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

M. Assou, A. Madinzi, A. Anouzla, M.A. Aboulhassan, S. Souabi, M. Hafidi

Abstract—In this work we examined the application of coagulation–flocculation for the treatment of stabilized leachates. Jar-test experiments were employed in order to determine the optimum concentration conditions for the removal of organic matter. Coagulant–flocculant combination, effective dosage Ferric chloride, Aluminum sulphate and Alginate were tested as conventional coagulants, whereas five commercial polyelectrolyte were co-examined: tree anionic and two cationic. The results indicate a coagulant FeCl₃ remove of 67.3% and 87% of COD and turbidity respectively from the stabilized landfill leachate for optimal concentration of 3000 mg/l and a variable concentration of flocculant Astral, removal efficiencies of COD and turbidity varies respectively by 64% and 100%. We obtained for variable concentrations of FeCl₃ and an optimal concentration of 198 mg/l of flocculant Astral yields disposal respectively 70.5% and 84% for COD and turbidity. These results are comparable to those obtained with FeCl₃ alone. Furthermore, it is noted that the amount of sludge produced by the FeCl₃ is less than that produced in the case of mixing and Astral + FeCl₃ and in the case of the other mixtures studied.

Index Terms—coagulation–flocculation, landfill leachate, turbidity, COD, sludge.

I. INTRODUCTION

One of the most important problems in designing and maintaining a landfill is managing the leachate that is generated when water passes through the waste. The leachates consist of many different organic matters (biodegradable, but also refractory to biodegradation), where humic-type constituents consist of an important group (Kang et al., 2002, Lou and Zhao 2007) [1, 2], as well as ammonia-nitrogen, heavy metals, chlorinated organic and inorganic salts (Tatsi and Zouboulis, 2002, Jing and al.2013) [3,4]. The characteristic of landfill leachates is a combination result of a complex number of factors including soil properties, weathering conditions, garbage composition. The composition and concentration of contaminants are influenced by the type of deposited wastes, the quality of refuse, hydrogeological factors and mainly by the age of landfill (Bu and al.2010) [5]. Regardless of the nature of the compounds, they constitute a potential pollution problem for local ground and surface waters. Landfill leachate, a high-strength typical wastewater, may be characterized by four groups of pollutants including dissolved organic matter (CH₄, volatile fatty acid, fulvic-like and humic-like compounds), inorganic macro components (Ca, Mg, Na, K, NH₄ +, Fe, Mn, Cl−, SO₄ ²− and HCO₃ −), heavy metals (Cd, Cr, Cu, Pb, Ni and Zn), and xenobiotic organic compounds (aromatic hydrocarbons, phenols and chlorinated aliphatics) (Jing and al.2013) [4]. However, there is no information regarding the treatment of landfill leachate using deep shaft bioreactor in literature (Jing and al.2013) [4]. Moreover, the varied quality and quantity of leachate subjected to seasonal variations refuse compositions; waste ages and landfilling techniques caused a challenge to meet the increasingly stringent discharge standards in many countries (Lou and al. 2007) [2]. Analysis of leachate after membrane filtration can qualify and quantify the dissolved pollution as was shown (Lou and al. 2007) [2]. The treatment processes of leachate are very complicated, expensive and generally require various process applications because of their high loading, complex chemical composition and seasonally variable volume (Bu et al. 2010, Kurniawan and al.2006b)) [5,6]. At present, no single unit process is available for proper leachate treatment simply because of the high concentrations of COD and nitrogen as well as color. Many technologies have been applied to remove the pollution of leachates biological treatment (Jing and al.2013) [4], activated carbon (Aziz and al. 2011) [7]. Advanced chemical oxidation, absorption, etc. (Renou and al. 2008, Foo and Hameed 2009) [8, 9]. Most of the treatment processes for wastewater treatment could be adapted for leachate treatment. Coagulation/flocculation is a commonly used process in wastewater treatment in which compounds such as ferric chloride Aluminum Sulphate and/or polymer are added to landfill leachate in order to destabilize the colloidal materials and cause the small particles to agglomerate into larger settle able flocs. Several studies have reported the examination of this process for the treatment of industrial wastewater, especially with respect to performance optimization of coagulant/flocculant, determination of experimental conditions, assessment of pH and investigation of flocculant addition (Renou and al.2008, Anouzla and al.2014). [8, 10]. The advantages of the proposed physico-chemical method for the treatment of leachates are mainly simplicity, low cost, good removal efficiencies and easy onsite implementation. This method could be used for pre- or post-leachate treatment in combination with biological treatment process. As a result of the apparent inability of the method for sufficient pollutant removal, the cost of the high chemical dosages that are required, and the associated problems of the chemical sludge that is generated, it could be suggested that no single leachate
treatment method, biological or physicochemical, is able to produce an effluent with acceptable quality, and that both approaches should be appropriately combined. The leachate characteristics vary with time and from site to site because they depend on the type of wastes disposed, rainfall, age of the landfill and design of the landfill etc (Renou and al. 2008) [8]. Physical chemical parameters of the landfill leachates can usually be represented in terms of the parameters such as COD, BOD, ratio of BOD/COD, color, pH, alkalinity, oxidation reduction potential and heavy metal (Kjeldsen and al.2002) [11]. The main objective of coagulation and flocculation process is the removal of organic compounds and heavy metal from the stabilized landfill leachate.

II. MATERIALS AND METHODS

A. Sampling Procedures

Leachate: Leachate samples were collected from the city of Moham media landfill (figure 1). Samples were collected in 50 L plastic containers, transported to the laboratory and stored at 4°C. Leachate samples were removed from the refrigerator and left under ambient temperature for about 2 hrs before the jar test was performed. Then, the samples were thoroughly stirred to resuspend settled solids before any further tests were conducted. Figure 1 illustrates the sampling point leachate used for this study.

![Fig 1: Photograph of the leachate outflow into the collector.](image)

Note the color of the effluent

B. Reagents

The two principal inorganic coagulants used in landfill leachate treatment are salts of FeCl3. Ferric or Aluminum ion is the coagulant most often chosen to destabilize the colloidal and suspended solids. In this study, Ferric Chloride and Aluminum Chloride were used as coagulant. The addition of certain commercial polyelectrolytes was also examined, tree anionic polyelectrolytes Chemic 5161, 5162 et 5163. The flocculants in from of powder are supplied by CHEMIC Factory (Italy). Astral and Superfloc are two cationic polyelectrolyte in from of liquid. They are provided by Astral and Lesieur society (Casablanca).

C. Analysis Techniques

The turbidity was determined by an HI 93703 Microprocessor turbidity meter. Chemical oxygen demand: COD and other physicochemical parameters (TKN, total phosphate, etc.) for landfill leachate characterization were determined according to Standard Methods (Standard Methods 2005) [12]. Volume of sludge: At the end of the slow mixing stage, the beaker contents are transferred into Imh off cones and allowed to settle for one hour. The volume of settled sludge in the cone is recorded according to volumetric method (AD Eaton and al.1995) [13].

D. Experimental Procedure

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: initial rapid mixing at 160 rpm for 10 min, followed by slow mixing for 20 min at 30 rpm, the final settling step for 1 h. Coagulation-flocculation was conducted with optimized operational parameters determined earlier. Six polyethylene beakers of equal volume were used to examine the different dosages of coagulant and initial pH in each run. Sample bottles were thoroughly shaken to resuspend any settled solids, and the appropriate volume of sample 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 sulphate stock solution was added to a jar containing 1liter of landfill leachate at different pH values adjusted with H2SO4 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 settling, the supernatant was withdrawn for analysis. To assess the efficacy of coagulants and flocculants for leachate treatment, the following parameters were determined: turbidity, chemical oxygen demand (COD) and decanted sludge.

E. Characteristics of Landfill Leachate

The characteristics physico-chemical parameters the xamined stabilized leachates samples are presented in Table 1.

<table>
<thead>
<tr>
<th>Parameter</th>
<th>Range</th>
<th>Average</th>
</tr>
</thead>
<tbody>
<tr>
<td>pH</td>
<td>7.7-8.92</td>
<td>8.26</td>
</tr>
<tr>
<td>Conductivity ms/cm</td>
<td>25.6-35.9</td>
<td>102.2</td>
</tr>
<tr>
<td>Turbidity NTU</td>
<td>63-140</td>
<td>31.17</td>
</tr>
<tr>
<td>HPO42- mg/L</td>
<td>592.4-2128</td>
<td>1693.28</td>
</tr>
<tr>
<td>Sulphate mg/L</td>
<td>77.47-218.7</td>
<td>156.04</td>
</tr>
<tr>
<td>Tot phosphate mg/L</td>
<td>1226.6-2217</td>
<td>1879.5</td>
</tr>
</tbody>
</table>
Sometimes the pH value of old leachate was lower than the stabilized leachate. In addition, leachate samples displayed high concentrations of contaminants and as expected. The variation of leachate characteristics were attributed to a number of reasons, such as variations in the composition of deposited solid wastes, landfill age, hydrogeology of landfill site, specific climate conditions and moisture routing through the landfill (Chu et al., 1994) [14]... From this table, it can be deduced that COD removal rates, range between 20 to 25%, without lime addition. In addition, the removal of organic matter (as expressed by COD) was maintained almost constant with the coagulant dosage. The figure 2 shows the evolution of the settling (2 hours) of waste leachate for several samplings. Furthermore, Figure 3 shows the percentage removal of pollution leachate (COD). These results show that the removal of pollution discharges leachate stabilized yield varies between 5 and 33%.

### II. RESULTS AND DISCUSSION

#### A. Performance Evaluation

Once the optimum conditions had been established for each of the coagulants and flocculants aids studied, the following was determined: the COD, turbidity and the amount of sludge. Table 2 shows the optimal concentrations of different coagulants and flocculants used for the study and the values of COD and turbidity obtained for the optimum. The advantages of the proposed physico-chemical method for the treatment of leachates are mainly simplicity, low cost, good removal efficiencies and easy onsite implementation. As a result of the apparent inability of the method for sufficient pollutant removal, the cost of the high chemical dosages that are required, and the associated problems of the chemical sludge that is generated, coagulation/flocculation is a commonly used process in water and wastewater treatment in which compounds such as ferric chloride and/or polymer are added to wastewater in order to destabilize the colloidal materials and cause the small particles to agglomerate into larger settleable flocs. Several studies have reported the examination of this process for the treatment of industrial wastewater, especially with respect to performance optimization of coagulant/flocculant, determination of experimental conditions, assessment of pH and investigation of flocculant addition.

#### B. Removal of COD and turbidity

Figure 4 illustrates the comparative study for removal COD and turbidity by different coagulants and flocculants.
The optimum dose of a coagulant or flocculant is defined as the value above which there is no significant difference in the increase in removal efficiency with a further addition of coagulant or flocculant (Alkhafaj et al., 2014) [15]. Thus, the optimum doses of ferric chloride that enhanced COD removal were 3000 mg/l. In this work during the addition of ferric chloride to the partially stabilized leachates, a higher COD removal rate was observed, reaching up to 67.3% and 87% for turbidity with 700 mg/l of sludge production. The use of 3000 mg/l dose of ferric chloride at different doses of polyelectrolyte increased the removal of COD and turbidity at all the doses of the polyelectrolyte, hence, this would not be a suitable decisive system for determining optimum dose. For optimal concentration of 3000 mg/l and a variable concentration of flocculant Astral, removal efficiencies of COD and turbidity varies respectively by 64% and 100%. We obtained for variable concentrations of FeCl3 and an optimal concentration of 198 mg/l of flocculant Astral yields disposal respectively 70.5 and 84% for COD and turbidity. These results are comparable to those obtained with FeCl3 alone.

### Table 2: Dose and optimal for different coagulants and flocculants aids used

<table>
<thead>
<tr>
<th></th>
<th>Concentration</th>
<th>COD (mg/l)</th>
<th>Turbidity (NTU)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Al2(SO4)3</td>
<td>1000</td>
<td>940.07</td>
<td>20.04</td>
</tr>
<tr>
<td>FeCl3</td>
<td>3000</td>
<td>808.95</td>
<td>15.32</td>
</tr>
<tr>
<td>Alginate</td>
<td>128</td>
<td>2520</td>
<td>0.00</td>
</tr>
<tr>
<td>Astral Floculant</td>
<td>198</td>
<td>1296.30</td>
<td>30.65</td>
</tr>
<tr>
<td>Superfloc Floculant</td>
<td>200</td>
<td>1133.03</td>
<td>35.36</td>
</tr>
<tr>
<td>FeCl3 variable + Superfloc (200 mg/l)</td>
<td>3000</td>
<td>910.38</td>
<td>51.86</td>
</tr>
<tr>
<td>Superfloc variable + FeCl3 (3000 mg/l)</td>
<td>20</td>
<td>1649.32</td>
<td>18.86</td>
</tr>
<tr>
<td>FeCl3 variable + Astral (192 mg/l)</td>
<td>5000</td>
<td>727.31</td>
<td>15.32</td>
</tr>
<tr>
<td>Astral variable + FeCl3 (3000 mg/l)</td>
<td>600</td>
<td>890.59</td>
<td>0.00</td>
</tr>
<tr>
<td>Alginate variable + FeCl3 (3000 mg/l)</td>
<td>100</td>
<td>1301.25</td>
<td>22.40</td>
</tr>
<tr>
<td>Chemic 1</td>
<td>4</td>
<td>1608.01</td>
<td>29.47</td>
</tr>
<tr>
<td>Chemic 2</td>
<td>4</td>
<td>1520</td>
<td>31</td>
</tr>
<tr>
<td>Chemic 3</td>
<td>4</td>
<td>1484.32</td>
<td>41.25</td>
</tr>
</tbody>
</table>

**Fig 4**: Comparative study of removal COD and turbidity by different coagulants and flocculants

The optimum dose of a coagulant or flocculant is defined as the value above which there is no significant difference in the increase in removal efficiency with a further addition of coagulant or flocculant (Alkhafaj et al., 2014) [15]. Thus, the optimum doses of ferric chloride that enhanced COD removal were 3000 mg/l. In this work during the addition of ferric chloride to the partially stabilized leachates, a higher COD removal rate was observed, reaching up to 67.3% and 87% for turbidity with 700 mg/l of sludge production. The use of 3000 mg/l dose of ferric chloride at different doses of polyelectrolyte increased the removal of COD and turbidity at all the doses of the polyelectrolyte, hence, this would not be a suitable decisive system for determining optimum dose. For optimal concentration of 3000 mg/l and a variable concentration of flocculant Astral, removal efficiencies of COD and turbidity varies respectively by 64% and 100%. We obtained for variable concentrations of FeCl3 and an optimal concentration of 198 mg/l of flocculant Astral yields disposal respectively 70.5 and 84% for COD and turbidity. These results are comparable to those obtained with FeCl3 alone.

### C. Amount of Sludge Produced

In addition to pollution removal, sludge production is considered in this work, as it may affect the economic feasibility of the proposed method. In the solid separation, sludge dewatering has been pointed out as one of the most expensive processes. The volume of sludge produced during the coagulation/flocculation process is given in figure 5. The combined use of inorganic FeCl3 and organic polyelectrolytes was found very efficient, resulting in the production of relatively low sludge volumes (Figure 5), similar to the corresponding findings of literature studies (Aguilar et al., 2002) [16]. However, the handling and removal of the sludge generated during the coagulation–flocculation process are important aspects and have to be considered with caution for the optimum selection of reagents, which will be used as coagulants/flocculants.

**Fig 5**: Comparative studies for production sludge (ml/L) by different coagulants and flocculants
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 (Alkhafaji et al.(2014) [15]). The wet sludge volume at the bottom of the jar test beakers after 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 and flocculent type and dose (mg/l). The volume of sludge produced, reduced considerably with increasing dose of polyelectrolyte in the coagulation process. This may be due to the non-ionic 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 chemically combine 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 water content of the sludge produced when ferric chloride alone was used and that produced when combination of ferric chloride and polyelectrolyte was used were measured after centrifugation and it was found that combination of ferric chloride and polyelectrolyte produced much water and less sludge than when ferric chloride alone was used. Leachate and the high initial content of pollutants (especially COD and ammonia-nitrogen), indicates the incorporation of an appropriate physical–chemical method, such as coagulation–flocculation, before the application of a biological one, which will result in the partial removal of organic matter and of possible existing toxicants (heavy metals), therefore reducing the initial loading of pollutants and permitting the application of a more efficient secondary biological treatment. The same trend was also observed during the combined action of coagulants–polyelectrolyte in leachate samples. Furthermore, pH control resulted in higher removal efficiencies of organic matter, than in cases without any pH adjustment. In general, chemical coagulation/flocculation is a process, which is highly pH dependent.

### III. CONCLUSION

The application of coagulation/precipitation treatment for raw leachate (before composting process) collected from a composting plant was examined in this study. Leachate was characterized by low pH value and high concentration of pollutants; especially that organic matter was in the range of 22520 mg/l COD The present study proved a coagulant FeCl₃ remove 67.3% and 87% of COD and Turbidity respectively from the stabilized landfill leachate. The combination of Ferric Chloride, Alginate and Super floc produced much water and less sludge (ml/l) than when ferric chloride was used with flocculant Astral. The addition of Ferric Chloride or Alum coagulants to leachates resulted in the reduction of COD values. The addition of high amount of coagulants was found to affect slightly the process efficiency. Furthermore, comparing the results of coagulation experiments, it can be observed that ferric chloride was more efficient than alum for the removal of COD. In this work, during the addition of ferric chloride to the partially stabilized leachates, a higher COD removal rate was observed, reaching up to 67.3% and 87 % for turbidity with 700 ml/l of sludge production. For optimal concentration of 3000 mg / l and a variable concentration of flocculant Astral, removal efficiencies of COD and turbidity varies respectively by 64% and 100%. We obtained for variable concentrations of FeCl₃ and an optimal concentration of 198 mg/l of flocculant Astral yields disposal respectively 70.5 and 84% for COD and turbidity. These results are comparable to those obtained with FeCl₃ alone. Furthermore, it is noted that the amount of sludge produced by the FeCl₃ is less than that produced in the case of mixing and Astral + FeCl₃ and in the case of the other mixtures studied.

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