Experimental investigation of dairy biogas as fuel for a molten carbonate fuel cell

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Based on experimental investigation, the performance of a molten carbonate fuel cell (MCFC) run on dairy biogas is presented, as well as an analysis of dairy biogas as fuel for MCFC. The results are compared with hydrogen as a reference fuel. Dairy biogas is characterised by lower efficiency and lower fuel utilisation factors compared to natural gas. Samples of biogas based on dairy products were made for the study, and these were assayed for hydrocarbon content. Test results for MCFCs were carried out using a single MCFC with an area of 25 cm^2, and the results obtained were compared with hydrogen as a reference fuel. Dairy biogas gives poorer results in terms of power and efficiency but is a much easier fuel to obtain than hydrogen.

Keywords: Fuel cells, biogas, experimental investigation

INTRODUCTION

Fuel price inflation and a long-term increase in electricity consumption have provided added impetus to the search for ultra-effective power generation systems. Fuel cells generate power in electrochemical reactions with potentially ultra-high efficiency. In contrast to low temperature fuel cells (Perna, Minutillo and Jannelli, 2015; Raj and Chan, 2015), high-temperature fuel cells (solid oxide fuel cell—SOFC) (Buonomano *et al.*, 2015; Damo *et al.*, 2015; De Lorenzo and Fragiacomo, 2015; Ferrari, 2015; Lv *et al.*, 2015; Polverino *et al.*, 2015; Ranaweera and Kim, 2015; Majedi, Abbasi and Davar, 2016; Subotić, Schluckner and Hochenauer, 2016) and molten carbonate fuel cell—(MCFC) (Haghighat Mamaghani *et al.*, 2015; Huang *et al.*, 2015; Law, Lee and Tay, 2015; Nekrasov *et al.*, 2015; Rexed *et al.*, 2015; Roshandel, Astaneh and Golzar, 2015; Zhang *et al.*, 2015)) are considered as highly efficient electricity sources due to the low cost catalysts used (Ni vs Pt). As is the case with gas turbine technologies (Mondal and De, 2015), state-of-the-art MCFCs are being built in the tens of MW power range. Research and development in this field is predicted to result in an increase in the power of those kinds of systems in the future. Additionally, MCFCs can be considered as a very efficient way to separate CO_2 from flue gases compared to other methods (Bartela, Skorek-Osikowska and Kotowicz, 2012; Janusz-Szymańska, 2012) to reduce emissions from fossil fuel power plants.

On the other hand, distributed generation of electricity in decentralised systems plays an increasingly vital role in Europe. This poses a challenge to the development of new technologies with the balanced use of energy from renewable sources and cogeneration systems (Chmielewski *et al.*, 2016). Energy policy takes into consideration micro-cogeneration as an element of a more effective use of electric energy from the electric power network (Farahnak *et al.*, 2015), as illustrated by the example of households for which biofuel is more suitable than compressed hydrogen (Dutka, Ditaranto and Løvås, 2015; Yang *et al.*, 2015). Office buildings (McLarty, Brouwer and Ainscough, 2016; Romero Rodríguez *et al.*, 2016) are potential distributed generation sources, as they are already equipped with emergency units (mainly internal combustion piston engines (Rey *et al.*, 2016)), and connected to the municipal networks (Ramírez-Elizondo *et al.*, 2015) (natural gas, electricity, heating (Grzebielec *et al.*, 2015)). It should be noted that the building sector uses about 40% of total primary energy and contributes to 35% of global greenhouse gas emission (Hesaraki, Halilovic and Holmberg, 2015). This shows the importance of improving heat and power systems in the residential sector. Using renewable energy such as solar energy, and biomass or low-carbon heat sources, such as district heating, heat pumps or heat storage instead of fossil fuels would greatly reduce CO_2 emissions and the environmental loads of fossil fuels.

Hydrogen and natural gas are currently considered to be the main fuels for fuel cells. Hydrogen is an ideal fuel with respect to fuel cell working conditions. Unfortunately, hydrogen is not present in the environment in an uncombined form and there are difficulties with production, transportation, and storage. Natural gas, meanwhile, is an interim fuel due to limited resources.

The most plausible future scenarios in the power markets are as follows:

1. Abandoning gas/liquid/solid fuels in favour of electricity generated by renewable sources and/or nuclear plants. In this case, the energy distribution role will be provided by the power grid, and the storage role by consumers.
2. Production of plant-derived gas/liquid fuels (Buonomano *et al.*, 2015) based on the cultivation of plants and shrubs, such as *e.g.* Salix Viminalis and their conversion into fuel, e.g. alcohols.

While exclusive use of electricity can be problematic (e.g. airplanes), the cultivation of 'energy' seems to be one of the most promising scenarios for the future.

The use of biogases (Budzianowski, 2016) in the MCFC has been relatively poorly investigated. Kivisaari (Kivisaari, Björnbom and Sylwan, 2002) evaluated MCFCs by deploying a biomass-fuelled integrated gasification combined cycle (IGCC) system in which biomass was converted into biogas by pressurised gasification with subsequent treatment. The results showed that the IGCC-MCFC systems fuelled by biomass have a maximum efficiency of 43%. Landfill-gas as a by–product of waste fermentation in landfills was considered as a fuel for the MCFC in (Lunghi, Bove and Desideri, 2004). Experimental comparison of MCFC performance using three different biogas types and methane is presented in (Bove and Lunghi, 2005). The biogases considered are derived from the following processes: steam gasification in an entrained flow gasifier, steam gasification in a dual interconnect fluidised bed gasifier, and biogas from an anaerobic digestion process. In (Wu, Lin and Ho, 2015) the operation of MCFC with different biogas sources was investigated. Two mobile MCFC test beds were operated with biogas from different origins, and quality and electrical efficiency of the MCFC stack of 50% was achieved. It was also found that there was no major interaction between biogas and fuel cell system components. In the town of Ahlen (Germany) a MCFC was operated in the municipal sewage system in 2005. It was the first project of its kind in Europe (Krumbeck, Klinge and Doging, 2006). In (Ciccoli *et al.*, 2010) an anaerobic digestion process of organic residues was used for electrochemical conversion to electricity and heat through an MCFC. It was shown that during biogas production H_2S has multiple negative effects on the performance and durability of the MCFC. Three general approaches to solving the sulphur problem in the MCFC are suggested. A much more comprehensive analysis about this issue is presented in (Zaza *et al.*, 2010, 2011).

Dairy wastes are plentiful in the dairy industries. The wastes contain high levels of organic matter and disposal of the effluents may cause serious environmental pollution (Patil *et al.*, 2012).

In (Hesaraki, Halilovic and Holmberg, 2015), the performance of a MCFC and micro gas turbine (MGT) hybrid power system bi-fuelled by city gas and biogas was evaluated. The MCFC/MGT hybrid power system and MFCF/MGT hybrid power system responses have been investigated experimentally and numerically in terms of performance. The MFCF/MGT hybrid power system can have manifest operation with the addition of biogas at a flow rate of 50% of the overall input heat value.

Sewage and waste from the dairy industry are the ideal raw material for bioenergy production because they contain significant amounts of easily degradable organic pollutants. In addition, they are characterised by a high content of water and trace elements necessary for the life of microorganisms. Ever-increasing energy prices and the growing requirements regarding CO_2 emissions have led to increased interest in alternative energy sources. Biogas plants based on anaerobic processes are an ideal solution for dairy waste utilisation while producing energy that can be used universally - to produce electricity, heat, motor fuel and gas. Anaerobic digestion is attractive because of the increased interest in the recovery of energy from waste. Due to various advantages, anaerobic digestion is preferable to aerobic methods. Anaerobic decomposition of organic compounds is conducted in close cooperation with various groups of bacteria (Lastella *et al.*, 2002). These include hydrolysing, acetogenic, homoacetogenic, sulphate-reducing and methanogenic bacteria (Chynoweth, Owens and Legrand, 2001). Up to 95% of the organic load in a waste stream can be converted to biogas (methane and carbon dioxide) and the remainder is utilised for cell growth and maintenance.

In the present study, numerical investigation is applied to simulate the behaviour of an MCFC run on dairy biogas.

EXPERIMENTAL INVESTIGATION

Diary biogas

During the investigation the following parameters were determined: total solids (TS, drying at a constant temperature of 105°C), volatile solids (VS, mineralisation in an oven at a constant temperature of 550°C). The biogas content: O_2, N_2, CH_4 and CO_2 was measured by GC analysis (gas chromatograph SRI 8610C, SRI Instruments, USA equipped with TCD detector; column temperature 60°C, detector temperature 150°C, carrier gas He at a flow rate of 10 mL/min).

Table I. Description of the samples used in the investigation

Sample	Description
1	100 mL of clean cheese whey + 1 g of fat + 350 mL of digested sludge
2	100 mL of clean cheese whey + 2 g of fat + 350 mL of digested sludge
3	100 mL of clean cheese whey + 3 g of fat + 350 mL of digested sludge
4	75 mL of clean cheese whey + 1 g of fat + 25 mL of sediment + 350 mL of digested sludge
5	75 mL of clean cheese whey + 2 g of fat + 25 mL of sediment + 350 mL of digested sludge
6	75 mL of clean cheese whey + 3 g of fat + 25 mL of sediment + 350 mL of digested sludge
7	100 mL of polluted cheese whey + 2 g of fat + 350 mL of digested sludge
8	Blank—350 mL of digested sludge

Table II. Characteristics of the individual substrates used in the investigation

Substrate	Total Solids, %	Volatile Solids, %
Clean cheese whey	7.42	6.45
Polluted cheese whey	5.44	4.73
Refused fat	6.38	5.99
Sediment	0.17	0.07
Digested sludge	22.93	16.03

The waste samples obtained from a dairy manufacturer located in the Lodz region were mixed with 350 mL of digested sludge obtained from the group wastewater treatment plant in Lodz, Poland. The waste samples included clean or polluted cheese whey, refused fat and sediment from the decanter. The description of the samples used in the experimental step is shown in Table I. The characteristics of the individual substrates are given in Table II. After mixing, the pH of the mixture was adjusted to 7 using $NaHCO_3$ or H_2SO_4 and the flasks were placed at 37°C and shaken once per day. Volumes of biogas evolved were measured by the liquid displacement method with the use of 33% NaCl solution. The concentrations of methane, carbon dioxide, oxygen and nitrogen content in the biogas produced were measured by gas chromatography analysis.

Table II. Biogas content for each sample used in form simulations with MCFC

Sample	O_2, %	N_2, %	CH_4, %	CO_2, %	H_2, %	H_2O, %
1	0.816	3.08	72.2	23.9	–	–
2	0.249	1.35	72.7	25.7	–	–
3	1.04	3.70	69.6	25.6	–	–
4	0.941	3.51	71.0	24.6	–	–
5	3.06	10.4	62.6	24.0	–	–
6	1.15	4.31	69.2	25.3	–	–
7	0.180	0.749	70.6	28.5	–	–
8	0.467	25.7	41.1	32.8	–	–
Reference Point	–	–	–	23	69	8

Biogas with the optimal methane content was used in the investigation into use of the MCFC. It was tapped on the seventh day of the anaerobic digestion process. The biogas content used in the MCFC tests for each sample is shown in Table III.

Molten Carbonate Fuel Cell

An experimental investigation was conducted to measure the performance of a single MCFC cell fuelled by biogas. The MCFC tested has a planar area of 20 cm^2 (see Fig. 1), where the anode was a porous (55%) Ni structure with a thickness of 0.76 mm, the cathode a porous (60%) nickel oxide structure with a thickness of 0.7 mm and the electrolyte a lithium carbonate and potassium carbonate—$(Li_2CO_3)_{0.62}(K_2CO_3)_{0.38}$—mixture (three matrices of 0.3 mm each, in total 0.9 mm). The electrode-electrolyte matrix was sandwiched between the two opposing separator plates and the fuel and oxidant flow concurrently in opposing channels. The cathodic and anodic current collectors were made in the form of stainless-steel embossed sheets. The cell was tested in the experimental facility shown in Fig. 2,

where the cell was held in a vessel and it was possible to set up and control each operating parameter. The vapouriser system provides the measurement and control of the water vapour directed to the anode and cathode.

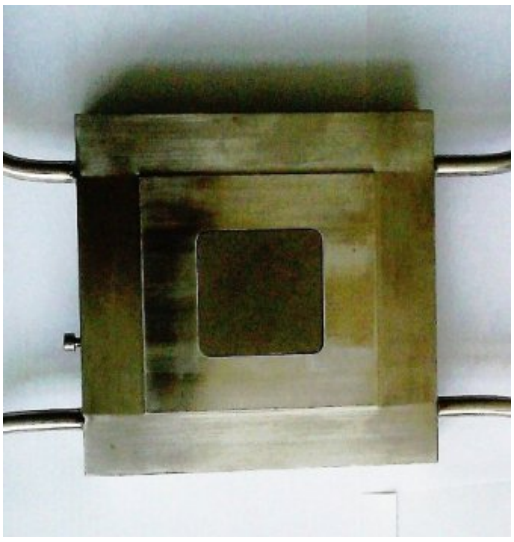

Figure 1. 20 cm² MCFC single cell used for tests.

The temperature of the system is subject to external control, and local temperatures collected by thermocouples, present in various positions on the cell, are processed by the PC board. Proprietary software allows one to easily view and adjust the condition and performance of the system.
The cell fabrication was concluded by the first heat up ('cell conditioning') during which the components assumed their final form. The cells were operated at atmospheric pressure with the same reference point gas compositions and flow rates. The fuel gas $(H_2)_{80}(CO_2)_{20}$ and the oxidant gas $(Air)_{70}$ $(CO_2)_{30}$ were fed.

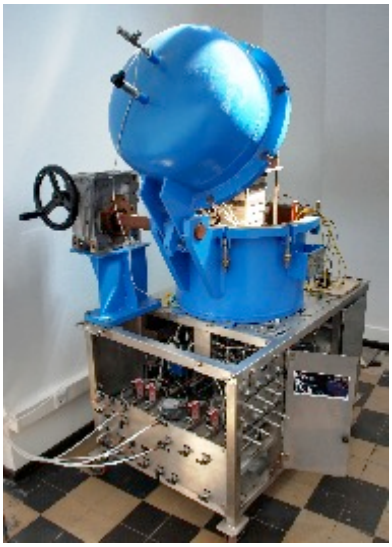

Figure 2. MCFC single cell test setup.

Cell voltage is directly measured at the two electrodes and its value is processed by a national instruments board. Cell resistance was measured by using a HIOKI 3560 AC mΩ HiTESTER (four wires, 1 kHz). The gas composition and flow rates were controlled by a set of mass flow controllers. The gases and water flow rate were measured and controlled by Brooks 5850E digital mass flow controllers,

chosen for their high accuracy and for their ability to be managed by software through serial PC ports. For load demand, a DC electronic load (SAE electronic conversion SRL) was used.
Eight series of tests were performed at 650°C, the temperature kept constant on the cell plane using heating plates equipped with three electric heaters each.

USING DAIRY BIOGAS AS FUEL FOR A MOLTEN CARBONATE FUEL CELL

Methane and diary biogas are mixed with steam to avoid carbon deposition. It was assumed that an adequate steam-to-carbon ratio required to prevent the carbon deposition was 2 (Herle *et al.*, 2004). The steam-to-carbon ratio (s/c ratio) defines steam molar flow in relation to methane and carbon monoxide molar flows delivered to the reformer.

$$\frac{s}{c} = \frac{n_{H_2O}}{n_{CH_4}+n_{CO}} \quad (1)$$

Three cases are given for reference: reference point, maximum flows and CH_4, s/c=2. The first is composed of reference conditions provided by Prof. Lee from Hanbat National University (see Acknowledgments) and is based on hydrogen as a fuel. This case is not the most convenient to compare against diary biogas for two reasons: firstly, there is no methane in the fuel flow; secondly, the amount of hydrogen delivered to the area of fuel cell is quite small, which creates difficulties with supplying methane with the same energy equivalency. Steam reforming of methane creates additional hydrogen from splitting water:

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

Thus, for delivering the same amount of chemical energy, the methane flow needs to be reduced by a factor of 4. To avoid very small fuel flows, two additional references are provided: (i) reference point composition with increased total volumetric flow for both sides: anode and cathode; (ii) methane as fuel with volumetric flow the same as hydrogen in the reference point (which means four times more chemical energy delivered to the fuel cell area).

Dairy biogas was delivered with the volumetric quantity energy equivalent to the reference point, as methane is not the only component in the fuel flow.

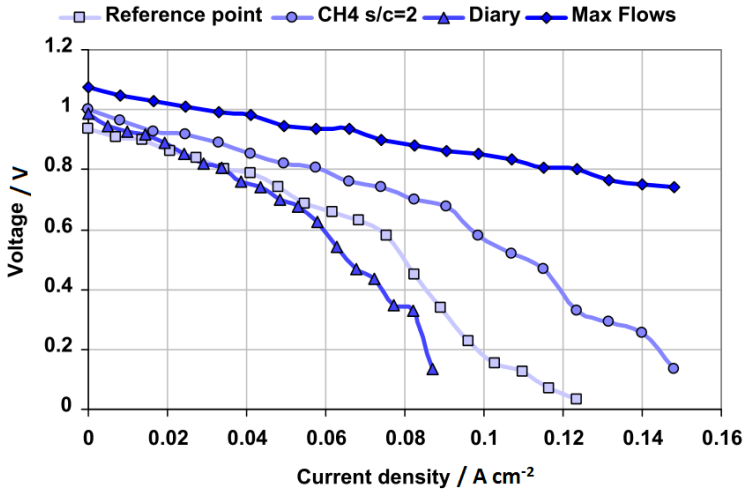

Figure 3. Polarisation curves for analysed cases.

Fig. 3 shows the polarisation curves obtained for the following analysed cases:

1. Maximum fuel and oxidant flow at the reference point composition
2. Reference point
3. Methane used as fuel at a steam–to–carbon ratio of 2
4. Dairy biogas used as fuel

Dairy biogas is characterised by the lowest voltages of all the analysed cases, but does not depart greatly from the reference point. Figure 3 shows the polarisation curves obtained for the four cases. The polarisation curve is a graphical representation of the cell voltage as a function of the current density. The maximum fuel and oxidant flow at the reference point composition and the reference point cases show similar voltage curves, indicating that they have similar performance. The methane case shows a slightly lower voltage curve, indicating a slightly lower performance compared to the reference cases. However, the dairy biogas case shows the lowest voltage curve, indicating the lowest performance of all the analysed cases.

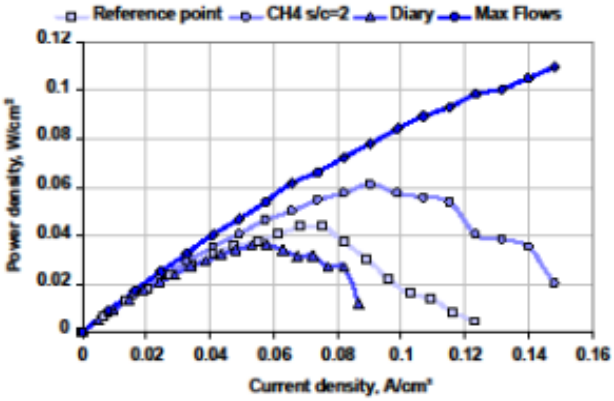

Figure 4. Power densities obtained for the analysed cases.

Fig. 4 presents power densities obtained for the analysed cases. Dairy biogas gives the lowest power density (4 mW/cm^2), but it should be noted that the efficiency and costs do not correspond with the power density here. The performance differences between four cases (maximum fuel and oxidant flow at the reference point composition, reference point, methane used as fuel at a steam-to-carbon ratio of 2, and dairy biogas used as fuel) based on the sample data presented in Tables 1 and 3, as well as the results obtained in Figures 3 and 4. Figure 4 presents the power densities obtained for the analysed cases. The power density is the amount of power produced per unit area of the cell. The dairy biogas case has the lowest power density (4 mW/cm2), indicating the lowest power output among all the analysed cases. However, the text notes that the efficiency and costs do not correspond with the power density, meaning that the power density alone does not provide a complete picture of the overall performance.

It should be noted that dairy biogas was delivered with the volumetric quantity energy equivalent to the reference point, as methane is not the only component in the fuel flow. This suggests that the lower performance of the dairy biogas case may be due to factors other than the energy content of the fuel. Finally, the MCFC fuelled by dairy biogas gives about 20% lower efficiency (2 percentage points) than the reference hydrogen fuel. This suggests that while the dairy biogas case may have a lower power output and voltage curve, its overall performance is significantly impacted by its lower efficiency compared to the reference case.

In summary, the sample data presented in Tables 1 and 3, as well as the results obtained in Figures 3 and 4, show that the dairy biogas case has the lowest performance among the analysed cases, with a lower voltage curve, power density, and efficiency compared to the reference case. However, the power density alone does not provide a complete picture of the overall performance, and the lower performance of the dairy biogas case may be due to factors other than the energy content of the fuel.

CONCLUSIONS

An experimental investigation was conducted to determine the working conditions of a laboratory-size MCFC working on biogas produced from dairy waste. Adequate samples of biogas were obtained by

aerobic digestion, and then experiments for simulated biogas with similar composition were performed. Two other fuels were taken for reference (hydrogen and methane). For both fuels, the experiments were done based on the same fuel cell.

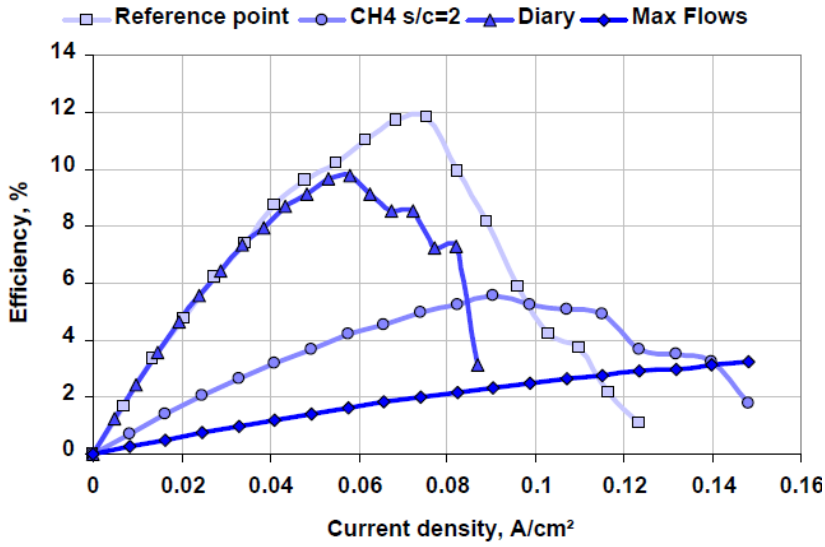

Figure 5. Efficiencies obtained for the analysed cases.

For all investigated fuels, moderate efficiencies were obtained (up to 12%). The highest efficiency power is generated with hydrogen as fuel but, surprisingly, dairy biogas took second place. Dust particles were not investigated in the simulated biogas, as they can be easily removed by conventional dedusting systems, such as bag filters, electrostatic precipitators, and electric-bag composite dust collectors, which can efficiently extract coarse particles (Wu *et al.*, 2016).

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