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Biogas purification with biomass ash

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ABSTRACT

The aim of the study was to investigate the option to purify biogas from small-scale biogas plants by entrapping CO₂ and H₂S with regionally available biomass ash. Connected to the existing biogas plant Neustift (Tyrol) wood ash placed in a 1 m³ container was used as a trap for CO₂ and H₂S in the biogas. With the process conditions chosen, for a period of a few hours CO₂ was trapped resulting in pure methane. The removal of H₂S was much longer-lasting (up to 34 d). The cumulative H₂S uptake by the biomass ash ranged from 0.56 to 1.25 kg H₂S per ton of ash. The pH of the ash and the leachability of Lead and Barium were reduced by the flushing with biogas, however toxicity towards plants was increased thus reducing the potential of ash use in agriculture. It can be concluded that biomass ash may be used for removal of hydrogen sulphide from biogas in small and medium biogas plants. The economic evaluation, however, indicated that the application of this system is limited by transport distances for the ash and its potential use afterwards.

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1. Introduction

The upgrading of gas from landfills by using incineration residues is a technique that is being employed to eliminate undesirable compounds such as sulfur compounds, siloxanes, aromatic compounds and halogenated volatile organic chlorinated compounds which are present in the gas (Chavez and Guadarrama, 2015). Already in 2007 it was proposed that municipal solid waste incineration ash (MSWI bottom ash) could be used for removal of CO₂ and H₂S from landfill gas (LFG) or biogas. Several experiments at the technical scale demonstrated that ash-based enrichment of CH₄ in biogas and lean landfill gas (Mostbauer and Lenz, 2007) is technically feasible. Recently Chavez and Guadarrama (2015) concluded that wood and dry coal fly ash from power plants are more appropriate to reduce the carbon dioxide in LFG due the higher calcium content than ashes from conventional and coupled kilns. The results have indicated several ecological benefits of the ash-based removal process, particularly a positive CO₂ balance and a significant reduction of leachability of ash (Mostbauer et al., 2014; Mostbauer et al., 2012; Starr et al., 2012). However, the focus is often put on the removal of CO₂ and an enrichment of CH₄, and less attention has been paid to the reaction of H₂S with ash constituents. Removal of H₂S from biogas is of utmost importance

since H₂S may induce corrosion in power systems through production of sulfur oxides when gas is combusted, furthermore H₂S is also known to affect the efficiency more innovative technologies such as solid oxide fuel cell (SOFC) systems (Papurello et al., 2015).

Many industrial by-products and incineration residues contain manganese, zinc, copper and iron compounds that may form sulfides of low solubility. Some of them also contain sulfites, a chemical species which also reacts with H₂S at ambient temperature. Alkaline or pH-neutral waste containing one of these reagents, either sulfites or much Fe-II (or Fe-III, Mn-II, Cu-I, Cu-II, Zn-II) or even both of these chemical species can be regarded as a potential H₂S-trap material. Removal of H₂S from biogas was attributed to the reaction of iron oxides with H₂S when steel slag is used for purification of biogas in a fixed bed reactor (Asaoka et al., 2013; Sarperi et al., 2014a, 2014b). Iron sulfidation, the formation of FeS and Fe₂S₃ was suggested by Sarperi et al. (2014b) as the mechanism relevant for H₂S uptake. Contrarily, Wallace et al. (2014) have observed potassium and sodium polysulfides, iron sulfite, iron sulfate and sulfide as the products of reactive adsorption.

Few publications are available on H₂S fixation by waste ash, coal ash or biomass ash. Lin et al. (2001) noticed the possibility of H₂S removal by coal ash. Parker et al. (2009) report removal of H₂S from LFG in a small experimental device, using MSWI bottom ash. The removal efficiency for H₂S decreased from close to 100% to 20% within a 37 day trial. Ducom et al. (2009) prepared a MSW bottom ash sample from a French mass incinerator by open air drying

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and adjusting the water content to 15%. The content of H₂S, CO₂ and mercaptanes was monitored in the LFG and purified gas. The removal efficiency for H₂S ranged from 50% to nearly 100%, depending on the flow rate of LFG and the time from the start of the experiment, with a clear decrease over time (Ducom et al., 2009). Other authors (Papurello et al., 2016) indicated the clear influence of water content of the ashes in the removal of H₂S and alcohols from biogas up coming from the organic fraction of municipal solid waste. Recently, Fontseré Obis et al. (2017) drew similar conclusions, and found that H₂S removal by MSWI bottom ash was dependent on the initial humidity of the gas and the water content of the employed ashes.

The main aim of the present study was to investigate the option to purify biogas from small-scale biogas plants by entrapping CO₂ and H₂S with locally available biomass ash. A second aim was to study the potential of the biomass ashes employed for gas upgrading as nutrient source in agriculture, and this way recovering essential nutrients for plant development, which are contained in the ashes, such as K, Mg, K and P (Insam and Knapp, 2011). However, it is known that when applied to the soil, ashes might have a strong effect on the flora and fauna if they are not previously stabilized (Huotari et al., 2015). In line with this, other authors (Lapa et al., 2002; Römbke et al., 2009; Tsiroidis et al., 2012) consider it necessary to test not only the ashes' chemical composition, but also their ecotoxicological potential so they can be considered either as a waste or as a raw material.

Ash stabilization, for example before application in forestry (Steenari and Lindqvist, 1997), has often been achieved by adding water and/or by leaving the ash react with atmospheric CO₂, where, due to carbonation processes, highly reactive hydroxide components are transformed to carbonates. Nowadays in Austria, the application of biomass ashes from combustion of untreated biomasses in forestry and agriculture is allowed, considering the ecological principle of bringing back to nature the nutrients presented is the ashes (Bundesministerium für Land- und Forstwirtschaft, 2011), in this context, the application of less reactive ashes can be of advantage since the application of unstabilized ashes into soils may lead to nutrient losses via leaching; and to an increase in soil solution pH and salt concentration (Zimmermann et al., 2002; Ozolinčius et al., 2005, Fernández-Delgado Juárez et al., 2015). We expected that the faster carbonation process due to the high concentration of CO₂ in the crude biogas, would accordingly reduce ash toxicity, improving its suitability as a soil amendment, e.g. as a replacement for lime and as a micronutrient source for plants.

Moreover, prior studies conclude the use of ashes for LFG or biogas upgrading using locally available ashes as a H₂S adsorbent, would reduce costs compared to other technologies (Fontseré Obis et al., 2017), and that economic assessment methods provide, together with LCA, useful insight for application of waste management strategies and planning (Finnveden et al., 2007), albeit no studies on the economic viability of their use have been done yet (e.g. Hao et al., 2008).

The main goals of this study were: (i) the use of regionally available wood ash is an appropriate method to purify biogas from small-scale biogas plants by entrapping CO₂ and H₂S; (ii) employing biomass ash as CO₂ trap was expected to improve its chemical properties and thus enhance its potential use as a nutrient source for plants, and (iii) moreover it was expected that this biogas upgrading technique would be economically feasible at a regional scale compared to other gas upgrading techniques.

The present paper describes the background, the setup of a demonstration plant, and the experiments performed with this demonstration unit. Further, a valorisation of the potential of the ashes as soil amendments, and an economical assessment for this upgrading technology is presented.

2. Material and methods

2.1. Neustift demonstration pilot-plant

A demonstration plant (2014) was set up in connection with the small-scale biogas plant of Armin Hofer (Neustift, Tyrol, Austria, 1030 m above sea level), where mainly source-separate collected domestic organic wastes are treated. Five test runs, to study the suitability of wood ash as material for biogas upgrading, were performed.

The design of the pilot plant is shown in Fig. 1. An intersection for flow partitioning was constructed at the existing biogas plant to enable a splitting of the biogas stream, and connected with a speed-regulated pump. The volume flow to the filter was monitored with a gas flowmeter. Temperature sensors and loggers enabled continuous registration of the gas composition.

Gas analysis was performed with a portable gas analyzer GA 5000 (GSI landfill gas analyser, GA 5000; Geotech, Leamington, UK) equipped with infrared detector for CO₂ and CH₄ (0–100% range), electrochemical sensor for O₂ (0–25% range) and an electrochemical sensor for H₂S (0–1000 ppm range), temperature of gas, outside air (gas and outside air loggers: Testo 175T3 Datalogger) and ash (3 loggers in the ash) (ash loggers: Tinytag Plus 2 – TGP-4017, Tinytag Gemini Data Loggers Ltd., UK), as well as pressure (external air, gas).

The steel tank reactor of 1 m³ for the biomass ash (wood ash) was equipped with a lid at the top and a front door. The inlet for the crude biogas was placed at the base of the reactor and the gas was distributed horizontally, to achieve a homogeneous gas flow through the ash-filled tank. After the experiments, the ash was sampled in three layers and the remainder removed through the front door.

At the beginning of each test run, the whole bypass system and the ash were flushed with N₂ to avoid formation of explosive gas mixtures (methane + air). During flushing, the gas was not fed back into the main biogas pipe, but discharged by opening the flushing valve (green valve in Fig. 1) until O₂ concentration dropped to less than 1.0% (v/v). During the subsequent main phase of the tests, the purified gas was fed back into the main biogas pipe and utilized – together with the main biogas stream – in a gas engine.

2.2. Experimental materials and performed test runs

The selection of the ash type for the five pilot-scale test runs was based on the results of previous laboratory scale test (data not shown; Mostbauer et al., 2015) done according to (Mostbauer, 2014). During the laboratory tests (using 20 kg of ash for each test) it has been observed that the H₂S uptake and carbonation velocity of dry biomass ash is very low. Humidified and coarse biomass ashes had clearly higher removal rates, than fly ashes and dried cyclone ashes, (e.g. wet bottom ashes showed a H₂S removal capacity of 0.4 kg H₂S kg/T t-1 WS, from biogas containing 700 ppm H₂S) (Mostbauer et al., 2015).

The ash used was a mixture of fly ash (10% f.w.) and bottom ash (90% f.w.) from an incineration plant in Kufstein (Austria) operated with untreated wood chips, saw dust and bark. Biomass is combusted in a fluidized-bed system, with a thermal power: 28.4 MW, and an electrical power of: 6.5 MW. Main composition and chemical properties of the used wood ash are summarized in Table 1. FTIR (Fourier Transformed Infra Red) spectral analysis (Supp mat. Fig. 1) indicated that the ash employed in the trial contained Ca(OH)₂.

A large grate ash sample (approx. 20 t) was obtained from the biomass combustion facility Kufstein (Austria) and stored in water-tight bigbags (1 m³ volume) until the experiments were per-

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