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Treatment of Leachate from the Technical Landfill Centre Ain Defla (Algeria) by Oxidation and Biosorption Process

B. Feraoun,^a A.-E. Belhadji,^a K. Otmanine,^{b,*} and M. Hammoudi^c^a Biomaterials and Transport Phenomena Laboratory (LBMPT), Yahia Fares University, Department of Chemical Engineering and Environment, Médéa, 26 000, Algeria^b Bioressources Naturelles Locales LBRN, Hassiba Benbouali University of Chlef, Faculty of Technology, Department of Process Engineering, Chlef, Algeria^c Biomaterials and Transport Phenomena Laboratory (LBMPT), Yahia Fares University, Department of Chemical Engineering and Environment, Experimental Biology and Pharmacology Team, Médéa, 26 000, Algeria

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Abstract

The aim of this study was the treatment of leachate from the Technical Landfill Centre Ain Defla in Algeria, which presents a serious threat to the environment. Elimination of organic matter (expressed as chemical oxygen demand (COD) and biological oxygen demand (BOD₅)), suspended matter (SM), mineral matter (phosphates and ammoniacal nitrogen), and heavy metals (zinc and iron) were experimentally studied using the coupling of oxidation (OP) and biosorption processes (BS). The analysis results showed that the leachate studied at pH 8.8 was very loaded in organic matter (turbidity of 553 NTU, SM = 820 mg l⁻¹, COD = 9669 mg O₂ l⁻¹, and BOD₅ = 8875 mg O₂ l⁻¹), in salts (EC = 19.4 mS cm⁻¹), in ammoniacal nitrogen (2027 mg l⁻¹), in phosphates (22.9 mg l⁻¹), and in sulphates (750 mg l⁻¹). It also contained significant amounts of heavy metals, notably zinc (4.21 mg l⁻¹) and iron (47.5 mg l⁻¹). The evolution of the physicochemical parameters during the treatment showed that, under the optimal conditions (T = 45 °C, [H₂O₂] = 1.6 mol l⁻¹, volume fraction φ(H₂O₂) = 5 %, and [Fe³⁺] = 0.5 mmol l⁻¹), the reduction in COD was about 99 %, the reduction in BOD₅ was 100 %, the elimination of colloidal particles (SM) could reach 95 %, reduction in phosphates was 78 %, reduction in ammonium was 98 %, reduction in sulphates was 96 %, reduction in zinc was 92 %, and the reduction in iron was 98 %.

Keywords

Leachate, landfill, oxidation process, biosorption

1 Introduction

Technical landfills present a prime solution for waste storage. However, this option creates different problems, among which is the production of leachate, an effluent with a high polluting potential (high organic matter content and salinity), which poses a serious threat to the environment and human health.^{1,2}

Their structure differs from one landfill to another based on the nature of the waste, age of the traps, age of the leachate, climatic circumstances, and site topography.^{3,4}

The literature recognises three kinds of leachate: *youthful leachates* defined by the elevated and relatively biodegradable organic burden; *intermediate leachates* with reduced organic burden, and *stable leachates* consisting mainly of humic substances immune to biodegradation. Because of their variable structure over time, they must undergo varying procedures before being discharged into the recipient setting.⁵

The Technical Landfill Centre (TLC) Ain Defla was commissioned in 2013. Approximately 41 Mt per year of household waste was entering the TLC Ain Defla and the major-

ity consisted of a high organic load. In the first four months of 2019, the volume of leachate produced was 20 m³ per day and the volume stored in three lagoons at the end of 2019 was 80,000 m³.⁶

In order to decrease the leachate composition, a scheme of recirculation and suction was set up, but efficiency remained very low. The physicochemical analyses performed on these leachates enabled the determination of their size and chemical composition, particularly their degree of contamination.^{3,4}

Leachate is composed of 83 % to 92 % of water, 4 % to 16 % of organic compounds, and 1 % to 20 % of inorganic compounds. The colour of the scrap varies from white to dark brown. It includes elevated levels of hanging fluid. In addition to visual annoyance and unpleasant odours, the elevated organic burden of leachate kills fauna and aquatic vegetation by capturing dissolved oxygen that has damaging impact on water, soil, micro-organisms, and crops.^{3,4}

More than 20 processes or technologies applicable for treatment of leachate have been identified in scientific works. In most instances, these are basic or combined operations that have been conducted on a laboratory or pilot-plant scale with no real industrial projection. The present methods were defined as the most widely used and

* Corresponding author: Khaled Otmanine
Email: otmaninegpp@gmail.com



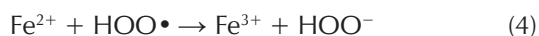
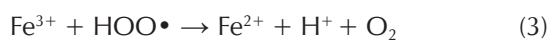
Fig. 1 – Location of leachate sampling point

they can possibly be applied. These methods of treatment can be categorised by the technique used:³

- *thermal* – by evaporation, incineration,
- *physicochemical* – by filtration/ultrafiltration, ozonation, and coagulation,
- *biological* – by anaerobic and aerobic treatment.⁷

However, when it comes to treating the (mature) leachate with a high proportion of bio-refractory compounds, these technologies face some limitations.⁸ To overcome this problem, in order to increase the efficiency of purification, biofiltration (BF) can be coupled with other non-biological systems.⁹ In recent studies, many combinations have been tested, including BF and chemical coagulation,^{10,11} BF membrane technologies,^{12,13} bio-process (BP) and advanced oxidising processes,^{14,15} BF and electrochemical processes, etc.¹⁶ Among these combinations, the homogeneous advanced oxidation process (HAOP) could be an interesting approach to support a biological system in a treatment process.^{17,18}

The concept of HAOP comprises the generation of free radicals, very unstable species of oxidising substances that can degrade chemical compounds found in effluents.^{19,20} In chemicals that are hard to degrade, the Fenton-reaction has shown excellent outcomes. In this response, the following sequence of Fenton reactions was suggested, where hydrogen peroxide (H_2O_2) is used effectively in place of ferrous salts for leachate:



The initial response is the overall Fenton reaction. In order to obtain the important quantities of radical $HO\bullet$, the cycle must be optimised. Ferric ion (Fe^{3+}) and a range of oxidising species ($HO\bullet$, $HOO\bullet$, HOO^- , and H_2O_2) will be available. Furthermore, the ferric ions (Fe^{3+}) will interact with the hydroperoxide radicals ($HOO\bullet$) and will be reduced to Fe^{2+} . However, Fe^{2+} will react with extra $HOO\bullet$ to generate Fe^{3+} .^{21,22}

As seen during the Fenton reaction, the hydrated ferrous oxide will eventually be formed. For the Fenton reaction to be effective, the pH must be adjusted between 2 and 4. The primary benefits of HAOP are small sludge output contrary to the chemical products.^{19,20}

A BF method can be used to achieve the extra suppression of contaminants following the advanced oxide treatment. The frequently used absorber is active carbon (AC), whose effectiveness is high (20 %). However, the impact from palm oil and empty fruits on extraction of phenol has been explored under various production circumstances for active carbon. These parameters include temperature, ignition moment, and speed of CO_2 stream.^{19,20}

Activated charcoal is made from various fresh products, such as coconut marrow activated with $ZnCl_2$, pecan and caps tile nuts shells, jackfruit bark, oak bowl pulp, H_3PO_4 acid treatment, and palm seeds shell. Coffee grounds and sawdust have been used as adsorbents because of their structure and characteristics.^{19–30}

The main objective of this study was to optimise the conditions for the elimination of organic and nitrogenous matter by coupling the oxidation process (OP) with biosorption (BS). The experiments were carried out on the leachate from the TLC Aïn Defla. After characterisation of the leachate, treatment trials made it possible to determine the optimal treatment doses. Finally, a process coupling performance analysis was conducted.

2 Materials and methods

2.1 Sampling and characterisation of leachate

Samples of coarse leachate were acquired from TLC Aïn Defla, Algeria. The leachate was collected and analysed between 1/9/2018 and 1/3/2019, and stored at 4 °C until its use within 48 h or as a note that the climate in this area is semi-arid.

The multi-parameter water analyser model HI19629 (HANNA Instruments, USA) was used to measure the electrical conductivity (EC), dissolved solids (DS), pH, and temperature *in situ*. A PPOxiTop® A6 thermostat was

used to measure BOD. The COD was evaluated using a HI839800 reactor model (HANNA Instruments, USA) with sulphochromic oxidation. Atomic absorption spectrometer (AAS) 700 was used to determine the heavy metals Al, Ca, Cd, Cu, Pb, Fe, K, and Mn. Turbidity was measured by the turbidity meter model HI88703 (HANNA Instruments, USA) for nephelometric turbidity units (NTU).

These leachate samples were forwarded in iceboxes to the Environment and Cleanliness Laboratory, Khemis Miliana Ain Defla, Sustainable Development Laboratory of the Region, University of Djilali Bounaama Khemis Miliana, Microbiology Laboratory and the Materials Laboratory of Yahia Farès, University of Médéa, with respect to the circumstances of conservation needed by the norms in order to slow down the biological, chemical, and physical processes.

2.2 Oxidation of the leachate

2.2.1 Determination of the optimal oxidation temperature

To ensure the complete homogenisation of the reactionary milieu, leachate was poured into volumetric glaze and shaken by a magnetic agitator for 30 min. The reactor thermostat was started and the set-up temperature was adjusted to the desired value. The reactor temperature was set to the following temperatures: 40, 45, 50, 55, and 60 °C. The pH of leachate was raised to 2.6 with a few drops of 10 % H_2SO_4 (for the homogeneous Fenton response the optimum pH is around this level). After stabilisation of reactor temperature, the first sample was drawn at time zero with the addition of hydrogen peroxide to start a method for oxidation. To monitor turbidity reduction, regular samples were obtained. About 3 ml of fluid samples were taken during specified periods (after 15, 30, 45, 60, 75, 90, 115, and 120 min) during 2 h of oxidation.

2.2.2 Determination of the optimal concentration of H_2O_2

In this part, the pH of liquid was decreased to 2.6 and the temperature was set at 45 °C because this temperature gave a reduction of 50 %. In this process, the amount of oxidising agent of varying degrees was used (0.8, 1.6, 2.4, and 3.2 mol l⁻¹). A sample was taken every 15 min for 2 h.

2.2.3 Determination of the optimal volume fractions of H_2O_2

The following processes are quite comparable to the past two. The reactor contained the same concentration of H_2O_2 , which was equivalent to 1.6 mol l⁻¹. The investigated hydrogen peroxide volume fractions (φ) were 1, 2.5, 5, and 10 %.

2.2.4 Effect of soluble catalyst on treatment

The strong catalytic agent to study was Fe(III). Therefore, under the current working conditions, it was planned to

examine the impact of metal oxide – $FeCl_3 \cdot 6H_2O$ (99 %) which is the source of Fe(III).

The experiment was carried out under optimal conditions, at 45 °C and pH 2.6, with a volume fraction of 5 % of H_2O_2 at concentration of 1.6 mol l⁻¹.

2.2.5 Effect of the ratio $R = [H_2O_2]/[Fe^{3+}]$ on processing

In the attempt to optimise the operating circumstances of leachate oxidation, the proportions between the oxidising agent and the catalyst were investigated. These proportions were $R = 10$ to $R = 60$. The operating conditions were $T = 45$ °C, $[H_2O_2] = 1.6$ mol l⁻¹, $pH = 2.6$, and $[Fe^{3+}] = 0.5$ mmol l⁻¹.

2.3 Treatment of leachate with biomaterials

Sand or activated carbon filtration or filter media are water filtration systems used to solve many problems in the treatment of liquid effluents. Each filter medium has a specific function in the liquid effluents treatment:

- Sand filters for the sediments.
- Coffee grounds and sawdust transformed into activated carbon to absorb chlorine and organic matter.

2.3.1 Preparation of the constituents for biosorption column

A – Sand: The sand was screened after drying in an oven at 150 °C for 24 h. The diameter of the used particle ranged from 0.040 to 0.500 mm.

B – Sawdust: Sawdust or chips are scraps of material after wood is cut or formed. It was crushed and sieved after washing the scrub thoroughly with distilled water, and put in an oven at a temperature of 105 °C for 24 h. There were two kinds of particle magnitude chosen. The first one was between 0.2 and 1 cm in diameter. The second diameter was between 0.040 and 0.025 cm. The resulting particles were cured in free air at 300 °C for 4 h, and then treated for 1 h in sodium hydroxide (3 mol l⁻³) baths, thoroughly washed out with distilled water, and dried for 2 h at 105 °C.

C – Coffee grounds: After washing the coffee grounds of any impurities, they were baked at 300 °C for 4 h in open air. They were then washed in sodium hydroxide (3 mol l⁻³) baths, washed to a neutral pH with distilled water, and dried for 2 h at 105 °C.

D – Peat: Peat is a mix between the Ain Defla landfill pot soil and the water retrieved from the lagoon floor.

2.3.2 Biosorption column

The column was made of PVC, 150 cm high and 10 cm in diameter. The packing of the column was mainly composed of sawdust, sand, coffee grounds, and peat. These four components were arranged in five layers (Fig. 2). The

column was formed from top to bottom as soot: a vacuum of 20 % was maintained to ensure the atmospheric pressure on the leachate, and then a first course upper layer composed only of sawdust with a diameter between 0.2 and 1 cm, which had the role of facilitating the passage of liquid and limiting clogging. The second layer was made up of 5 % sand, 35 % sawdust of diameter 0.04 to 0.25 mm, 35 % coffee grounds, 20 % peat, and 5 % calcite by volume percentage. Calcite (CaCO_3) added to these layers made it possible both to stabilise the pH of the effluent thanks to the buffering capacity of carbonates, and to optimise nitrification with the availability of a mineral carbon source for the autotrophic bacteria responsible for the nitrification. To facilitate the percolation of the liquid in the filter medium, another 20-cm layer of 50 % sand and 50 % wood chips with a diameter of 0.2 to 0.1 cm was placed between the second and fourth layer. The fourth layer was 50 cm high and composed of 50 % sawdust with a diameter of 0.04 to 0.25 mm, 40 % peat, 5 % sand, and 5 % calcite. The fifth layer was placed on 5-cm high coffee grounds, and finished with a 5-cm layer of green beads to allow the filtrate to drain.



Fig. 2 – Biosorption column

3 Results and discussion

3.1 Characterisation of the leachate of Aïn Defla landfill

Every landfill has a specific chemical composition of leachate. This differs widely depending on the nature and age of the deposit, waste type and decomposition, the method of deposit, the nature of the landfill, the climate, etc.^{3,4,31}

Table 1 – Average values of the physicochemical parameters of the leachate from TLC Aïn Defla

Parameter	Unit	Value
pH	–	8.8
total dissolved solids (TDS)	mg l^{-1}	10
EC	mS cm^{-1}	19.4
turbidity	NTU	533
SM	mg l^{-1}	820
O_2 dissolved	mg l^{-1}	3.12
BOD_5	$\text{mg O}_2 \text{ l}^{-1}$	8875
COD	$\text{mg O}_2 \text{ l}^{-1}$	9669
BOD_5/COD	–	0.91
NH_4^+	mg l^{-1}	2027
NO_2^-	mg l^{-1}	2.73
NO_3^-	mg l^{-1}	5021
PO_4^{3-}	mg l^{-1}	22.9
SO_4^{2-}	mg l^{-1}	750
Ca^{2+}	mg l^{-1}	12.80
Mg^{2+}	mg l^{-1}	350
K^+	mg l^{-1}	2.85
Na^+	mg l^{-1}	2.5
As	mg l^{-1}	0.19
Cd	mg l^{-1}	1.25
Cr	mg l^{-1}	2.0
Cu	mg l^{-1}	1.45
Fe	mg l^{-1}	47.5
Mn	mg l^{-1}	1.10
Ni	mg l^{-1}	3.43
Pb	mg l^{-1}	0.9
Zn	mg l^{-1}	4.21

Table 1 presents the results of physicochemical analysis carried out during the characterisation of the raw leachate from the Aïn Defla landfill. For the different parameters analysed in leachate, the highest level needed refers to the existing Algerian legislation on the release of industrial liquid effluents, since discharge juices or leachates are often equivalent to the complicated agricultural discharges of organic and inorganic pollutants.¹³

Presuming that the release or juices of leachate can be compared to the complicated manufacturing releases comprising both organic and inorganic contaminants, the previously found sand results of physicochemical parameters of Aïn Defla landfill coarse leachate with a faecaloid odour release are presented in Table 1.

Aïn Defla landfill leachate is essential. The mean pH-value registered is 8.8. The characteristic of acidic conditions in the initial aerobic waste degradation phase is generally increased in order to solubilize chemical elements (hydrox-

ide acids and carbonate species) and to decrease the waste sorption capacities. However, the pH is essential in this situation, probably because of the recycling system between the lagoons and lockers.

The analysis of the results showed that the leachate was rich in natural material and that elevated BOD₅ and COD numbers (8875 mg O₂ l⁻¹, 9669 mg O₂ l⁻¹, respectively) were true. COD in the leachate exceeded appropriate values. On average, it was more than 80 times higher than the prescribed level of Algerian industrial discharges¹, which is about 120 mg O₂ l⁻¹, with regard to the BOD₅, detected in the various leachate samples after 5 days of incubation, thus exceeding the approved norm by approximately 35 mg O₂ l⁻¹. However, the real level of BOD₅ continued to be superior to the actual figures since the substrate was packed with toxic materials. Furthermore, the BOD₅/COD proportion is the indication of organic matter biodegradation and leachate maturation.^{32,33} Therefore, for young rejects from which the biological activity corresponds to the acid phase of anaerobic degradation, this ratio reaches the value of 0.83. It was down to 0.05 for ancient sites, where the final phase of methanogens fermentation was achieved. The decreases in COD and specifically in BOD₅ characterise this formation phase in our situation. The TLC Ain Defla leachate appeared to be in the acidic phase (BOD₅/COD = 0.91).^{32–34}

In fact, the average EC was 19.4 mS cm⁻¹, indicating strong mineralisation of TLC Ain Defla leachate. This mineralisation was mainly attributable to the following parameters: ammonium (2027 mg l⁻¹), phosphate (22.9 mg l⁻¹), sulphate (750 mg l⁻¹), potassium (2.85 mg l⁻¹), and sodium (2.5 mg l⁻¹). Dissolved solids (DS = 10 mg l⁻¹) refer to all the minerals, salts, metals, cations or anions dissolved in the water. This includes anything present in the water other than H₂O molecules, including suspended solids. Turbidity of leachate was significant. This was because the sun had not penetrated the algae that show biological imbalance, confirmed by the elevated level of suspended material (SM).

For nitrites (Table 1), it was noticed that, on several occasions, the quantity passed the obtained level. The existence of nitrites showed the identification of our samples as comparable to industrial discharges.

The most responsible ingredients of leachate smell are sulphates, and in this model, they were concentrated at 750 mg l⁻¹.³⁵ The sulphates were then reduced to sulphides in the form of H₂S, the gas responsible for bad smells emanating from the landfill.³⁵

The evolution of the levels of salt, potassium, calcium, and magnesium in leachate, like most metallic components (Table 1), is linked to the quantity of organic matter dissolved.³⁶ Indeed, the high concentrations of the magnesium element is due to the fact that, during the dry period, the organic matter responsible for the complexity of these elements is very low.

Table 1 shows the heavy metal composition of the leachate. The metal load in these percolates was significant. Iron was the metal in larger amounts with concentration levels of 47.5 mg l⁻¹, which is compatible with the bibliographic data. The metal products of the waste dump are primarily composed of iron.³³

Toxic metals were present in relatively low concentrations (nickel – 3.43 mg l⁻¹, cadmium – 1.25 mg l⁻¹, copper – 1.45 mg l⁻¹), and they were lower than leachate generated by other landfills.^{18,37} The household waste treatment scheme positioned before the container was responsible for these values, but most heavy metals were stuck in the waste.^{34,35} The following rates were classified: Fe > Zn > Ni > Cu and Mn > Cd > Cr > Pb > As in increasing orders. Therefore, in these leachates, there was an obstruction of microbial development and interference with the competitive BOD₅ outcomes by the existence of heavy metals.

3.2 Study of the oxidation process

The oxidation study revealed that the sophisticated oxidation method under consideration was dependent on the HAOP. What treatment of this leachate was needed to prevent any type of environmental pollution?

3.2.1 Temperature effects

The turbidity evolution of crude leachate alone (witness 1) with oxidising agent (witness 2), and with oxidising agent plus pH = 2.6 adjustment (sample) was monitored during 120 min corresponding to the standard reaction time (Fig. 3).

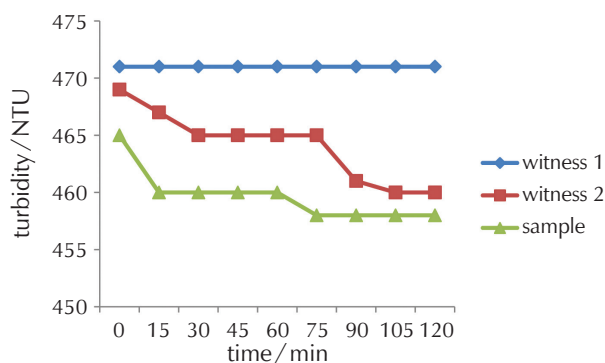


Fig. 3 – Stability of turbidity at 20 °C

The results showed that H₂O₂ hardly decreased leachate during the reaction time at 20 °C (less than 3 % transformation within 120 min). Fig. 4 shows that turbidity had decreased with time at various temperature conditions (T from 40 to 60 °C).

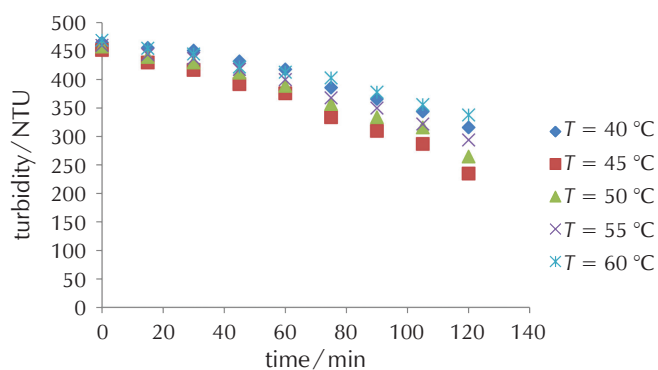


Fig. 4 – Influence of temperature on turbidity

At the start of responses, turbidity usually amounted to about 460 NTU and above, and the study was carried out within a 120-min period. Following the increase in temperature (40, 45, 50, 55, and 60 °C), it was noted that the impact of H_2O_2 in acidic environment ($pH = 2.6$) became more efficient than the findings in the neuter environment ($pH = 7$).

The leachate turbidity from the TLC Ain Defla had a value of 465 ± 5 NTU. After studying the influence of temperature on the turbidity, it was found that the value of 235 NTU was reached with a yield of 50 %. Once the temperature rises, it starts to demineralise because of the degradation of hydrogen peroxide at high temperature.

3.2.2 Determination of the optimal volume fraction of H_2O_2

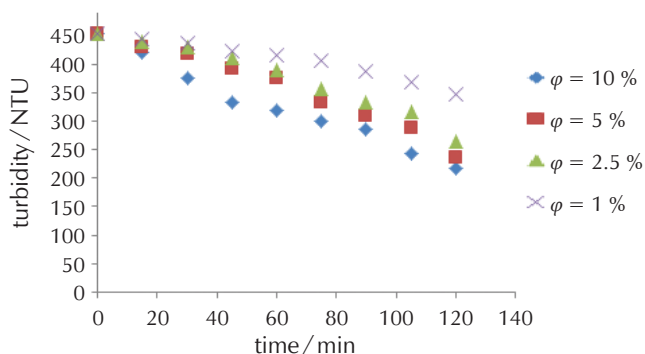


Fig. 5 – Influence of H_2O_2 volume fraction on turbidity

3.2.3 Determination of the optimal concentration of H_2O_2

According to the obvious first-order kinetics, when a leachate solution is subjected to HAOP, original turbidity reduces linearly with a moment. After 2 h of treatment, a decrease frequency for an H_2O_2 level was more than 61 %, equivalent to 1.6 mol l^{-1} (Fig. 5). Fig. 6 also demonstrates the turbidity image. Therefore, low concentration of H_2O_2 (0.8 mol l^{-1}) led to a reduction in reaction rate.

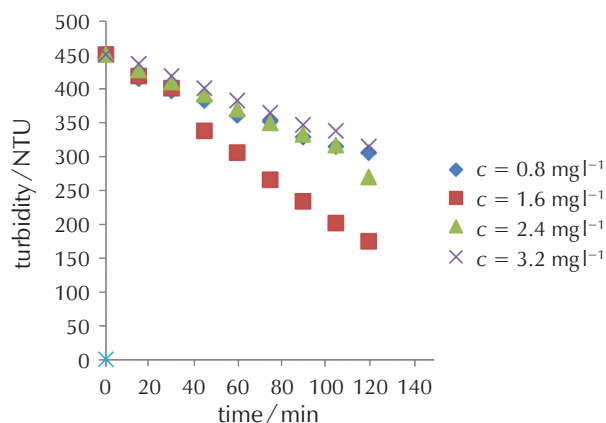


Fig. 6 – Influence of H_2O_2 concentration on turbidity

According to Eq. 2, at high concentrations, the hydroxyl radicals preferentially react with the hydrogen peroxides of the leachate.

3.2.4 Effect of soluble catalyst (Fe^{3+}) on the treatment

For removing leachate turbidity, Fig. 7 shows the initial Fe^{3+} level at varying concentrations.

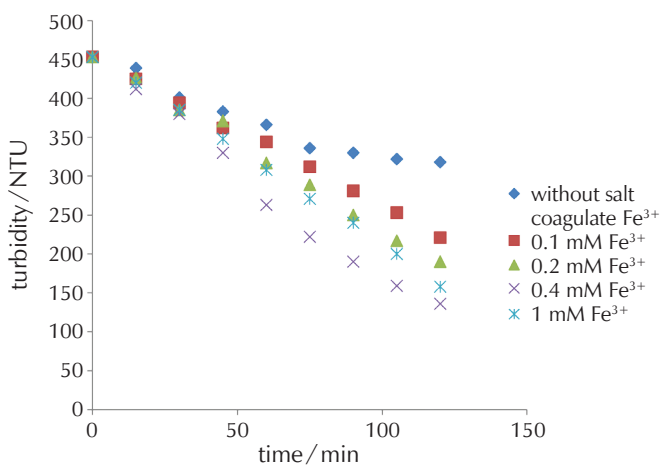


Fig. 7 – Influence of catalyst Fe^{3+} concentration on turbidity

Fig. 7 shows that adding the ferric ions improved the system's effectiveness, and resulted in the rise of the initial catalyst concentration from 0.1 to 0.5 mmol l^{-1} (Eqs. (5) and (6)):^{38,39}



Table 2 – Performance of leachate treatment with OP. Conditions: $T = 45\text{ }^{\circ}\text{C}$, $[\text{H}_2\text{O}_2] = 1.6\text{ mol l}^{-1}$, $[\text{Fe}^{3+}] = 0.5\text{ mmol l}^{-1}$, $R = 40$.

Characteristics of leachate	pH	BOD ₅	COD	EC	Turbidity	SM	NH ₄ ⁺	NO ₂ ⁻	NO ₃ ⁻	PO ₄ ³⁻	Ca ²⁺	Mg ²⁺	Fe
unit	–	mgO ₂ l ⁻¹	mgO ₂ l ⁻¹	μS cm ⁻¹	NTU	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹
before treatment	8.8	8875	9669	19.4	533	820	2027	2.73	5021	22.9	12.80	350	47.5
after treatment	6.39	0	1933.8	0.024	53.3	235	1479	1.52	1300	5.04	1.64	27.94	3.25
output/%		100	80	99.99	90	71	24	44	74	78	87.18	92	93

3.2.5 Influence of the ratio $R = [\text{H}_2\text{O}_2]/[\text{Fe}^{3+}]$

An experiment was carried out in order to determine the effect of the ratio $R = [\text{H}_2\text{O}_2]/[\text{Fe}^{3+}]$ on leachate mineralisation as a function of time. Fig. 8 shows the results obtained by the Fenton process.

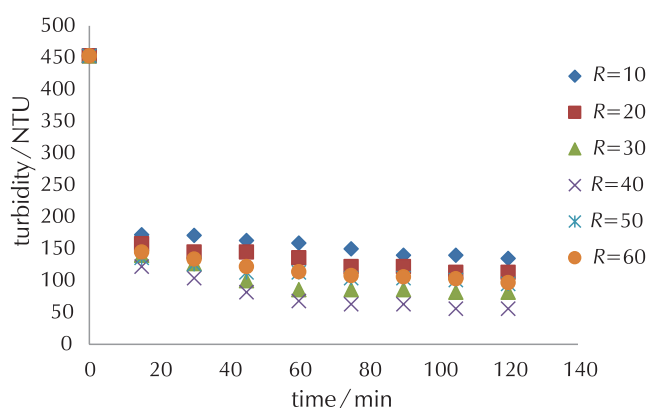


Fig. 8 – Assessment of turbidity during leachate mineralisation as a function of time for variable R values for $[\text{Fe}^{3+}] = 0.5\text{ mmol l}^{-1}$

Fig. 8 shows the evolution of turbidity during the leachate mineralisation as a function of time for R -ratio values ranging from 10 to 60 at a fixed concentration of $\text{Fe}^{3+} = 0.5\text{ mmol l}^{-1}$.

The achieved findings indicated that the kinetics of mineralisation improved when R increased. The level of leachate mineralisation in aspects of turbidity reduction increased quickly when R increased from 10 to 40 (from 65 % to 70 %) after 15 min of treatment. An 87 % decrease in turbidity was achieved with $R = 40$ after 120 min of treatment or almost complete elimination of the pollutant. This elevated mineralisation speed is related to the highly elevated radical hydroxyl formation (Eqs. (2) and (7)). For the Fenton reaction,⁴⁰ an unnecessary rise in the amount of hydrogen peroxide ($R = 60$) may be a limiting factor in the Fenton process by consuming hydroxyl radicals according to Eqs. (2) and (7).^{36–41}



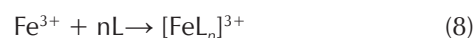
Thus, when treated leachate by the Fenton process, $R = 40$ was selected as the ideal value for the initial Fe^{3+} concentration (0.5 mmol l^{-1}).

3.2.6 OP performance on the leachate treatment of Ain Defla landfill

Table 2 summarises the efficient removal of pollutants under the best operating conditions by the oxidation process ($45\text{ }^{\circ}\text{C}$, $[\text{H}_2\text{O}_2] = 1.6\text{ mol l}^{-1}$, $[\text{Fe}^{3+}] = 0.5\text{ mmol l}^{-1}$, $R = 40$). It should be noted that the values in this table are the average values obtained through analyses of leachate treated for feeding the biosorption unit under the optimum conditions. It can be seen that ammonia and siltation were insignificant. It was anticipated that OP would have poor ammonia suppression effectiveness. Other studies have also reported low ammonia removal in such landfill leachate treatment processes.^{42–44} After treatment, turbidity, colour, and metals had been efficiently removed. Ninety percent of the turbidity was withdrawn, likely because of the humic acid removal. In fact, despite the low proportion of humic acids in the waste leachate of landfills, it had a high colouring capacity. Metals, zinc, and iron, among others, have been effectively removed (87 % Zn and 93 % Fe removed).^{42–44}

The good removal of metals by the OP could be attributed to the precipitation and co-precipitation. The total phosphorus concentration was reduced by 78 %.⁴⁵

Finally, in terms of BOD₅, the biodegradable organic matter was eliminated 100 % with a COD efficiency of 80 % for the non-biodegradable organic matter. This is explained by the existence of non-oxidising compounds by OP, since it is hard to degrade carboxylic acids that were produced as intermediate. Some carboxylic acids, especially polyacids, form stable complexes with iron, which, in the absence of UV/Vis irradiation, inhibit the reaction by immobilising the metal in an inactive form (Eq. (8)):⁴⁶



where L are the mono- and dicarboxylic acids.

3.2.7 Biosorption

The biosorption unit was supplied with oxidised leachate or mineralised leachate previously acquired after adaptation to activate the microorganisms.

3.2.7.1 Elimination of organic matter

The performance of biological treatment with regard to the removal of COD is shown in Fig. 9. Compared to the total removal of COD (99.87%), BOD₅ was fully removed after treatment with OP. Following biological treatment, this value was maintained. The overall suppression of COD can be clarified by the presence of organic refractory content in leachate that had been adsorbed by the biosorption components and even biodegraded.

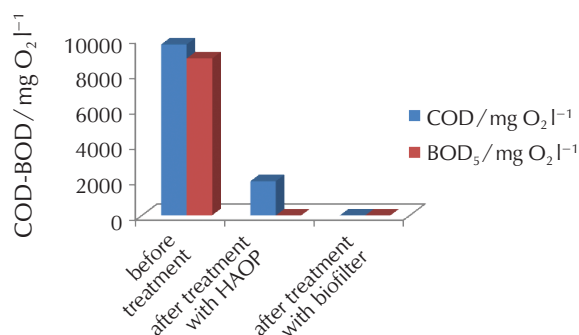


Fig. 9 – Assessment of organic matter before and after biosorption of mineralised leachate

Biosorption used to treat leachate from landfills, that were pre-treated under oxidation process, reported similar findings.^{47,48} After biosorption treatment of landfill leachate, a good rate of removal of the Hyl fraction and an insignificant rate of removal of humic substances (humic acid – HA and fulvic acid – FA) was observed.

3.2.7.2 Removal of ammonia

As stated in Table 5, at 27 % of the initial load during OP, the removal of ammonia was low. Therefore, the primary task of treatment was the effective treatment of ammonia pollution. Average inlet flux of 500 ml d⁻¹ was provided for the biosorption column. During the biological treatment, nitrogen variations were formed, as shown in Fig. 10. After biosorption, ammonia was almost entirely removed. The average outlet ammonia concentration was 0.68 mg l⁻¹, i.e., well below the Algerian standard norms.¹ Nitrate (NO₃⁻) and nitrite (NO₂⁻) levels were also monitored during the biosorption. The results were 99.57 % and 99.96 % elimination effectiveness. These results showed that nitrification was the principal mechanism for removing the ammonia from biosorption.

However, other biological and physicochemical events, such as NH₃-removal, assimilation, denitrification, and adsorption could also occur in a biosorption column.^{49,50}

Despite the small phosphorus level and significant quantity of remaining SS acquired after OP treatment, the nitrification method was very efficient. Taking into account the sensitivity of nitrifying bacteria, inhibition of toxicity was a problem. These critical parameters cause the nitrification

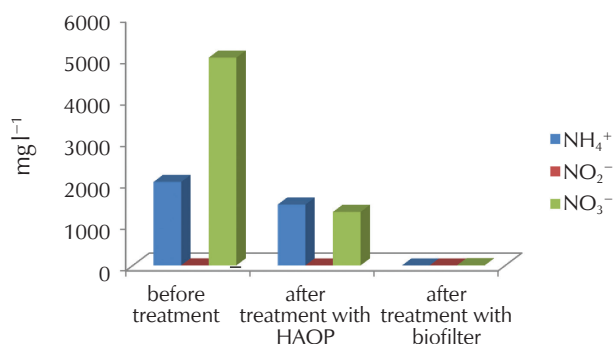


Fig. 10 – Evaluation of the nitrogenous material before and after biosorption of the mineralised leachate

method. With respect to phosphorus, the suitable item, the amount needed for microorganisms is a contentious topic.

The traditional proportion of C/N/P : 100/5/1 is usually recognised for suitable biosorption treatment. However, amid phosphorus deficiency,¹⁸ several researchers recorded excellent nitrogen extraction rates, describing the small phosphorus demand by the feasible biofilm recycling of phosphorus, arising from biomass decomposition. No negative effects on the nitrifying bacteria were found with respect to the effect of residual SM. This was in contradiction with some studies that argued that the ions present in OP's remaining SM had an adverse effect on the biological treatment.⁵¹ In fact, metal ions can create dormant bonds and associations with certain bacterial enzymes and cause inhibition of their biological functions.⁵²

Moreover, it has been stated that microorganisms are more susceptible to the toxicity of metals when they are in the shape of bare ions. On the other hand, metals' toxicity decreases when they are chelated by certain organic ligands such as humic substances.⁵³ This may clarify the negligible effect of ions on the method of nitrification, according to this research using Ain Defla landfill leachate.

3.2.8 Biosorption performance on mineralised leachate

Table 3 summarises the efficacy of mineralised and oxidised leachate biosorption treatment. The very low BOD₅ and COD concentrations of organic matter at the outlet indicated that all biodegradable organic matter had been removed. The pH adjustment was associated with the use of inorganic carbon during the nitrification phase by nitrifying autotrophic bacteria.

The leachate became transparent with a minimum turbidity value of 0.7 NTU, likely due to the double effect of biosorption: the adsorption and the nitrification of pollutants. The oxidation of the SM gave the yield of 71 %. This value improved with biosorption and reached the yield of 95.6 %. There was a slight increase in EC, which could be explained by the salt ion adsorption (NaCl) present in the physiological water used to wash the biosorption. The column was washed in abundance with physiological and demineralized water to remove all the salts and metals

Table 3 – Performance of treatment of oxidised leachate by biosorption

Characteristics of leachate	pH	COD	EC	Turbidity	SM	NH ₄ ⁺	NO ₂ ⁻	NO ₃ ⁻	PO ₄ ³⁻	Ca ²⁺	Mg ²⁺	Fe
unit	–	mg O ₂ l ⁻¹	μS cm ⁻¹	NTU	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹	mg l ⁻¹
oxidised leachate	6.39	1933.8	0.024	53.3	235	1479	1.52	1300	5.04	1.64	27.94	3.25
biosorption effluent	6.85	2.51	0.032	0.7	10.34	0.68	0.006	5.59	1.2	2.71	0.74	0.05
output / %		99.87	–	98.73	95.6	99.95	99.96	99.57	78	–	97.35	98.2

present in the components of the biomaterials. This technique has demonstrated an efficiency or no increase in the concentration of metals justified in other works by leaching the sludge used.^{18,54} Iron and other metals were also removed either by adsorption into pores of sawdust and activated coffee grounds or by the use of the bacterial sludge.

4 Conclusion

Relating to the characterization and treatment of the leachate from the TLC Ain Defla, it can be concluded that these effluents are at neutral pH 8.8 and have high content of organic matter (COD of 9669 mg O₂ l⁻¹, BOD₅ of 8875 mg O₂ l⁻¹) and suspended matter of 820 mg l⁻¹. These contents greatly exceed the permitted chemical element contents of industrial effluents according to the Algerian standard. Analysis of the mineral fraction of the leachate showed a dominance of magnesium. The treatment of leachate by coupling oxidation and biosorption under optimal conditions (45 °C, pH 2.6, with H₂O₂ volume fraction of 5 %, H₂O₂ concentration of 1.6 mol l⁻¹, Fe³⁺ concentration of 0.5 mmol l⁻¹, and the ratio [H₂O₂]/[Fe³⁺] is 40) is an efficient treatment process with BOD₅ reduction of 100 % and COD reduction of 99 %. Removal of colloidal particles (SM) was up to 95.6 %. Reductions of phosphates, ammonium, sulphates, zinc, and iron were 78, 98, 96, 92, and 98.2 %, respectively. This methodology could be transposed to other national and international landfill sites with the same intrinsic and extrinsic characteristics.

Conflict of interests

The corresponding author states that there is no conflict of interest.

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SAŽETAK

Obrada procjednih voda iz odlagališta otpada Ain Defla (Alžir)
procesom oksidacije i biosorpcije*Brahim Feraoun,^{a,*} Abd-Elmouneim Belhadj,^a Khaled Otmanine^b i Mounir Hammoudi^c*

Tema ovog istraživanja je obrada procjednih voda odlagališta otpada Ain Defla u Alžiru, koje predstavljaju ozbiljnu prijetnju po okoliš. Ispitivano je uklanjanje organskih tvari (izraženo preko kemijske potrošnje kisika (KPK) i biološke potrošnje kisika (BPK₅)), suspendiranih čestica (SČ), mineralnih tvari (fosfati i amonijačni dušik) i teških metala (cink i željezo) iz procjednih voda kombiniranjem procesa oksidacije i biosorpcije. Rezultati analiza pokazali su da je pH-vrijednost procjedne vode iznosila 8,8 te da je bila izrazito opterećena organskom tvari (zamućenje = 553 NTU, SČ = 820 mg l⁻¹; KPK = 9669 mg O₂ l⁻¹, te BPK₅ = 8875 mg O₂ l⁻¹), u solima (EC = 19,4 mScm⁻¹), amonijačnim dušikom (2027 mg l⁻¹), fosfatima (22,9 mg l⁻¹) i sulfatima (750 mg l⁻¹). Također sadržavala je i značajne količine teških metala, osobito cinka (4,21 mg l⁻¹) i željeza (47,5 mg l⁻¹). Iz fizikalno-kemijskih parametara praćenih tijekom obrade vidljivo je, pri optimalnim uvjetima (45 °C, [H₂O₂] = 1,6 mol l⁻¹), volumni udio φ(H₂O₂) = 5 % i [Fe³⁺] = 0,5 mmol l⁻¹), smanjenje KPK vrijednosti od 99 %, BPK₅ vrijednosti od 100 %, smanjenje fosfata, amonijaka, sulfata, cinka i željeza za 78 %, 98 %, 96 %, 92 %, odnosno 98 %. Također, 95,6 % suspendiranih čestica uklonjeno je tijekom obrade procjedne vode.

Ključne riječi

Procjedne vode, odlagalište, proces oksidacije, biosorpcija

^a Biomaterials and Transport Phenomena Laboratory (LBMPT), Yahia Fares University, Department of Chemical Engineering and Environment, Médéa, 26 000, Alžir

^b Bioresources Naturelles Locales LBRN, Hassiba Benbouali University of Chlef, Faculty of Technology, Department of Process Engineering, Chlef, Alžir

^c Biomaterials and Transport Phenomena Laboratory (LBMPT), Yahia Fares University, Department of Chemical Engineering and Environment, Experimental Biology and Pharmacology Team, Médéa, 26 000, Alžir

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