



# Analysis of an innovative process for landfill gas quality improvement



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## ABSTRACT

Low methane content landfill gas is not suitable for feeding engines and is generally flared. This type of landfill gas may be enriched by removing the inert carbon dioxide. An innovative process, based on the carbon dioxide captured by means of accelerated carbonation of bottom ash was proposed and studied for the above purpose. The process was investigated at a laboratory scale, simulating different landfill gas compositions. The enrichment process is able to decrease the carbon dioxide concentration from 70 to 80% in volume to 60% in volume, requiring about 36 kg of bottom ash per Nm<sup>3</sup> of landfill gas. Using this result it was estimated that an industrial scale plant, processing 100–1000 Nm<sup>3</sup>/h of low methane content landfill gas requires about 28,760–2,87,600 t of bottom ash for a one year operation. The specific cost of the studied enrichment process was evaluated as well and ranges from 0.052 to 0.241 Euro per Nm<sup>3</sup> of entering landfill gas. The energy balance showed that about 4–6% of the energy entered with the landfill gas is required for carrying out the enrichment, while the use of the enriched landfill gas in the engine producing electricity allows for negative carbon dioxide emission.

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## 1. Introduction

Landfill gas (LFG) is generated from biodegradable waste anaerobic degradation in landfills and its main portion is comprised of methane and carbon dioxide with some trace gases. As a matter of fact, European directives about waste management and landfills (2008/98/EC [1] and 1999/31/EC [2]) defined a rigorous strategy in which the landfill has a residual role in the waste management systems. Only pre-treated waste and waste from which it is not possible to recover material or energy can be landfilled. In particular biodegradable waste landfilling must be strongly reduced, being this type of waste responsible for methane emissions, contained in the escaped LFG to atmosphere. In this view biodegradable waste will be diverted from landfills and a reduction of LFG production and quality is expected for future years. However, the biological degradation of the already landfilled waste will take several decades to complete and it will be worth it to exploit the produced LFG for energy recovery, since this is a beneficial effect in terms of overall greenhouse gas balance and primary energy saving [3–5].

The typical composition of LFG is 50–65% in volume methane

and 35–50% in volume carbon dioxide, with a Low Heating Value (LHV) in the range of 17.5–22.7 MJ/Nm<sup>3</sup>. As a landfill ages, gas generation deteriorates, producing gas with reduced methane content and sometimes with an intermittent flow. The amount and rate of production of LFG, as well as its methane concentration along with time, may change for a variety of reasons, including [6]: i) dilution with air that 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, resulting in a gas mixture containing nitrogen, carbon dioxide, oxygen and methane; ii) dilution with nitrogen from air; the oxygen present in the entered air may be used by aerobic micro-organisms to oxidise methane produced in an anaerobic area, resulting in a gas mixture of nitrogen, carbon dioxide and residual methane; iii) the amount of generated methane 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 types of waste in the landfill.

LFG with a high methane content (40–65% in volume) is relatively straightforward to burn, and the most common way for recovering energy from LFG is combustion in reciprocating internal combustion engines (ICEs) for combined heat and power production. ICEs are often chosen for their reliability, low specific cost and high electrical efficiency, especially in the power range of hundreds

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of kW to few MW [7].

However, once the methane content of the LFG falls below approximately 35–40% in volume difficulties sustaining combustion in engines can occur. At this point LFG would normally be flared through a high temperature flare. When the methane 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<sub>2</sub> [7].

As a matter of fact several processes are commercially available for removing the carbon dioxide from LFG or biogas and increasing the methane content [8–10]. These include absorption by chemical solvents, physical absorption, cryogenic separation, membrane separation and CO<sub>2</sub> fixation by biological or chemical methods [11]. 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 [12,13]. In this frame, Tippayawong and Thanompongchart [14] investigated the possibility of chemically absorbing CO<sub>2</sub> and H<sub>2</sub>S by aqueous solutions of sodium hydroxide, calcium hydroxide and mono-ethanolamine. Montanari et al. [15] investigated the use of 4 A and 13X zeolite adsorbents. However, in all these types of processes, the CO<sub>2</sub> removed from the biogas, being of biogenic origin, is successively vented to atmosphere.

More in general, several Carbon Capture and Storage (CCS) technologies have been proposed and investigated over the last decades in connection with their application to large point emissions sources, namely thermal power plants. Existing capture technologies include physical and chemical absorption, in this last case the most commonly used solvents are aqueous solutions of alkanolamines; adsorption, through PSA, Temperature Swing adsorption (TSA) and Electrical Swing adsorption (ESA); cryogenic separation; membrane technologies diversified according to gas separation and gas absorption membranes [16,17]. Emerging technologies include enzyme based separation; facilitated transport membrane; hydrate based separation [18–20]; mixed matrix membrane; calcium looping CO<sub>2</sub> capture [21].

Additionally, accelerated carbonation of natural materials has been considered an interesting process for CCS since a long time [22].

The basic principle behind mineral carbonation is natural weathering of rocks containing compounds capable of forming carbonates, such as calcium or magnesium, when they are exposed to CO<sub>2</sub> dissolved in rain water. Metal ions are released into the mildly acidic water and may react with the dissolved CO<sub>2</sub>, forming mineral carbonates. Natural weathering is a very slow process, but the reaction can be promoted – at industrial level – by using appropriate conditions realizing the accelerated carbonation. Several studies are concerned with the aqueous carbonation route, precipitating carbonates from aqueous solutions [22–25]. Other investigations chose the gas-solid carbonation process that aims at utilising the heat released from the exothermic carbonation reaction at elevated temperatures [26].

Beside natural rocks, some industrial residues have appropriate characteristics – i.e. calcium or magnesium contents – to be suitable for capturing CO<sub>2</sub> through accelerated carbonation [27,28]. Among these, we focused the attention on bottom ash (BA), which is the solid residue remaining in the furnace after, mainly consisting of un-burnable, i.e. inert, materials. BA is generally classified as industrial non-hazardous waste from the European Waste Catalogue. Talking about municipal solid waste (MSW) incineration, the produced BA is about 15–25% in mass of the incinerated waste, according to its content of inert [29–31]. Accelerated carbonation

of BA has been investigated in the past mainly as a treatment process for reducing the leaching of some metals [27,32–36].

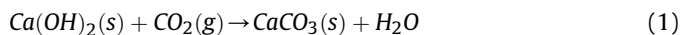
As an application of accelerated carbonation of BA for capturing CO<sub>2</sub>, the production of biomethane from landfill gas or biogas – i.e. upgrading – was originally proposed [37] and further investigated as gas-solid ambient condition process [38,39]. Also the use of steel slag was proposed as a low-cost solution to produce biofuel from biogas at farm scale level [40]; or the use of air pollution control residues to produce biomethane from landfill gas [41].

After testing the process of accelerated carbonation of BA for capturing CO<sub>2</sub> on pilot scale [38], we concluded that the upgrading of biogas or landfill gas has a rather high BA specific requirement per unit of processed gas volume, thus the process appeared more suitable for small scale upgrading plants, in order to contain the overall annual amount of required BA.

On the basis of this previous experience, we looked at the possibility of applying the gas-solid carbonation of BA for capturing CO<sub>2</sub> for a slightly different type of application, possibly with lower BA specific requirement.

In particular, in this work, we investigate the possibility of applying the gas-solid carbonation of BA for capturing CO<sub>2</sub> from LFG characterized by high CO<sub>2</sub> and low CH<sub>4</sub> contents, as it may be generated in old landfills, by the processes previously described. In this case the CO<sub>2</sub> capture is aimed at enriching the CH<sub>4</sub> concentration in the LFG, up to values suitable for feeding ICEs, assumed here as CH<sub>4</sub> higher than 40% in volume and CO<sub>2</sub> lower than 60% in volume.

We apply for this type of process, a direct gas/solid contact method, by flowing the CO<sub>2</sub>-rich gas through the BA fixed bed. CO<sub>2</sub> is quickly removed by direct carbonation of readily reactive Ca-oxide phases (e.g. Ca(OH)<sub>2</sub>) contained in the BA, as it is summarized in the reaction 1:



CO<sub>2</sub> is thus fixed in solid and stable form of carbonate.

The investigated process has some innovative features, which are mainly: using a waste stream (BA) as material to capture CO<sub>2</sub>, in place of natural rocks; using a simple gas/solid fixed bed contact reactor at ambient conditions. Additionally, to the best of the authors' knowledge, the application of CO<sub>2</sub> capture from LFG characterized by high CO<sub>2</sub> and low CH<sub>4</sub> contents was not investigated before up to now.

Moreover, some additional environmental benefits targeted by this process are: permanently capture and store from the LFG the CO<sub>2</sub>, hence the process acts as a CO<sub>2</sub> sink, since the stored CO<sub>2</sub> is of biogenic origin; improving the BA leaching behaviour with respect to some metals [27,32,34–36]. The use of a type of industrial residues – in this case BA – as input material to another industrial process – in this case LFG enrichment – fits the industrial ecology approach and the industrial symbiosis concepts [42].

The enrichment process was first investigated at laboratory scale by setting up a facility for realizing the direct contact between the BA and the simulated LFG in a fixed bed reactor. The results obtained from the experimental phase were used for evaluating the process from the economic, energy and environmental points of view.

## 2. Materials and methods

### 2.1. Experimental setup

The experimental facility mainly consists of the BA fixed bed reactor and the measuring systems.

The BA fixed bed reactor is a 27 l stainless steel cylindrical tank

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