipitation and adsorption reactions of ferric cation in leachate samples are greatly affected by the presence of humic substances. Specific interactions may appear between the humic substances, the surface of flocculates and the dissolved ferric species, influencing the efficiency of coagulation–flocculation process.

6. Removal of heavy metals

The removal of metallic elements by ferric chloride FeCl₃ is given in **Table 6**. The concentration of heavy metals (Zn, Cu, Cd, Cr and Ni) in the leachate exceeds the maximum authorized values.

The flocculation coagulation made it possible to eliminate a yield varying between 80 and 99% (**Table 6**) for the various metallic elements studied (Cu, Zn, Cr, Cd, Ni, Pb and Sb). The results concerning the elimination of Chromium obtained during the present study are comparable with the results obtained for the treatment of leachate with FeCl₃ [35] with a removal efficiency of 71% during the treatment with FeCl₃ alone, whereas treatment with the latter in the presence of lime resulted in a yield of 90% of chromium [35, 36]. The same authors have shown that the aluminum sulphate allowed an elimination yield of 83% (**Table 7**).

Landfill leachate	Cu	Zn	Cr	Cd	Ni	Pb	Sb
Before Traetment	1.20	2.30	4.20	0,02	0.35	0.30	0.90
After treatment	0.10	0.01	0.21	—	0.12	0.01	0.03
Removal	91.70	99.6	95	—	80	97	97

Table 6.
Analysis of metals in stabilized leachate before and after coagulation flocculation-using FeCl₃ (concentration mg/l).

	As mg/l	Cd mg/l	Cr mg/l	Cu mg/l	Fe mg/l	Mn mg/l	Ni mg/l	Pb mg/l	Sb mg/l	Zn mg/l
Raw leachate	0.14	4.36	2.04	0.4	33.72	2.58	3.74	0.18	0.63	4.68
FeCl ₃	< 0.01	<0.01	0.6	0.03	232.4	—	0.33	0.07	0.44	0.53
FeCl ₃ + lime	< 0.01	—	0.18	0.03	4.50	0.42	0.23	<0.01	—	0.14
Al ₂ (SO ₄) ₃	< 0.01	—	0.33	0.03	4.91	—	0.19	<0.01	—	0.32
Al ₂ (SO ₄) ₃ + lime	< 0.01	—	0.20	0.02	0.22	0.04	0.12	<0.01	—	0.15

Table 7.
Comparative study of the removal of metals by coagulation flocculation [35].

Lime has been used traditionally as a coagulant in the treatment of leachate for many years, requiring doses of around 1 to 15 g/l [35]. Amokrane et al. [37] have shown that the removal of heavy metals, such as Fe, Cd, Cr, reaches up to 90% efficiency during lime treatment. In fact, the removal of COD and heavy metals can be obtained at a pH of 10, which results in a yield of 28% and 86% respectively for COD and heavy metals, with the addition of 2,000 mg/l of ferric chloride [38–40]. In this study, the results indicate that iron (III) chloride [L5] coagulation-flocculation is very effective in reducing metallic elements; the reduction reaching 80 and 99.5% at optimal concentrations of 18.5 mmol/l Fe^{3+} achieved at a pH of 6.5. Similar results were obtained during the coagulation flocculation of leachate discharges from the city of Fez landfill by FeCl_3 [37].

Previously, the low removal of heavy metals in the leachate landfill was observed by Silva et al. [41] during the coagulation/flocculation process.

7. Effects of coagulation flocculation on sludge production

In general, the amount and characteristics of the sludge produced during the coagulation flocculation process are highly dependent on the specific coagulating and flocculating agent used and the operating conditions. The volume of wet sludge settled after the coagulation flocculation process is an interesting parameter to take into account when choosing the coagulant used for the treatment. The volume (ml/l) of the settled sludge can be is represented as a function of the dose of the coagulant.

The effect of the optimal concentrations of FeCl_3 coagulant and flocculants (Astral, Supefloc, Alginate) alone or in mixture in the production of sludge is given in Table 6.

The addition of organic flocculants has been investigated for improving the removal of organic material. The FeCl_3 + Superfloc mixture showed a removal efficiency of 30 and 66% respectively of COD and turbidity. These values are higher than those obtained in the case of the FeCl_3 + Astral or FeCl_3 + alginate mixture.

From Table 8, it is observed that the volume of sludge produced was considerably reduced with increasing dose of polyelectrolyte in the coagulation process. This may be due to the cationic nature of the polyelectrolyte used in this study. In fact, the high molecular weight of the polyelectrolyte allowed an improvement in the growth of the particles, which facilitates the settling. The cationic flocculant also has the ability to attract and retain colloidal particles at the polar sites of the molecule. In general, organic polymers generate less sludge than inorganic salts since it does not add weight or combine chemically with other ions in the water to form a precipitate. Thus, the sludge produced by the use of ferric chloride in combination with a polyelectrolyte is greatly reduced.

Reagents	Optimal concentrations mg/l	Optimal concentration in the FeCl_3 + flocculants mixture mg/l	Sludge in the mixture ml/l FeCl_3 + flocculants
FeCl_3	2500–2400	2500	—
Astral flocculant	192	500	800
Superfloc	200	140	230
Aliginate	120	20	80
Cactus ml/l	32	25	195

Table 8.
Effect of optimal concentrations of coagulants and flocculants alone or in mixture on sludge production.

The combination of Ferric Chloride, Alginate and Superfloc produces less sludge (ml/l) than when Ferric Chloride is used with Astral Flocculant.

8. Effect of the order of introduction of coagulants and flocculants

The effect of the order of introduction of the flocculant FeCl_3 and Astral on the removal of organic matter (COD), turbidity and sludge production are shown in (Figure 21).

Beyond the type of tests we performed in coagulation, we tested the influence of the method of adding the coagulant and flocculant on the properties of the leachate destabilized by coagulation/flocculation. In practice, in addition to the procedure described above where the reagents were added separately (coagulant + flocculant, flocculant + coagulant), we carried out a simultaneous addition of the coagulant and the flocculant. The protocol is then similar to that of coagulation on Jar test, but instead of the coagulant alone, the flocculant is added after the coagulant.

The results obtained show that when ferric chloride (FeCl_3) is introduced first, followed by the flocculant, the yield of COD and turbidity varies around 81% and 79% respectively. When FeCl_3 is introduced at the same time as the flocculant, the turbidity is considerably reduced (56%), while the COD is around 78% which does not show a significant difference between when FeCl_3 is introduced first, followed by the flocculating agent. In addition, the results obtained when the flocculant is introduced before FeCl_3 showed a significant decrease in COD (59%) and turbidity (58%). However, the reduction in organic matter (COD) during the addition of coagulant-polyelectrolyte mixtures is greater than the results obtained by the addition of similar and single doses of FeCl_3 . In addition to the removal of pollutants, sludge production was considered in this work, as it may affect the economic feasibility of the proposed treatment method. The sludge produced during the physico-chemical treatment of landfill leachate (Table 9) is composed by the amount of colloidal organic matter and suspended solids, as well as by the compounds formed due to the possible addition of chemical reagents.

The quantity of sludge produced by the introduction of FeCl_3 followed by the flocculant is 200 ml/l (compacted sludge) which remains low compared to the results obtained by introducing the Astral flocculant the first followed by FeCl_3

Figure 21.
Effect of the order of introduction of the dose dose of coagulant and flocculant FeCl_3 and Astral on the elimination of organic matter (COD) and turbidity.

Order of introduction	Sludges ml/l
FeCl ₃ + Flocculant	200
9Flocculant + FeCl ₃	300
Mixture (FeCl ₃ + Flocculant)	750

Table 9.
Effects of the order of introduction of the dose of FeCl₃ and Astral flocculant on sludge production. Optimal concentrations: FeCl₃: 2500 mg/l, Flocculant: 500 mg/l and pH = 7.

(300 ml/l). In addition, the study of the coagulation of the mixture (flocculant + FeCl₃) showed a large quantity of uncompacted sludge (750 ml/l). In conclusion, the addition of FeCl₃ first, followed by the flocculant results in a small amount of compacted sludge volume.

9. Discussion

Knowing the composition of the waste (household waste + industrial waste) makes it possible to assess the risks of percolation through the soil (leaching). During the solubilization by rainwater, metal oxides and chemicals, often highly toxic contained in the waste, pass into the leachate water. Ecologically, the alarm bells have already been sounded, because in addition to the air pollution, the waters of the Oued Al Maleh which is close to the landfill (10 m) are heavily polluted due to the leachate produced by the old public dump of Mohammedia [1, 4].

To assess the impact of a landfill on the environment, it is necessary to characterize the effluents it generates. Indeed, whatever the mode of operation of a landfill, the leachate constitutes, if it is not treated before its discharge, a source of nuisance which is added to the numerous problems of contamination of the surrounding environment. These liquids loaded with mineral and organic substances resulting from the decomposition of waste can be carried away by runoff and reach surface water, or infiltrate through the substratum of the landfill and contaminate the water of the water table which is not deep (case of the city of Mohammedia). The results of **Tables 1** and **2** have shown that the leachate is characterized by high concentrations of organic matter, ammonium and nitrogen compounds..... The leachate from these separate stages contains different constituents and therefore different characteristics.

In the coagulation - flocculation process [4]. It is very important to adjust the pH since coagulation occurs within a specific pH range for each coagulant and flocculant and depending on the type and characteristic of the raw effluent to be treated.

Coagulation/flocculation is a commonly used process for wastewater treatment in which reagents such as ferric chloride and/or ferric polymer are added to landfill leachate in order to destabilize colloidal materials. This causes the agglomeration of the small particles into easily settling flocs. Several studies have reported the examination of this process for the treatment of landfill leachate, in particular in relation to the optimization of the performance of coagulant/flocculant, the determination of the experimental conditions, the evaluation of the pH, and the search for the addition of flocculant [4, 24].

To improve the coagulation/loculation process, the addition of some commercial polyelectrolytes has been investigated, including two cationics (Astral and Superfloc). The treatment of the leachate by coagulation flocculation using several coagulants has shown that the percentage of elimination of various pollution parameters such as COD, BOD5, turbidity and sludge formed during the treatment vary with the type of coagulant or flocculant chosen for study.

The optimal process variables for landfill leachate coagulation using Fe (III) and Al (III) were found at pH 6.5 for Fe (III) and 5.3 for Al (III) at coagulant doses of 18.5 mmol/l Fe (III) and 5.82 mmol/l Al (III). Although the doses required were identical (0.035 mol/l of Fe^{3+} or Al^{3+}), with an initial COD concentration of 4100 mg/l, FeCl_3 led to a higher elimination of organic compounds (55%) than 'aluminum (42%). Indeed, the results obtained during our study made it possible to show that FeCl_3 is more interesting in terms of reducing pollution, in particular the elimination of COD and turbidity with a yield of 95 and 67% respectively and the COD. In addition, the results showed that the pH considerably affects the coagulation and flocculation by the different coagulants and flocculants tested. The decrease in pH due to Fe + Al cations which present acidic characteristics; therefore, hydrolysis took place under acidic conditions caused by precipitation of metal hydroxides. Several studies have examined coagulation-flocculation for the treatment of landfill leachate, aimed at optimizing performance, namely selection of the most suitable coagulant, determination of experimental conditions, evaluation of the effect of pH, and the choice of flocculant [42]. In addition, during the precipitation process, coagulation, flocculation and adsorption appear to contribute to the removal of organic matter from the leachate [43, 44].

As for other coagulants and flocculants the results showed the yield of COD and turbidity varies from one reagent to another. The Astral and Lesieur flocculants in a mixture with FeCl_3 were able to improve the reduction of leachate pollution. No significant difference was observed in the reduction of COD and turbidity by the flocculants supplied by the Italian company (Chimic 1, 2, 3) during flocculation coagulation. In addition, the order of introduction of FeCl_3 and Astral flocculant showed that the reduction of pollution (COD and Turbidity) is better when introducing FeCl_3 first (elimination of 81% of COD and 79% of turbidity) while the introduction of the flocculant before the FeCl_3 resulted in a yield of 59% of the COD and 58% of the turbidity.

It was shown by Hamzeh et al. [45] that at an initial concentration of 5690 mg/L and at a pH of 4.8, a maximum COD removal of 56% was obtained with 0.8 g/l of FeCl_3 , compared to 39% with 0.4 g/l of $\text{Al}_2(\text{SO}_4)_3$. In addition, Ez Zoubi et al. [46] showed during the treatment of leachate produced by the public landfill of the city of Fez with FeCl_3 that the COD content goes from 53199.6 mg/l to 41933 mg/l with a yield of 21% while Bakraouy et al. [25] showed that FeCl_3 could remove a yield of 65% of COD. This shows that the pollution removal efficiency varies with the coagulant used, the age and the quality of the leachate.

The treatment of leachate is a difficult and expensive process [25]. Although, the leachate can be treated biologically, the COD removal efficiency is generally low due to the high ammonium ion content and the presence of toxic compounds such as metal ions. The results suggest that FeCl_3 may be a viable coagulant in managing COD problems associated with landfill leachate. In addition, the treatment of the leachate by coagulation-flocculation has led to a reduction in the physicochemical (COD, BOD5, ...) and suitable microbiological parameters while the coagulation flocculation combined with a biological treatment of SBR type has allowed eliminate 99% of the COD from leachate produced by the public landfill in the city of Fez [47].

As regards the study of the elimination of metals by coagulation flocculation with FeCl_3 , the results have shown that the efficiency of elimination of the metallic elements by the various coagulants and flocculants varies between 80 and 99.6. This proves that the flocs formed during coagulation flocculation allow most of the metal elements to be adsorbed in the leachate.

Khalil et al. [35] have shown that flocculation coagulation is a very effective technique for reducing metal pollution from leachate produced at the landfill in the

city of Fez. However, the efficiency of removing the metallic elements varies from one coagulant to another and the combination of lime on the one hand with ferric chloride and on the other hand with Alumina Sulfate improves the efficiency of elimination of the elements Cr (90%), Fe (96%) and Mn (99%) [35].

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However, it should be noted that the precipitation and adsorption of ions in leachate samples is strongly affected by the amount of humic compounds present, which impairs the efficiency of coagulation.

Tatsi et al. [26] in a study of coagulation flocculation for the removal of COD, showed that the addition of Aluminum to fresh leachate resulted in a 25–38% reduction in COD using a dose of 3 g/l d'Aluminum. Amokrane et al. [37] have shown that ferric chloride is more effective than Aluminum Sulphate with a yield of 94 and 87%, respectively, in removing turbidity from landfill leachate. The removal rates of COD and color are 41% and 70% respectively by adding 2.5 g/l of ferric chloride as Fe^{3+} [45].

The addition of Superfloc polyelectrolytes did not significantly affect the elimination of organic matter, which does not exceed 30%, compared with the results obtained with the mixture FeCl_3 + Astral (62%), and FeCl_3 + alginate (50%). Similar results were observed during the addition of polyelectrolytes K1370 and A321 for the treatment of stabilized leachate without pH correction [25]. However, the removal of pollutants was improved by the addition of polyelectrolytes to the samples. It should be noted that at this stage the hydrolysis, ferric cation precipitation and adsorption reactions in the leachate are greatly affected by the presence of humic and fulvic substances. Indeed, specific interactions can appear between humic substances, the surface of flocculants and dissolved ferric species, influencing the efficiency of the coagulation-flocculation process. The effect of addition of coagulant and flocculant mixture on COD removal and partially stabilized leachate turbidity could improve leachate remediation performance.

10. Conclusion

The establishment of landfills in sites that are not suitable or not specifically designed for this purpose increases the risk of contamination of surface and groundwater, and consequently human health, knowing that water is the source of life. Following a bio-physico-chemical evolution of this piled up waste and the action of the rains, a juice called leachate is generated. This juice constitutes a source of pollution for the environment.

In this study, the treatment of leachate from the public landfill in the city of Mohammedia by a flocculation coagulation process was evaluated. Landfill leachate was characterized by a low pH value and a high concentration of pollutants. Organic matter varies between 2153 and 2707 mg/l of COD, and from 1080 to 1405 mg/l of total nitrogen. As for turbidity, however, the elimination of turbidity remains dependent on the efficiency of the coagulation, ie the coagulant used for the study.

In addition, the results obtained showed that ferric chloride is more effective than aluminum sulphate in removing COD, especially when the pH is above 9. Hydrated iron hydroxides precipitate more easily than flocs formed by aluminum, which results in a more efficient removal of pollutants than that obtained at lower pH values. The optimal doses of the various coagulants and flocculants chosen for the study vary from one reagent to another. However, the reduction in pollution by the flocculants alone does not show an efficiency of elimination whereas the coagulant and flocculant mixture allows a good elimination of the pollution. FeCl_3 remains the most suitable coagulant to further eliminate organic and metal pollution. In addition, flocculation coagulation has significantly reduced metal pollution.

It can be concluded that the physicochemical treatment by coagulation flocculation proposed for the treatment of leachate is effective. The technique has several advantages:

- low cost,
- good pollution elimination efficiency, ease of implementation.

It can be concluded that FeCl_3 could have a considerable impact on reducing pollution compared to other results obtained with other coagulants and flocculants.

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