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Landfill leachate treatment by a coagulation–flocculation process: effect of the introduction order of the reagents

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ABSTRACT

The physicochemical process of coagulation–flocculation was highly effective and economically suitable for leachate treatment. The leachate is characterized by a high chemical oxygen demand (COD), between 2,153 and 2,707 mg/L. The organic matter was not easily biodegradable (BOD5/COD: 0.2–0.135). Metal concentrations ranged between 0.1 and 4.2 mg/L for Cr, 0.005–0.04 mg/L for Cd, and 0.3–0.8 mg/L for Pb. The formation of sludge and its aptitude for decantation were measured. Several parameters were selected to check the purification of the landfill leachate, and these include turbidity, COD, metals, and sludge volume. Treatment with $FeCl_3$ proved to be effective at pH 6.5, and for $Al_2(SO_4)_3$ the optimal pH was 5.3. The results indicate that coagulation–flocculation by $FeCl_3$ and $Al_2(SO_4)_3$ is very effective in the reduction of turbidity, with abatement reaching 95 and 98%, respectively. For COD, removal by $FeCl_3$ and $Al_2(SO_4)_3$ reached 67 and 60%, respectively, at optimal concentrations of 18.5 mmol/L Fe^{3+} and 5.82 mmol/L Al^{3+}. Aluminum sulfate produced less sludge than ferric chloride. The volume of sludge produced by $FeCl_3$ remained around 800 ml/L, while the volume of sludge produced by the aluminum sulfate was 230 ml/L.

Keywords: Coagulation; Flocculation; Landfill leachate; Solid waste

1. Introduction

Leachate is a byproduct of the passing of water through waste and it poses a problem when designing and maintaining a landfill. Landfill leachates have been identified as potential sources of ground and surface water contamination, as they may percolate through soils and subsoils, causing extensive pollution of streams, wells, and the water table itself, if they are not properly collected, treated, and safely disposed of [1,2].

Climate and the landfill site are the main factors that affect the production and the composition of leachate. Clearly, where the climate is prone to higher levels of precipitation, there will be more water entering the landfill and, therefore, more leachate will be generated. Another factor is the topography of the site, which influences the runoff patterns and the water balance within the site.

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Landfill leachate treatment is a complicated process because of the type of contaminants which it contains and the variation in volume. The percolation of rainwater through municipal landfill waste lixiviates the products of the biological and chemical process taking place in waste. The combination of the previous factors results in a dark effluent whose properties also largely depend on the age of the landfill [3].

Municipal solid waste landfill leachate is considered dangerous and profoundly polluting. It contains huge amounts of organic matter, some of which are biodegradable whereas others are not, where humic-type constituents account for an important group. The optimization of the factors controlling the treatment of this liquid effluent may significantly increase process efficiency. Coagulation–flocculation process can be applied to landfill leachate without being affected by the leachate's toxicity and could constitute a simple, selective, and economically acceptable alternative to traditional methods.

The chemical treatment methods that are most widely applied in leachate management are coagulation and precipitation [4,5] chemical oxidation, reduction, and ammonia stripping. Successful and cost-effective leachate treatment methods are difficult to find. Therefore, various advanced physicochemical treatment processes have been developed [6,7]. Collecting and recycling are the most common ways of dealing with landfill leachate, but these methods of eliminating contaminants are neither effective nor economically attractive. Biological treatment processes such as activated sludge is problematic due to the low kinetics of degradation and to foam production [8]. In leachate treatment, many factors can influence efficiency, such as the type and dosage of the coagulant/flocculant and the pH [5,9]. The sanitary landfill method for the ultimate disposal of solid waste material continues to be widely accepted and used due to its economic advantages [10].

The main objective of this study is the optimization of the conditions of the effectiveness of the coagulation precipitation process in the treatment of landfill leachate, and determining the most appropriate dose of ferric chloride and aluminum sulfate at various pH values. These can remove especially the organic matter (chemical oxygen demand (COD)), suspended solids, and metals.

2. Material and methods

2.1. Sampling procedures

Leachate samples were collected from the city of Mohammedia landfill. Samples were collected in 50-L plastic containers, transported to the laboratory, and stored at 4˚C. The leachate samples were placed for 2 h at room temperature before the jar test was performed. Then, the samples were thoroughly stirred to resuspend settled solids before any further tests were conducted.

2.2. Coagulants/Flocculants assay

Ferric chloride ($FeCl_3{\cdot}6H_2O$) and aluminum sulfate ($Al_2(SO_4)_3{\cdot}18H_2O$) are two coagulants used to destabilize the colloidal and suspended solids. The effect of Astral and Superfloc cationic polyelectrolytes from Casablanca Astral and Lesieur factories were also examined. The physicochemical parameters are shown in the Table 1.

Cactuses were collected from Mohammedia city and processed at laboratory. They were washed, milled, and then sieved to obtain cactus juice. The cactus juice was then used as a raw material to treat leachate samples.

A laboratory-scale evaluation of chemical coagulation and flocculation was performed using a six-place jar test apparatus. The experimental process consisted of three subsequent stages: an initial rapid mixing at 160 rpm for 10 min, followed by a slow mixing for 20 min at 30 rpm, and then a final settling step for 1 h. Coagulation–flocculation was conducted with the optimized operational parameters (COD, Turbidity … etc.) determined earlier. Six polyethylene beakers of equal volume were used to examine the different dosages of coagulant and initial pH in each run. The sample bottles were thoroughly shaken to resuspend any settled solids and the appropriate volume of sample was transferred to the corresponding jar test beakers. First, the optimum pH for the activity of ferric ion was determined. A known volume of ferric chloride or aluminum sulfate stock solution was added to a jar

Table 1
Physicochemical features of flocculants

Typical	Superfloc	Astral
Type	Cationic	Cationic
Appearance	Opaque liquid	Opaque liquid
Degree of charge en%	40	–
Relative	High	High
Specific gravity at 25˚C	1, 01–1, 05	–
Freezing point ˚C	18	–
Flash point closed cup ˚C	<93	–
Viscosity at 25˚C	290	–
−0.5%	570	
−1%	185	
−2%	0	

containing 1 L of landfill leachate at different pH values adjusted with H_2SO_4 and NaOH. To investigate the optimum coagulant dose, the pH of the leachate was maintained at the optimum as determined above, and varying doses of ferric ion were then added. After 60 min of settling, the supernatant was withdrawn for analysis. To assess the efficacy of ferric chloride or aluminum sulfate for leachate treatment, the following parameters were determined: turbidity, COD, decanted sludge, and metal elements.

2.3. Analysis techniques

Turbidity of landfill leachate was determined by an HI 93703 Microprocessor turbidity meter. For other physicochemical parameters (pH, Total phosphorus, Conductivity, Turbidity, HPO_4^{2-}, Sulfate, TKN, COD, BOD, O_2, and NO_3^-) characterization were determined according to Standard Methods (AFNOR 1999).

3. Results and discussion

3.1. Characteristics of the landfill leachate

Different physicochemical parameters (pH, Total phosphorus, Conductivity, Turbidity, HPO_4^{2-}, Sulfate, TKN, COD, BOD, O_2, and NO_3^-) of the landfill leachate are given in Table 2. The results of metal assays (Cu, Zn, Cr, Ni, Pb, Sb, and Sn) in the leachate at three different dates are shown in Table 3. The leachate is seen to contain a high inorganic as well as organic pollution load.

The characteristics of a landfill leachate can usually be represented in terms of the basic parameters such as COD, BOD, ratio of BOD/COD, colour, pH, oxidation–reduction potential, and heavy metal content [11]. In this study, the leachate is characterized by high levels of organic matter (Table 2) and high concentrations of ammonium and nitrogenous compounds. Organic matter, in terms of COD values, reached 2,153–2,707 mg/L (leachate stabilized) in leachate from samples deposited in 2,000, whereas high concentrations of ammonium–nitrogen were measured (587–1,410 mg/L, TKN (1,080–1,405 mg/L), while the principal heavy metal concentrations ranged between 4.2 and 0.1 mg/L for Cr, 0.04–0.005 mg/L for Cd and 0.8–0.3 mg/L for Pb. The characterization of the average leachate indicated that its mean annual BOD/COD ratio was between 0.2 and 0.135 with little further biological degradation likely to occur corresponding to partially stabilized samples.

The variation in leachate characteristics was attributed to several factors, such as the variations in the composition of the solid waste, the age of the landfill, the hydrogeology of the landfill site, the rainfall and the specific weather conditions, and the moisture routing through the landfill [11–13].

A landfill can be classified into three categories based on age: young, middle, and old. A landfill that has accumulated for less than five years is termed as "young." It usually consists of a large amount of biodegradable matter and its leachate has a high COD of the order of 20 000 mg/L. At 5–10 year-old landfill is known as "middle age" and its leachate presents a COD between about 3,000–15,000 mg/L [14]. After 10 years, a landfill contains much less biodegradable matter and leachate COD is less than 2,000 mg/L at this age, it is designated as an "old landfill" [14,15].

The results of the diagnoses performed at the leachate treatment station showed fluctuations of pH, flow, and levels of organic matter vs. time. This can influence the effectiveness of the elimination of the pollutants and justifies the installation of a homogenization basin. The age of the landfill is one of the

Table 2
Physicochemical parameters of the landfill leachate

Parameter	Range	Average	Moroccan guide level
pH	7.7–8.92	8.3	6.5–8.5
Conductivity (ms/cm)	25.6–35.9	31.2	2.7
Turbidity (NTU)	63–140	102.2	–
HPO_4^{2-} (mg/L)	592.4–2,128	1,693.3	–
Sulfate (mg/L)	77.47–218.7	156.04	–
Tot phosphate (mg/L)	1226.6–2,217	1,879.5	10
TKN (mg/L)	1,080–1,405	1,289.75	30
COD (mg/L)	2,153–2,707	2,473.9	500
BOD (mg/L)	526–290	399	–
BOD/COD	0.2–0.14	0.16	–
O_2 (mg/L)	0–0.2	0.03	
NO_3^- (mg/L)	36.3–453.9	173.2	–

Table 3
Metallic elements analysis in landfill leachate

Date of landfill deposit	Cu (mg/L)	Zn (mg/L)	Cr (mg/L)	Cd (mg/L)	Ni (mg/L)	Pb (mg/L)	Sb (mg/L)	Sn (mg/L)
25 March 2000	0.7	0.2	0.1	0.035	0.3	0.5	1.3	–
10 August 2002	1.2	2.3	4.2	0.02	0.4	0.3	0.9	–
5 October 2007	1.2	1.9	3.2	0.005	0.2	0.8	0.5	0.6

main factors affecting the characteristics of the leachate. As the landfill becomes older, the biological decomposition of the waste shifts from a relatively short initial aerobic period to a longer decomposition period, which has two distinct, subphases, i.e. an acidic phase and a methanogenic phase. Leachate from these distinct stages contains different constituents and therefore has different characteristics.

In terms of BOD, there have been reported values of as much as 8,000 mg/L for fresh leachate samples and only 4,200 mg/L for old leachate samples, whereas concentrations of ammonium–nitrogen remained high [11]. It should be noted that fresh leachate presented relatively low pH values (around 6), a rather low BOD:COD ratio (about 1:3), high COD levels, and a very high ammonium-nitrogen content.

Landfill leachate contains chemicals including metal ions such as iron. Successful and cost-effective leachate treatment methods are difficult to find [16].

Rivas and Gimento [17] pointed out that old landfills produce stabilized leachates with relatively low COD in the range of 500–5,000 ppm, slightly basic pH 7.5–8.5, low biodegradability (ratio BOD/COD below 0.1), and a significant amount of heavy metals and high-molecular weight compounds (humic substances). Leachate presents considerable variations in both volumetric flow and chemical composition [18]. The composition and concentration of contaminants are influenced mainly by the age of the landfill and also by the type of waste deposited and other hydrogeological factors [19,20].

Leachate may contain large amounts of organic matter (biodegradable but also matter refractory to biodegradation), where humic-type constituents are an important group [21], as well as ammonium–nitrogen, heavy metals, chlorinated organic, and inorganic salts [21].

3.2. Leachate settling

The study of the removal of pollution from leachate by sedimentation is shown in Table 4. Variations in settling times between 30 and 120 min were studied to see the influence of time on leachate settlement. Supernatant was collected after a settling time of 120 min.

The reduction in COD by simple decantation varies from 5 to 34.5%. This depends essentially on the quality of collected leachate. Indeed, sedimentation is still essential to reduce leachate pollution with the least cost. It is necessary to clarify that the natural settling of leachate is based on the physicochemical characteristics of the effluent (Table 2). Indeed, more leachate loaded with colloidal material performance is more important. Decantation was carried out for six samples having different COD values. For the companion 4, the performance of the removal of pollution is around 5%. This is due to the fact that the sample having a COD of 2,688 mg/L is rich in soluble or dissolved solids and has very little colloidal material which normally promotes natural decantation.

Settling and decanting is a method to reduce turbidity by letting the water sit for 2–24 h so that the particulates settle to the bottom of the container. The benefit of settling and decanting is that it requires no equipment besides the containers. However, the drawbacks of this method are the need for multiple containers, the time it takes the water to settle, and the difficulty in observing the effect of settling, if the containers are opaque. In laboratory studies, the use of settling and decanting significantly reduced both the turbidity and the chlorine demand of turbid waters.

The removal of suspended particles by sedimentation depends upon the size and specific gravity of those particles. Settleable solids are measured as the visible volume accumulated at the bottom of an Imhoff cone after the water has settled for one hour. (Standard Methods for the Examination of Water and Wastewater (1975). Unhindered settling is a process that removes discrete particles in a very low concentration without interference from nearby particles. In general, if the concentration of the solution is lower than 500 mg/L of the total suspended solids, sedimentation is considered to be discrete.

Table 4
Study of the settling of leachate (COD mg/L) time on leachate settling 120 min

Leachate (COD mg/L)	Leachate settling (COD mg/L)	Abatement (%)
2,707	1,800	34.5
2,301	1,632	29
2,153	1,536	29.5
2,688	2,544	5
2,240	1,728	23
2,540	1,866	26.5

3.2.1. *Effect of Fe^{3+} and Al^{3+} on the removal of leachate pollution*

The results of the Fe^{3+} and Al^{3+} coagulation of leachate are presented in Figs. 1 and 2. The comparative study of coagulation–flocculation by Fe^{3+} and Al^{3+} is given in Table 5.

Test for COD removal efficiency using $FeCl_3$ was carried out at different pH values from 2 to 12 (Fig. 3), while the optimum pH of the raw leachate, before the addition of the coagulant, was pH (6.5). At this pH, COD and turbidity fell by 84 and 96%, respectively. This finding is in agreement with that of Renou et al. [12]. Coagulation/flocculation is a commonly used process in water and wastewater treatment in which compounds such as ferric chloride and/or polymers are added to landfill leachate in order to destabilize the colloidal materials. This causes the smaller particles to agglomerate into larger settleable flocs. Several studies have reported the examination of this process for the treatment of landfill leachate, especially with respect to the optimization performance of coagulant/flocculant, the determination of experimental conditions, the assessment of pH, and the investigation of flocculant addition [16]. To enhance the proposed coagulation/flocculation, process the addition of certain commercial polyelectrolytes was also examined, including two cationic (Astral and Superfloc). After the settling period, the supernatant was withdrawn from the beaker and was used for chemical analysis.

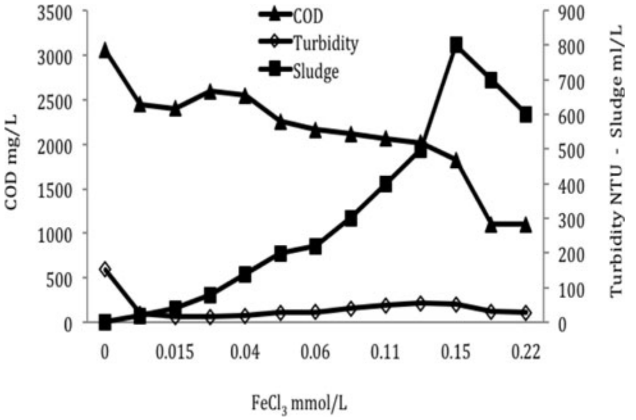

Fig. 1. Effect of $FeCl_3$ dosage on the removal of leachate pollution. Experimental conditions: Initial COD of Leachate = 3,175 mg/L, initial turbidity = 128 NTU, pH 7.8, time on leachate settling 120 min.

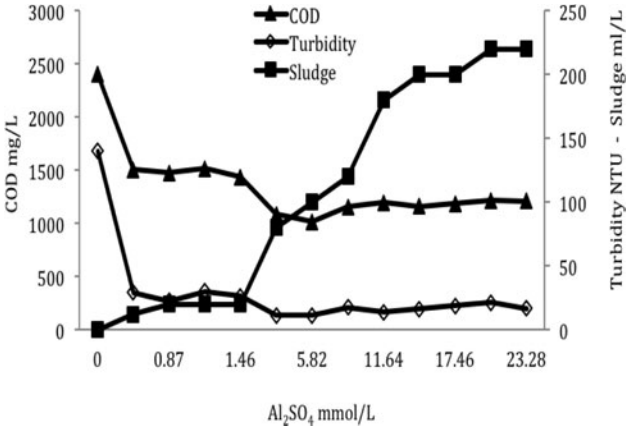

Fig. 2. Effect of Al_2SO_4 dosage on the removal of COD, turbidity, and decanted sludge. Experimental conditions: initial COD of leachate = 2,400 mg/l, initial turbidity = 140 NTU, pH 7.8.

Fig. 4 shows the effect of pH on the coagulation of leachate using $Al_2(SO_4)_3$. It shows a maximum of COD and a turbidity removal of 60 and 98%, respectively compared to the initial value at pH 5.3. The maximum sludge volume, generated at optimum pH, was 375 ml/L.

The flocs generated under highly acidic or basic conditions were significant, but very few in number, especially in an acidic environment; pH reduction was due to the acidic character of Al^{3+} from $Al_2(SO_4)_3$. For the other coagulant doses tested, the flocs were microscopic and of similar size.

Table 5
Comparative study of coagulation–flocculation by Fe^{3+} and Al^{3+}

	Optimal concentration (mmol/L)	Turbidity removal (%)	COD removal (%)	Sludge (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

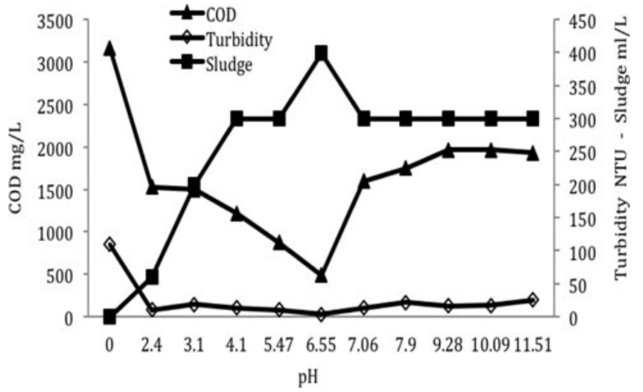

Fig. 3. Effect of pH on the coagulation of leachate using $FeCl_3$. Experimental conditions: (optimal concentration of $FeCl_3$: 3,500 mg/L), initial COD of leachate = 3,168 mg/L, initial turbidity = 110 NTU, pH 7.8.

The optimum process variables of the coagulation of landfill leachate using Fe^{3+} and Al^{3+} were found at pH 6.5 for Fe^{3+} and 5.3 for Al^{3+} at coagulant dosages of 18.5 mmol/L Fe^{3+} and 5.82 mmol/L Al^{3+}. Although the doses required were identical (0.035 mol/L of Fe^{3+} or Al^{3+}), with an initial COD concentration of 4,100 mg/L, $FeCl_3$ was found to give a higher removal of organic compounds (55%) than Al^{3+} (42%). At an initial concentration of 5,690 mg/L and at pH 4.8, maximum COD removal of 56% was achieved with 0.8 g/L of $FeCl_3$ as compared to 39% with 0.4 g/L of Al $(SO_4)_2$ [22]. The effect of coagulant dosages showed a similar trend for COD and turbidity removal. This suggests that the COD in the landfill leachate was mainly accounted for by organic matter with some insoluble forms that exhibited turbidity [16].

In the field, leachate treatment is a difficult and expensive process. Although, leachate can be processed biologically, COD removal efficiency is usually low due to high ammonium ion content and the presence of toxic compounds such as metal ions [23]. The output of this physicochemical treatment shows a strong reduction in the polluting load of landfill leachate. Tchobanoglous et al. [24] reported that the levels of COD in leachate younger than 2 years of age may reach 3,000–60,000 mg/L. It is well known that the COD is mainly due to the presence of humic substances. The difference in removal rates could be attributed to the age of the landfill sites. During

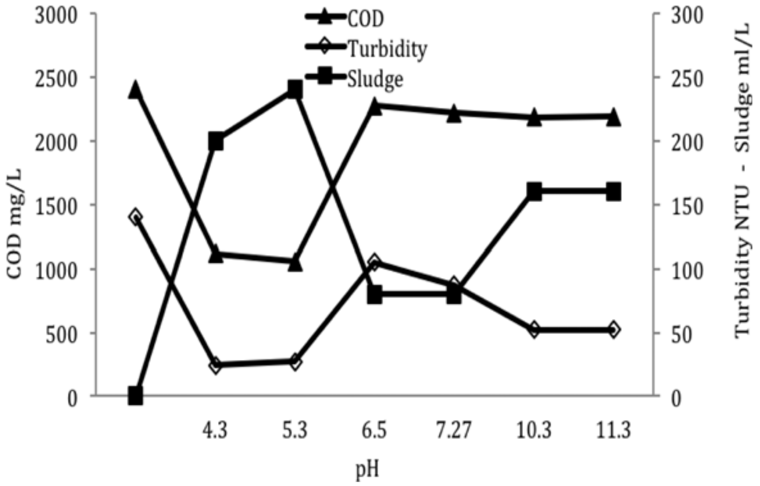

Fig. 4. Effect of pH on the coagulation of leachate using Al_2SO_4. Experimental conditions: (Optimal concentration of Al_2SO_4: 1,000 mg/L), initial COD of leachate = 2,400 mg/l, initial turbidity = 140 NTU, pH 7.8, time on leachate settling 120 min.

coagulation, the amount of sludge produced depends on the characteristics of the leachate and the pollutant removal efficiency.

Coagulation–flocculation is a relatively simple and frequently applied technique in water and wastewater treatment. This technique could be successfully employed for the treatment of stabilized landfill leachate. The removal of pollutants mainly involves charge neutralization of negatively charged colloids by cationic hydrolysis products, followed by the incorporation of impurities in an amorphous hydroxide precipitated through flocculation. However, it should be noted that precipitation and adsorption of ferric cation in leachate samples is highly affected by the amount of humic compounds present, which alters the efficiency of the coagulation. Therefore, the dosage of ferric salts required in wastewater treatment is primarily specified by the concentration of naturally occurring organic compounds which are know by their high load in the leachate than in the wastewater. Tatsi et al. [16] in a study of COD removal by coagulation reported that the addition of aluminum coagulants to fresh leachate resulted in a 25–38% reduction of COD at a dosage of 3 g/L aluminum. Amokrane et al. [25] similarly found ferric chloride to be more effective than aluminum sulfate (94 and 87%, respectively) for coagulation–flocculation pre-treatment turbidity removal from landfill leachate. Maximum COD in leachate and colour removal rates of 41 and 70%, respectively have been achieved by the addition of 2.5 g/L of ferric chloride as Fe^{3+} [22]. The addition of ferric chloride or alum coagulants to leachates resulted in a reduction of COD values; the optimum removal was found during the addition of 1.4 g/L alum and 2 g/L ferric chloride to the samples [26].

Several studies have examined coagulation–flocculation for the treatment of landfill leachates, aiming at performance optimization, i.e. the selection of the most appropriate coagulant, the determination of experimental conditions, the assessment of pH effect, and the investigation of flocculant addition [27]. Moreover, during the precipitation–coagulation process, adsorptive micellar flocculation seems to contribute to the removal of organic matter from the leachate [28].

The combined action of ferric chloride–polyelectrolyte mixtures was examined. Flocculant addition at a ratio coagulant:flocculant (Astral 2.5:0.5, Superfloc 2.5:0.14 Alginate 2.5:0.02) (g), resulted in COD and turbidity removal capacities (Fig. 5). The addition of polyelectrolytes superfloc did not substantially affect organic matter removal, which did not exceed 30%, as compared with the results $FeCl_3$ + Astral (62%), and $FeCl_3$ + Alginate (50%). Similar results were observed by Tatsi et al. [16], during the addition of polyelectrolytes K1370 and A321 to stabilized leachate samples without pH correction. However, the removal of pollutants was enhanced by the addition of polyelectrolytes to the samples. It should be noted that at this point of 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 the coagulation–flocculation process. The effect of a coagulant and flocculant mixture addition on removal of COD and turbidity from partially stabilized leachates is important

3.3. Removal of heavy metals

The removal of metallic elements by $FeCl_3$ is shown in Table 6. Concentration of heavy metals (Zn, Cu, Cd, Cr, and Ni) in leachate exceeded the maximum values allowed.

Lime was used traditionally as a coagulant in leachate treatment over many years, requiring dosages of about 1–15 g/L and was found well suited for the removal of heavy metals, such as Fe, Cd, Cr, reaching up to 90% efficiency [25]. Leachates may contain large amounts of organic matter with humic-type constituents making up an important group [21], but there are also ammonium–nitrogen, heavy metals, chlorinated organic and inorganic salts [12,29,30].

The experimental results show that an 18% removal of COD and 90% removal of heavy metals can be attained at pH 6.5 (optimum for Aluminum) with the addition of 1,400 mg/L of aluminum and a 28% removal of COD and 86% removal of heavy

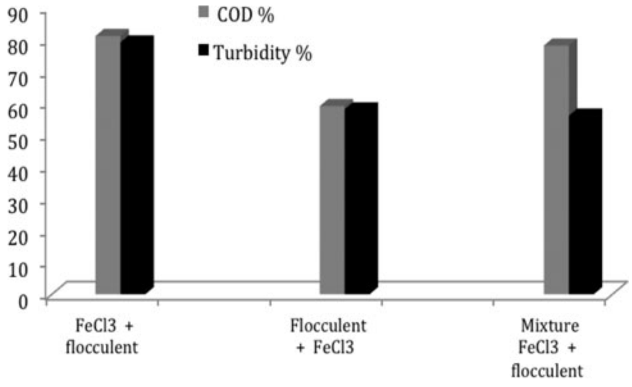

Fig. 5. Effect of introduction order of coagulant $FeCl_3$ and flocculant Astral dose on the removal organic matter (COD) and Turbidity.

Table 6
Metal assay in the stabilized landfill leachate before and after coagulation–flocculation using $FeCl_3$

Landfill leachate	Cu (mg/L)	Zn (mg/L)	Cr (mg/L)	Cd (mg/L)	Ni (mg/L)	Pb (mg/L)	Sb (mg/L)
Before treatment	1.20	2.30	4.20	0,02	0.35	0.30	0.9
After treatment	0.10	0.01	0.21	–	0.12	0.01	0.03
Removal (%)	91.70	99.6	95	–	80	97	97

metals can be attained at pH 10 (optimum for Ferric Chloride) with the addition of 2,000 mg/L Ferric Chloride [26].

In this study, the results indicate that coagulation–flocculation by iron(III) Chloride is very effective in the reduction of metallic elements; abatement reaching 80 and 99.5% at optimal concentrations of 18.5 mmol/L Fe^{3+} can be attained at pH 6.5. Previously, lower heavy metals removal in landfill leatchate was observed by Silva et al. [23] during the coagulation/flocculation process.

3.4. Effects on solid production

In general, the amount and the characteristics of the sludge produced during the coagulation/flocculation process are highly dependent on the specific coagulant used and on the operating conditions. The wet sludge volume at the bottom of the jar test beakers after the coagulation/flocculation process was used to quantify the volume of sludge generated in this study. The volume (ml/L) of the settled sludge is shown as functions of coagulant, flocculant type, and dose (mg/L).

The effect of optimal concentrations of coagulant $FeCl_3$ and flocculants (Astral, Supefloc, Alginate) alone or mixed in the production of sludge produced during the coagulation/flocculation process coagulant type and dose volume of sludge is given in Table 7.

The addition of organic flocculants was examined for the enhancement of organic matter removal. Mixing $FeCl_3$ + Superfloc showed a removal efficiency of 30 and 66%, respectively, for COD and Turbidity. These values are higher than the results obtained in the case of mixing $FeCl_3$ + Astral or $FeCl_3$ + Alginate.

According to Table 7, the volume of sludge produced was reduced considerably with an increasing dose of polyelectrolyte in the coagulation process. This may be due to cationic nature of the polyelectrolyte employed in this study, which has high-molecular weight, thus, providing long bridges between small flocs to enhance particle growth. It also has the ability to attract and hold colloidal particles at polar sites on the molecule. Generally, organic polymers generate less sludge than inorganic salts since they do not add weight or combine chemically with other ions in the water to form precipitate. Thus, the sludge produced by the use of ferric chloride in combination with polyelectrolyte was compact and reduced in volume.

The combination of ferric chloride, alginate, and Superfloc produced much water and less sludge (ml/L) than when ferric chloride was used with flocculant Astral.

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

The effect of the introduction order $FeCl_3$ and Astral flocculant dose on the removal organic matter (COD), turbidity and production of sludge are presented in (Fig. 5).

The results obtained shows that when $FeCl_3$ is introduced first, followed by the flocculant the reduction of COD and turbidity varies around 81 and 79%, respectively. When $FeCl_3$ is introduced along with the flocculant, turbidity is greatly reduced (56%) while the

Table 7
Effect of optimal concentrations of coagulant and flocculants alone or mixed on the production of sludge

Reagents	Optimal concentrations (mg/L)	Optimal concentration in mixture $FeCl_3$ + flocculants (mg/L)	Sludge in mixture (ml/L) $FeCl_3$ + flocculants
$FeCl_3$	2,500	2,500	–
Astral	192	500	800
Superfloc	200	140	230
Aliginate	120	20	80
Cactus (ml/L)	32	25	195

Table 8
Effects of the introduction order $FeCl_3$ and Astral flocculant dose on the production of sludge. Optimal concentrations: $FeCl_3$: 2,500 mg/L, Astral Flocculent: 500 mg/L, pH 7

Introduction order	Sludge (ml/L)
$FeCl_3$ + Flocculant	200
Flocculant + $FeCl_3$	300
Mixture ($FeCl_3$ + Flocculant)	750

COD is around 78%, which shows no significant difference from when $FeCl_3$ is introduced first followed by the flocculant. Moreover, the results obtained when the flocculent is introduced before the $FeCl_3$ shows a significant decrease in COD (59%) and turbidity (58%). However, the values of organic matter removal during the addition of coagulant–polyelectrolyte mixtures are more important than the values achieved by the addition of similar dosages of single $FeCl_3$. In addition to pollutant removal, sludge production was considered in this work, as it may affect the economic feasibility of the proposed method. The sludge produced during the physical–chemical treatment of landfill leachate (Table 8) is composed of the amount of originally suspended organic matter and solids, as well as by the compounds formed due to the possible addition of chemical reagents.

The amount of sludge produced by first introducing the $FeCl_3$ followed by the flocculant is 200 ml/L (Compacted sludge) which remains low compared to results obtained by introducing the first Astral flocculant and $FeCl_3$ (300 ml/L). In addition, the study of coagulation flocculation mixture $FeCl_3$ and flocculant showed a significant amount of non-compacted sludge (750 ml/L). In conclusion, the introduction of $FeCl_3$ in the first place followed by the flocculant can lead to a less compacted sludge volume.

4. Conclusion

The application of coagulation/precipitation to leachate treatment was examined in this study. Landfill leachate was characterized by low pH and a high concentration of pollutants. Organic matter was in the range of 2,153–2,707 mg/L COD, TKN 1,080–1,405 mg/L

Furthermore, from the results of the coagulation experiments, it can be observed that ferric chloride was more efficient than aluminum sulfate for the removal of COD, especially when the pH was greater than 9, hydrated iron hydroxides precipitate more easily than the corresponding aluminum flocs, resulting in a more efficient removal of pollutants than that obtained at lower pH values. It can be concluded that the advantages of the proposed physicochemical technique to treat the hazardous pollutant of leachate are mainly simplicity and good removal efficiencies along with easy onsite implementation.

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