Available online at www.sciencedirect.com
SCIENCE DIRECT®

Journal of Power Sources 145 (2005) 588–593

JOURNAL OF POWER SOURCES

www.elsevier.com/locate/jpowsour

Short communication

Experimental comparison of MCFC performance using three different biogas types and methane

Roberto Bove*, Piero Lunghi

University of Perugia, Industrial Engineering Department, Italy

Accepted 3 January 2005
Available online 4 June 2005

Abstract

Biogas recovery is an environmentally friendly and cost-effective practice that is getting consensus in both the scientific and industrial community, as the growing number of projects demonstrate.

The use of fuel cells as energy conversion systems increases the conversion efficiency, as well as the environmental benefits. Molten carbonate fuel cells (MCFC) operate at a temperature of about 650 °C, thus presenting a high fuel flexibility, compared to low temperature fuel cells.

Aim of the present study is to compare the performance of an MCFC single cell, fuelled with different biogas types as well as methane. The biogases considered are derived from the following processes: (1) steam gasification in an entrained flow gasifier; (2) steam gasification in a duel interconnect fluidized bed gasifier; (3) biogas from an anaerobic digestion process.

The performances are evaluated for different fuel utilization and current densities.

The results are an essential starting point for a complete system design and demonstration.

Keywords: MCFC; Experimental tests; Biogas; Performance comparison

1. Introduction

Conventional fossil fuels currently represent the most widely used source of energy for electricity generation. As can be observed in Fig. 1 [1], both hydropower and nuclear power account for about 17% of the total energy production, while renewable (excluding hydropower) accounts only for a small share of the total energy production. Although fossil fuel shortage is not going to happen in the short term, the use of non-renewable fuels as the primary energy sources is a practice that cannot last forever. Due to the growing gross domestic product (GDP) of developed and of some developing countries, such as China and India, energy demand increases year by year, while fossil extraction, in the mid-long term, will not be able to fully satisfy the demand. The effect of the increasing energy requirements and, at the same time, of the fossil fuels extraction reduction, will result in a tremendous price increase of conventional fuels.

The contribution of nuclear power, however, will hardly increase. There are at least two main issues that will slow down nuclear power employment. First, public opinion is worldwide hostile to nuclear-based energy conversion systems, and, secondly the cost per kilowatt-hour is quite high [2]. Elevated reliability and waste disposal issues, in fact, increase the production cost, so that nuclear energy is currently more expensive than many other practices, including, for example, wind energy [2].

The other consistent contribution in electricity generation is represented by hydropower, i.e. a renewable, clean and relatively low environmental intrusive practice. However, most of the rivers that present potentiality for power plant realization have been exploited. In Switzerland, for

---

* Corresponding author at: Connecticut Global Fuel Cell Center, 44 Weaver Road, Storrs, CT 06269-5233, USA. Tel.: +1 860 486 5407.

E-mail addresses: rbove@unipg.it (R. Bove), lunghi@unipg.it (P. Lunghi).

0378-7753/$ – see front matter © 2005 Elsevier B.V. All rights reserved.
doi:10.1016/j.jpowsour.2005.01.069

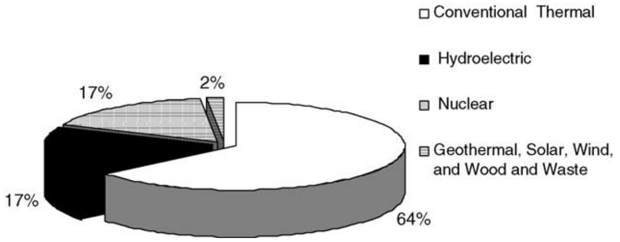

Fig. 1. World energy sources for electricity production in 2002 [1].

example, 59% of the electricity is produced by using hydropower, as well as in Sweden about 50% [1]. Excluding few developing countries, it is possible to foresee that no new installations will be created in the future. In any case the contribution of hydropower will not be able to increase significantly.

For the reasons above, it is clear that the future scenario for power generation relies on renewable energy sources, like wind, solar energy, biomass and waste.

High temperature fuel cells, i.e. solid oxide fuel cells (SOFC) and molten carbonate fuel cells (MCFC) present the peculiarity of being able to tolerate the presence of impurities in the fuel, more than other fuel cell technologies. As for the other FCs, both MCFC and SOFC present very high conversion efficiency, thus a reduction of primary energy demand is associated with their employment.

Environmental benefits of using MCFC to produce electricity from landfill gas has been shown in a previous study of the authors, where the entire life-cycle system is analyzed [3]. Although the study is conducted for landfill gas, similar results are expected also for other biogases.

In a previous publication [4], the authors presented the first experimental results for an MCFC single cell running on a steam gasification derived gas. In the present study, more detailed results are provided, together with experiments conducted on a lab-scale fuel cell, operating with different biogases.

2. Biogas types and usage

The term “biogas” is referred to a gas, generally rich in CH_4 and CO_2, derived by animal waste, human sewage or crop residues. For water treatment facilities, farms or factories, for examples, biogas is just a by-product and is generally discharged into the atmosphere after an eventual specific treatment. Since the energetic content is often relevant, the energy recovery allows one to couple the benefits of energy savings with the reduction of waste disposal.

Biogas composition is mainly dependent on the raw material and the production process that generated it. Table 1 provides USA typical compositions of landfill gas, anaerobic digestion product gas (from wastewater treatment facilities) and gasification synthesis gas [5].

Table 1 clearly shows that the energy content of each gas, associated with the high CH_4 and/or H_2 content, makes biogas an interesting energy source. Because of the CH_4 presence (that is a greenhouse gas), when energy recovery is not conducted, biogas is generally flamed. Other energy recovery forms, like chemical energy storage and thermal energy production, are alternatives to electricity production. The option is usually selected on the basis of specific needs, or economical factors. Different technologies are currently available for biogas energy conversion. For electricity generation, internal combustion engines are currently the most employed. This is because internal combustion engines have been used for decades, thus they present high reliability, despite their low price.

A case study for the optimal energy conversion system for electricity generation from landfill gas is presented by Iannelli and Moreno [6] for an Italian landfill. A multi-criteria approach is used to determine, under different economical and technical conditions, the optimal choice.

3. Biogas and fuel cells

Due to the high environmental benefits, and the high potentiality of fuel cells, the possibility of using fuel cells as energy conversion systems for biogas has caused interest in both the scientific and industrial community.

First attempts to use biogas as a fuel for fuel cells originated in the 90’s, when phosphoric acid fuel cells (PAFC) had already reached high performance and reliability [7–10]. The successful experience with PAFC [11], encouraged developers of other fuel cell technologies to consider

Table 1
Typical chemical composition of biogases in United States

	Landfill gas	Anaerobic digestion product gas (dry basis)	Biomass gasification synthesis gas (dry basis)
CH_4	40–45%	55–65%	1–20%
CO_2	35–50%	30–40%	10–20%
CO	–	–	10–45%
H_2	–	–	10–30%
H_2O	1–10%	–	–
N_2	0–20%	1–10	40–50% (for air blown)
O_2	–	<0.5%	–
Other hydrocarbons	250–3000 ppm	Trace	1–10%
H_2S	<200 ppm	<200 ppm	<1.3%

biogas as a valuable alternative to hydrogen or conventional hydrocarbons.

Due to the presence of noble metals as the catalyst, PEM fuel cell compatibility with biogas is very limited [12].

The high operating temperature of SOFC and MCFC makes them particularly suitable for biogas usage. The presence of CO, that is a harmful gas for low temperature fuel cells, is not only harmfulness, but represents an additional fuel. Moreover, the high temperature of the outlet gas from the fuel cell allows the gas-processing unit to be well integrated with the power section.

According to the biogas considered, particular units for contaminants removing, humidification and gas reforming (when needed) are required. Several system solutions have been proposed for integrating fuel-processing systems with high temperature fuel cells [13–19] and the resulting performances are very high, compared to traditional systems for biogas energy conversion.

Together with system configurations, fundamental studies and tests on small-scale fuel cells are conducted for validating biogas potentiality in SOFC [20–26] and MCFC [4,13,27–32].

4. Biogas types considered

In the present study, the performance of an MCFC, running on four different biogases are experimentally evaluated. Biogases considered are:

(1) biogas from anaerobic digester (ADG);
(2) landfill gas (LFG);
(3) syngas from an entrained bed steam gasifier (EBG);
(4) syngas from a dual interconnected fluidized bed steam gasifier (DBG).

As previously mentioned, all the biogases contain impurities that limit fuel cells lifetime and performance. In order to evaluate the biogas potential for an MCFC, the gas must be tested “pollutant free”. At this aim, a gas mixture, reproducing the chemical composition of the gas under study is provided to the cell, changing the cell operating conditions. The effect of the pollutants is then conducted in a different test campaign, where the maximum allowable concentration of each pollutant is pointed out.

All the performances are compared to natural gas (NG), since this is currently the most used fuel for MCFC.

The chemical composition of each gas, after the steam reforming process, is computed. A CH_4 conversion factor of 97% is supposed. The reactions considered for methane conversion are:

$$CH_4 + H_2O \rightarrow CO + 3H_2 \tag{1}$$

$$CO + H_2O \leftrightarrow H_2 + CO_2 \tag{2}$$

The starting compositions are chosen according to the data provided by technologies suppliers or other research centers.

Table 2
Chemical composition of the tested-simulated biogases and the reformed methane

Anodic gas (%)	ADG	RNG	LFG	EBG	DBG
H_2	39.17	54.92	38.9	42.85	31.3
CO_2	18.59	6.9	19.74	7.29	10.15
CO	9.1	8.58	9.04	17.92	13.8
CH_4	0.27	0.4	0.27	2.76	4.75
H_2O	32.14	29.2	31.92	29.18	40
O_2	0.1	0	0.03	0	0
N_2	0.61	0	0.1	0	0

ADG: anaerobic digester gas; RNG: reformed natural gas; LFG: landfill gas; EBG: entrained bed gasifier; DBG: dual bed gasifier.

The final compositions after the steam reforming (i.e., the gas composition provided to the MCFC) are reported in Table 2.

Due to the presence of carbon monoxide and carbon dioxide, carbon deposition at the MCFC anode is possible. The most significant reactions that can occur are:

$$CH_4 \rightarrow 2H_2 + C \tag{3}$$

$$2CO \rightarrow CO_2 + C \tag{4}$$

$$CO + H_2 \rightarrow H_2O + C \tag{5}$$

Fig. 2 depicts the combinations of C, H and O content that lead to carbon formation, according to reactions (3–5). If the gas composition is in the region below the limit line, no carbon deposition occurs. On the other hand, a gas mixture inside the “carbon deposition” zone does not necessarily lead to carbon formation. This is because the line of Fig. 2 is determined on the basis of the equilibrium theory, while kinetics of reactions (3–5) are very slow and the activation energy is not always exceeded. For this reason, gas compositions of Table 2 are considered safe from carbon formation.

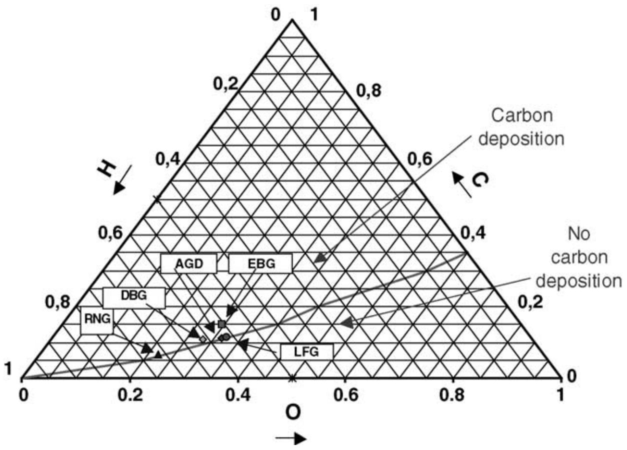

Fig. 2. Carbon deposition equilibrium.

5. Experimental tests apparatus

The experimental tests on the single fuel cell are conducted at the University of Perugia, Italy. The single cell is constructed, assembled and provided by Ansaldo Fuel Cells S.p.A. (AFCo). The cell is composed of an Ni–Cr anode, an NiO lithiate cathode, and an Li_2CO_3–K_2CO_3 and γ-$LiAlO_2$ electrolyte. The iso-thermal condition of 650 °C is ensured by two electric heaters. The cell voltage is directly measured at the two electrodes and its value is processed by a National Instruments board, which yields other analogical measurements, such as hydrogen and the carbon monoxide environment concentrations. The board is a PCI-6035E NI, characterized by two analog outputs, 16 analog inputs with an acquisition range between 610 V and 650 mV and a sampling rate of 200 kS s^{-1}. The gases and water flow rate are measured and controlled by the Brooks 5850E Digital Mass Flow Controllers, chosen for their high accuracy, and for their characteristic of being able to be managed by software through serial PC ports.

The vaporizer system provides the measurement and control of the water vapor directed to the anode and, through a serial port, it is possible, through the software, to manage the temperature and the flow rates. The electric load bank is an Agilent N3301A, able to work in constant current, constant voltage or constant resistance mode. The load is also remotely controlled by the serial PC port.

Finally the temperature of the system is subjected to external control, and local temperatures collected by thermocouples, present in various positions on the cell, are processed by the PC board. A proprietary software, realized in Visual Basic language, allows one to easily view and manipulate the system condition and performance.

A complete description of the apparatus test is illustrated in a previous publication [33].

6. Results

Tests are conducted so that most of the possible systems operating conditions are reproduced. The temperature of the cell is always kept constant at 650 °C, because this is the designed stack temperature. A first performance comparison is conducted providing the cell with a constant inlet flow rate, and varying the current density of the cell. The gas compositions of Table 2 are provided. Performances obtained using LFG and ADG are extremely close, thus only ADG test results are reported. Fig. 3 depicts the voltage comparison, while the relative power density is reported in Fig. 4.

As can be noted, the performances of all the biogases are very close to those of reformed natural gas. In particular, biogas derived from entrained bed gasifier (EBG) allows the fuel cell to reach performances that are very close to that of reformed natural gas. This is due to the very low water content of this gas. As can be seen in Fig. 2, in fact, EBG presents the lowest steam to carbon ratio. The high performances obtainable, however, are counter-paid by the risk of carbon formation in specific zones of the fuel cell, where particular temperature or concentration gradients can arise. Although no carbon formation is believed to occur in the single fuel cell tested, in a full-scale stack, different conditions can take place.

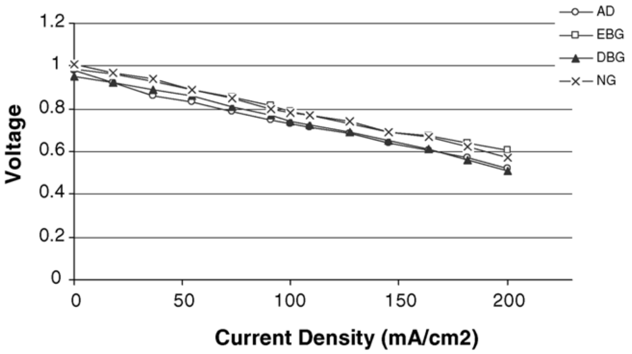

Fig. 3. Voltage comparison at constant inlet flow rate.

The tests reported in Figs. 3 and 4 represent a very good way of comparing different gas potentials, when used in an MCFC. However, it is not easy to understand the effect of different operating conditions on the fuel cell. When the current is increased, in fact, the voltage reduction is due to both the increased overvoltage and to the increased fuel utilization. When the fuel utilization is increased, the Nernst equation (Eq. (6)), that provides the ideal fuel cell voltage, presents a lower value, since hydrogen, oxygen and the cathodic carbon dioxide partial pressure decreases, and the water and anodic carbon dioxide increase with respect to the fuel utilization.

$$E = E_0 + \frac{RT}{2F} \ln \frac{P_{H_2} \times P_{O_2}^{0.5} \times P_{CO_2,c}}{P_{H_2O} \times P_{CO_2,a}} \tag{6}$$

In expression (6), E is the ideal voltage at open circuit voltage, E_0 is E evaluated at standard pressure, R the universal gas constant, T the cell temperature, F the Faraday constant, and P_i the partial pressure of the ith gas species.

In order to investigate the effect of the current density, and that of the fuel utilization, separately, further tests are

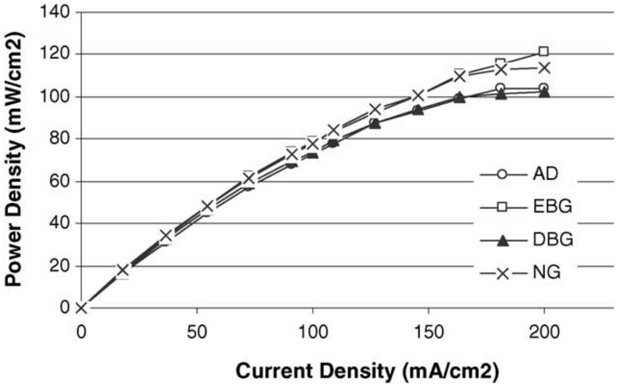

Fig. 4. Power density comparison at constant inlet flow rate.

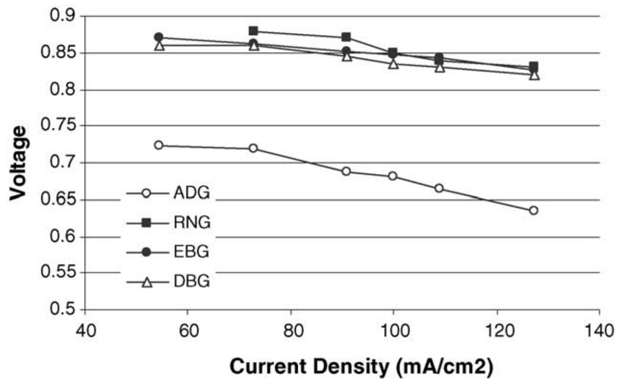

Fig. 5. Performance comparison at constant fuel utilization.

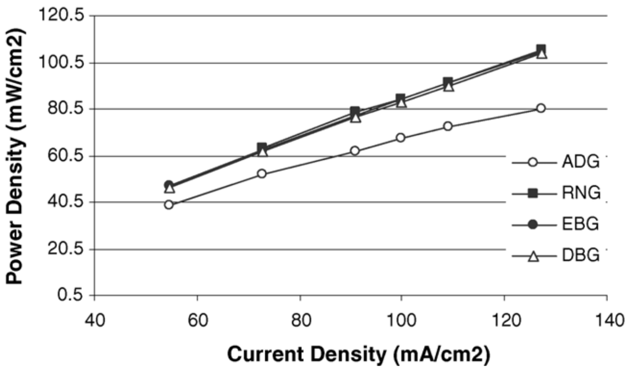

Fig. 6. Power density at constant fuel utilization.

conducted. Fig. 5 shows the effect of the current density variation on the voltage, when the fuel utilization is kept constant at 40%. The relative power densities are reported in Fig. 6. Despite what is deducted for the test at constant flow rate (Figs. 3 and 4), ADG shows very poor performance, compared to other biogases. Since the H_2 and CO content (i.e., the useful fuel for the FC) is of the same magnitude of the other gases considered (Table 2), the poor performance is probably due, not to the gas itself, but to problems incurred in the fuel cell tests. Additional experimental data are indeed needed, and those of Figs. 5 and 6 must be considered preliminary.

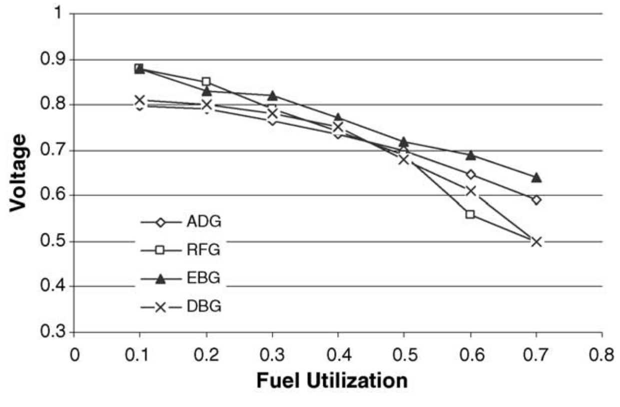

Fig. 7. Voltage comparison at constant current density.

Finally, Fig. 7 reports the voltage variation, when the current density is constant and the fuel utilization is varied (i.e., the inlet flow rate is varied). As for the tests at constant inlet flow rate performances are comparable.

7. Conclusions

In the present study different anodic gas compositions, representing different biogas types, have been tested on an MCFC single cell. Performances are compared to those obtained when reformed natural gas is used as the fuel. This comparison is performed because natural gas is currently the most used fuel for MCFC. Tests have been conducted at different fuel cell operating conditions, in terms of current density, fuel utilization and total flow rate. Results clearly show the high potentiality of all the gas compositions considered.

Acknowledgements

The authors would like to thank Arturo Torazza, Paolo Capobianco and all the Ansaldo Fuel Cells people for the valuable support given during the tests and for the contribution given in this manuscript preparation.

References

[1] International Energy Annual, Energy Annual 2002, International Energy Agency final report, 2004.

[2] R. Rechsteiner, Ten Steps to a sustainability energy future, in: B. Ulf (Ed.), Proceedings of the Fuel Cell World, 28 June–2 July 2004, Lucerne, Switzerland, 2004, pp. 3–28.

[3] P. Lunghi, R. Bove, U. Desideri, Life-cycle-assessment of fuel cells based landfill-gas energy conversion technologies, J. Power Sources 131 (1–2) (2004) 120–126.

[4] R. Bove, P. Lunghi, A. Lutazi, N.M. Sammes, Biogas as fuel for a fuel cell system: investigations and first experimental results for a molten carbonate fuel cell, Fuel Cell Sci. Technol. 2 (1) (2005) 9–13.

[5] D.C. Dayton, Fuel cell integration—a study of the impacts of gas quality and impurities, National Renewable Energy Laboratory (NREL) final report, 2001.

[6] R. Iannelli, A. Moreno, Application of molten carbonate fuel cells for exploitation of landfill gas. A feasibility study in southern Italy, in: Biofuels for Fuel Cells: Biomass Fermentation Towards Usage in Fuel Cells, IWA Publishing, June 2005.

[7] G.J. Randelli, R.J. Spiegel, Fuel cell energy recovery from landfill gas, J. Power Sources 37 (1–2) (1992) 255–264.

[8] G.J. Randelli, J.C. Trocciola, R.J. Spiegel, Landfill gas pretreatment for fuel cell applications, J. Power Sources 49 (1–3) (1994) 143–149.

[9] R.J. Speigel, J.C. Trocciola, J.L. Preston, Test results for fuel cell operation on landfill gas, Energy 22 (8) (1996) 777–786.

[10] S.T. Naumann, C. Myren, Fuel processing of biogas for small fuel cell power plants, J. Power Sources 56 (1995) 45–49.

[11] R.J. Spiegel, J.L. Preston, Technical assessment of fuel cell operation on landfill gas at the Groton, CT, landfill, Energy 28 (2003) 397–409.

[12] Xenergy, Towards a renewable power supply: the use of bio-based fuels in stationary fuel cells, Xenergy final report, 2002.

[13] P. Lunghi, R. Bove, E. Cerluini, A. Moreno, Energy recovery from waste water treatment biogas: integration of an anaerobic digester with a high temperature fuel cell, in: Proceedings of the second International Conference on Fuel Cell Science Engineering and Technology, June 14–16, 2004, Rochester, NY, 2004.

[14] K.V. Lobachyov, H.J. Richter, An advanced integrated biomass gasification and molten carbonate fuel cell system, Ener. Convers. Manage. 39 (16–18) (1998) 1931–1943.

[15] P. Lunghi, R. Burzacca, U. Desideri, Celle a Combustibile e Gassificazione per la Trigenerazione da Scarti Industriali: Criteri di Progettazione, Prestazioni e Test Case in un'Industria Alimentare, in: Proceedings of the XXVII Convegno Nazionale ANIMP 2001, Spoleto (PG), Italy, 2001 (in Italian).

[16] P. Lunghi, R. Burzacca, R. Bove, Assessment and analysis of gasification and fuel cell integration to enhance performances of hybrid plants for electricity production, Fuel Cell Seminar, November 18–21, Palm Springs, CA, 2002.

[17] A.O. Omosun, A. Bauen, N.P. Brandon, C.S. Adjiman, D. Hart, Modelling system efficiencies and costs of two biomass-fuelled SOFC systems, J. Power Sources 131 (1–2) (2004) 96–106.

[18] T. Kivisaari, P. Björnbom, C. Sylwan, Studies of biomass fuelled MCFC systems, J. Power Sources 104 (2002) 115–124.

[19] W.A. Amos, Analysis of two biomass gasification/fuel cell scenario for small-scale power generation, National Renewable Energy Laboratory (NREL) final report, 1998.

[20] J. Van herle, Y. Membrez, O. Bucheli, Biogas as fuel source for SOFC co-generators, J. Power Sources 127 (2004) 300–312.

[21] S. Baron, N. Brandon, A. Atkinson, B. Steele, R. Rudkin, The impact of wood derived gasification gases on Ni-CGO anodes in intermediate temperature solid oxide fuel cells, J. Power Sources 126 (2004) 58–66.

[22] J. Van herle, F. Maréchal, S. Leuenberger, D. Favrat, Energy balance model of a SOFC cogenetrator operated with biogas, J. Power Sources 118 (2003) 375–383.

[23] S.C. Singhal, Advances in solid oxide fuel cell technology, Solid State Ionics 135 (2000) 305–313.

[24] M. Kübel, R. Berger, K.R.G. Hein, Biomass gasification: an option for SOFC fuel? in: J. Huijsmans (Ed.), Proceedings of the fifth European Solid Oxide Fuel Cell Forum, 2002, pp. 995–1002.

[25] J. Staniforth, K. Kendall, Cannock landfill gas powering a small tubular solid oxide fuel cell—a case study J. Power Sources 86 (2000) 401–403.

[26] Y. Matsuzaki, I. Yasuda, The poisoning effect of sulfur-containing impurity gas on a SOFC anode. Part I. Dependence on temperature, time, and impurity concentration, Solid State Ionics 132 (2000) 261–269.

[27] U. Desideri, P. Lunghi, R. Burzacca, State of the art about the effects of impurities on MCFCs and pointing out of additional research for alternative fuel utilization, in: Proceedings of the first International Conference of Fuel Cell Science Engineering and Technology, April 21–23, Rochester, NY, 2003.

[28] US Department of Energy (DOE) and Electric Power Research Institute (EPRI), Landfill gas clean up for carbonate fuel cell power generation, CRADA MC95-031 final report, 1998.

[29] M. Kawase, Y. Mugikura, T. Watanabe, Electrochemical studies on the high performance electrodes and the effect of impurity gas in molten carbonate fuel cells, CRIEPI report, 2001.

[30] M. Kawase, Y. Mugikura, Y. Izaki, T. Watanabe, Y. Ito, Effects of fluoride on the performance of MCFCs, J. Power Sources 124 (2003) 52–58.

[31] M. Kawase, Y. Mugikura, T. Watanabe, Y. Hiraga, T. Ujihara, Effects of NH_3 and NOx on the performance of MCFCs, J. Power Sources 104 (2002) 265–271.

[32] S. Takahashi, T. Mizukami, T. Kahara, Effects of H_2S and HCl in coal gas on MCFC performance, in: Proceedings of the Fuel Cell Seminar, Palm Spring, CA, November 18–21, 2002.

[33] P. Lunghi, R. Burzacca, An effective and reliable method for single cells experimental testing and results comparison. Advance control technique, in: Proceedings of the World Renewable Energy Congress VII and Expo, Cologne Congress Centre, 29 June–5 July, Germany, 2002.