Study of Coagulation Process with Lime in Treatment of Landfill Leachate from Fkih Ben Salah City (Morocco)

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Abstract
The leachates are the seat of complex processes which give them a heterogeneous character. Their compositions vary according to several factors: nature of the waste, conditions of their deposition, climatic conditions, their durations of stay, etc. They contain important quantities of organic, mineral matters even of bacteria, which require their treatment in order to safeguard the environment. To do this, several methods are used, such as membrane techniques (reverse osmosis, nanofiltration, etc.), biological techniques (activated sludge, SBR, etc.) and physicochemical techniques (Coagulation-flocculation, adsorption on activated carbon, etc.). Among these techniques, the leachate treatment by coagulation process with the lime showed interesting reduction of the various pollutants: 92.95% of turbidity, 88.23% of suspended matter, 89.89% of COD, 90.83% of BOD5, 78.39% of Fe, 77.78% of Mo, 38.29% of Cd, 48.75% of Al, 50.24% of S2−, 20.57% of K+, 27.24% of phosphorus and 19.53% of Cl−. Based on these results, the coagulation with the lime reveals interesting because it allows to reduce at a lesser cost the pollutants present in leachates.

Keywords
Leachate Treatment, Pollutants, Biological Techniques, Physicochemical Techniques

1. Introduction
The Fkih Ben Salah landfill generates leachates with a high polluting load (COD
= 74,530 mg O₂/L and Conductivity = 24.90 ms/cm). They are a source of contamination for groundwater and superficial water, Idlahcen, Souabi, Taleb et al. (2014) and Chtioui, Khalil, Souabi et al. (2008). They must be imperatively treated before being rejected in the natural environment, Renou, Poulain, Givaudan et al. (2009), by using simple and little expensive technologies, Berradi, Chabab, Arroub et al. (2014).

The main objective of this work is to study the coagulation with the lime of young leachates. It aims to determining the optimal conditions of this treatment for a significant reduction of TSS, the organic and mineral load, Merzouki, Hanine, Lekhlif et al. (2016). The use of the lime is opportune, because it plays the coagulant role (Ca²⁺), and at the same time it allows the neutralization of the leachate (acid pH of the young leachate) thanks to its ions OH⁻. Its effectiveness compared with other coagulants (Fe³⁺, Al³⁺) has been shown by several authors, Merzouki, Hanine, Lekhlif et al. (2016), Hamidi, Salina, Mohd Nordin et al. (2007), Benradi, El Yahyaoui, Bouhlassa et al. (2013), El Bada, Assobhei, Kebbabi et al. (2010), and Khalil, Bouaouine, Chtioui et al. (2015).

In this study, we present the results of the coagulation tests using the following parameters: pH, turbidity, COD, BOD5, conductivity, quantity of the generated sludge, mineral and metallic elements (Al, Fe, Cd, Mg, Ca, Cl, Mo, P, K, Si, S), as well as abatement rates of present pollutants in the leachate.

2. Materials and Methods

2.1. Sampling Zone

The landfill of Fkih Ben Salah city covers an area of 20 ha. It is located in 12 km north of the city on the R11 national road (Figure 1). It receives more than 85.51 tons per day of waste and generates 2730.72 L/day of leachate, Merzouki, Hanine, Lekhlif et al. (2016), corresponding to rate of 3.49% liter of leachate by ton of waste.
The leachate samples were collected directly at the level of collection trucks bringing household waste to the landfill. Volumes of leachate (5 L) were collected and mixed to form a homogeneous sample. The pH, the electrical conductivity and the temperature were measured in situ. In order to determine the other parameters, the samples were kept at a temperature below 4˚C and transmitted to 4˚C and transmitted to the laboratory within 24 hours.

2.2. Preparation of Coagulant

For coagulation, a stock solution of coagulant was prepared by adding in one liter of distilled water 60 g of the hydrated lime in powder form of Ca(OH)$_2$. From this solution, increasing volumes are taken and added to six beakers, each containing a volume of 500 ml of leachate. The concentration of Ca(OH)$_2$ obtained ranges from 2 to 12 g/L.

2.3. Procedure Jar Test

The coagulation tests were accomplished in a Jar test apparatus (Figure 2). The mixture was agitated quickly at a speed of 250 rpm for 5 minutes and then to a speed of 25 rpm for 20 minutes. After 120 min of decantation, supernatant fraction samples are then recovered to analyze the following parameters: pH, DCO, total suspended solids (TSS), quantity of sludge and concentrations of mineral and metallic elements.

2.4. Measurement of Biological and Physicochemical Parameters

The potential of Hydrogen (pH), the electrical conductivity (CE), the temperature (˚C), TSS and the salinity were measured using a multi-parameter probe type Consort C933. The suspended solids (MS) are determined by filtration on membranes at 0.45 μm in diameter (AFNOR T 90-105). The biological oxygen demand (BOD5) is determined according to the respirometric method in an enclosure thermostat at 20˚C (AFNOR, T 90-103). Chemical oxygen demand (COD) was determined using the standard method. The turbidity is measured by a turbidity

Figure 2. Jar-test device.
meter of the Palintest 7000 type. Calcium and magnesium were determined by complexometric titration with EDTA. Mineral and metallic elements (Fe, Cd, Cl, Mo, Si, S, K ...) were measured by a portable analyzer of the fluorescence Ray-X.

The estimation of the sludge produced after treatment by the lime coagulation technique was realized by measuring its volume and weight after decantation, filtration and drying in an oven at 105°C until constant weight.

3. Results and Discussion
3.1. Characterization of the Studied Leachate

The leachates studied have a brownish color and a fecal odor. Their contents are presented in Table 1.

The physicochemical analyzes of the raw leachate have shown that it has an acidic character (pH = 4.05), an electric conductivity of 24.90 ms/cm and contains a concentration in SM of 3400 mg/L and a turbidity of 8018.33 NTU. The acid character is due to the fact that the leachate is young. This observation is noted by several authors, Jirou, Harrouni, Arroud et al. (2014), Benyoucef, EL Ghmari, & Ouatmane (2015).

Table 1. Processes with special significance during SEA treatment.

| Parameters | Minimum  | Maximum  | Average  | Standard deviation | *GVRD | Unit |
|------------|----------|----------|----------|--------------------|-------|------|
| Ph         | 3.99     | 4.02     | 4.05     | 0.08               | 5.5 - 9.5 | -    |
| T          | 28.16    | 33.15    | 30.65    | 0.12               | 30    | °C   |
| Turbidity  | 7895.00  | 8180.00  | 8018.33  | 146.32             | -     | NTU  |
| Salinity   | 13.02    | 14.23    | 13.45    | 0.68               | -     | PSU  |
| CE         | 24.71    | 25.02    | 24.90    | 0.16               | 2.7   | ms/cm|
| TSS        | 3090.00  | 4150.00  | 3613.33  | 53.01              | -     | mg/L |
| COD        | 72150.00 | 76124.68 | 74530.00 | 209.96             | 500   | mg O₂/L |
| BOD5       | 37690.00 | 39420.00 | 38313.33 | 96.10              | 100   | mg O₂/L |
| SM**       | 2703.20  | 3906.00  | 3400.40  | 62.39              | 100   | mg/L |
| Cl         | 33.25    | 34.26    | 33.81    | 0.5                | 0.2   | mg/L |
| Ca         | 2075.00  | 2275.00  | 2175.00  | 100.00             | -     | mg/L |
| Mg         | 198.16   | 215.38   | 204.85   | 9.23               | 2     | mg/L |
| Al         | 2.49     | 3.04     | 2.78     | 0.27               | 10    | mg/L |
| P          | 799.21   | 883.25   | 853.93   | 47.43              | 10    | mg/L |
| K          | 813.57   | 865      | 844.96   | 27.53              | -     | mg/L |
| Cd         | 23.43    | 24.98    | 24.00    | 0.85               | 0.25  | mg/L |
| Mo         | 9.06     | 10.12    | 9.47     | 0.57               | -     | mg/L |
| Iron       | 213.62   | 268.32   | 241.087  | 27.35              | 5     | mg/L |
| Si         | 3.168    | 4.86     | 3.74     | 3.74               | -     | mg/L |
| S          | 498.32   | 585.12   | 553.243  | 47.769             | -     | mg/L |

*General limit values of rejection of discharge; **SM: Suspended Matter.
Concerning the electrical conductivity and the concentration in SM, they are very high in comparison to the limit values, Jirou, Harrouni, Arroud et al. (2014) (CE = 2.7 ms/cm and SM = 100 mg/L). Nevertheless, they are in the range of values usually encountered for leachate, Jirou, Harrouni, Arroud et al. (2014), and Bouaouine, Khalil, Chtioui et al. (2015). The turbidity presents, as for it, a relatively high value, compared with those found by other authors: 2053 NTU, Khalil, Bouaouine, Chtioui et al. (2015), 1360 NTU, Toklo, Josse, Topanou et al. (2015), and 840 NTU, Benradi, El Yahyaoui, Bouhassa et al. (2016).

The COD and the BOD5 are equal respectively to 74,530 mg/O₂/L and 38313.33 mg/O₂/L. They exceed the discharge standards (DCO = 500 mg/O₂/L, BOD5 = 100 mg/O₂/L), Jirou, Harrouni, Arroud et al. (2014). They are indicative of a strong organic load; they are comparable with those of landfill leachate of Agadir city (Morocco): COD = 72,000 mg O₂/L, BOD5 = 44,000 mg O₂/L, Jirou, Harrouni, Arroud et al. (2014), and superior to those obtained in Fez (Morocco): COD = 5400 mg O₂/L, BOD5 = 1700 mg O₂/L 5, Bouaouine, Khalil, Chtioui et al. (2015).

Concerning the mineral and metallic pollution, there is a high load of Ca (2175 mg/L), Al (2.78 mg/L), Mg (204.85 mg/L), P (853 mg/L), K (844.96 mg/L); Cd (24 mg/L), and Fe (241.087 mg/L). These values exceed the standards, Jirou, Harrouni, Arroud et al. (2014). They are greater than the values found in the Fez landfill (Morocco) (Fe = 33.72 mg/L, Cd = 4.36 mg/L), Benyoucef, EL Ghmari, & Ouatmane (2015), and lower than those of the Oran landfill (Algeria) (Ca = 5216 mg/L, Mg = 4800 mg/L), Bennama, Younsi, Zoubir et al. (2010) and Larache Landfill (Morocco) (Fe = 650 mg/L), Er-raiou, Bouzid, Khannous et al. (2011). These results indicate pollution by several types of industrial solid waste rich in mineral and metallic elements. The concentrations of the various pollutants found at the Fkh Ben Salah landfill are generally different from those of other landfills. This is due to the nature of the waste, the age of the landfill and the climatic conditions, Bikash, Khet, & Sanjay (2014), and Kurniawan, Lo, & Chan (2006).

### 3.2. Results of the Leachate Coagulation

#### 3.2.1. Visual Observations

During coagulation, it is observed that the sludge separates progressively depending on the coagulant dose. During decantation, the leachate color changes. It gradually changes from brown to light brown (Figure 3). This change of color by coagulation has also been observed by several authors, El Bada, Assobhei, Kebbabi et al. (2010), and Shabiimam & Anil (2011).

#### 3.2.2. Evolution of the pH

The pH of the young leachate is acid character (4.05). This denotes the installation of the anaerobic process, in particular in its acidogenesis phase producing the AGV. When the lime concentration increases, the supernatant pH value increases (Figure 4). This can be explained by the release of the OH⁻ ions in solution.
Figure 3. Color of leachate before and after coagulation.

Figure 4. Evolution of pH according to the concentration of the lime (g/L).

This evolution has also been observed by other authors, Renou, Poulain, Givaudan et al. (2009), and Shabiimam & Anil (2011).

3.2.3. Evolution of the Quantity of Sludge Generated by Coagulation
As shown in Figure 5 and Figure 6, the volume of sludge increases with the lime concentration. It varies from 30.33 ml/L to 27,967 ml/L (Figure 5). These volumes of sludge correspond to the quantities of dry matter ranging from 0 to 47.60 g/L (Figure 6).

The quantity of sludge obtained in these tests is lower compared to other studies, such as the one carried out, Khalil, Bouaouine, Chtioui et al. (2015), which found 647 ml/L of sludge after introduction 12 g/L of the lime on a leachate, agitated at a rotation speed of 200 rpm for 10 min and then at a slow speed of 60 rpm for 30 min. Merzouki, Hanine, Lekhlif et al. (2016), found almost identical results under the same coagulation conditions (302 ml/L of sludge). That can be due to the nature of the leachate, the rheology of the sludge and the retention of water within the solid matrix.

3.2.4. Evolution of Electrical Conductivity
According to the figure (Figure 7), we note a slight decrease of the electrical conductivity, it passes from 24.08 mS/cm to 21.91 mS/cm to 6 g/L of Ca(OH)₂, then an increase to 24.16 mS/cm from 10 g/L. At the beginning, the added lime eliminates the ions and molecules, contributing to the conductivity of leachate,
by coagulation up to pH = 6, which probably explains the decrease in the conductivity. Its increase then could be attributed to the concentration of added Ca^{2+} and OH⁻ ions, Renou, Poulain, Givaudan et al. (2009). Renou, Poulain, Givaudan et al. (2009), and El Bada, Assobhei, Kebbabi et al. (2010) have observed the same conductivity behavior. On the other hand, Renou, Poulain, Givaudan et al. (2009) have noted that the lime doses corresponding to the conductivity minimum increase when its initial value in the leachate increases.
3.2.5. Evolution of Turbidity and Suspended Matter

The results of turbidity (Figure 8) show a decrease in the addition of lime. It passes from 8031.67 NTU to 566.27 NTU in a concentration of 10 g/L, which corresponds to a removal efficiency of about 92%. This value is similar to that found by Renou, Poulain, Givaudan et al. (2009) for an initial turbidity of 5190 NTU and by Slater, Uchrin, & Ahlert (1983), and it is very high than that found by El Bada, Assobhei, Kebbabi et al. (2010) who carried out tests on an initial leachate turbidity of 130 NTU during methanation (65%). As for Shabiimam & Anil (2011), having studied a leachate presenting an initial turbidity of 317 NTU, they were able to obtain a 99.9% removal of the turbidity at pH = 8 with a coagulant dose of 25 g/L.

According to Figure 9, the suspended matter abatement rate reaches 88.23% at 10 g/L of lime. This rate is in the range of values found by numerous authors, Melike & Kadir (2007). Merzouki, Hanine, Lekhlif et al. (2016) have found successively a reduction of 82% and 71.6% of the suspended matter on the raw leachate.

![Figure 8. Evolution of turbidity reduction as a function of the lime concentration (g/L).](image)

![Figure 9. Evolution of suspended matter reduction as a function of the lime concentration (g/L).](image)
3.2.6. Evolution of the Organic Matte

The addition of the lime has reduced the COD of the leachate (Figure 10). The abatement rate is 89.76%. It is comparable to that obtained by Merzouki, Hanine, Leklif et al. (2016) in the same conditions (90.27%) and Shabiimam & Anil (2011) (86%), after adding 25 g/L of the lime on an initial COD of 2451 mg/L, and higher compared to those obtained by other authors. Tatsi, Zouboulis, Matis et al., (2003) found COD removal rates ranging from 30% to 45% when adding lime at a concentration of 7 g/L in fresh and partially stabilized leachate samples with an initial COD of 5350 mg/L, El Bada, Assobhei, Kebabi et al. (2010) found a yield of 31% at a concentration of 10 g/L of lime for an initial COD of 138.76 mg/L, as for Shabiimam & Anil (2011), they found a yield of 69% after addition of 25 g/L of lime for an initial COD leachate of 2451 mg/L.

The addition of the lime allowed to reducing also the BOD5 of the leachate (Figure 11). The removal is 83.48%.

The elimination of COD and BOD5 of the leachate is due to the coagulation by the lime. It is also probably due to the adsorption on metal precipitates, including those of Ca²⁺, which are formed in the basic pH range, such as hydroxides, phosphates, carbonates, etc., or by entrainment during the coagulation of

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Figure 10. Evolution of COD reduction as a function of the lime concentration (g/L).

Figure 11. Evolution of BOD5 reduction as a function of the lime concentration (g/L).
the suspended matter. They can be further adsorbed by the positive charge of the Ca\(^{2+}\) cations, which could constitute a bridge in the image of clay-humic complexes occurring clay coagulation.

COD and BOD\(_5\) decrease with lime increase simultaneously, giving a biodegradability ratio ranging between 1.21 and 3.27 with an average of 2.17 (Table 2), which denoting the biodegradability of the studied leachate, Kouassia, Ahossik, Koffiy et al. (2014). We note that at the first addition of 2 g/L of lime, the ratio COD/DBO5 increases and then decreases (Table 2, Figure 12). The increase is probably due to hydrolysis reactions or reactions leading to the release of entities more or less biodegradable. The decrease afterwards can be explained by the coagulation, precipitation and adsorption reactions which may occur as a result of the increase in the lime quantity; the mineral and refractory fractions tend to decrease also.

3.2.7. Evolution of Mineral and Metallic Elements

The concentration of the main heavy metals presents in the leachate decreases. The elimination rates are 77.14% for the iron (Figure 13), 77.78% for molybdenum (Figure 14), and 41.67% for cadmium (Figure 15). They are eliminated either by precipitation when the solubility product is reached or by simple adsorption on the various precipitates or on the coagulated suspension material, forming in solution. For the majority of them (Fe, Zn, Ba, Cr, Ni, Mn…), precipitation occurs when the pH increases, however their optimal pH do not coincide, Salem & Allia (2011). They precipitate in the form of hydroxides, carbonates, phosphates, etc.

![Figure 12](image_url). Evolution of the DCO, the DBO\(_5\) and the DCO/DBO\(_5\) according to increasing doses of the lime g/L.

**Table 2.** Ratio COD/BOD\(_5\) according to doses of the lime.

| Dose of lime (g) | 0 | 2 | 4 | 6 | 8 | 10 | 12 |
|-----------------|---|---|---|---|---|----|----|
| COD (g O\(_2\)/L) | 74.5 | 44.8 | 31.8 | 28.8 | 19.2 | 7.63 | 7.5 |
| BOD\(_5\) (g O\(_2\)/L) | 38.3 | 13.7 | 12.7 | 12.3 | 10.3 | 6.33 | 3.6 |
| COD/BOD\(_5\) | 1.95 | 3.27 | 2.50 | 2.34 | 1.86 | 1.21 | 2.08 |
Many researchers having realized the tests of coagulation have found results more or less similar by coagulation with the lime, El Bada, Assobhei, Kebabi et al. (2010), and Khalil, Bouaouine, Chtioui et al. (2015). According to Steeve, (1998), the treatment with the lime allows almost complete precipitation of numerous metals: Fe$^{3+}$, Al$^{3+}$, Fe$^{2+}$ at pH 7.4 - 7.5. Dean, Bosqui, & Lanouette (1972),
showed that the lime allows the removal of many metals such as copper, zinc, iron, manganese, nickel and cobalt.

**Figures 16-18** show the evolution of phosphorus, $P$, $K^+$ and $S$. The abatement rates are respectively 15.60% (Phosphorus), 20.57% ($K^+$) and 20.77% (S). They are probably eliminated by adsorption. Phosphorus and S may also be removed by precipitation as phosphate or metal sulphide.

**Figure 16.** Evolution of phosphorus abatement according to the lime concentration (g/L).

**Figure 17.** Evolution of potassium abatement in function of increasing doses of the lime (g/L).

**Figure 18.** Evolution of sulfur abatement as a function of increasing doses of the lime (g/L).
4. Conclusion

The physicochemical study of the leachate characterization of the public landfill of Fkih Ben Salah city showed strong levels of pollution which widely exceed the legal limit values. According to this study, the leachate treatment by coagulation process with the lime showed interesting reduction of the various pollutants: turbidity, organic matter, mineral matter, etc. That is due to the coagulation by Ca²⁺, the precipitation of heavy metals in form of hydroxides, phosphates, carbonates, etc. when the pH increases, and their adsorption rates. The yields obtained were: 92.95% of turbidity, 88.23% of suspended matter, 89.89% of COD, 90.83% of BOD5, 78.39% of Fe, 77.78% of Mo, 38.29% of Cd, 48.75% of Al, 50.24% of S²⁻, 20.57% of K⁺, 27.24% of Phosphorus and 19.53% of Cl⁻. On the other hand, the biodegradability ratio COD/BOD5 presents an average value of 2.17, indicating the biodegradability of the leachate, which is similarly obtained with the pretreated leachate. Thus, a complementary biological treatment can be envisaged to further reduce the polluting load.

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Conflicts of Interest

The authors declare no conflicts of interest regarding the publication of this paper.

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