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Effect of nano-ZnO on biogas generation from simulated landfills

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ABSTRACT

Extensive use of nanomaterials in commercial consumer products and industrial applications eventually leads to their release to the waste streams and the environment. Nano-ZnO is one of the most widely-used nanomaterials (NMs) due to its unique properties. It is also known to impact biological processes adversely. In this study, the effect of nano-ZnO on biogas generation from sanitary landfills was investigated. Two conventional and two bioreactor landfills were operated using real MSW samples at mesophilic temperature (35 °C) for a period of about 1 year. 100 mg nano-ZnO/kg of dry waste was added to the simulated landfill reactors. Daily gas production, gas composition and leachate Zn concentrations were regularly monitored. A model describing the fate of the nano-ZnO was also developed. The results obtained indicated that as much as 99% of the nano-ZnO was retained within the waste matrix for both reactor operation modes. Waste stabilization was faster in simulated landfill bioreactors with and without the addition of nano-ZnO. Moreover, the presence of the nano-ZnO within the waste led to a decrease in biogas production of about 15%, suggesting that the nano-ZnO might have some inhibitory effects on waste stabilization. This reduction can have potentially significant implications on waste stabilization and the use of biogas from landfills as a renewable energy source.

1. Introduction

Nanotechnology, which is the utilizing of matters at nanoscale (1–100 nm) is considered today one of the most significant advancements in science and technology (Abdelsalam et al., 2016). As a result of rapid development in nanotechnology in recent years, the number of commercially available nanotechnology products has exceeded one thousand (Vance et al., 2015). The concern about the impacts of NMs on the environment and public health is increasing because of the wide usage of NMs in different applications and therefore their potential discharge to the environment. There exist many recent studies in literature that predict the release of NMs to air, soil, water and landfills (Keller and Lazareva, 2014). In particular, recent work reported about the presence of colloids and nanoparticles in 25 different leachate samples (Hennebert et al., 2013). It is estimated that 50% of NMs used in numerous commercial products will ultimately be sent to landfills for final disposal after their useful lives are over (NanoTech Project, 2007). Most of the published studies found in the literature have mainly focused on the ecotoxicity of NMs (Bystrzejewska-Piotrowska et al., 2009; Donaldson and Golyasnya, 2004; Handy et al., 2008; Lovern and Klaper, 2006; Maynard and Kuempel, 2005; Nel et al., 2006; Roberts et al., 2007). However, after their disposal the fate and impact of NMs during waste stabilization in landfills which play a crucial role in integrated waste management systems, is still unknown.

The fate of NMs in landfills depends on the properties of landfill leachate since leachate is site-specific and varies according to solid waste composition and landfill design. It is also depended on landfill age and the environmental factors including the precipitation rate and the change in temperature (Bolyard et al., 2013; Plocoste et al., 2016). It is therefore important to investigate the fate of NMs in landfills in order to evaluate their impacts on waste stabilization, biogas generation and control their possible discharge to the groundwater system through landfill leachate.

Among various NMs, nano-ZnO gained a lot of attention because of their wide implementation in industrial, military and medical applications (Nguyen et al., 2015). Nano-ZnO can find their way to the landfills through disposal of cosmetics, UV protection, coating materials, food additives, rubber manufacture and catalysts (Bystrzejewska-Piotrowska et al., 2009; Demirel, 2016).

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Nomenclature

C	Zn concentration in leachate function of time, M/L^3
$C_{zn,l}(t)$	concentration of Zn in the weekly collected leachate, function of time
D	dispersion coefficient, L^2/T
K_{dep}	deposition rate constant, 1/T
K_{det}	detachment rate constant, 1/T
$m_{zn,in}(t)$	mass of Zn inside the reactor, function of time
$m_{zn,r}(t)$	mass of Zn added to Reactors 1 and 2 due to leachate recirculation, function of time
$m_{zn,b}$	background mass of Zn in the waste (from Table 3)
$m_{zn,}n(t)$	mass of nano-Zn added to Reactors 1 and 4 at the beginning of the experiments
$m_{zn,l}(t)$	mass of Zn in the collected leachate, function of time
q	time-dependent water flux in the reactor due to moisture addition and leachate recirculation (if present), L/T
S	Zn concentration on the solid waste as a function of time, M/M
t	time
$V_{zn,l}$	volume of weekly collected leachate samples
z	vertical coordinate, L
ρ_b	dry bulk density of the waste, M/L^3
θ	moisture content of the waste, L^3/L^3

Luna-del Risco et al. (2011) investigated the short-term impacts of bulk and nano-ZnO on biogas and methane production from mesophilic anaerobic digestion (AD) of cattle manure. It was reported that the biogas production decreased by 43% and 74% in the presence of 120 and 240 mg/L of nano-ZnO. In a long-term study, the effect of nano-ZnO on anaerobic digestion of waste activated sludge was reported. It was observed that methane production was not affected by the addition of 1 mg ZnO/g total suspended solids (TSS). On the other hand, presence of higher concentrations of 30 and 150 mg ZnO/g TSS in the system represented inhibitory impacts which is showing that the amount of nano-ZnO added to the system plays the crucial role in inhibition mechanism (Mu and Chen, 2011). It was also concluded that the adverse impact of nano-ZnO was mainly associated with the release of Zn^{2+} from ZnO and its effect on the activity of key enzymes such as acetate kinase, protease and coenzyme F_{420} during the hydrolysis and methanogenesis stages of the AD process (Mu and Chen, 2011). The authors also reported that up to 150 mg/g TSS of nano-TiO_2, nano-Al_2O_3 and nano-SiO_2 concentrations did not adversely affect the AD of waste activated sludge. This finding indicated that nano-ZnO posed inhibitory effects on the biological activity even at much lower amounts in comparison to nano-TiO_2, nano-Al_2O_3 and nano-SiO_2 (Mu et al., 2011). The adverse impacts of nano-ZnO, on the acetoclastics and hydrogenotrophic methanogens in anaerobic granular sludge were also reported (Gonzalez-Estrella et al., 2013). In addition, the impacts of nano-ZnO on aerobic wastewater treatment processes have been extensively studied (Chauque et al., 2014; Hou et al., 2014; Mei et al., 2014; Puay et al., 2015; Tan et al., 2015; Yu et al., 2015; Zhou et al., 2015).

The above literature shows that the impact of nano-ZnO on anaerobic digestion and wastewater treatment systems has been widely studied. Yet, only a handful of studies have examined the fate of nano-ZnO within integrated solid waste management systems. The first study to focus on the fate of NMs in relation to municipal solid waste (MSW) was conducted by Bolyard et al. (2011), who investigated the behavior of nano-TiO_2, nano-Ag, nano-ZnO in different leachates acquired from real landfills. It was observed that coated NMs in leachate had no effect on biological processes, in particular on the five-day Biochemical Oxygen Demand (BOD_5) and methane generation processes. This was due to the low concentration of free metal ions generated from the interaction of NMs with leachate. Moreover, it was also reported that nano-TiO_2 and nano-ZnO aggregated into larger particles (Bolyard et al., 2011). In another study, Yang et al. (2013) investigated the effect of nano-Ag on waste stabilization in bioreactor landfill. It was reported that the addition of 10 mg/kg of nano-Ag to the municipal solid waste led to a 80% reduction in methane production. Dülger et al. (2016) and Sakallıoğlu et al. (2016) examined the leaching potential of nano-TiO_2 and nano-ZnO with different surface coatings from fresh MSW and they both reported that the NMs were mostly retained within the solid waste matrix rather than moving with the leachate.

The above literature review indicates that little information about the fate and impacts of NMs on waste stabilization in landfills is available. Moreover, no study was encountered in the literature that investigated the long-term impact of nano-ZnO in simulated landfills despite the fact that nano-ZnO had reported impacts on biological processes. Therefore, the aim of this study was to fill the lack of information about the impact of nano-ZnO on waste stabilization including measuring landfill gas generation. In order to capture the overall effect on produced biogas, landfill lysimeter experiments were conducted (Krause et al., 2016) to understand and evaluate the behavior of nano-ZnO in waste disposal sites. Two types of landfill operation systems were considered: conventional and bioreactor. Conventional landfills are operated on dry tomb basis where leachate is collected for external treatment, leading to waste decomposition in several decades. However, bioreactor landfills implement leachate recirculation to the waste to enhance the decomposition rate by creating more uniform environment (Benson et al., 2007; Onay and Pohland, 1998). A model simulating the fate of the nano-ZnO in the waste was also developed in this study.

2. Materials and methods

2.1. Materials

Real MSW samples were obtained from a sanitary landfill located in Izmit, Turkey. The samples were homogeneously mixed, shredded to a diameter of 8–10 cm, characterized and stored at 4 °C prior to use. The waste characterization of the MSW was conducted by manual separation and weighing of the components (Table 1). The organic content of the waste was high, with an

Table 1
Waste composition.

Waste component	Weight percent, %
Organic waste	52.00
Plastic	13.50
Glass	7.40
Textile	6.29
Paper	15.84
Yard waste	2.50
Others	2.47

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