Waste Management xxx (2016) xxx–xxx

Contents lists available at ScienceDirect

Waste Management

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

Experimental evaluation of two different types of reactors for CO_2 removal from gaseous stream by bottom ash accelerated carbonation

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ARTICLE INFO

Article history:
Received 1 May 2016
Revised 26 September 2016
Accepted 26 September 2016
Available online xxxx

Keywords:
Landfill gas
Carbon dioxide
Accelerated carbonation
Alkaline residues
Bottom ash
Fixed bed reactor
Rotating drum reactor

ABSTRACT

Low methane content landfill gas may be enriched by removing carbon dioxide. An innovative process, based on carbon dioxide capture and storage by means of accelerated carbonation of bottom ash is proposed and studied for the above purpose. Within this research framework we devoted a preliminary research activity to investigate the possibility of improving the way the contact between bottom ash and landfill gas takes place: this is the scope of the work reported in this paper. Two different types of reactors – fixed bed and rotating drum – were designed and constructed for this purpose. The process was investigated at laboratory scale. As the aim of this phase was the comparison of the performances of the two different reactors, we used a pure stream of CO_2 to preliminarily evaluate the reactor behaviors in the most favorable condition for the process (i.e. maximum CO_2 partial pressure at ambient condition). With respect to the simple fixed bed reactor concept, some modifications were proposed, consisting of separating the ash bed in three layers. With the three layer configuration we would like to reduce the possibility for the gas to follow preferential paths through the ash bed. However, the results showed that the process performances are not significantly influenced by the multiple layer arrangement. As an alternative to the fixed bed reactor, the rotating drum concept was selected in order to provide continuous mixing of the solids. Two operating parameters were considered and varied during the tests: the filling ratio and the rotating speed. Better performances were observed for lower filling ratio while the rotating speed showed minor importance. Finally the performances of the two reactors were compared. The rotating drum reactor is able to provide improved carbon dioxide removal with respect to the fixed bed one, especially when the rotating reactor is operated at low filling ratio values and slow rotating speed values. Comparing the carbon dioxide specific removal obtained by using the rotating reactor (35–37 g/kg_{BA}), in the best operating conditions, with that measured for the fixed bed reactor (21–23 g/kg_{BA}), an increase of about 61–66% is observed.

1. Introduction

Anaerobic biodegradation processes of biodegradable waste in landfills generate landfill gas (LFG) which is mainly constituted by methane and carbon dioxide with some other trace gases. A rigorous strategy is defined by the European directives about waste management (2008/98/EC EC, 2008 and 1999/31/EC (EC, 1999) stating that landfill use must be minimized, ensuring that no landfilled waste is biologically active or contains mobile hazardous substances. Even if in sanitary landfills LFG extraction and flaring is compulsory, landfilling of biodegradable waste must be strongly reduced, to contain the methane emissions escaping to the atmosphere. According to this framework, biodegradable waste will be diverted from landfills and a reduction of LFG production and quality is expected in the next future. However, biological degradation in landfills is a very slow process and LFG generation from the already landfilled waste will take several decades to complete. The LFG exploitation for energy recovery should be pursued, since this is a beneficial effect in terms of overall greenhouse gas balance and primary energy saving (Hao et al., 2008; Manfredi et al., 2009; Lombardi et al., 2006).

LFG has a typical composition with 50–65% in volume of methane (CH_4) and 35–50% in volume of carbon dioxide (CO_2), with a Lower Heating Value (LHV) in the range of 17.5–22.7 MJ/Nm^3. As landfill aging takes place, gas generation decreases and often gas quality deteriorates showing a reduced CH_4 content

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http://dx.doi.org/10.1016/j.wasman.2016.09.038
0956-053X/

and sometimes an intermittent flow. The amount and rate of production of LFG, as well as its CH_4 concentration along with time, may change for a variety of reasons (Browe et al., 2009). Air may enter into a landfill as a result of gas extraction, changes in the relative pressure within the landfill, or deterioration of the extraction infrastructure, diluting the internal gas resulting in a gas mixture containing nitrogen, carbon dioxide, oxygen and CH_4. The oxygen present in the air may be used by aerobic micro-organisms to oxidise CH_4 produced in anaerobic area, resulting in a gas mixture of nitrogen, carbon dioxide and residual CH_4. Additionally, the amount of generated CH_4 decreases as the organic content of the waste is reduced according to the microbial action, resulting in a carbon dioxide-rich gas mixture. The time horizon for such LFG deterioration may change depending on several site-specific factors, such as moisture content and the type of waste.

When the CH_4 content in the LFG is quite high (40–65% in volume), its combustion is relatively straightforward, and the most common devices used for recovering its energy content are reciprocating internal combustion engines (ICEs), for combined heat and power production. ICEs have several positive features as high reliability, low specific cost and high electrical efficiency, and are available in the power range of hundreds of kW to few MW (IPCC, 2007).

On the contrary, when the CH_4 content of the LFG falls below approximately 35–40% in volume, difficulties in sustaining combustion in traditional engines can occur. In these conditions LFG would normally be flared through a high temperature flare. When the CH_4 content falls even further, to approximately 20% in volume, difficulties can occur in sustaining combustion even within the flare. In this case LFG is currently vented untreated into the atmosphere, contributing to global warming with a factor equal to 25 kg of equivalent CO_2 (IPCC, 2007).

Considering low CH_4 and high CO_2 content LFG, it is possible, in principle, to remove at least part of the diluting CO_2, in order to increase the CH_4 content to levels suitable for combustion. Presently, several processes are commercially available for removing carbon dioxide from LFG or biogas and increasing the methane content (Yang et al., 2014; Ryckebosch et al., 2011; Rasi et al., 2014) and they are usually applied for the so called process of up-grading aimed at producing biomethane, in which CH_4 concentration needs to be higher than 95–95% in volume. Biogas or LFG upgrading processes include absorption by chemical solvents, physical absorption, cryogenic separation, membrane separation and CO_2 fixation by biological or chemical methods (Makaruk et al., 2010). The most commonly applied methods are high pressure water scrubbing (HPWS), amine scrubbing, and pressure swing adsorption (PSA) and are generally applied to treat normal quality LFG or biogas with the aim of producing biomethane to substitute natural gas (Browne et al., 2011; Patterson et al., 2011).

Among the several possibilities studied worldwide to remove CO_2, accelerated carbonation has been considered an interesting process for Carbon Capture and Storage (CCS) since a long time (Lackner et al., 1995). Mineral carbonation is based on the natural weathering of rocks containing compounds capable of forming carbonates, such as calcium or magnesium, when they are exposed to CO_2 dissolved in rain water. The natural process is very slow, thus at industrial level appropriate conditions can be applied to promote accelerated carbonation. One possibility is to apply the aqueous carbonation route, precipitating carbonates from aqueous solutions (Lackner et al., 1995; Rasul et al., 2014; Wang and Maroto-Valer, 2013; Yanagisawa, 2001). Alternatively the gas-solid carbonation process can be applied (Fagerlund et al., 2012).

In addition to natural rocks, some industrial residues are suitable for capturing CO_2 through accelerated carbonation, thanks to some appropriate characteristics, i.e. calcium and magnesium oxide contents (Fernández Bertos et al., 2004). In this work the attention is focused on bottom ash (BA), which is the solid residue remaining in the furnace after solid waste incineration, mainly consisting of un-burnable, i.e. inert, materials. BA is generally classified as industrial non-hazardous waste from the European Waste Catalogue. In reference to municipal solid waste (MSW) incineration, the produced BA is about 15–25% in mass of the incinerated waste, depending on its content of inert fractions (Grosso et al., 2011; Birgisdóttir et al., 2007; Karagiannidis et al., 2013). Accelerated carbonation of BA has been investigated in the past mainly as a treatment process for reducing the leaching of some metals (Fernández Bertos et al., 2004; Van Gerven et al., 2005; Arickx et al., 2006; Rendek et al., 2006; Baciocchi et al., 2010; Costa et al., 2007).

As an application of accelerated carbonation of BA for CO_2 capture and storage, the production of biomethane from landfill gas or biogas – i.e. upgrading – was originally proposed (Mostbauer et al., 2008) and further investigated (Mostbauer et al., 2014; del Valle-Zermeño et al., 2015). In our previous work, we tested the process of accelerated carbonation of BA for removing CO_2 from LFG with the aim of producing biomethane on pilot scale, by directly contacting wet BA and LFG in simple fixed bed reactor (Mostbauer et al., 2014).

The readily reactive Ca-oxide phases (e.g. $Ca(OH)_2$), contained in the BA, and gaseous CO_2 dissolve in the liquid phase surrounding the solid particles and then form carbonates that precipitate, as it is summarized in Reaction (1):

$$Ca(OH)_2(s) + CO_2(g) \rightarrow CaCO_3(s) + H_2O(l) \quad (1)$$

CO_2 is thus fixed in solid and stable form of carbonate.

After an initial phase of complete CO_2 removal, the BA ability of capturing and storing the CO_2 depletes. We stop the process when the exiting stream contains the maximum allowable CO_2 concentration that, in the biomethane application case, is about 4–6% in volume. To run the process semi-continuously, the BA must be replaced with a fresh amount. We concluded that such an application requires a rather high BA amount per unit of processed gas volume, suggesting that this type of application is more suitable for small scale upgrading plants, in order to contain the overall annual amount of required BA (Mostbauer et al., 2014).

On this basis, we suggested the possibility of applying accelerated carbonation of BA for capturing CO_2 from low-CH_4 rich-CO_2 LFG, which may be generated in old landfills (Lombardi and Carnevale, 2016). In this case CO_2 capture and storage is aimed at enriching the CH_4 concentration in the LFG, up to values suitable for feeding traditional ICEs, assumed as CH_4 higher than 40% in volume. This process requires a lower BA amount per unit of processed gas volume than the upgrading to biomethane (about 36 vs. 50 kg of BA per Nm^3 of feed LFG) (Lombardi and Carnevale, 2016). Within this research framework, we devoted a preliminary research activity to investigate the possibility of improving the way the contact between BA and LFG takes place, proposing some modifications to the simple fixed bed configuration reactor and proposing the use of a different type of reactor based on a rotating drum.

Thus in this work we report the investigation of the possibility of using two different types of reactors – fixed bed and rotating drum that were designed and constructed for this purpose – to directly contact the gas containing the CO_2 and BA, in order to remove the CO_2 from the gas phase and store it in solid form. The process was investigated at laboratory scale. As the aim of this research phase is the comparison of the performances of the two different reactors, we used a pure stream of CO_2 to evaluate the reactor behaviors in the most favorable condition for the process (i.e. maximum CO_2 partial pressure at ambient condition).

Please cite this article in press as: Lombardi, L., et al. Experimental evaluation of two different types of reactors for CO_2 removal from gaseous stream by bottom ash accelerated carbonation. Waste Management (2016), http://dx.doi.org/10.1016/j.wasman.2016.09.038