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Coupling ARB-based biological and photochemical (UV/TiO_2 and UV/$S_2O_8^{2-}$) techniques to deal with sanitary landfill leachate

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ABSTRACT

The aim of this study was to provide an alternative way to remove bio-refractory organics and ammonical-nitrogen from mature municipal solid waste (MSW) landfill leachate by combining biological and photochemical processes. To achieve this objective, the effectiveness of anoxic aged refuse-based bioreactor (ARB) for biological leachate pretreatment followed by Advanced Oxidation Processes (AOPs) by heterogeneous photocatalysis (TiO_2/UV) and persulfate ($S_2O_8^{2-}$) oxidation were tested. The results obtained after ARB based pre-treatment demonstrated a mean 72%, 81% and 92% degradation of COD, NH_4—N and TN, respectively. However, this treated leachate cannot be discharged without another treatment; hence, it was further treated by UV-mediated TiO_2 photocatalysis and $S_2O_8^{2-}$ oxidation. An average 82% of COD was abated at optimum condition (1 g L^{-1} TiO_2; pH 5) whereas, using an optimum 1.5 g L^{-1} persulfate at pH 5, 81% COD reduction occurred. Acidic and alkaline pH favored COD and NH_4—N removal respectively. The results of this study demonstrated that coupling ARB with AOPs is potentially applicable process to deal with bio-recalcitrant compounds present in mature landfill leachate.

1. Introduction

Sanitary landfilling is the widely practiced approach throughout the world for disposal of municipal solid waste (MSW) especially in developing and underdeveloped countries owing to its low immediate costs; but improper landfilling is a matter of concern due to generation of heavily polluted leachate as a result of percolation of rainwater and moisture content through solid waste (Hassan and Xie, 2014; Hassan et al., 2016). Based on strength of pollutants concentration and age, landfill leachate can be classified into young, intermediate and stabilized/mature leachate (Table 1). Young landfill leachates containing high COD values and BOD_5/COD ratio or greater than 0.4, is comparatively easier to be treated by biological process (Chamarro et al., 2001), while mature landfill leachates typically contain a high strength ammonium and non-biodegradable organic matter content (dominated by humic and fulvic acids), which leads to low BOD_5/COD and BOD_5/TN ratio (Harsem, 1983; Renou et al., 2008).

Certain biological treatment methods have shown satisfactory results were applicable only for young landfills, which typically generate more readily biodegradable leachate. Mature landfill leachate is characterized by low biodegradability due to the release of refractory organics with high molar masses and greater concentration of ammonical nitrogen, making it difficult to be treated by conventional biological processes (Calace et al., 2001). In such cases there is an urgent need of certain processes that offer better treatment efficiency. In this context, Advanced Oxidation Processes (AOPs), which are oxidation methods relying on the action of highly reactive species such as hydroxyl ($HO^\cdot$) radicals, could be a viable alternative for the removal of recalcitrant organic pollutants difficult to treat using conventional methods owing to their high chemical stability and/or low biodegradability (Meeroff et al., 2012). However, in order to enhance pollutants removal efficiency and reduce operating costs associated to the application of AOPs, their combination with conventional biological processes should be considered (Cesaro et al., 2013). AOPs can be used as pre and/or post-treatment of wastewater and landfill leachate. In the former case, AOPs aim to improve biodegradability of wastewaters, (Bila et al., 2005); whereas in the latter case, the oxidation step is directed towards the removal of those pollutants which could not be completely degraded by biological treatment techniques (Poole, 2004).

Over the past decade, photo-assisted TiO_2 catalysis (Batkhande et al., 2001) and peroxydisulfate oxidation (Abu Amr et al., 2013) have attracted much attention due to their outstanding efficiency for environmental purification. In TiO_2 photocatalysis, $^\cdot OH$ radicals

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Table 1
Landfill leachate classification versus age (Chian, 1977; Mcbean et al., 1995; Cortez et al., 2011; Renou et al., 2008; Lema et al., 1988; Kurniawan et al., 2006).

Parameters	Young leachate	Medium leachate	Mature leachate
Age (years)	<10	5–10	>10
pH	6.5	6.5–7.5	>7.5
COD ($mg\ L^{-1}$)	>10000	5000–10,000	<5000
BOD_5 ($mg\ L^{-1}$)	10,000–20,000	–	50–100
TOC ($mg\ L^{-1}$)	9000–15,000	–	100–1000
BOD/COD	>0.5	0.1–0.5	<0.1
TOC/COD	<0.3	0.3–0.5	>0.5
Organic compounds (%)	80% VFAs	5–30% VFAs + HAs and FAs	HAs and FAs
Biodegradability	High	Medium	Low

VFA: volatile fatty acids; HAs: humic acids; FAs: fulvic acids.

and others can be generated, which can oxidize a wide variety of organic pollutants to harmless inorganics such as CO_2, H_2O and other minerals (Saien et al., 2011; Bauer et al., 1999).

Peroxydisulfate ($S_2O_8^{2-}$, commonly known as persulfate) is a commonly used compound for the production of sulfate radicals (SRs), and has received a significant consideration in landfill leachate treatment owing to its effectiveness in degrading refractory organics and ammonical nitrogen content (Deng and Ezyske, 2011). After activation by heat energy, UV irradiation or ultrasound, persulfate anions are transformed into SRs ($SO_4^{-\cdot}$, $E° = 2.6$ V), which subsequently might be converted into $^{\cdot}OH$ ($E° = 2.80$ V) and hydrogen peroxide ($E° = 1.78$ V), which are strong oxidizing agents (House, 1962).

So far, a generalized treatment method does not exist as a consequence of diverse landfill leachate composition. Therefore, a combination of biological and physico-chemical methods is essential to treat mature leachate. In this study an aged refuse-based bioreactor (ARB) has been employed as biological pretreatment followed by UV-assisted titanium dioxide (UV/TiO_2) and sodium persulfate ($UV/Na_2S_2O_8$) for the post-treatment of mature landfill leachate. ARB has been proved to be a promising biological treatment method to remove various pollutants from landfill leachate; and it validates the principal of waste control by waste (Hassan and Xie, 2014). Many studies have showed significant removal of organic pollutants using ARB, e.g., 90–99% CODr (Zhao et al., 2002), 80% CODr (Xie et al., 2010), and 80–91% CODr (Wang et al., 2014). Despite better removal efficiency most of the studies were unable to meet the stringent maximum permissible discharge limits for leachate, i.e. 100 $mg\ L^{-1}$ (COD), 30 $mg\ L^{-1}$ (BOD_5) and 25 $mg\ L^{-1}$ (NH_4—N) (National Emission Standard, 2008). For example, using ARB to deal with landfill leachate, the COD effluent concentration attained were 226–335 $mg\ L^{-1}$ (Wang et al., 2014), 239 $mg\ L^{-1}$ (Lei et al., 2007), and 520 $mg\ L^{-1}$ (Xie et al., 2010) from initial concentrations of 2257, 2758 and 5800 $mg\ L^{-1}$, respectively. In this study, AOPs based post treatment is meant to apply in order to remove the pollutants meeting the discharge standard for leachate.

The objectives of this study were (i) to evaluate the removal efficiency of ARB + UV/TiO_2 or $UV/S_2O_8^{-2}$ with respect to COD and NH_4—N, and (ii) to evaluate the effects of removal efficiency by pH, UV irradiation time, chemical dose and air flow rate.

2. Materials and methods

2.1. Leachate sampling and reagents used

Raw leachate (mature) samples were collected from Laogang sanitary disposal treatment plant, located on the outskirts of shanghai, China (°52′46.10″E, 31°3′25.22″N). Laogang landfill is the largest landfill in China, which has been operated since 1989. It receives around 10,000 tons MSW per day and generates about 3000 m^3 leachate per day. The landfill is separated into many small pits (400 m × 125 m). Banks, waterways, docks, roads, bridges and separating bands are equipped in the landfill. Reaching 4 m in height, MSW is covered with 30 cm thick layer of soil. Leachate is collected to ponds from every pit, which after anaerobic digestion and oxidation in the ponds is directly released to the East China Sea, 1 km away from the landfill site.

The leachate samples were shipped in polyethylene containers directly from the landfill site to the lab, and stored at 4 °C in the dark immediately after being received to minimize any further physico-chemical or biological change until completion of the experiments. The characteristics of leachate were as follows: COD of 1700–2300 $mg\ L^{-1}$, BOD_5 of 320–500 $mg\ L^{-1}$, NH_4^+—N of 1040–1237 $mg\ L^{-1}$, NO_2^-—N of 0.5–4.7 $mg\ L^{-1}$, NO_3^-—N of 13.7–46.1 $mg\ L^{-1}$, TN of 2000–2250 $mg\ L^{-1}$ and pH of 7.9–8.2. The photocatalyst used in this study was Degussa P-25 TiO_2 (80% anatase and 20% rutile) with particle size of 21 nm. $Na_2S_2O_8$ with ⩾ 99.5% purity was obtained from Shanghai Lingfeng Chemical Reagent, China. All reagents were used as received without further purification. Distilled water was used throughout the experiments.

2.2. Experimental setup

2.2.1. Biological procedure

The refuse in landfills becomes aged or stabilized after 8–10 years of placement, and the resultant partly or fully stabilized refuse thus obtained in this work is referred to as aged refuse (AR). The lab scale AR bioreactor used for this study was made of PVC pipe with dimensions (30 cm inner diameter; 50 cm height) and effective volume of 30 L, as employed in our previous study (Xie et al., 2010, 2012). The AR collected from landfill cell of Shanghai Laogang sanitary disposal site was used as filter material after being shredded into smaller particles ($\Phi < 50$ mm). In order to avoid the clogging of the water outlet, the bottom of the reactor was filled with 5 cm layer of gravel followed by shredded AR. The column reactor was placed in a temperature controlled room, and the experiment was conducted at 30 °C. The influent hydraulic load was about 20 $L\ m^3\ d^{-1}$, corresponding to a hydraulic retention time of 10 d.

The AR has special characteristics different from other biological media like soil, activated sludge, etc. It has little volumetric weight, high porosity, high organic content, cation exchange capacity, and adsorption ability (Zhao et al., 2002; Sachs, 1999). Moreover, AR is a suitable media for microbial attachment and growth. It contains a diverse and populous community of microorganisms which is adapted to the conditions. Pyrosequencing analysis indicated that the predominant bacterial groups in samples at family level are *Pseudomonadaceae*, *Flavobacteriaceae*, *Xanthomonadaceae*, *Alcaligenaceae*, *Planctomycetaceae*, etc. (Xie et al., 2012).

2.2.2. Photochemical procedure

Effluent collected from ARB was irradiated with the photoreactor system A (Fig. 1A) and tubular-shaped photoreactor B (Fig. 1B). Photoreactor A (15 × 15 × 15 cm in dimension) consists of a UV lamp (model GGZ-125, 125W; λ 365 nm) located on the top of the reactor at a height of around 10 cm from the leachate sample kept in a glass container. The working volume of the sample was 1 L with surface area of 120 cm^2 exposed to UV lamp from the top. The titania suspension was continously agitated by a magnètic stirrer to homogenise the solution. The reactor was wrapped with aluminum foil so as to reflect any scattered UV light back into the photoreactor and to prevent the system from room lights. The