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Original Article

Effects of nickel addition during different phases of solid waste decomposition

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Abstract

This study examined the effects of nickel on solid waste decomposition as well as its soluble concentrations during different times of addition. Three landfill reactors, loaded with 20 kg of waste, were used. Leachate recirculation was applied to all reactors, starting from Day 148. Nickel in the form of $NiCl_2$ was added to the acid and methane reactors on Day 152 and 181, respectively. Leachate and gas parameters from all reactors exhibited similar trends during the first 151 days. Cumulative biogas production was356, 269, and 305 L for the control, acid, and methane reactors, respectively, indicating inhibition in the last two. Residence time of nickel in the acid reactor was more than two times that in the methane reactor (179 vs. 71 hours). Shorter residence time of soluble nickel during the methane phase addition may be responsible for better waste decomposition and could be beneficial for field application.

Keywords: nickel, solid waste, decomposition, inhibition, landfill

1. Introduction

Nickel (Ni) is the 24th most abundant element in the Earth's crust, exhibiting an approximate concentration of 0.008% (Kerfoot, 2005).Ni as well as its compounds and alloys have been utilized in numerous industrial and commercial applications such as machineries, precision electronics, and energy storage materials (Marafi & Stanislaus, 2008; Perosa & Tundo, 2005; Younkin *et al.*, 2000). During the past twenty years, huge amounts of discarded nickel compounds has been found in rechargeable batteries, mainly in the forms of Ni–cadmium (Ni–Cd) and Ni metal hydride (Ni–MH) (Shukla *et al.*, 2001). Mean Ni content in Ni–Cd batteries was 16.4% (Huang *et al*., 2009) and between 25 and 49% in Ni–MH batteries (Scott, 2009). Exposure to highly Ni-polluted environments can induce a variety of pathological effects in humans, varying from contact dermatitis to lung fibrosis, cardiovascular and kidney diseases, and even cancer (Denkhaus & Salnikow, 2002; Kasprzak *et al*., 2003). However, trace concentrations of nickel were reported to be beneficial for anaerobic digestion process since it was required for the growth of all methanogens and the synthesis of cofactor F_{430}.(Choong *et al*., 2016; Geeta *et al.*, 1990)

To date, landfills and open dumps are the most commonly practiced method for solid waste disposal in Thailand (Chinda *et al*., 2012; Pollution Control Department [PCD], 2015). This is primarily due to its ability to be designed, constructed and operated at a lower cost than other alternatives. Unfortunately, all landfills generate leachate that

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is known to be a potential source of groundwater contamination (Akinbile & Yusoff, 2011; Han *et al*., 2016; Park *et al*., 2016). Such situation could be even more aggravated if solid waste disposed in landfill cells contains heavy metals. In 2015, Thai people discarded approximately 206,894 tons of batteries, lamps, and chemical containers (PCD, 2015). If not properly managed, these wastes would end up in landfills and undergo complex transformation processes that may eventually cause leaching of hazardous constituents to receiving groundwater bodies.

Decomposition of organic solid waste in municipal landfills can be normally characterized by five sequential phases namely; initial adjustment, transition, acid formation, methane fermentation, and maturation. Among all five phases, the ones that may have direct influence on metal mobility in landfills are the acid formation and the methane fermentation. During the acid formation phase, the system pH can be as low as 4.5 due to high volatile fatty acid content (Asian Institute of Technology and Tongji University, 2004). Low pH environment enhances solubilities of most metal species in leachate. Soluble heavy metals in leachate can either be stimulatory or inhibitory to organic decomposition processes in landfills. This is dependent on several factors like total metal concentrations, chemical forms of metals and other environmental-related factors such as pH and redox potential (Chen *et al*., 2008). In the methane fermentation phase, methanogens consume intermediate acids produced and convert to methane and carbondioxide. Sulfate and nitrate are reduced to sulfide and nitrogen gas, respectively. The leachate pH values then rise. It should be noted that, in the presence of sulfides, most heavy metals, except chromium, form extremely insoluble sulfide salts and therefore were stripped out from the liquid phase (Fairweather & Barlaz, 1998).

Operational practices of landfill, especially with leachate recirculation, can inevitably affect leachate quality and rate of water decomposition since a landfill consists of cells with different ages (Asian Institute of Technology and Tongji University, 2004; Tchobanoglous *et al*., 1993). New or young cells may be in the acid formation while old cells are most likely in the methane fermentation stage. Young landfill leachate in most municipal solid waste (MSW) facilities have been found to be acidic and possibly contain high content of heavy metals. This does not only threaten environmental quality but also demand expensive and complex systems for leachate treatment. Proper landfill operation and practice could alleviate this problem. Unfortunately, understandings of the behavior of heavy metals such as nickel added to cells with different decomposition stages as well as their impacts on landfill parameters for Thailand's solid wastes are still lacking. The focus of this study was to examine the effect of nickel on decomposition of organic solid waste as indicated by leachate and gas parameters, as well as decomposition effects to the nickel mobility at different phases of landfill.

2. Materials and Methods

2.1 Reactor configuration

Three landfill reactors were constructed using PVC material as shown in Figure 1.

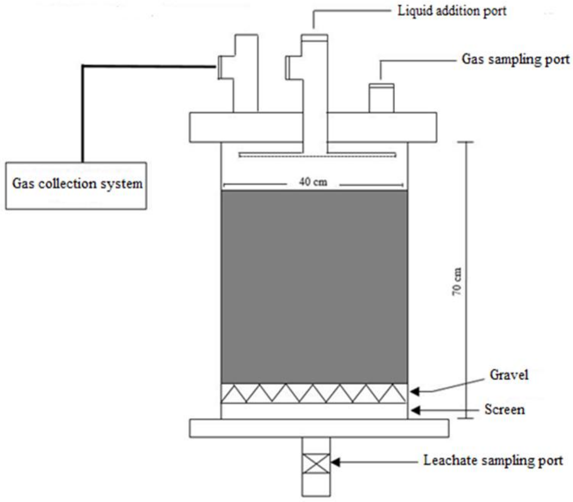

Figure 1. Landfill reactor configuration.

Each reactor had a diameter of 40cm and a height of 70 cm. The total volume of each reactor was 87.9 liters. Columns were assembled with two 50-cm PVC flanges at both ends to provide support for top and bottom lids. Reactors were equipped with four inlet/outlet ports; one at the bottom for leachate sampling and collection and three at the top for gas collection and liquid addition.

2.2 Reactor loading

The reactors were loaded with 20 kg of shredded solid waste. The solid waste mixture consisted of 88.5% vegetable and 11.5% fruit by weight and represented a typical composition of solid waste from a wholesale market at Pathumthani province (Table 1).

The market was a major contributor to a nearby landfill. One liter of anaerobic digested sludge from a Bangkok wastewater treatment plant was added as an innoculum and mixed with the waste for all three reactors. Characteristics of sludge used were as follows: Total solids (TS) = 1,000 mg/L, Volatile solids (VS) = 126 mg/L, pH = 7.73, and alkalinity = 1,550 mg/L.

Table 1. Composition of solid waste used in this study.

Type	Wet weight (kg)	Percent by weight (%)
Eggplant	6.3	31.5
Chinese white cabbage	2.5	12.5
Cow pea	2.5	12.5
Morning glory	2.3	11.5
Kale	2.0	10.0
Water mimosa	0.6	3.0
Cabbage	0.6	3.0
Bitter cucumber	0.5	2.5
Chinese cabbage	0.4	2.0
Banana	1.0	5.0
Orange	1.3	6.5
Total	20	100

2.3 Reactor operation

Preliminary analysis indicated that initial moisture content of the solid waste and sludge mixture was 90% by weight. After reactor loading, liquid collected at the bottom of all reactors was recycled back until field capacity was reached and the date was marked as Day 0. There was no significant moisture addition to all reactors from Day 1to Day 147 to ensure natural progress through decomposition phases of organic waste. Average leachate recirculation rate of 2 liters/day was applied to all three reactors from Day 148 to Day 195, which was the last day of the experiment.

2.4 Nickel addition

Reactors 1, 2, and 3 were designated as control, acid, and methane reactors, respectively. All three reactors were treated similarly from Day 1 to Day 151. Then, introduction of nickel solution to the acid reactor was performed. Approximately two grams of $NiCl_2$in 1,500 mL solution were introduced to Reactor 2 on Day 152 to see the effect of inclusion during the acid formation phase. On Day 181, addition of the same amount of nickel to Reactor 3 was performed to see the effect on the methane fermentation phase. The onset of the methane phase was indicated by methane percentage of approximate 40% or more (Pohland *et. al*., 1987). Residence time of nickel in the bioreactors was computed according to (Pohland & Rachdawong, 1996) as following

$$T_R=\sum M_iT_i/\sum M_i$$

Where T_R is the retention time (in hours), M_i is the mass of nickel in leachate sample, (in grams), and T_i is the time when the nickel mass M_i came out (in hours).

2.5 Sampling and analytical procedure

Leachate samples were collected from the bottom of all reactors and were analyzed for chemical oxygen demand (COD), oxidation reduction potential (ORP), pH, sulfide and nickel concentrations. All parameters, except Ni, were measured according the procedures outlined in the Standard Methods for the Examination of Water and Wastewater (APHA, 1999). Leachate samples containing nickel were collected every hour after addition, microwave-digested, and determined by inductive coupled plasma (ICP). Data on daily temperature, daily gas production rate, and gas composition were also recorded. Methane composition in biogas, measured in percent by volume, was determined by a gas chromatography (Agilent Technology Model 6890 with total conductivity detector).Determination of ORP and methane percentage was started on Day 99 and Day 148, respectively, due to technical problems.

3. Results and Discussion

3.1 Leachate parameters

3.1.1 Chemical oxygen demand

Chemical oxygen demand values in Figure 2 indicate organic strength of the leachate generated throughout the phases of solid waste decomposition. At the beginning of the experiment, initial COD concentrations were 36,134, 44,800, and 37,466 mg/L for the control, acid, and methane reactors, respectively. Until Day 55, COD values from all reactors fluctuated between 36,800 to 63,035 mg/L. Due to insufficient water content at top section, all reactors experienced decreasing COD concentrations from Day 63 to Day 147.After recirculation began on Day 148, leaching caused COD values for all reactors to increase. However, COD concentrations from the acid reactor declined to the range of 26,342 to 29,792 mg/L during Day 155 to Day 168 and may be from $NiCl_2$ addition. The COD trend was found to decrease towards the end of the experiment for all reactors due to active conversion of COD to methane.

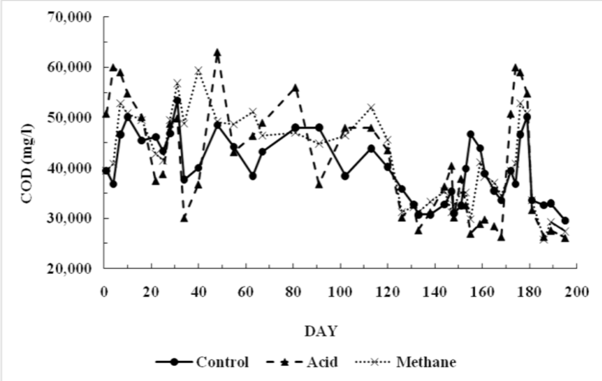

Figure 2. Chemical oxygen demand of leachate samples.

3.1.2 pH

From Figure 3, pH values for the control, acid, and methane reactors showed an increasing trend from 3.88 on Day 1 to 5.24 on Day 20. From Day 21 to Day 158, all three bioreactors pH values were found to vary within a narrow band of 5.0 to 5.35, exhibiting the characteristics of the acid formation phase (Tchobanoglous *et al*., 1993). Effects of leachate recirculation was seen from Day 159 to Day 195 since the pH values had sharply increased from 5.31 to 7.1 and were an indication of active volatile acids conversion to methane gas during the methane fermentation phase.

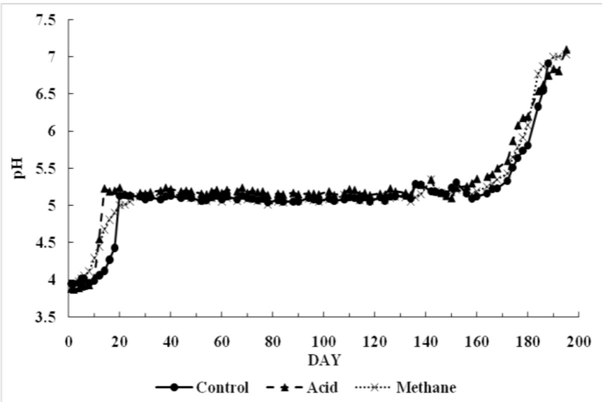

Figure 3. pH values of leachate samples.

3.1.3 Oxidation reduction potential

Due to a technical problem, measurement of ORP values was not commenced until Day 99. From Day 100 to Day 147, the oxidation reduction potential for all reactors was in the range of -79 to -170 as shown in Figure 4.After recirculation began on Day 148, decreasing trend was observed for a few days for the three reactors. Then, the ORP values of leachate from the acid reactor rose and remained in the range of -128 to -180 from Day 159 to Day 185.This was probably an effect of nickel addition on Day 152.Addition of nickel solution to the methane reactor on Day 181 did not produce the same response since the leachate ORP values remained almost the same in the range of -207 to -340 during the last period of the study.

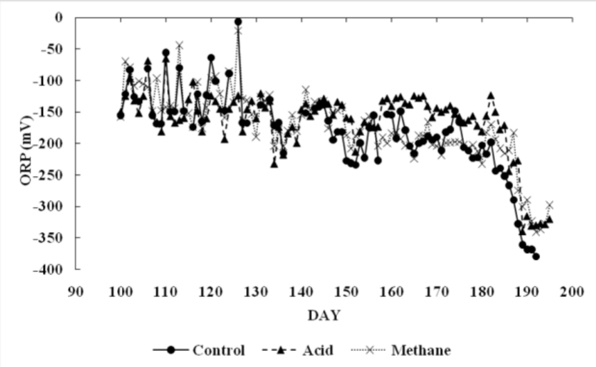

Figure 4. ORP values of leachate samples.

3.1.4 Sulfide

Soluble sulfide in leachate from the acid and methane reactors was determined for the entire period of the study as shown in Figure 5.During the first 120 days, sulfide in both reactors expressed a slow declining trend. Addition of nickel on Day 152 to the acid reactor did not reduce soluble sulfide concentration significantly since the system pH was still acidic as seen in Figure3.Most sulfide species at the acidic pH range were in the form of HS^- or H_2S that cannot form insoluble nickel sulfide precipitates (Chen *et al*., 2008). On the other hand, inclusion of nickel chloride to the methane reactor on Day 181 resulted in the reduction of sulfide level to about 0.3 mg/L on Day 188.

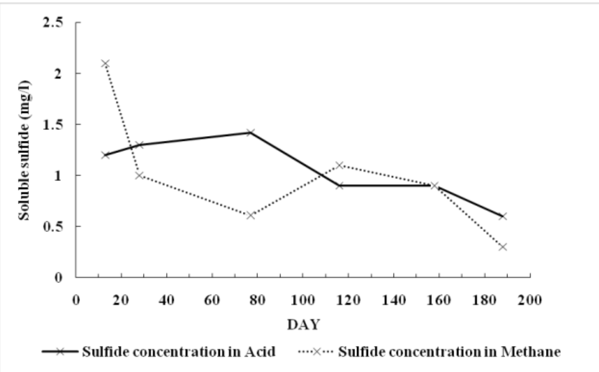

Figure 5. Soluble sulfide concentrations of leachate samples.

3.2 Gas parameters

3.2.1 Cumulative gas

Cumulative biogas production could be used as an indicator for the degree of organic waste stabilization. Slopes of cumulative biogas production were quite steep for the first 27 days for all reactors as in Figure 6. This could be inferred to rapid rates of biogas production. From Day 28 to Day 147, the rates of biogas production for all reactors were slow down due to inadequate moisture content in the reactors. After Day 148, cumulative gas production for all three reactors began to differentiate. The control reactor exhibited the highest cumulative gas production that reached 356 L on Day 195. Total biogas production of the methane reactor was 305 L, indicating certain degree of inhibition. However, the gas production rate of the methane reactor was obviously stimulated as seen from Day 181, although the percent methane was reduced (Figure 7).This could be a consequence of$NiCl_2$ addition. Since nickel, in a range of 0.1 to 1 mg/L, has shown stimulatory effect for both biogas production and methane percentage in anaerobic digestion in a few studies (Abdelsalam *et al*., 2016; Choong *et al*., 2016; Facchin *et al*., 2013).Cumulative biogas production for the acid reactor was the lowest, at 269 L, exhibiting the highest degrees of biogas inhibition as compared to the other two.

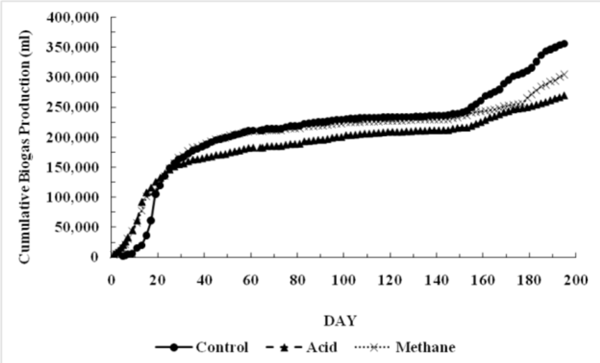

Figure 6. Cumulative biogas production.

3.2.2 Percent methane

Measurement of percent methane in the biogas started on Day 148.During Day 148 to Day 176, percent methane from the three reactors was in the range between 2.2 to 38.3%, an indication of transition from active acid production toward methane fermentation phase (Figure 7). Addition of nickel solution to the acid reactor may be responsible for low methane percentages in the gas (11.5%-39%) during Day 155 to Day 194. Similarly, addition of nickel to the methane reactor on Day 181 has resulted in slight reduction in percent methane from 49.65% on Day 186 to 42.66% on Day 194.

3.3 Residence time of nickel

From Figure 8, nickel concentration from the acid reactor reached its highest point at 85.3 mg/L after 124 hours or 5 days of addition. In contrast, the peak (68.2 mg/L) for the

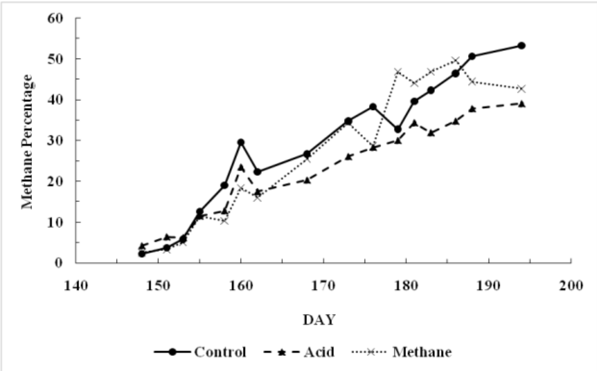

Figure 7. Methane percentage in biogas.

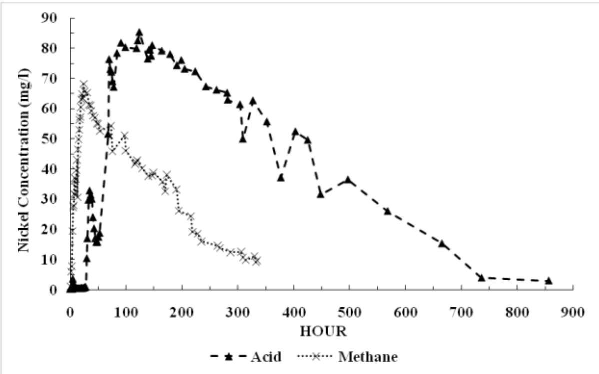

Figure 8. Nickel concentration in leachate.

methane reactor was obtained only after 24 hours or one day of addition. Water distribution in the solid waste mass may play an important role here since the addition of nickel to the acid reactor was done after only four days of leachate recirculation to the system. Another important factor was the low pH values, 5.0 to 5.35, as shown in Figure 3 that enabled nickel, which was added during the acid formation phase, to remain longer soluble (Chen *et al*., 2008). Average residence time of nickel in the acid reactor was more than two times greater than that in the methane reactor (179 vs. 71 hours).Lasting soluble nickel at concentration of greater than 1 mg/L in the system was a cause of inhibition for methanogenesis in both acid and methane reactors (Choong *et al*., 2016; Facchin *et al*., 2013). Sulfide precipitation may be responsible for nickel removal in the methane reactor as indicated bylow soluble sulfide concentrations remained (0.3 mg/L) on Day 188 as observed in Figure 5.

4. Conclusions

Inclusion of high concentration of nickel to solid waste landfill can inhibit organic waste decomposition as indicated by cumulative biogas production of 356, 269, and 305 L for the control, acid, and methane reactors, respectively. Decomposition stages also impacts nickel mobility. Addition of nickel during the acid formation phase of landfill can result in a longer average residence time of soluble nickel (179 vs. 71 hours) for the methane phase reactor. Soluble nickel can be toxic to microbial activities. To solve this problem, nickel may be added during the methane phase since nickel sulfide can be easily formed and precipitated out. This concept may lead to a field application. For example, the methane phase cell could act as a supporting and reactive layer for removal of heavy metals in young leachate produced in other cells of a landfill.

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