

## Treatment of partially stabilized landfill leachate using combinations of coagulation, Fenton oxidation and granular activated carbon (GAC) adsorption

F. Boumechhour\*, R. Kerbachi, A.E. Rechoum, M.S. Benmenni

Department of Environmental Engineering, Faculty of Science of Engineers, University of Boumerdes. Avenue du 1er Novembre 35000 Boumerdes, Algeria.

\*Corresponding author: fatimaboumechhour@gmail.com ; Tel.: +213 58083004.

### ARTICLE INFO

#### Article History:

Received : 05/09/2016

Accepted : 24/11/2016

#### Key Words:

Coagulation;  
Fenton oxidation;  
Activated Carbon;  
adsorption;  
Landfill leachate;  
Treatment.

### ABSTRACT/RESUME

**Abstract:** This study investigated the treatment performances of coagulation, Fenton oxidation, granular activated carbon adsorption and combinations between them aiming to provide an efficient method for the treatment of partially stabilized leachates. Leachates were collected from Ouled Fayet landfill (Algeria), samples were characterized with a low biodegradability ( $BOD_5/COD$  ratio about 0.15), COD of 8294 mg/l and pH around 8. The sequence of stages implemented was: (a) Coagulation/flocculation (COD removal  $\approx$  71 % with  $FeCl_3$  dosage of 1.6 g/l at pH 5); (b) Fenton oxidation (COD removal  $\approx$  74.6% using 5g/l of  $H_2O_2$ ,  $H_2O_2/Fe(II)$  molar ratio = 6 at pH 3.5); (c) Coagulation/flocculation followed by Fenton oxidation (>86 % of COD removal was achieved); (d) Fenton oxidation followed by coagulation/flocculation (COD removal  $\approx$  81.7% ) and (e) Granular activated carbon adsorption after coagulation and Fenton process ( very good reduction in COD : 94.2% ).

### I. Introduction

Leachates are generated at any landfill by the contact of water with wastes and they represent potential hazard for the environment. The quantity and quality of produced leachates depend on a number of factors: the type of deposited wastes, hydro-geological conditions, the age of the landfill and the phase of waste decomposition [1-3,4]. Leachate generated from landfills is a high-strength wastewater that may contain large amounts of organic matter and inorganic matter, with humic-type substances which represent an important part, as well as ammonia nitrogen, heavy metals, chlorinated organics and inorganic salts. Untreated leachate can permeate ground water or mix with surface waters and contribute to the pollution of soil, ground water, and surface water [5].

Coagulation was used to remove suspended solids (SS) and colloids from the leachate. The operating cost of coagulation is low, and the process is widely used in wastewater treatment scenarios [6]. The coagulation process destabilizes colloidal particles by the addition of a coagulant. To increase the

particle size, coagulation is usually followed by flocculation of the unstable particles into bulky flocculants so that they can settle more easily [7].

The Fenton process has been proposed and studied extensively in recent years [8-15] and it has been confirmed as a highly effective alternative for degrading recalcitrant organic matter in a variety of wastewaters, including landfill leachates. With the Fenton process, the high fraction of high molecular weight organics is degraded and partially removed, enhancing biodegradability. The potential of prior chemical oxidation is to convert initially recalcitrant compounds to more readily biodegradable intermediates [16, 17].

Adsorption process, a surface phenomenon by which a multi-components fluid (gas or liquid) mixture is attracted to the surface of a solid adsorbent and form attachments via physical or chemical bonds, is recognized as the most efficient and promising fundamental approach in the wastewater treatment processes [18]. A notable trend in the development of activated carbon, an adsorbent with its large porous surface area, controllable pore structure, thermo-stability and low

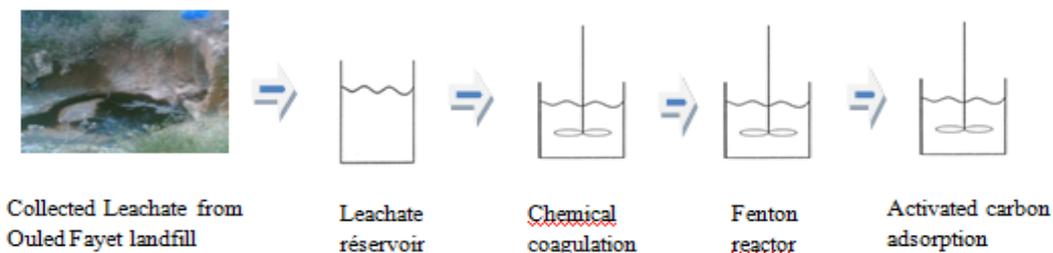
acid/base reactivity has been promulgated [19], owing to its superior ability for removal of a wide variety of organic and inorganic pollutants dissolved in aqueous media [20].

The primary aim of the work was to study the effectiveness of the Fenton's process and coagulation for the treatment of partially stabilized leachate, more specifically to investigate the optimum operational conditions for each method separately and the appropriate combinations between them. The second objective was to evaluate the potential of activated carbon adsorption process as a polishing (post-treatment) step after coagulation and Fenton process.

## II. Materials and methods

Samples of leachate were collected from Ouled Fayet site (South-West of Algiers), which is the first controlled landfill site in Algeria operating since 2002. Partially stabilized leachate was collected between May and June 2010 with a frequency of one sample per week. Leachate samples were collected in plastic bottles, transported to the laboratory, stored at 4°C and

subsequently characterized. Physico-chemical analyses were performed over the two days following sampling. The pH value, turbidity and conductivity of samples were measured with a pH meter, turbidimeter and conductivimeter, respectively. COD, total suspended solids (TSS) and volatile suspended solids (VSS), chloride and Metal values of samples were determined by using closed reflux method, gravimetry, AgNO<sub>3</sub> method and atomic absorption (AA) spectrophotometric method described in the standard methods [21], respectively. BOD<sub>5</sub> measurements were performed via an Oxi-top BOD analyzer. Total Kjeldahl nitrogen (TKN), NO<sub>3</sub>-N, NH<sub>3</sub>-N, NO<sub>2</sub>-N, PO<sub>4</sub>-P and sulphate (SO<sub>4</sub><sup>2-</sup>) were analyzed in accordance with standard methods [21]. The experimental apparatus of the present investigation consisted of three major parts: chemical coagulation, Fenton reactor; and activated carbon adsorption unit. The three treatment units could be operated independently or in combination. The batch mode was adopted for determining the individual operating conditions. The experimental schematic is shown in figure 1.



**Figure 1.** Flow process of the three treatment units of landfill leachate (coagulation, Fenton and activated carbon adsorption)

### II.1. Coagulation process

Coagulation /Flocculation experiments were conducted at ambient temperature (30-33°C) by using a conventional Jar-test apparatus, equipped with 6 beakers of 1l volumes. Leachate samples were removed from the refrigerator and were remained for about 2h under ambient temperature (30-33°C). Then, sample bottles were thoroughly shaken, for re-suspension of possibly settling solids, volume of 500 ml of sample was transferred to the corresponding Jar-test beakers and the pH values of samples were adjusted to the desired levels by the addition of appropriate amounts of H<sub>2</sub>SO<sub>4</sub> 95-97% (w/w). Pre-determined concentrations of Ferric (III) Chloride (FeCl<sub>3</sub>) were added first and mixed for 1 min under vigorous mixing conditions (300 rpm), and under slow mixing conditions (50 rpm) during other 20 min. Finally, the sample was allowed to settle without stirring for an hour and the supernatant was withdrawn from the beaker and was used for chemical analysis. Supernatant was

withdrawn from the beaker and was used for chemical analysis.

### II.2. Fenton process

Fenton process was performed in the same Jar-test used for the Coagulation /Flocculation experiments. Fenton treatment was carried out according to the following procedure: First, every beaker was filled with 500 ml of leachate and its pH was adjusted to the designed value by H<sub>2</sub>SO<sub>4</sub> 95-97% (w/w). The second step was the addition of Ferrous Sulphate (FeSO<sub>4</sub>, 7H<sub>2</sub>O). Subsequently hydrogen peroxide solution (H<sub>2</sub>O<sub>2</sub>, 30% w/w) was added rapidly under vigorous stirring at 300 rpm. After 2 hours of Fenton's treatment, the solution was allowed to settle for 45 min and the supernatant was withdrawn for analyses.

### II.3. Granular activated carbon adsorption

The precedent jar-test was employed for activated carbon adsorption treatment at room temperature

(30-33°C). Accurately weighted granular activated carbon (GAC) doses (2, 4, 6, 8, 10, 12, 14 and 16 g/l) were added to 300 ml of leachate with optimized 100 rpm stirring rate to keep agent in suspension for few minutes.

### III. Results and discussion

#### III.1. Leachate characterization

The average physico-chemical characteristics of the leachate are summarized in table1. It was an alkaline mixture of black colour with a low BOD<sub>5</sub>/COD ratio of 0.15, indicating that it contained highly refractory substances and slowly biodegradable organic materials. The effluent was classified as a partially stabilized leachate according to its characteristics. Other major compounds present in this effluent were ammonium and chloride. Several authors have found that High ammonia content is the primary cause of acute toxicity of municipal landfill leachate [22, 23].

*Table1. Landfill leachate composition*

Parameters	Mean	Range	Ecartype	Standard <sup>a</sup>
pH	7.7	7.6-7.7	0.03	6.5-8.5
Conductivity (ms/cm)	19.2	7.7-35.3	13.83	/
Total suspended solids (TSS) mg/l	972.78	675.6-1186.7	250.35	40
Volatile suspended solids (VSS) mg/l	492.8	200-771.1	249.67	/
COD mg/l	8294.4	7515.4-9073.3	652.73	120
BOD <sub>5</sub> mg/l	1270	931-1609	479.42	35
BOD/COD	0.15	0.11-0.19	0.05	/
NO <sub>3</sub> <sup>-</sup> mg/l	32.9	28.9-36.2	3.22	/
NO <sub>2</sub> <sup>-</sup> mg/l	4.9	3.4-6.7	1.63	/
NH <sub>4</sub> <sup>+</sup> mg/l	4051.6	3456.8-4683.2	517.93	/
TKN mg/l	3007.2	627.2-6204.8	2747.47	40
SO <sub>4</sub> <sup>-2</sup> mg/l	657.8	549-758	109.73	/
Orto PO <sub>4</sub> <sup>-3</sup> mg/l	156	115.1-220	45.18	10
Cl <sup>-</sup> mg/l	4816.9	1846-7810	2482	/
Fe mg/l	20.2	NA		3
Cd mg/l	0	NA		0.2
Cu mg/l	4.3	NA		0.5
Pb mg/l	2.16	NA		0.5

<sup>a</sup>Standard of the Environmental Quality (Sewage and Industrial Effluents), regulation 2006 (Algeria).

Among the metals that were identified in the leachate, Iron, copper and lead exhibited

dangerously high concentrations. The poor biodegradability and high ammonia concentration imply that the application of a biological treatment process will be inappropriate. Therefore, alternative treatment processes must be considered, particularly physico-chemical treatments and advanced oxidation technologies.

#### III.2. Leachate treatment

Five treatment schemes were examined:

- coagulation/flocculation,
- Fenton oxidation,
- coagulation/flocculation followed by Fenton oxidation,
- Fenton oxidation followed by coagulation/flocculation,
- Coagulation, Fenton oxidation and GAC adsorption

##### III.2.1. Coagulation /flocculation

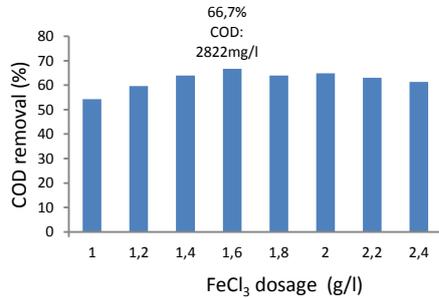
###### III.2.1.1. Effect of coagulant dose on coagulation

Once the partially stabilized leachate pH was adjusted to 4.5, various quantities of Ferric Chloride (from 1 to 2.6 g/l) were added to and the COD removal determined. As it can be seen in figure 2, increasing the concentration of coagulant improved the COD removal efficiency achieving 66.7% COD removal at concentration of 1.6 g/l of ferric chloride. Then, we observed a slow decrease in COD removal up to 61.4 %. This could be attributed to the re-stabilization of colloidal particulates when coagulants were used in excess of optimum dosage value. Which means colloids can absorb the cations and become positively charged, hence, may be stable again as a result of electrical repulsion, diminishing the capacity of organic matter removal.

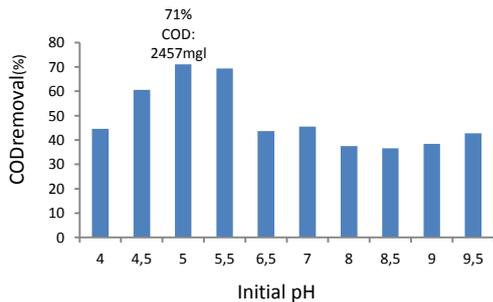
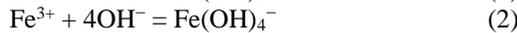
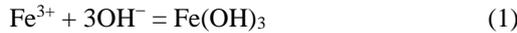
###### III.2.1.2. Effect of initial pH on coagulation

Destabilisation is a primary driving force in flocculation of wastewater and the suspension stability is related with pH according to the zeta potential pH relationship; once the optimum dosage of FeCl<sub>3</sub> had been obtained, a series of experiments were then carried out employing the optimum dosage of 1.6 g/l of FeCl<sub>3</sub> while pH values were varied from 4 to 9.5. Figure 3 indicated that the most effective removal of organic substances (71%) was achieved at pH 5. This result can be explained by the different hydrolyzed species of iron. In a basic medium, iron in oxidation state Fe<sup>3+</sup> can react with hydroxyl (OH<sup>-</sup>) to form Fe(OH)<sub>3</sub> or Fe(OH)<sub>4</sub><sup>-</sup>, using equations (1) and (2). In acidic medium, ferric iron can hydrolyze and form polynuclear cations as shown in equation (3). In this equation, Fe<sub>x</sub>(OH)<sub>y</sub><sup>(3x-y)</sup> may represent

Fe (OH)<sub>2</sub><sup>+</sup>, Fe<sub>2</sub>(OH)<sub>2</sub><sup>+4</sup>, Fe<sub>3</sub>(OH)<sub>4</sub><sup>+5</sup> and other positively charged species [24]. Obviously, the polynuclear cations are preferable to Fe (OH)<sub>3</sub> and Fe (OH)<sub>4</sub><sup>-</sup> as almost all colloidal impurities in the water are negatively charged [25].



**Figure 2.** Effect of coagulant dosing on COD removal efficiency (pH=4.5; original COD =8474.7 mg/l)



**Figure 3.** Effect of initial pH on COD removal efficiency (FeCl<sub>3</sub>=1.6g/l; original COD 8474.72 mg/l)

### III.2.2. Fenton's oxidation

The effects of H<sub>2</sub>O<sub>2</sub> to Fe (II) molar ratio, the dosage of hydrogen peroxide (H<sub>2</sub>O<sub>2</sub>), initial pH and reaction time on the effectiveness of the Fenton oxidation process were studied.

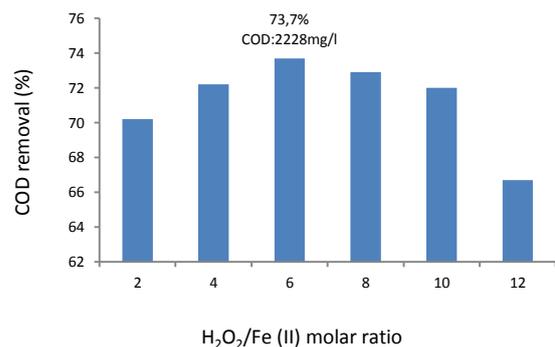
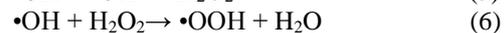
#### III.2.2.1. Effect of H<sub>2</sub>O<sub>2</sub> to Fe (II) molar ratio

Referring to the Fenton process, it is well known that higher hydrogen peroxide to substrate ratios result in more extensive substrate degradation, while higher concentrations of iron ions yield faster rates. However, in order to maximise the effectiveness of the process, it is preliminarily necessary to determine the optimal operational H<sub>2</sub>O<sub>2</sub>/Fe<sup>2+</sup> molar ratio. In other words, an optimal ratio between H<sub>2</sub>O<sub>2</sub> and Fe<sup>2+</sup> must be fixed in order to minimize scavenging effects. To investigate the optimum H<sub>2</sub>O<sub>2</sub>/Fe(II) molar ratio, six molar ratios of [H<sub>2</sub>O<sub>2</sub>]/[Fe<sup>2+</sup>] (2, 4, 6, 8, 10 and 12) were tested

with a fixed dosage of H<sub>2</sub>O<sub>2</sub> (5g/l), the initial pH was adjusted to 3. As shown in Figure 4, COD removal efficiencies increased with the increase of H<sub>2</sub>O<sub>2</sub>/Fe (II) molar ratio. Further increase in H<sub>2</sub>O<sub>2</sub>/Fe (II) molar ratio over 6 produced less efficient improvement in removals. This may be explained by the fact that if one of the reagents is not in the optimum ratio, the elimination efficiency of the organic material decreases due to the OH<sup>-</sup> scavenging either by H<sub>2</sub>O<sub>2</sub> or Fe<sup>2+</sup>.

#### III.2.2.2 Effect of H<sub>2</sub>O<sub>2</sub> dosage

Hydrogen peroxide plays a very important role as a source of •OH generation in Fenton's reaction .Figure 5 presents the effect of H<sub>2</sub>O<sub>2</sub> dosage on COD removal with the optimum molar ratio H<sub>2</sub>O<sub>2</sub> to Fe (II) of 6 at pH 3. Increasing H<sub>2</sub>O<sub>2</sub> dosage from 2g/l to 5 g/l improved the COD removal efficiency and the optimum value of 72.8 % was obtained for 5g/l dosage. Further increase of hydrogen peroxide dosage over 5g/l produced reduction of COD removal. Generally, under the critical concentration, the oxidation removal efficiency increases with increasing hydrogen peroxide dosage due to the increment of •OH radical, which produces through the decomposition of increasing hydrogen peroxide (Eq. (4)). Beyond the critical dosage, the degradation rate of organic compounds decreases with the increase of hydrogen peroxide due to scavenging effect of •OH by H<sub>2</sub>O<sub>2</sub>, as well as the incremental generation of •OOH also consumes •OH (Eqs. (5) and (6)) [26].



**Figure 4.** Effect of H<sub>2</sub>O<sub>2</sub> / Fe (II) molar ratio on COD removal efficiency (conditions: initial pH= 3; H<sub>2</sub>O<sub>2</sub>= 5g/l; original COD=8474.7 mg/l)

#### III.2.2.3 Effect of the initial pH

An essential characteristic of the Fenton process is that the acidic range strongly favours oxidation. For landfill treatment, the optimal pH values reported for conventional Fenton process range

between 2.0 and 4.5 [27]. Figure 6 demonstrates the effect of pH on the COD removal of the leachate treated by Fenton process at  $H_2O_2$  concentration of 5g/l and a molar ratio  $H_2O_2/Fe(II)$  of 6. Low pH between 2 and 3.5 has been found effective for Fenton's reagent, and best removal efficiency (74.6 %) was obtained at pH 3.5. The results may be due to that at extremely low pH (<2.0), oxidation removal decreased sharply due principally to the formation of complex species  $[Fe(H_2O)_6]^{2+}$ , which reacted slower with peroxide when compared to that of  $[Fe(OH)(H_2O)_5]^{2+}$  [28]. In addition, the peroxide gets solvated in the presence of high concentration of  $H^+$  ion to form stable peroxy ion  $[H_3O_2]^+$ . The peroxy ion leads to an electrolytic behavior on the part of hydrogen peroxide improving its stability and substantially reducing the reactivity with ferrous ion [29]. Moreover, exceptionally low pH can inhibit reaction between  $Fe^{3+}$  and  $H_2O_2$  [30]. On the other hand, oxidation removal rapidly decreased with increasing pH above 5.0. The reasons for this inhibition might be explained not only by the decomposition of hydrogen peroxide, but also by the desactivation of a ferrous catalyst with the formation of ferric hydroxide complexes leading to a reduction of  $\bullet OH$  radical [26].

#### III.2.2.4. Effect of the reaction time

The effect of Fenton reaction time on COD removal in leachate sample was investigated. Figure 7 shows that within the first 120 minutes, the removal of COD, improved with an increasing treatment time. After the first 120 minutes, a further increase in the reaction time led to a slow decrease in the removal of COD. Based on these results, the reaction time for the Fenton's treatment was determined to be 120 min. The results indicated that in many cases, it is unnecessary to achieve complete mineralization of the organic compounds into carbon dioxide and water because partial oxidations of intermediate compounds can minimize the consumption of chemicals reagents, and often result in substantial reductions of COD and toxicity. Additionally, higher biological activity of degraded low molecular weight components were in favor of the further biological treatment [31-33].

#### III.2.3. Combined treatment by coagulation/flocculation followed by Fenton's process

Considering the insufficient COD removal obtained with the coagulation/ flocculation step, a further treatment process must be applied. Therefore, it was decided to test the combination between coagulation/flocculation (using ferric chloride under the best operating conditions

previously selected) and Fenton reagent. Based on the optimization of coagulation and Fenton process, the initial leachate was firstly subjected to coagulation process, and subsequently the supernatant water was withdrawn and treated by Fenton. The final COD removal after this combined treatment attained 86.1% in comparison to the 74.6 % obtained by coagulation/flocculation alone. This can be explained by the fact that the use of Fenton process as a post treatment of coagulation treated leachate has been found to be an effective method for the removal of recalcitrant organics not eliminated during coagulation process.

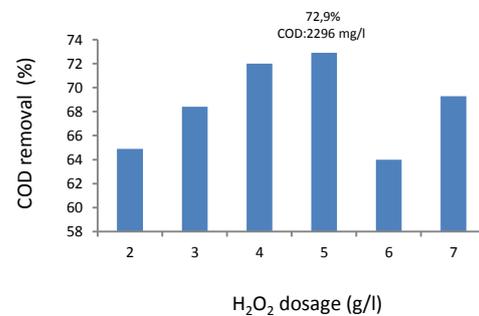


Figure 5. Effect of  $H_2O_2$  dosing on COD removal efficiency (conditions: Initial pH 3;  $H_2O_2/Fe(II)$  molar ratio=6; original COD 8474.7 mg/l)

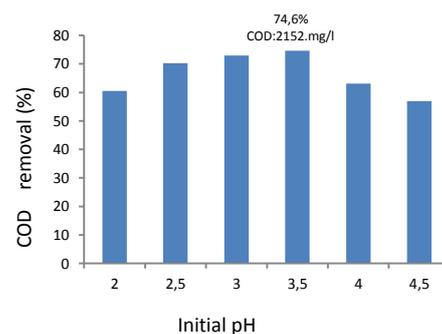
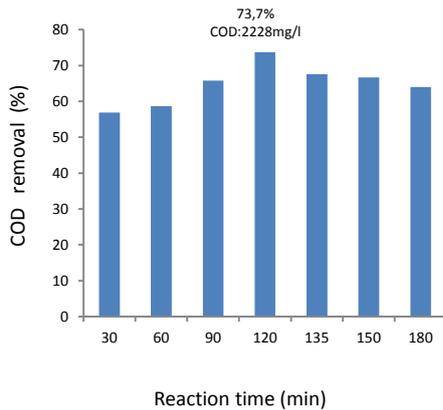


Figure 6. Effect of initial pH on COD removal efficiency (Conditions:  $H_2O_2=5g/l$ ,  $H_2O_2/Fe(II)$  molar ratio=6; original COD =8474.7mg/l)

#### III.2.4. Combined treatment by Fenton process followed by coagulation/ flocculation

To ascertain if the coagulation/ flocculation followed by Fenton's process is the best combination of treatment when compared to the reverse route, it was decided to conduct a new sequence by implementing the coagulation /flocculation step after Fenton oxidation. The

combined treatment took place by the pre-oxidation of leachate followed by the application of coagulation by use of ferric chloride according to the optimum conditions previously found. The COD removal efficiencies (up to 81.7%) were lower than that obtained in coagulation followed by Fenton's process. This may be explained by the fact that most of the particles that contribute to the COD are removed during the Fenton process while the residual dissolved COD is difficult to be removed through destabilization and cross-linking mechanism of coagulation process.



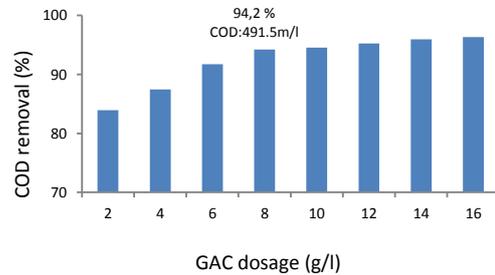
**Figure 7.** Effect of reaction time on COD removal efficiency (conditions: initial pH= 3.5; H<sub>2</sub>O<sub>2</sub>= 5g/l; H<sub>2</sub>O<sub>2</sub>/Fe (II) molar ratio=6, original COD=8474.7 mg/l)

### III.2.5. Combined treatment using coagulation, Fenton oxidation and granular activated carbon (GAC) adsorption.

Experiment with adsorption activated carbon was carried out after coagulation and Fenton process. Various amounts of granular activated carbon were added (2, 4, 6, 8, 10, 12, 14 and 16g/l). As it can be seen in figure8, the results indicated that the removal efficiency of COD increased with increasing GAC dosage, achieving 94.2 % COD removal at concentration of 8 g/l of ferric chloride but it scarcely increase with increasing GAC dosage over 8g/l. Based on the high cost of GAC, the optimal dosage for the activated carbon adsorption post treatment was determined to be 8g/l. this COD removal efficiency can be explained by the production of smaller and more easily adsorbable compounds during Fenton oxidation.

As indicated in figure 9, the photos inserted in the figure confirm the high removals of colour by chemical coagulation and Fenton oxidation.

However a total removal of color, odor and turbidity was achieved by the treatment train. The coagulation/sedimentation stage led to the humic acids precipitation, promoting a marked change in leachate color, from dark-brown to yellowish-brown (related to fulvic acids). The overall performances of leachate treatment before and after the various stages of treatment are listed in Table2. It is seen in this table that The COD, color, Turbidity and NH<sub>4</sub><sup>+</sup> removal is consistently very good. The COD concentration of leachate effluent after combined treatment (491.7) exceeds the standard for direct discharge to a natural water body but it is improved enough to be released into public sewage where the COD legal limit is 1000mg / l<sup>-1</sup>.



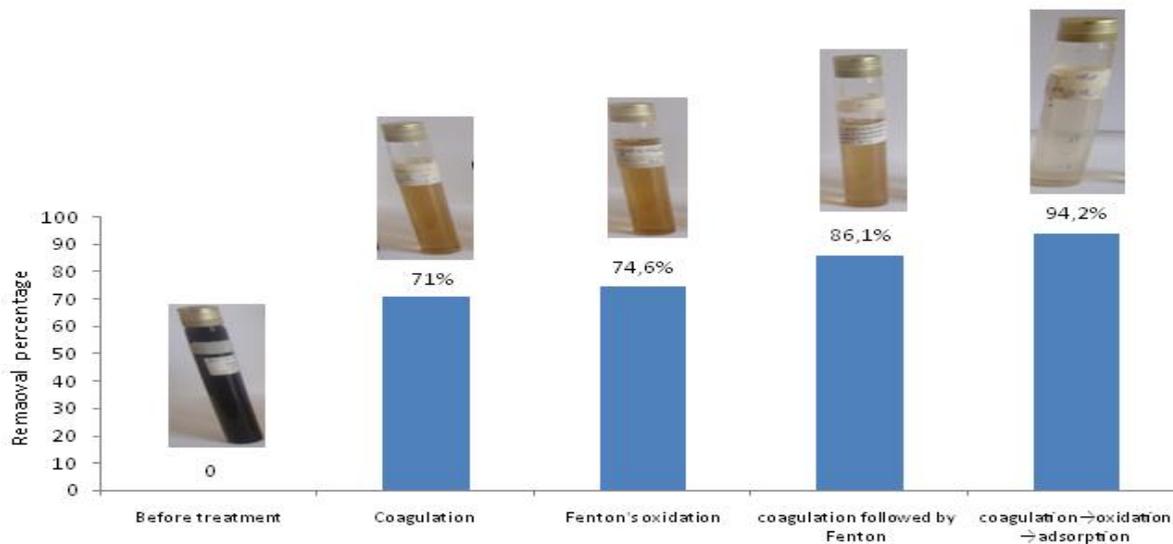
**Figure 8.** Activated carbon adsorption as post treatment after coagulation and Fenton oxidation

### III.3 Economic analysis

An estimated economic analysis of the operating costs associated with each treatment separately and the cost of the proposed treatment train was realized. The operating costs were estimated through the costs of reagents, It is important to note that this analysis is just an approximate tool to differentiate the trends in the operating costs associated to the use of different treatment processes. A rigorous economic analysis should consider the costs of reagents required to adjust the pH, energy, the initial investment, prices at plant scale, maintenance and labour costs, etc. The calculated costs are listed in Table3. Note that this economic survey is based on prices of industrial grade reagents. Table 3 shows that the total reagent cost of combined treatment process was 1165.8 DZD/m<sup>3</sup>. This high cost may be considered acceptable because of the complete treatment of landfill leachate.

**Table 2.** Characteristics of leachate before and after the various stages of treatment

Parameters	Before treatment	Coagulation	Fenton	Coagulation followed by Fenton	Coagulation→oxidation →adsorption
Color (Hazen)	221	85	250	/	15
Turbidity (NTU)	750	7.3	158	93	29.8
COD mg/l	8474.7	2457.7	2152.6	1178	491.7
NH <sub>4</sub> <sup>+</sup> mg/l	4193.6	2115.7	2455.7	915.1	190.7



**Figure 9.** Chemical oxygen demand (COD) removal efficiencies against treatment stages

**Table 3.** Chemical reagents cost in each treatment unit and combined treatment process

Items	Coagulation	Fenton oxidation		Combined treatment process coagulation→oxydation →adsorption			
	FeCl <sub>3</sub>	H <sub>2</sub> O <sub>2</sub>	FeSO <sub>4</sub> , 7H <sub>2</sub> O	FeCl <sub>3</sub>	H <sub>2</sub> O <sub>2</sub>	FeSO <sub>4</sub> , 7H <sub>2</sub> O	GAC
Price (DZD/kg)	58.1	100.9	21.9	58.1	100.9	21.9	109.7
Dosage (kg/m <sup>3</sup> )	1.6	5	6.8	1.6	1.5	2	8
Operation cost (DZD/m <sup>3</sup> )	93	653.4		1165.8			

#### IV. Conclusions

Our work was focused on investigation of different treatment procedures for removal of pollution from a partially stabilized leachate collected from Ouled Fayet landfill (Algiers). It was poorly biodegradable, contained significant amounts of ammonium nitrogen and chloride. Some metals identified in the leachate, Iron, copper and lead exhibited dangerously high concentrations. The present study clearly shows the added value of combined treatment trains for the purification of partially stabilized leachate. The following conclusions can be drawn:

- After coagulation with  $\text{FeCl}_3$  dosage of 1.6 g/l and an initial pH of 5, COD removal efficiency reached as high as 71%.
- In Fenton process, 73.7 % of COD removal was obtained at the following conditions: initial pH= 3.5;  $\text{H}_2\text{O}_2= 5\text{g/l}$ ;  $\text{H}_2\text{O}_2/\text{Fe}$  (II) molar ratio=6, reaction time=120 min)
- Coagulation/Flocculation followed by Fenton process has proven to be effective in reducing organic matter, it resulted in a final COD removal greater than 86%.
- The adsorption of pollutants onto activated carbon after coagulation and Fenton process, leads to very good reduction in COD (94.2 %). Under these conditions the leachate treated with remaining COD 491.7 g/l can not be discharged in natural water bodies but it is improved enough to be released into public sewage where the COD legal limit is 1000mg/l-1.
- Economic analysis shows that the overall operating cost of the combined treatment was 1165.8 DZD/m<sup>3</sup>. This multistage process is useful because of the complete treatment of landfill leachate

#### V. Références bibliographique

1. Lema, L.M.; Mendez, R.; Blazquez, R. Characteristics of landfill leachates and alternatives for their treatment: a review, *Water Air Soil Pollut.* 40 (1988) 223–250.
2. Christensen, T.H.; Kjeldsen, P.; Albrechtsen, H.-J.; Heron, G.; Nielsen, P.H.; Bjerg, P. I.; Holm, P.E. Attenuation of landfill leachate pollutants in aquifers. *Crit. Rev. Environ. Sci. Technol.* 24 (1994) 119–202.
3. Kjeldsen, P.; Barlaz, M.A.; Rooker, A.P.; Braun, A.; Ledin, A.; Christensen, T.H. Present and long-term composition of MSW landfill leachate: a review, *Crit. Rev. Environ. Sci. Technol.* 32 (2002) 297–336.
4. Scott, J.; Beydoun, D.; Amal, R.; Low, G.; Cattle, J.; Landfill Management, Leachate Generation, and Leach Testing of Solid Wastes in Australia and Overseas, *Crit. Rev. Environ. Sci. Technol.* 35 (2005) 239–332.
5. Deng, Y.; Englehardt, J.D. Treatment of landfill leachate by the Fenton process. *Water Res.* 40 (2006) 3683–3694.
6. Bigda; Bigda, R.J. Consider Fenton's chemistry for wastewater treatment. *Chem. Eng. Prog.* 91 (1995) 62–66.
7. Cheng, R.C.; Liang, S.; Wang, H.C.; Beuhler, M.D.; Enhanced coagulation for arsenic removal. *J. AWWA* 86(1994) 79–90.
8. Atmaca, E. Treatment of landfill leachate by using electro-Fenton method. *J. Hazard. Mater.* 163 (2009) 109–114.
9. Altin, A. An alternative type of photoelectro-Fenton process for the treatment of landfill leachate. *Sep. Purif. Technol.* 61 (2008) 391–397.
10. Kochany; Lipczynska-Kochany, E. Utilization of landfill leachate parameters for pretreatment by Fenton reaction and struvite precipitation—a comparative study. *J. Hazard. Mater.* 166 (2009) 248–254.
11. Primo, O.; Rivero, M.J.; Ortiz, I. Photo-Fenton process as an efficient alternative to the treatment of landfill leachates. *J. Hazard. Mater.* 153 (2008) 834–842.
12. Burbano, A.A.; Dionysiou, D.D.; Suidan, M.T. Effect of oxidant-to-substrate ratios on the degradation of MTBE with Fenton reagent. *Water Res.* 42 (2008) 3225–3239.
13. Lopez, A.; Pagano, M.; Volpe, A.; Dipinto, A.; Fenton's pretreatment of mature landfill leachate *Chemosphere* 54 (2004) 000–1005.
14. Zhang, H.; Choi, H.J.; Canazo, P.; Huang, C.P. Multivariate approach to the Fenton process for the treatment of landfill leachate. *J. Hazard. Mater.* 161 (2009) 1306–1312.
15. Urtiaga, A.; Rueda, A.; Anglada, Á.; Ortiz, I. Integrated treatment of landfill leachates including electrooxidation at pilot plant scale. *J. Hazard. Mater.* 166 (2009) 1530–1534.
16. Otal, E.; Arnaiz, C.; Gutierrez, J.C.; Lebrato, J. Anaerobic degradation of p-coumaric acid and pre-ozonated synthetic water containing this compound. *Biochem. Eng. J.* 20 (2004) 29–34.
17. Lu, X.J.; Yang, B.; Chen, J.H.; Sun, R. Treatment of wastewater containing azo dye reactive brilliant red X-3B using sequential ozonation and upflow biological aerated filter process. *J. Hazard. Mater.* 161 (2009) 241–245.
18. Daifullah, A.; Girgis, B.; Gad, H. A study of the factors affecting the removal of humic acid by activated carbon prepared from biomass material. *Colloids Surf. A* 235 (2004) 1–10.
19. Li, W.; Zhang, L.B.; Peng J.H.; Li N.; Zhu X.Y. Preparation of high surface area activated carbons from tobacco stems with  $\text{K}_2\text{CO}_3$  activation using microwave radiation. *Indus. Crops Prod.* 27 (2008) 341–347.
20. Chingombe, P.; Saha, B.; Wakeman, R.J. Surface modification and characterization of a coal-based activated carbon. *Carbon* 43 (2005) 3132–3143.
21. Standard Methods for the Examination of Water and Wastewater, twenty first ed., 2005. American Public Health Association, Washington.
22. Cotman, M.; Zgajnar-Gotvajn, A. Comparison of different physico-chemical methods for the removal of toxicants from landfill leachate. *J. Hazard. Mater.* 178 (2010) 298–305.

23. Wang, F.; Smith, D.W.; El-Din, G. Application of advanced oxidation methods for landfill leachate treatment – a review, *J. Environ. Eng. Sci.* 2 (2003) 413–427.
24. Ching, H.W.; Tanaka, T.S.; Elimelech, M. Dynamics of coagulation of kaolin particles with ferric chloride. *Water Res.* 28: 3 (1994) 559–569.
25. Duan, J.M.; Gregory, J. Coagulation by hydrolyzing metal. *Salts Adv. Colloid Interface Sci.* 100-102 (2003) 475–502.
26. Yanyu Wua,C.; Shaoqi Zhoua, B.C.; Fanghui Qina,C.; Huaping Penga,C; Yanglan Laia,C; Yiming Lina,C. Removal of humic substances from landfill leachate by Fenton oxidation and coagulation. *Process Safety and Environmental Protection* 88 (2010) 276–284.
27. Deng, Y.; Englehardt, J.D. Treatment of Landfill Leachate by the Fenton Process. *J. Hazard.Mater.* 125 (2005)166-174.
28. Gallard, H.; de Laat, J.; Legube, B. Effect of pH on the noxidation rate of organic compounds by Fe-II/H2O2. Mechanisms and simulation. *New J Chem*, 22(1998):263–268.
29. Tang, W.Z.; Huang, C.P. Stoichiometry of Fenton's reagent in the oxidation of chlorinated aliphatic organic pollutants. *Environ. Technol.* 18(1997) 13–23.
30. Pignatello, J.J.; Oliveros, E.; MacKay, A. Advanced oxidation processes for organic contaminant destruction based on the Fenton reaction and related chemistry. *Crit Rev Environ. Sci. Technol.* 36 (2006) 1–84.
31. Zhang, H., Choi, H.J.; Huang, C.P. Optimization of Fenton process for the treatment of landfill leachate. *J Hazard Mater*, 125(2005) 166–174.
32. Gotvajn, A.Z.; Tisler, T; Zagorc-Koncan, J., Comparison of different treatment strategies for industrial landfill leachate. *J Hazard Mater*, 162 (2009) 1446–1456.
33. Sun, J.H.; Li, X.Y.; Feng, J.L; Tian, X.K., Oxone/Co<sup>2+</sup> oxidation as an advanced oxidation process: comparison with traditional Fenton oxidation for treatment of landfill leachate. *Water Res.* 43(2009) 363–4369.

#### Abbreviations:

COD, chemical oxygen demand.  
BOD, biological oxygen demand.  
AOPs, advanced oxidation processes.  
TKN, total Kjeldahl nitrogen.  
GAC, granular activated carbon  
NTU, nephelometric turbidity units

#### Acknowledgement

This work was supported by the Algerian Ministry of Higher Education and Scientific Research.

#### Please cite this Article as:

Boumechhour F., Kerbach R., A.E. Rechoum A.E., Benmenni M.S ., *Treatment of partially stabilized landfill leachate using combinations of coagulation, Fenton oxidation and granular activated carbon (GAC) adsorption, Algerian J. Env. Sc. Technology, 2:3 (2016) 249-257*