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Combination of advanced oxidation and biological processes for the landfill leachate treatment

R. Chemlal [a], L. Azzouz [a], R. Kernani [a], N. Abdi [a], H. Lounici [a], H. Grib [a], N. Mameri [a,c], N. Drouiche [b,*]

[a] Ecole Nationale Polytechnique, URIE, B.P. 182-16200, El Harrach, Algiers, Algeria
[b] Centre de Recherche en Technologie des Semi-Conducteurs Pour l'Energétique (CRTSE), Division CCPM, 2, Bd Dr. Frantz Fanon, P.O. Box 140, 7 merveilles, Alger 16038, Algeria
[c] University of Technology of Compiègne, Département Génie Chimique, B.P. 20.509, 60205 Compiègne Cedex, France

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ABSTRACT

This study's main purpose is to contribute to the Oued Smar landfill leachate decontamination. To achieve this aim, the advanced oxidation process (AOP) via heterogeneous photocatalysis (TiO_2/UV) was coupled with seeded bioreactors with different inoculums types (raw leachate, soil extract and activated sludge). The results obtained after heterogeneous photocatalysis show that the reduction is comprised between 50 and 84% of the initial COD maintained at pH 5. However, this treated leachate cannot be reused or discharged without another treatment into the environment. The new BOD_5/COD ratio ranging between 0.045 and 0.18 are favorable for biological treatment. The AOP–bioreactor coupling allowed an abatement of 90% of the initial BOD_5 and 87% of the initial COD leachate surface reduction with a final value of 1000 mg O_2/L. However, this value is not yet conformed to the norm. The biological treatment has shown the landfill indigenous microorganism's ability to degrade the irradiated leachate since, the mineralization dissolved organic carbon (DOC) rate is almost identical to the value obtained by activated sludge. These results encourage the supposition to recycle the irradiated leachate in the landfill which now possess a biopile behavior (loop AOP + landfill).

1. Introduction

The main problem encountered in the Oued Smar landfill rehabilitation is the generated leachate accumulation. Indeed, from 1978 to 2010, the discharge Oued Smar has received a large urban and industrial waste amount accumulating to 40 million tons. The leachate as a consequence of its heterogeneous composition (highly rich in organic and mineral matter) can contaminate the surrounding environment, in particular groundwater, affecting the water quality and compromising the human health. In addition to groundwater contamination, a soil quality alteration and an ecosystem imbalance was observed. The removal of the persistent leachate pollutants is a significant challenge (Deng and Ezyske, 2011; Cortés-Lorenzo et al., 2014; Zhang et al., 2013).

The conventional landfill leachate treatments can be classified in three major groups: (a) leachate transfer, (b) biodegradation and (c) chemical oxidation. These conventional leachate treatment methods are often expensive because of the initial outlay plant equipment, the energy requirements and the additional chemicals frequent use (Renou et al., 2008a; Oller et al., 2011). The leachate complexity, principally old leachate, implies more efficient and inexpensive treatment search, in order to reduce the landfill leachate negative impact on the environment (Vilar et al., 2011a; Fang et al., 2014).

During the last few decades, AOPs have been an intensive research theme for the mature or biologically stabilized leachate treatment, with the following purposes: (a) to increase the organics biodegradability for subsequent biological treatment; (b) to remove organic constituents; (c) to degrade organics as a post-treatment unit for other technologies (Deng, 2009; Poblete et al., 2011); and (d) reduce the toxicity. The AOP use is then justified if the resulting products in the reaction are assimilated by microorganisms in biological treatment (Lapertot et al., 2008; Yahiat, 2010).

The AOPs are characterized by the highly reactive $HO^{\bullet}$ radicals presence, which are suitable for a rapid and indiscriminate reaction with an organic compounds inducing its almost total mineralization (Bauer, 1994; Rodriguez et al., 2002; Tiburtius et al.,

* Corresponding author. Tel.: +213 21 433511; fax: +213 21 433511.
E-mail address: nadjibdrouiche@yahoo.fr (N. Drouiche).

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2005). Among the several processes that can be used to generate hydroxyl radical as a powerful oxidizing species, the photocatalytic oxidation by titanium dioxide (TiO_2) has attracted much attention as a promising chemical procedure for environmental cleanup (Ohko et al., 2001; Guo et al., 2010). TiO_2 is widely used as a photocatalyst because of its non-toxicity, photochemical stability, and low cost. The scientists predict that it will soon be recognized as one of the most effective means to deal with various wastewater kinds (Hoffmann et al., 1995; Wu et al., 2009).

In the initial step in TiO_2 – mediated photocatalysis degradation is used an (e−/h+) pair generation, leading to the hydroxyl radicals (•OH) formation as shown below:

To reduce the photocatalysis high cost, the inexpensive biological treatment was coupled with the photocatalysis (Sarria et al., 2002; Yahiat, 2010).

This study's aim is the coupling feasibility examination of an advanced oxidation process (TiO_2 supported/UV) with a biological treatment of leachate heavily laden with Oued Smar landfill pollutants. In the biological treatment, two microbial inoculum types were used: one was from the Oued Smar landfill raw leachate and the other is extracted from the Oued Smar landfill soil. These results are compared to those observed in the case of a conventional activated sludge treatment. The interest in the use of Oued Smar landfill raw leachate and soil extract as inoculum is to test the landfill indigenous microorganisms potential to metabolize organic matter fractions irradiated by AOP.

2. Materials and methods

2.1. Leachate characteristics

The leachates were collected during the 2010–2011 period at landfill distinct locations. The leachates were stored in obscurity at freezing temperature to minimize microbial activity on the leachate composition. It was characterized using the standard methods (APHA, 1999). The pH, conductivity were respectively measured using HANNA pH-meter (pH 211) and HANNA conductivity-meter (EC 214). Suspended solids (SS) were obtained by centrifugation then drying at 105 °C (NFT 90-105), BOD_5 analysis was carried out by respirometric method at constant volume (MA.315 BOD_5-1.1) and the heavy metals (Fe, Pb, Hg, Cu) were measured by atomic absorption spectrophotometer type SOLAAR UNICAM M series as described in standard methods (APHA, 1999).

The ammonia–nitrogen, nitrite and nitrate were determined using the ISO 7150/1, NFT 90-013 and NFT 90-102, respectively. The total nitrogen, chloride and PO_4^{3-} were dosed using NF EN 25,663, NFT 90-014 and NF EN 1189, respectively.

COD was measured by using thermoreactor closed reflux colorimetric method (MA.315 DCO-1.1).

2.2. Photocatalytic reactor design

The photocatalysis experiments were conducted in batch mode with a photocatalytic reactor operating like falling-film flow reactor along a glass plate with a recycle loop. The latter consists of a 3 L capacity tank steel chassis menu, an inclined glass plate is coated with the photocatalytic media 1048 (Ahlstrom–France) on which runs a thin layer of leachate to be treated and a cover under which are deposited three UV-C lamp OSRAM HNS 15 Watts – which allow continuous irradiation to the media. The media is a fibrous cellulose support coated with TiO_2, silica and zeolite. The silica plays the role of inorganic binder that ensures the TiO_2 adhesion on the support surface, the zeolite incorporation to the mixture TiO_2–SiO_2 increases the media adsorption capacity.

The treated leachate flow and slope inclination are adjustable. The leachate even distribution over the catalyst support entire surface is provided by a distributor valves equipped. The solution to be treated agitation is provided by a helical stirrer and the leachate oxygenation is supported by an air pump.

2.3. Biological treatment

The biological treatment consisted in the establishment of three bioreactors inoculated with different inoculums namely raw leachate, extract of soil from the Oued Smar landfill and finally the activated sludge.

Before beginning the biological treatment, a microorganism's adaptation phase in the irradiated leachate is necessary in order to reduce the bioreactors processing time. The raw leachate, the Oued Smar landfill soil extract and the activated sludge were mixed with leachate irradiated at 50% of the total volume, to which nutrients with a ratio C/N/P fixed at 100/10/1 were added. The inoculate were incubated at ambient temperature under stirring for 3 days and aeration is provided by an air pump. The precultures prepared and transferred at a rate of 10% in flasks 2 L containing irradiated leachate. The cultures were incubated at ambient temperature under stirring and aeration is provided by a compressor. During the treatment, urea and K_2HPO_4 were added as source of nitrogen and phosphorus according to the ratio C/N/P fixed (100/10/1) in the purpose to stimulate and improve the organic matter biodegradation. pH, NO_3^-, NO_2^-, NH_4^+, COD parameters have been monitored regularly to evaluate growth and microbial metabolism. The microbial growth was evaluated turbidimetrically by measuring the biomass concentration optical density at 600 nm using a spectrophotometer Shimadzu UV-240. The CO_2 emitted during the organic matter biodegradation in the bioreactors was determined by using the previous technique reported by Namkoong et al. (2002).

Where DOC_i is the initial dissolved organic carbon concentration for irradiated leachate, the C–CO_2 is the carbon amount for CO_2 generated by the organic matter mineralization.

Finally TOC and DOC (samples were pre-filtered through 0.45 mm Millipore filter) were measured by a TOC analyzer type TOC-VCPH, Shimadzu (MA.300-C1.0.

The AOS (the average oxidation state), which is an important parameter very useful to estimate the mixed solutions oxidation degree and gives indirect information on its biodegradation probability, was calculate using Eq. (1) (Sarria et al., 2003).

$$AOS = \frac{4(TOC - COD)}{TOC} \quad (1)$$

Where TOC and COD are expressed in mg of carbon per liter and mg of O_2 per liter, respectively. AOS takes values between +4 for CO_2,

Table 1
Physico-chemical characteristics of the landfill leachate.

Parameters	Unity	L1 (03/2010)	L2 (05/2010)	L3 (01/2011)
pH	–	7.4	7.8	7.3
Conductivity	mS/cm	6.6	8.7	5
Chloride	mg/L	1302	3184	1782
S.M	mg/L	413	1142	39,125
COD	mg O_2/L	1233	16,500	4417
BOD_5	mg O_2/L	220	750	250
BOD_5/COD ratio	–	0.178	0.045	0.056
NTK	mg/L	5.8	8.5	3.5
NH_4^+	mg/L	166	392	356
NO_3^-	mg/L	15	54	95
NO_2^-	mg/L	0.11	1.9	0.3
PO_4^{-3}	mg/L	0.9	1.22	26.41
Fe	mg/L	10	2.8	83
Cu	mg/L	0.04	0.08	0.14
Pb	mg/L	0.22	0.34	0.46
Hg	mg/L	ID	ID	ID
Color λ = 450 nm	–	0.198	0.840	0.094
λ = 254 nm	–	0.748	0.960	0.611