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Leaching of nano-ZnO in municipal solid waste

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HIGHLIGHTS

- Leaching potential of 3 different types of nano-ZnO in real fresh MSW was investigated.
- Batch tests were conducted at different pH, ionic strength and ZnO concentrations.
- Most of the added nano-ZnO mass was retained within the solid waste matrix.
- The pH and IS conditions did not significantly influence the leaching behavior of ZnO.
- A kinetic particle deposition/detachment model was developed to analyze ZnO behavior.

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ABSTRACT

Despite widespread use of engineered nanomaterials (ENMs) in commercial products and their potential disposal in landfills, the fate of ENMs in solid waste environments are still not well understood. In this study, the leaching behavior of nano ZnO -one of the most used ENMs- in fresh municipal solid waste (MSW) was investigated. Batch reactors containing municipal solid waste samples were spiked with three different types of nano ZnO having different surface stabilization. The leaching of ZnO was examined under acidic, basic and elevated ionic strength (IS) conditions. The results of the 3-day batch tests showed that the percent of the added nano-ZnO mass retained within the solid waste matrix ranged between 80% and 93% on average for the three types of nano-ZnO tested. The pH and IS conditions did not significantly influence the leaching behavior of ZnO. To further analyze the behavior of ZnO in the MSW matrix, a kinetic particle deposition/detachment model was developed. The model was able to reproduce the main trends of the batch experiments. Reaction rate constants for the batch tests ranged from 0.01 to 0.4 1/hr, reflecting the rapid deposition of nano-ZnO within the MSW matrix.

1. Introduction

Engineered nanomaterials (ENMs) are used in numerous commercials products nowadays. Despite the rapid and massive development of ENMs, the fate and transport of these ENMs when released into the environment and their potential adverse effects on human health and other living organisms are not adequately understood. It was reported that silica, titanium, alumina, iron and zinc oxides dominated the ENM market in 2010 based on material mass flow analysis. Among these ENMs, zinc oxide (ZnO), the focus of this study, is commonly used in cosmetics, electronic and optics, coatings-pigments-paints, textiles, plastics, energy, environmental, and medical applications [1].

Large fractions of ENMs released from household or industrial commodities eventually find their way into the waste or wastewater treatment plants [2,3]. It was predicted that 63–91% or over 260,000–309,000 metric tons of global ENM production eventually ended up in landfills in 2010 [1]. A more recent study indicated that ZnO, along with TiO_2, represented 94% of ENMs released into the environment from personal care products [4]. More detailed discussion regarding environmental concentrations of ENMs, including ZnO, can be found elsewhere [5]. It is noted that, ZnO ENMs are emitted into the environment during both manufacturing and use, and a huge amount is estimated to end up in landfills. Furthermore, they are predicted to end up in soil and water as well [1]. Since Zn is known to have adverse impacts on biological activity, the form of Zn entering the environment coming from ZnO ENMs is very crucial.

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Nomenclature

$C(t)$	ZnO concentration in leachate function of time, M/L^3
ENM	Engineered nanomaterials
K_{dep}	Deposition rate constant, 1/T
K_{det}	Detachment rate constant, 1/T
MSW	Municipal solid waste
M_s	Mass of dry solid waste in the test, M
$S(t)$	ZnO concentration on the solid waste as a function of time, M/M
t	Time
V_w	Volume of water in the test (includes moisture in the waste and water added), L^3

Even though various studies predict that the majority of ENMs will end up in landfills either through direct disposal or as sludge and bottom and fly ash matrix from waste incineration [2,6–9], information about the fate of ENMs during waste stabilization is still scarce [10]. It is however reported that future research activities should at least include analytical techniques to characterize nano-waste, definition of acceptable limit values for exposure and to determine the release mechanisms of ENMs [10].

In the most relevant study covering the behavior of ZnO NPs in solid waste environment, the fate of coated nano-ZnO in real landfill leachates was investigated [11]. This study showed that the nano-ZnO which remained mostly in solid form did not affect the biological processes in landfill leachate and the dissolved Zn was mainly associated with dissolved organic matter. On the other hand, numerous studies in the literature have reported about the inhibitory/toxic effects of ZnO nanoparticles to aerobic [12–15] and anaerobic waste/wastewater treatment processes [16–22], indicating the potential adverse impacts of ZnO nanoparticles on biological systems.

To date, no study can be found in the literature that specifically examines the fate and leaching potential of the ZnO nanoparticles within the complex landfill matrix. Therefore, understanding the behavior of ZnO in landfill environment warrants further research. This study was designed to investigate the leaching behavior from municipal solid waste (MSW) of nano-ZnO with different types of surface stabilization. Specifically, the objective was to determine whether nano-ZnO tended to leach or stay within the solid waste matrix. Lab-scale batch reactors were set-up and MSW samples obtained from a landfill site were spiked using three different types of ZnO nanoparticles. The leaching behavior of uncoated nano ZnO (nano powder), coated nano ZnO (nano powder) and suspension nano ZnO (referred to as dispersion ZnO) was evaluated within the solid waste-leachate matrix under various experimental conditions by varying nano-ZnO concentrations, pH (acidic and basic) and ionic strength (IS) of the leachate. The results provide further insight to the potential fate of nano-ZnO in landfills during waste stabilization.

2. Materials and methods

2.1. Materials

Approximately 20 kg of mixed, fresh MSW samples were obtained from a sanitary landfill that receives household waste in the city of Izmit, Turkey. Prior to the experiments, the samples were homogeneously mixed, shredded and their size was reduced to 5–10 cm. Representative waste samples were taken for chemical analysis and the samples were then stored at 4 °C in the laboratory until use. The main characteristics of the MSW are presented in Table 1 and Table 2, respectively. For composition determination, the fresh MSW was manually divided into its components, and each component was weighed

Table 1
Characteristics of MSW used for leaching tests.

Parameter	Unit	Value
Total solids (TS)	%	35
Moisture content	%	65
Carbon (C)	%	36
Nitrogen (N)	%	6
Hydrogen (H)	%	16
Ti	mg/kg	1158
Ag	mg/kg	8.6
Si	mg/kg	160
Zn	mg/kg	115
Al	mg/kg	157
Cd	mg/kg	0.3
Co	mg/kg	3.6
Cr	mg/kg	97.4
Cu	mg/kg	76.4
Fe	mg/kg	107.7
Mn	mg/kg	247.7
Mo	mg/kg	2.4
Ni	mg/kg	20.7
Pb	mg/kg	17.2

Table 2
The composition of MSW used for leaching tests.

Component	Unit	Value
Kitchen waste	%	36.21
Paper	%	19.58
Glass	%	4.00
Textile	%	10.53
Plastic	%	15.16
HDPE-PET	%	2.63
Metal	%	3.79
Yard waste	%	1.47
Demolition waste	%	3.79
Inert	%	2.84
Total	%	100

Nano powder ZnO and dispersion ZnO were commercially obtained from Sigma-Aldrich. The average particle sizes provided by the manufacturer for the powder nano ZnO and dispersion nano ZnO were <100 nm and <35 nm, respectively. The coated nano powder ZnO (Z Cote HP1) was obtained from BASF and the average particle size reported by the manufacturer was <200 nm.

2.2. Experimental approach

The batch reactors were filled with 250 g MSW and 300 mL deionized water (DI) as reported previously [29]. The amount of MSW loaded in the batch reactors was on wet basis. The moisture content of the MSW was estimated to be about 40% (100 g of the 250 g of MSW) resulting in an initial dry MSW/liquid ratio of 0.38 in the batch reactors. The same initial MSW/liquid ratio was used for all of the experiments. As aqueous samples were collected from the system, the amount of moisture in the reactors decreased as the experiments progressed in time. No additional moisture was added to replace the collected aqueous phase samples in order not to dilute the system. No solid waste was removed from the system during this sampling procedure. However, the decrease in volume of moisture was accounted for in all mass balance calculations and in the developed particle attachment/detachment model. For each experiment, the reactors were spiked with known amounts of nano-ZnO suspensions. The selected concentrations were 0 (control reactor, without nano-ZnO addition), 25 and 100 mg/L nano-ZnO, respectively. 100 mg/L nano-ZnO stock solution was freshly prepared for each set of experiments by adding the nano-ZnO to