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Comparative study of leachate treatment by coagulation-flocculation process using iron-based coagulants: A case study on Souk-Ahras city

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GRAPHICAL ABSTRACT

Leachate lagoon basins at the landfill Technical Center of Souk-Ahras city.

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ABSTRACT

The objective of this study was to evaluate the coagulation-flocculation process in the clarification of leachate from the landfill Technical Center of Souk-Ahras city using three coagulants based on iron: ferrous sulfate $FeSO_4.7H_2O$; ferrous chloride $FeCl_2.4H_2O$ and ferric chloride $FeCl_3$. The influence of some parameters namely pH leachate, dose and nature of coagulant and nature of flocculant was studied. The best treatment efficiency was obtained at 20 % of $FeCl_3$ giving a turbidity of 4.09 NTU with pH adjustment of the raw leachate at acidic pH (3.5 ± 0.2) before coagulant addition and at a basic pH (7.5 ± 0.2) after addition of coagulant. The iron valence and the nature of anion at which is linked, played a determinant role in the clarification of leachate. The treatments made with ferric chloride in the presence of a flocculant have proved that the starch was more efficient than lime giving abatement rates of 99 % for COD and 85 % for BOD_5.

1. Introduction

The leachate represents the effluent of highly polluted water coming from the sanitary landfill. The leachate is contaminated with various products and pollutants present in this bioreactor that constitutes the landfill. Monitoring this effluent is necessary both to prevent health and environmental risks and to regularly collect data, which will be used to improve their treatment (Jinghuan et al. 2015; Majdy et al. 2015). The composition of the leachate depends on several parameters such as pH, site age, type of waste stored, plus the climate and the season. In order to choose the most suitable process for each effluent, it is necessary to consider a series of conditions such as the characteristics of the raw wastewater in terms of pollutants and also the quality of the final effluent required according to the specifications of treated water related to regulations (Sanphoti et al. 2006; Wang et al. 2011). Several techniques have been applied for the treatment of

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leachate, the most recent are the physico-chemical techniques which have experienced a great development, in particular the coagulation-flocculation process. Coagulation-flocculation is a water purification process, used for the treatment of drinking water or waste water. It is an attractive process because of its many advantages such as its environmental compatibility, its adaptability, its efficiency and its low cost (Önen et al. 2018).

The objective of this work was to treat leachate from the landfill Technical Center of Souk-Ahras city, by applying the coagulation-flocculation technique. The evaluation of the treatment was examined by using three coagulants based on iron; ferrous sulfate $FeSO_4.7H_2O$, ferrous chloride $FeCl_2.4H_2O$ and ferric chloride $FeCl_3$; in order to achieve a good reduction in turbidity and organic charge of the leachate as a function of the operational parameters. Each of these coagulants has a specific behavior with respect to leachate. Despite the various researches carried out on the treatment of leachate discharges by coagulation flocculation, a similar comparison between these three coagulants in the literature, particularly in the field of leachate treatment, has not been mentioned which constitutes the main novelty of this contribution.

2. Materials and methods

2.1. Implementation of coagulation-flocculation experiments

A volume of leachate was put in a beaker then placed on a magnetic stirrer. The hydrogen potential of leachate was adjusted to 3.5 ± 0.2 by adding a few drops of H_2SO_4. An equal volume of a coagulating solution prepared at a well-defined mass percentage was introduced into a burette and dispersed in the contaminated solution (the leachate). Then the mixture was stirred during a 15 min. At the end of this operation, the pH of leachate is readjusted between 7 and 8 using a concentrated NaOH solution in order to promote the formation of iron hydroxide precipitates. According to the experiments carried out, all the readjustment pHs were around 7.5 ± 0.2. At the end of coagulation, a flocculant i.e. starch (organic flocculant) or lime CaO (mineral flocculant) at 20 % was added to leachate and the mixture was still stirred for 15 min (flocculation time). The mixture obtained after this step was decanted for 6 h in a separating funnel in order to separate between the flocs formed and the supernatant. The flocs obtained after decantation were separated from supernatant by filtration through filter papers, dried in an oven at 105 °C for 6 h then weighed. In contrast, the supernatant was collected for analysis. All experiments were conducted at room temperature 18 ± 2 °C.

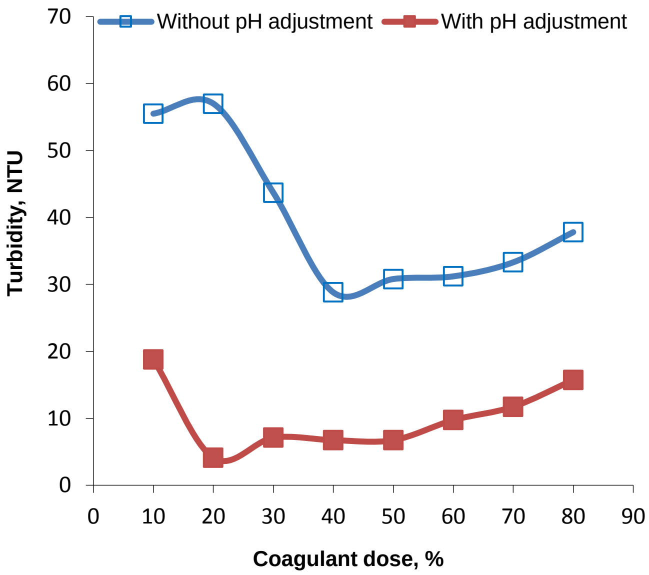
2.2. Analytical procedures

All leachate solutions have undergone some analyzes before and after treatment with the coagulant-flocculation process. The pH and conductivity were measured by a pH meter and conductmeter from Hanna Instruments while the refractive index was measured by a refractometer ("Zuzi Model-315 ABBE brand"). The chemical oxygen demand (COD) was determined using a COD-Hanna reactor (model C 9800) and the biological oxygen demand (BOD_5) was measured by an Oxitop (WTW). The sludge recovered after decantation was examined using Fourier transform infrared spectroscopy (FTIR) by means of a spectrophotometer of type IRAffinity-1S SHIMADSU in combination with a single reflection ATR.

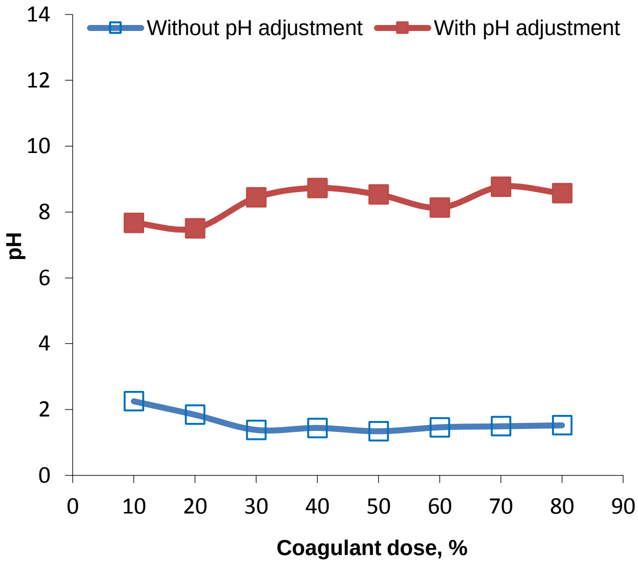
3. Results and discussion

3.1. Effect of pH adjustment of the medium

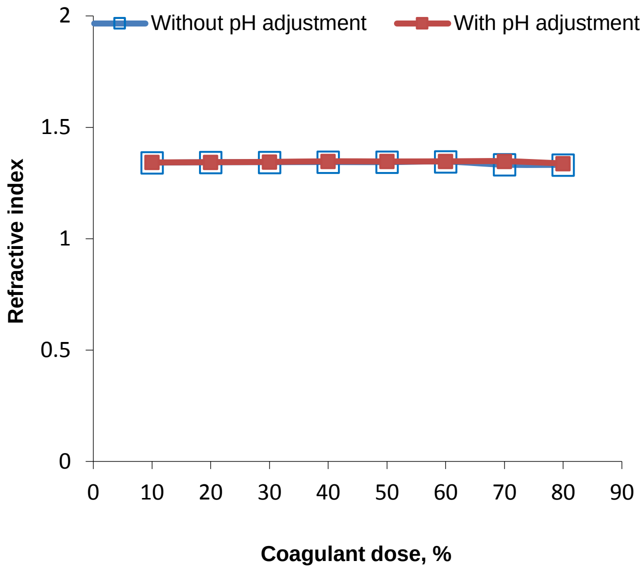
Figs. 1 a-e show the results of the analyzes i.e. turbidity, pH, refractive index, conductivity and mass of the flocs respectively; for the leachate treated with FeCl3 at different doses varying from 10 to 80 %. Coagulation tests carried out without pH adjustment (the empty symbols) showed that it has been a considerable reduction in the turbidity of the treated leachate, especially as the concentration of coagulant increases. For 40 % of ferric chloride the leachate turbidity was attenuated about 70 %, hence a value equal to 28.8 NTU. On the other hand, for a treatment carried out with pH adjustment (the full symbols) i.e. before adding coagulant the leachate pH was adjusted at 3.5 ± 0.2, once the coagulant is dissolved, the pH was readjusted to 7.5 ± 0.2, the turbidity attenuation is estimated at around 96 %, hence a turbidity of 4.09 NTU at 20 % of coagulant (Fig. 1a). From figure 1b, in the case of pH adjustment of the feed solution; the pH values of treated leachate are slightly basic for all the coagulant concentrations used; they are between 7.5 and 8.7 which confirms that the flocculation mechanism was stable (Bouranene et al. 2015). On the other hand, for the leachate which has not undergone a pH adjustment, the final pH values after treatment vary between 1.3 and 2.3. This result can be explained by the acidic nature of the added coagulants (Djeffal et al. 2019). By examining the values of the refractive indices (Figure 1c), it was found that the difference between the two curves is negligible varying from 0.01 % to 1.15 %. According to the measured values, the highest refractive indices are obtained with pH adjustment; this is due to the addition of the acid and / or base to adjust the medium. The same trend was observed for the results of electrical conductivity for the recovered supernatant (Fig. 1d). The two curves are very close so a negligible difference varying between 2.44 % and 17.11 %. These results are perfectly in agreement with those of the refractive index which shows that the supernatants are charged almost with the same composition; which has been reflected by close values of conductivity and refractive index.



(a)



(b)



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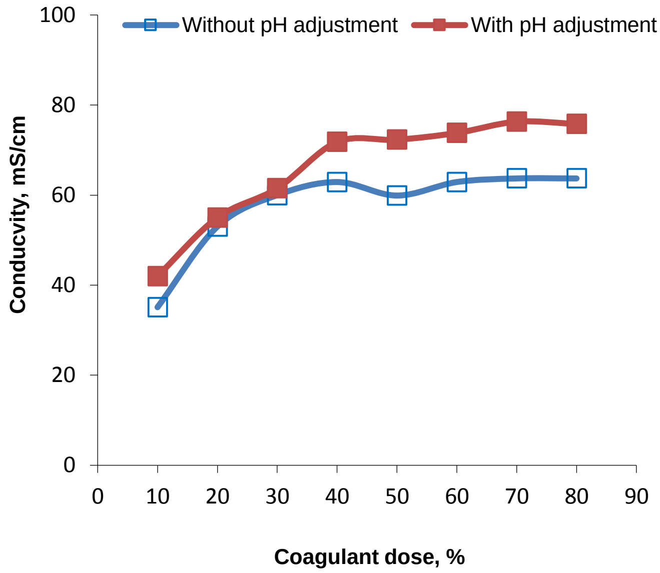

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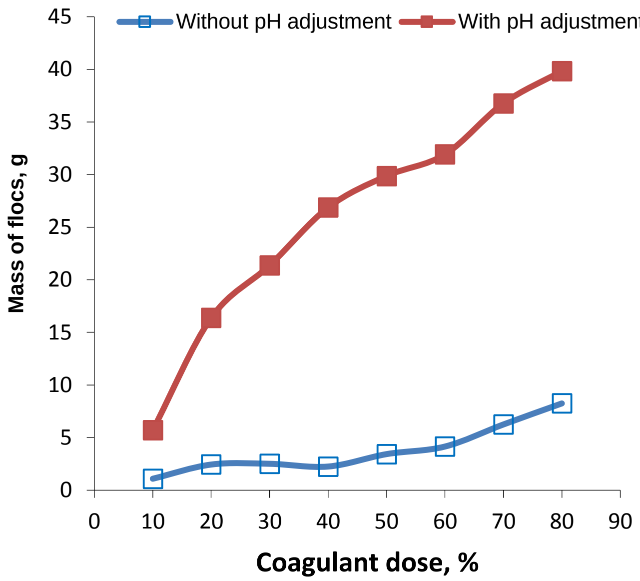

(e)

Fig. 1a-e. Variation of some physicochemical parameters of supernatants as a function of the $FeCl_3$ coagulant dose for two cases ((□) without pH adjustment of the leachate; (■) with pH adjustment of the leachate; Stirring speed = 400 rpm; Stirring time = 15 min).

Fig. 1e shows that the treatment of raw leachate without any adjustment of pH gives less sludge (flocs) in comparison with that carried out with adjustment of pH for the studied range of concentrations. This result is due to the appearance of iron hydroxide flocs following a pH favorable to their formation (pH = 7.5 ± 0.2). These flocs increase by increasing the coagulant dose which led to maximum clarification of leachate with maximum neutralization of colloidal particles. Indeed, the flocs formed, of larger size and concentration, allow the sweeping and filtration of the liquid, which contributes to a more efficient trapping of impurities present (Monette et al. 2000). Therefore the pH adjustment of leachate before and after addition of coagulant is very interesting to have a better clarity of supernatant.

3.3. Effect of dose and nature of coagulant

To investigate the influence of this parameter, the pH of row leachates was adjusted at 3.5 ± 0.2 before addition of coagulant and at 7.5 ± 0.2 after addition of coagulant. As depicted in Fig. 2, the optimal clarification of the leachate solution is obtained for a dose of 20% for the three chosen coagulants; hence final turbidities equal to 4.09 NTU (i.e. drop of 95. 7 %), 41.5 NTU (i.e. drop of 56.4 %) and 44.7 NTU (i.e. drop of 53.1 %) for $FeCl_3$, $4H_2O$ $FeCl_2.4H_2O$ and $FeSO_4.7H_2O$ respectively. Furthermore, the supernatants recovered after treatment with ferric chloride compared to ferrous chloride are the most clearer with a very low turbidity for the entire studied range of concentration. Given that these coagulants have the same anion (Cl^-) this phenomenon can be attributed to the difference in the valence of iron (Fe^{3+}/ Fe^{2+}), a higher charge leads to a more efficient neutralization of colloids (Al-Malack et al. 1999).

In contrast ferrous sulfate is generally more efficient than ferrous chloride; this is probably due to the nature of anion linked to iron. In fact, sulfates are usually more effective than chlorides since it is the same divalent cation (Fe^{2+}). This result is in agreement with previous works which have shown that the efficacy of sulfate ions used as coagulants for positively charged hematite particles was better than that of chloride ions (Bouranene et al. 2015; Liang and Morgan, 1990).

The sludge formed after treatment of the leachate with ferric chloride at a dose of 20 % was analyzed by FTIR (Fig. 3). The spectrum obtained shows a small peak at a wave number of 1000 cm^{-1} (characteristic of the C-O bond), a small peak at 1400 cm^{-1} (characteristic of the C-C bond), a small peak at 1600 cm^{-1} (characteristic of the bond C=C), a small peak at 2300 cm^{-1} which can be attributed to C≡N stretching vibration bond and a wide band between 3300 cm^{-1} and 3600 cm^{-1} which can include the OH bond, these OH can be hydroxyl groups linked to organic alcoholic or carboxylic molecules and/or simple hydroxyls belonging to the free water circulating in the mud. The pick observed at 3800 and 3900 correspond to the free O-H bond.

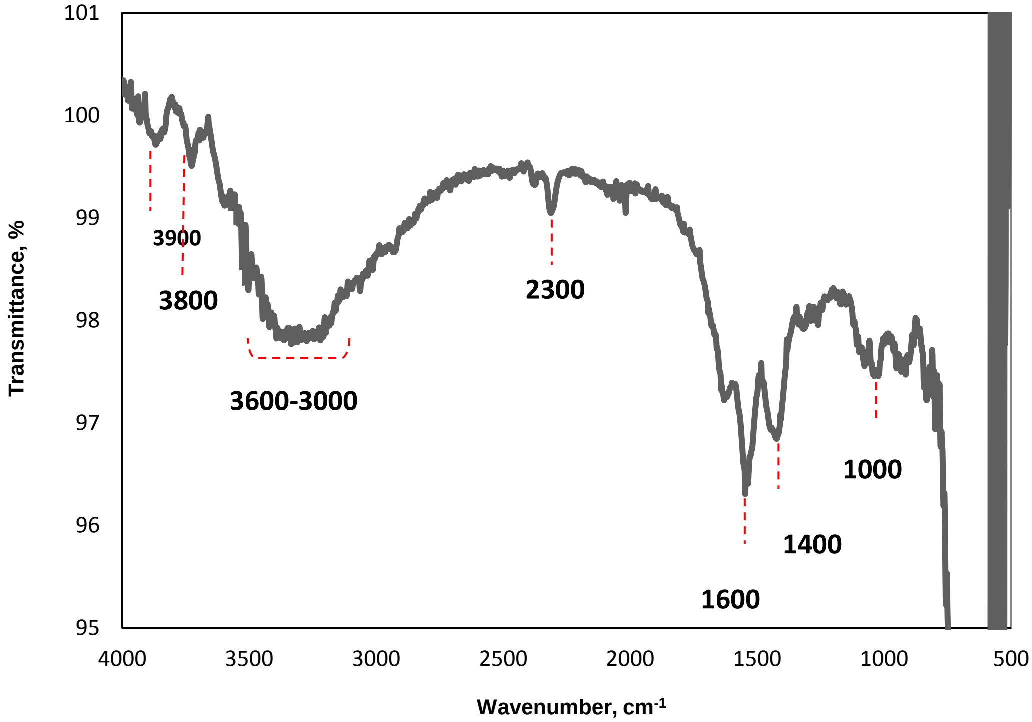

Fig. 3. Infrared spectra of sludge recovered after leachate treatment by coagulation-flocculation using ferric chloride FeCl3 at 20 % (pH of leachate was adjusted at 3.5 ± 0.2 before addition of coagulant and at 7.5 ± 0.2 after addition of coagulant, stirring speed=400 rpm, stirring time=15 min).

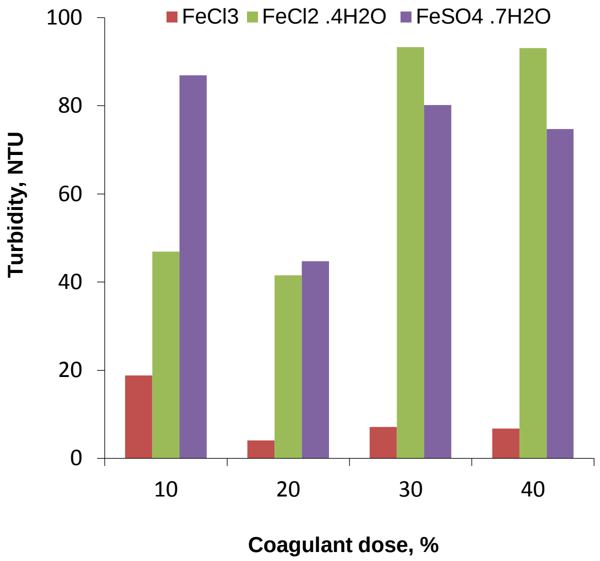

Fig. 2. Variation of the supernatant turbidity as a function of the dose and the nature of coagulant (pH of leachate was adjusted at 3.5 ± 0.2 before addition of coagulant and 7.5 ± 0.2 after addition of coagulant, turbidity of raw leachate = 95.2 NTU; stirring speed=400 rpm, stirring time =15 min).

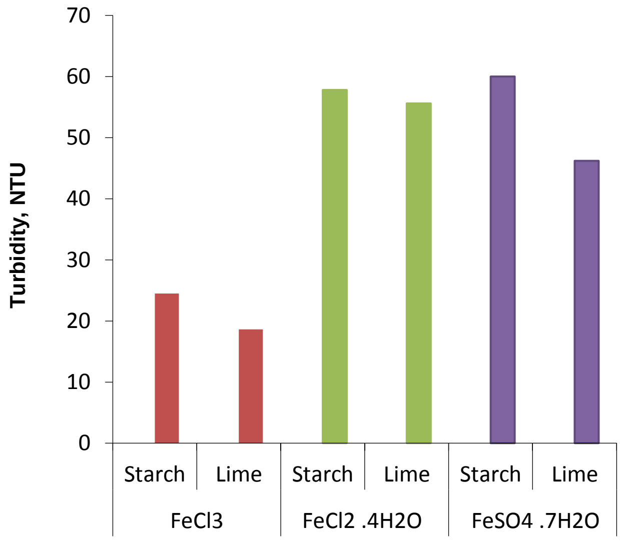

Fig. 4. Variation of the supernatant turbidity as a function of the nature of flocculant (starch or lime) in the presence of coagulant ($FeCl_3$ or $FeCl_2$.$4H_2O$ or $FeSO_4$.$7H_2O$) at 20 % (pH of leachate was adjusted at 3.5 ± 0.2 before addition of coagulant and at 7.5 ± 0.2 after addition of coagulant, turbidity of raw leachate=95.2 NTU, stirring speed=400 rpm, stirring time=15 min (coagulation or flocculation), flocculant dose=20 %).

3.4. Effect of addition of flocculant

The effect of adding a flocculant was investigated by following some analysis, namely turbidity, COD and BOD_5. Fig. 4 gives the turbidity values of the supernatants resulting from a treatment of leachate by coagulation-flocculation process, it can be clearly seen that the turbidity values are lower in the presence of lime than in the presence of starch whatever the coagulant used. This can be explained by the alkaline properties of lime which ensure a basic pH favorable for the formation of flocs and consequently, a better clarification of the leachates. CaO has been proven in the literature that helped to enmesh the colloidal particles (Barrington et al. 2004). The highest decreases in turbidity are obtained with a treatment carried out in the presence of ferric chloride with addition of lime (drop of approximately 80 %) and with addition of starch (drop of approximately 74 %).

Fig. 5 shows the results of the COD, it has been observed that starch ensures better removal of dissolved matter in the presence of ferric chloride giving the highest drop i.e. 99.4 % (final COD = 91.2 mg/L) unlike lime which has been proven better than starch in the presence of ferrous chloride, i.e. a drop of 98.7 % (final COD = 200 mg/L) and ferrous sulfate, i.e. a drop of 53.3 % (final COD =7000 mg/L). The COD reduction can be ascribed to the adsorption of organic and inorganic pollutants on the metal hydroxides involving Van Der Waals forces or hydrogen bonds (Djeffal et al. 2019; Achour et al. 2005; Wais-mossa et al. 1991). Indeed, it also appears that the flocculant effect depends on the coagulant nature. In the case of ferric chloride, starch which is an organic flocculant seems a good candidate to establish the link between the flocs than lime (Krentz et al. 2006; Vandamme et al. 2010).

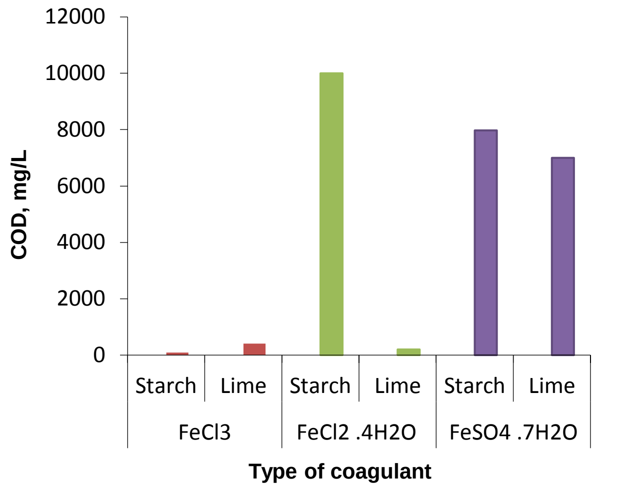

Fig. 5. Variation of the supernatant COD as a function of the nature of flocculant (starch or lime) and in the presence of coagulant ($FeCl_3$ or $FeCl_2 .4H_2O$ or $FeSO_4 .7H_2O$) at 20 % (pH of leachate was adjusted at 3.5 ± 0.2 before addition of coagulant and 7.5 ± 0.2 after addition of coagulant, COD of raw leachate > 15000 mg/L, stirring speed = 400 rpm, stirring time =15 min (coagulation or flocculation), flocculant dose = 20 %).

With regard to BOD_5 measurements (Figure 6), starch has been found to be effective whatever the coagulant used, the BOD_5 abatement is 85.7% (final BOD_5 = 100 mg/L). The starch has proven its effectiveness as a flocculant in the field of wastewater treatment, it exhibit an effective bridging flocculation effects à cause de son affinity for organic components besides its antibacterial properties (Wei et al 2018; Huang et al. 2017). Lime was also found to be good for the reduction of organic matter, giving a reduction of 85.7 % in BOD_5 with ferric chloride and 71 % with both ferrous chloride and ferrous sulfate. Some recent research has shown that addition lime to wastewater immediately produces abundant and insoluble precipitates capable of effectively sweeping away impurities, organic matter and any other contaminants (Prasad et al. 2019; Madeira et al. 2020).

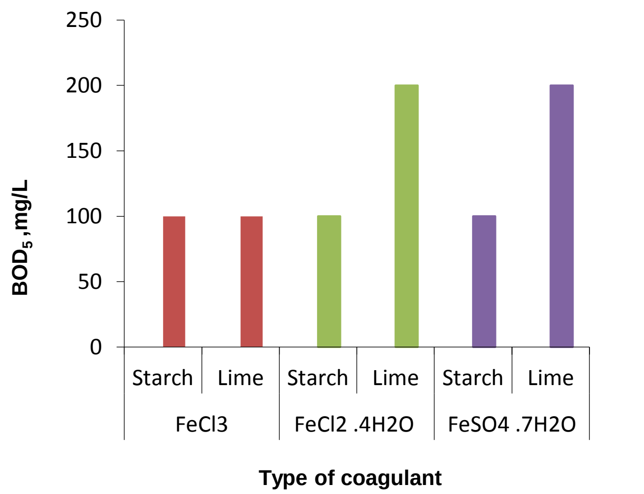

Fig. 6. Variation of the supernatant BOD_5 as a function of the nature of flocculant (Starch or Lime) and in the presence of coagulant ($FeCl_3$ or $FeCl_2 .4H_2O$ or $FeSO_4 .7H_2O$) at 20 % (pH of leachate was adjusted at 3.5 ± 0.2 before addition of coagulant and 7.5 ± 0.2 after addition of coagulant, BOD_5 of raw leachate = 700 mg/L, stirring speed = 400 rpm, stirring time = 15 min (coagulation or flocculation), flocculant dose =20 %).

Fig. 7 summarizes the results obtained following a treatment of leachate carried out with ferric chloride without any addition of flocculant and with addition of starch or lime as flocculants. It seems that adding the flocculant doesn't enhance the clarity of supernatants in terms of colloidal particles. However the presence of flocculant plays an important role in the elimination of dissolved matter in particular the starch polymer which presented high removal efficiencies for COD and BOD_5 (Teh et al. 2014; Fersi et al. 2018).

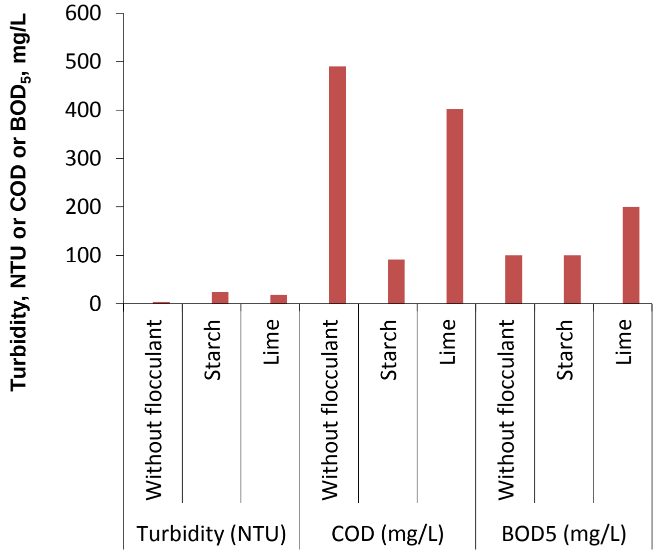

Fig. 7. Variation of some physicochemical parameters of supernatants recovered after treatment by ferric chloride coagulant ($FeCl_3$) at 20 % with and without flocculant addition (pH of leachate was adjusted at 3.5 ± 0.2 before addition of coagulant and 7.5 ± 0.2 after addition of coagulant, raw leachate: turbidity=95.2 NTU, COD > 15000 mg/L and BOD_5=700 mg/L, stirring speed=400 rpm, stirring time=15 min (coagulation or flocculation), flocculant dose =20 %).

4. Conclusions

The treatment of leachate from the public landfill of Souk-Aharas city by a coagulation flocculation process was investigated. Several experiments have been carried out with a view to improve the treatment efficiency of leachate in the presence of three iron-based coagulants (ferric chloride, ferrous chloride and ferrous sulfate) and in the presence of two flocculants lime and starch. The efficiency of the process was evaluated in terms of turbidity, sludge production, refractive index, final supernatant pH, electrical conductivity, chemical oxygen demand (COD) and the biological oxygen demand (BOD_5). Characterization of leachate sludge by infrared spectroscopy has shown the existence of functional groups proving the adsorption of organic matter on the hydroxides formed during flocculation. The pH adjustment of leachate revealed a better elimination of pollutants where an acid pH is favorable for neutralizing colloidal particles and a basic pH promotes the formation of flocs (metal hydroxides). The effectiveness of treatment depends strongly on the nature and dose of the coagulant. It has been found that 20% ferric chloride $FeCl_3$ with an adjustment of the leachate pH can lower significantly the turbidity to around 95.7 %. The treatment process by coagulation-flocculation depends on the valence of cation forming the coagulant and also on the nature of anion to which is linked. In this study, Fe^{3+} ions are more effective than Fe^{2+} and sulfates are more reliable than chlorides. The combination of starch with ferric chloride has shown great effectiveness in reduction of organic pollution and improving the visual appearance compared to lime with ferric chloride (a reduction of 99.4 % in COD and 85.7 % of BOD_5). Finally, this study paves the way for future research in the field of leachate treatment from landfill waste by the coagulation-flocculation process and other experimental studies are recommended for a better understanding of the mechanisms involved.

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