Waste Management xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

journal homepage: www.elsevier.com/locate/wasman

Assessment of methane production from shredder waste in landfills: The influence of temperature, moisture and metals

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ARTICLE INFO

Article history:
Received 23 August 2016
Revised 14 November 2016
Accepted 16 November 2016
Available online xxxx

Keywords:
End-of-life vehicles
Anaerobic digestion
Biogenic carbon
Biochemical methane potential (BMP)
Biocorrosion
First-order decay kinetic constant

ABSTRACT

In this study, methane (CH_4) production rates from shredder waste (SW) were determined by incubation of waste samples over a period of 230 days under different operating conditions, and first-order decay kinetic constants (k-values) were calculated. SW and sterilized SW were incubated under different temperatures (20–25 °C, 37 °C, and 55 °C), moisture contents (35% and 75% w/w) and amounts of inoculum (5% and 30% of the samples wet weight). The biochemical methane potential (BMP) from different types of SW (fresh, old and sieved) was determined and compared. The ability of metals (iron, aluminum, zinc, and copper) contained in SW to provide electrons for methanogens resulting in gas compositions with high CH_4 contents and very low CO_2 contents was investigated. The BMP of SW was 1.5–6.2 kg CH_4/ton waste. The highest BMP was observed in fresh SW samples, while the lowest was observed in sieved samples (fine fraction of SW). Abiotic production of CH_4 was not observed in laboratory incubations. The biotic experiments showed that when the moisture content was 35% w/w and the temperature was 20–25 °C, CH_4 production was extremely low. Increasing the temperature from 20–25 °C to 37 °C resulted in significantly higher CH_4 production while increasing the temperature from 37 °C to 55 °C resulted in higher CH_4 production, but to a lower extent. Increasing the moisture and inoculum content also increased CH_4 production. The k-values were 0.033–0.075 yr^{-1} at room temperature, 0.220–0.429 yr^{-1} at 37 °C and 0.235–0.488 yr^{-1} at 55 °C, indicating that higher temperatures resulted in higher k-values. It was observed that H_2 can be produced by biocorrosion of iron, aluminum, and zinc and it was shown that produced H_2 can be utilized by hydrogenotrophic methanogens to convert CO_2 to CH_4. Addition of iron and copper to SW resulted in inhibition of CH_4 production, while addition of aluminum and zinc resulted in higher CH_4 production. This suggested that aluminum and zinc contribute to high CH_4 production from SW by providing H_2 for hydrogenotrophic methanogens. Gas compositions with higher CH_4 and lower CO_2 observed in landfilled SW are thus most likely due to the consumption of existing CO_2 in the produced biogas and the produced H_2 by biocorrosion of aluminum and zinc by methanogens.

1. Introduction

An end-of-life vehicle (ELV) is a vehicle that is discarded by its owner as waste. Together with other metal-containing waste products, including white goods, ELVs are collected, dismantled and shredded by authorized shredding companies. The ferrous and non-ferrous metals are then recovered. The residual fraction after recovery of metals and dismantled parts is called shredder waste (SW). SW consists of mainly plastic, metals, rubber, textile, foam, glass and wood, and it constitutes approximately 20–25% of an ELV's weight (Ahmed et al., 2014; Fiore et al., 2012; Morselli et al., 2010).

According to EU-Directive 2000/53/EC, a maximum 10% of an ELV's weight may be incinerated, and a maximum of 5 wt% may be landfilled. However, the majority of SW is landfilled in most countries, including Denmark (Ahmed et al., 2014; Fiore et al., 2012). By deposition of SW in landfills, the biodegradable fractions produce landfill gas (LFG), which consists of methane (CH_4) and carbon dioxide (CO_2). Landfills are one of the main anthropogenic sources of CH_4 emission to the atmosphere. CH_4 is 28 times more powerful than CO_2 in terms of global warming potential (IPCC, 2013).

According to the European Pollutant Release and Transfer Register (E-PRTR), landfills—excluding landfills of inert waste—receiving

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http://dx.doi.org/10.1016/j.wasman.2016.11.023
0956-053X/

more than 10 tons of waste per day or with a total disposal capacity of 25,000 tons are required to report their CH_4 emission (CEC, 2006). The reporting of CH_4 emissions from landfills is based on modeling of CH_4 generation in most countries, including Denmark. These models are based on the first-order decay (FOD) of organic matter, as shown by Eq. (1):

$$m_t = m_0 \times e^{-kt} \quad (1)$$

where m_t is the mass of organic carbon (g) after time t, m_0 is the mass of organic carbon (g) at t = 0, t is the degradation time (yr) and k is the FOD kinetic constant (yr^{-1}). The k-value (FOD kinetic constant) and biochemical methane potential (BMP) are two important parameters for estimation of the CH_4 generation by FOD models (Mou et al., 2015). Currently, knowledge of k-values and the BMP of SW is limited. Thus there is a need for more research about these parameters.

Although SW has passed through metal-separation technologies, the efficiency of separation is not 100% and it still contains metals. The metal content of SW depends on the recovery technologies used and the initial amount of metals in the feed material. However, it is evident that due to an increase in metal scrap prices over time, metal recovery technologies are improving to recover more metals from SW (Ahmed et al., 2014).

Iron (Fe), aluminum (Al), zinc (Zn) and copper (Cu) are the most abundant metals in SW (Ahmed et al., 2014; Cossu et al., 2014; Fiore et al., 2012; Galvagno et al., 2001; Granata et al., 2011). Table 1 provides an overview of the metal content of SW. The fraction of these metals in SW varies significantly in previous studies. For instance, the percentage of Fe varied from 1.78 to 15.40%. However, it can be observed that Fe is always the most abundant metal.

After sieving, the coarse fraction of SW consists of high amounts of metals and combustibles (plastic, rubber, foam, wood and textiles), while the fine fraction consists of more minerals and inert materials (Ahmed et al., 2014). This suggests that sieving SW could be an option for higher material and energy recovery from the coarse fraction, in order to reach the defined goal of minimum 95% reuse and recovery of the ELV's weight by the EU-Directive 2000/53/EC. If shredding companies apply sieving SW, the fine fraction will be landfilled, which could change the LFG production from SW significantly. Thus it would be interesting to know more about gas generation from the fine fraction of SW.

Generally, SW consists of about 20–30 wt% of plastic, 15–20 wt % of rubber, 20–40 wt% of paper and wood, and 10 wt% of inert materials (e.g. glass, and soil) and metals (Fiore et al., 2012). This indicates that SW contains low fractions of biodegradable waste and that the biodegradable fractions have a high content of lignocellulosic components such as paper and wood. However, relatively high production of CH_4 from SW (0.020–0.031 g CH_4 h^{-1} ton^{-1} of SW) has been observed in previous studies (Mønster et al., 2015; Scheutz et al., 2011). The reason for these relatively high CH_4 production rates from SW is unknown.

Conventional LFG consists of 55–60% v/v of CH_4 and 40–45% v/v of CO_2. However, previous studies have shown gas compositions in SW monofills, which differed from conventional landfill gas—having a high CH_4 content and very low or no CO_2 (Olsen and Willumsen, 2013; Scheutz et al., 2011). Moreover, high temperatures were reported inside SW monofills; 59 and 40 °C at 10 and 20 m depths, respectively (Olsen and Willumsen, 2013). The reason for the unusual gas composition and high temperatures are unknown. However, it is known that no aerobic reaction was taking place as there was no O_2 present in the sampled gas.

During anaerobic digestion, a part of the organic material is converted to CO_2 initially by fermentation and acetogenesis. Part of this generated CO_2 can be converted to CH_4 by hydrogenotrophic methanogens converting CO_2, using H_2 as the electron source as shown in Eq. (2) (Wise et al., 1978).

$$4H_2 + CO_2 \rightarrow CH_4 + 2H_2O \quad (2)$$

However, only around 30% of the produced CH_4 is derived from this pathway due to limited H_2 supply (Gujer and Zehnder, 1983). Thus, theoretically it should be possible to increase CH_4 production by supplying H_2. A number of previous studies have investigated this approach in practice (Kim et al., 2013; Luo and Angelidaki, 2012; Luo et al., 2012). It was observed that supplying H_2 increased the CH_4 yield. H_2 can be produced by corrosion of the metals present in SW. For instance, H_2 production from Fe can be seen in Eqs. (3)–(5) (Lorowitz et al., 1992).

$$Fe^0 \rightarrow Fe^{2+} + 2e^- \quad (3)$$

$$2H_2O \rightarrow 2H^+ + 2OH^- \quad (4)$$

$$Fe^0 + 2H_20 \rightarrow Fe(OH)_2 + H_2 \quad (5)$$

The corrosion rate decreases as the corrosion products adhere to the surface of the metal, forming a protective layer. Microbial activity within the formed layer can influence the kinetics of the reactions. Accelerated deterioration of metals due to microbial activity is called biocorrosion or microbially influenced corrosion. In biocorrosion processes, the protective layer of H_2 formed on the surface of the metal can be used by biological reactions, such as hydrogenotrophic methanogenesis, and corrosion is accelerated (Beech and Gaylarde, 1999; Beech and Sunner, 2004; Lorowitz et al., 1992).

We hypothesized that high CH_4 production from SW and unusual gas composition could be due to H_2 production by biocorrosion of metals in the waste, supporting hydrogenotrophic methanogens to convert CO_2 to CH_4, resulting in higher CH_4 content and lower CO_2. The hypothesis is supported by microbial studies showing that biocorrosion of metals resulted in production of H_2 and enhancement of CH_4 yields by utilizing the produced H_2.

The objective of this study was to determine CH_4 production rates from SW with a focus on investigating the ability of metals contained in SW to provide electrons for methanogens. Moreover, the BMP of different types of SW (i.e., fresh, old and sieved SW) was determined, and the impact of the age of the sample on the determined BMP was discussed. Finally, the impact of temperature, moisture and inoculum addition on biogas production from SW was evaluated and k-values at different operating conditions were calculated and compared to the literature.

Table 1
Overview of the content (%) of Fe, Al, Zn and Cu in SW. The content is given per wet weight basis.

Reference	Fe (%)	Al (%)	Zn (%)	Cu (%)
Ahmed et al. (2014)	12.55–15.40	2.24–2.45	0.99–1.46	0.51–2.32
Fiore et al. (2012)	1.78	0.38	0.53	0.37
Granata et al. (2011)	9.30	1.50	1.10	1.50
Galvagno et al. (2001)	2.36–2.70	0.62–4.88	0.42–0.66	0.72–2.18

Please cite this article in press as: Fathi Aghdam, E., et al. Assessment of methane production from shredder waste in landfills: The influence of temperature, moisture and metals. Waste Management (2016), http://dx.doi.org/10.1016/j.wasman.2016.11.023