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An innovative multistage treatment system for sanitary landfill leachate depuration: Studies at pilot-scale

Tânia F.C.V. Silva [a,*], Petrick A. Soares [a], Diego R. Manenti [a], Amélia Fonseca [b], Isabel Saraiva [b], Rui A.R. Boaventura [a], Vítor J.P. Vilar [a,*]

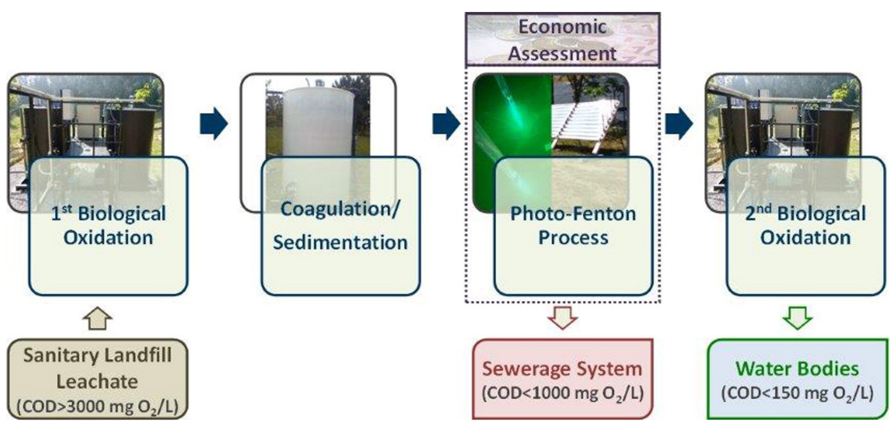
[a] *Laboratory of Separation and Reaction Engineering - Laboratory of Catalysis and Materials (LSRE-LCM), Faculdade de Engenharia, Universidade do Porto, Rua Dr. Roberto Frias, 4200-465 Porto, Portugal*

[b] *Efacec Engenharia e Sistemas S.A. (Unidade de Negócios Ambiente), SA, Rua Eng. Frederico Ulrich – Guardeiras, Apartado 3003, 4471-907 Moreira da Maia, Portugal*

HIGHLIGHTS

- Multistage treatment strategy for mature leachate from municipal sanitary landfill.
- Integration of bio-oxidation/coagulation/photo-Fenton/bio-oxidation.
- The pre-treatments led to a photo-oxidation 4× faster, with less energy and H_2O_2.
- The full combined treatment allows to obtain a total mineralization above 90%.
- The photo-Fenton cost was predicted in 6.4 €/m^3, using artificial/solar radiation.

GRAPHICAL ABSTRACT



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ABSTRACT

In this work, an innovative methodology for the treatment of landfill leachates, after aerobic lagooning, is proposed and adjusted at pilot-scale. This methodology involves an aerobic activated sludge biological pre-oxidation (ASBO), a coagulation/sedimentation step (240 mg Fe^{3+}/L, at pH 4.2) and a photo-oxidation through a photo-Fenton (PF) reaction (60 mg Fe^{2+}, at pH 2.8) combining solar and artificial light.

The ASBO process applied to a leachate after aerobic lagooning, with high organic and nitrogen content (1.1–1.5 g C/L; 0.8–3.0 g N/L) and low biodegradability (BOD_5/COD = 0.07–0.13), is capable to oxidise 62–99% of the ammonium nitrogen, consuming only the affluent alkalinity (70–100%). The coagulation/sedimentation stage led to the humic acids precipitation, promoting a marked change in leachate colour, from dark-brown to yellowish-brown (related to fulvic acids), accompanied by a reduction of 60%, 58% and 88% on DOC, COD and TSS, respectively.

The PF system promoted the degradation of the recalcitrant organic molecules into more easily biodegradable ones. According to Zahn-Wellens biodegradability test, a leachate with 419 mg DOC/L after coagulation, would have to be photo-oxidized until DOC < 256 mg/L, consuming 117 mM of H_2O_2 and 10.4 kJ/L of accumulated UV energy, to achieve an effluent that can be biologically treated in compliance with the COD discharge limit

* Corresponding authors.
E-mail addresses: tania.silva@fe.up.pt (T.F.C.V. Silva), vilar@fe.up.pt (V.J.P. Vilar).

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(150 mg O_2/L) into water bodies. The biological process downstream from the photocatalytic system would promote a mineralization >60%. The PF step cost to treat 100 m^3/day of leachate was 6.41 €/m^3, combining 1339 m^2 of CPCs with 31 lamps.

1. Introduction

In the last decades, the production of waste increased significantly due to excessive population growth and changing of the consumption habits (Hoornweg et al., 2013). Landfilling is the most used final waste disposal method around the world. However, one of the environmental problems associated with the disposal of municipal waste in landfills is the generation of leachate. This kind of wastewater is characterized by a high strength complex mixture containing dissolved hazardous organic compounds, ammonia, heavy metals and inorganic salts, which need to be removed due to their toxicity and consequent impacts on the environment. Besides that, the leachates also present high variability in their quantity and quality, along the year, which makes the definition of an efficient treatment line for all situations very difficult (Öman and Junestedt, 2008; Renou et al., 2008).

Due to their cost-effectiveness, biological processes are usually used to remove the biodegradable organic fraction present in wastewaters. These oxidation processes are very effective in the treatment of leachates from young landfills but are ineffective in the treatment of stabilized leachates due to the presence of a high fraction of recalcitrant organic compounds, mainly humic substances (O'Leary and Tchobanoglous, 2002; Renou et al., 2008). Advanced oxidation processes (AOPs) are able to degrade a wide range of compounds from stabilized landfill leachates. Despite their high effectiveness, they become quite expensive if applied alone (Cassano et al., 2011; Hermosilla et al., 2009; Umar et al., 2010; Vilar et al., 2011b). Having in mind that leachates present high contents of nitrogen and recalcitrant organic matter and aiming at a significant reduction on the treatment cost, the best strategy, for leachate remediation, seems to be the integration of biological and chemical oxidation processes.

The first pilot plant for stabilized leachate treatment, combining a solar photo-Fenton (PF) reaction with activated sludge biological oxidation (ASBO), was presented in our previous works (Silva et al., 2016; Silva et al., 2013a; Silva et al., 2013b; Silva et al., 2013c). However, it was observed that the photo-reaction efficiency was strongly affected by the (i) weather conditions, mainly due to low irradiances and temperatures in the winter season; (ii) presence of humic acids, related to the dark-brown colour intrinsic to leachates; (iii) high amount of total suspended solids (TSS), resulting from the precipitation of some organic compounds with ferric ions, after acidification and during reaction, and (iv) high amounts of sulphate ions provided by the sulphuric acid addition to perform the initial acidification of the PF reaction. The non-elimination of the produced acid sludge decreases the PF reaction efficiency (~30%), due to the low light transmissibility caused by the high amount of TSS that compete with H_2O_2 and iron species as photons absorbers. Besides, higher amounts of H_2O_2 and energy were required for the degradation of the additional particulate organic matter.

So, considering all drawbacks found in the previous works (Silva et al., 2016; Silva et al., 2013a; Silva et al., 2013b; Silva et al., 2013c) and the high treatment costs associated with the PF reaction (Silva et al., 2016), it was concluded that the best strategy to treat mature leachates would be: (i) initial ASBO under aerobic conditions, to promote nitrification and simultaneous removal of the biodegradable organic carbon fraction and alkalinity, thus decreasing the acid dose needed in the subsequent coagulation step; (ii) coagulation using ferric salts at acid pH to achieve the precipitation of humic acids (up to 50% of the recalcitrant organic content), resulting in a yellow colour leachate, mainly attributed to the presence of dissolved fulvic acids, with a low suspended solids content and high UV–visible transmissibility; (iii) PF oxidation reaction (Fe^{2+}/H_2O_2/UV–vis), using artificial and solar radiation, to degrade the most recalcitrant organic compounds, through the generation of powerful reactive chemical species, such as hydroxyl radicals (•OH), turning them into simpler and easily biodegradable organic compounds and (iv) subsequent biological oxidation step under aerobic/anoxic conditions, to promote the removal of the remaining nitrogen species and biodegradable organic fraction (TN_{final} < 15 mg/L; COD_{final} < 150 mg O_2/L; Decree law n°236/98).

Since it was not possible to fully adapt the existing pilot plant to operate according to this new methodology, it was decided to use a treatment strategy based on the first three steps and evaluate the last one by the Zahn-Wellens biodegradability test. Bearing all this in mind, the following objectives were established: (i) leachate characterization along all stages (biological oxidation, coagulation and photo-oxidation); (ii) individual efficiency assessment of the biological reactor (under aerobic regime), coagulation/sedimentation stage (for different values of pH and settling times) and PF reaction (using solar and/or artificial radiation and changing the number and rated power of the lamps); (iii) evaluation of the leachate treatment train; and (iv) economic analysis of the phototreatment step (based on the test conducted under the optimal operating conditions).

2. Experimental

2.1. Chemicals

Coagulation tests were accomplished by employing ferric chloride (40%, 1.44 g/cm^3) and sulphuric acid (98%, 1.84 g/cm^3). The PF experiments were performed using hydrogen peroxide (50% (w/v), 1.10 g/cm^3), iron sulphate heptahydrate, as iron source, and sulphuric acid and sodium hydroxide (30%, 1.33 g/cm^3) for pH adjustments. All commercial grade chemicals were purchased from Quimitécnica.

2.2. Sanitary landfill leachate samples

Leachate samples were collected after aerobic lagooning at a leachate treatment plant (LTP), from a municipal solid waste sanitary landfill located in northern Portugal.

Table 1 presents the main physico-chemical characteristics of the leachate.

2.3. Experimental setup

The pilot-scale plant, located at the LTP, comprises: (i) biological oxidation; (ii) coagulation/sedimentation and (iii) photo-oxidation systems. The schematic representation of the facility is shown in Fig. 1. Furthermore, the detailed description of the plant, as well as the main information about each component, is presented in Table SM-1 (see Supplementary data file).

2.4. Experimental procedure

During the trial period, 15 experiments were carried out, combining ASBO with coagulation/sedimentation and PF reaction, using solar/artificial radiation. Table SM-2 shows a brief description of the experiments performed along this work.