

Testing a PEM Fuel Cell System with Biogas Fuel

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ABSTRACT

The use of biogas in fuel cells advantageously combines a cost effective renewable energy source with a technology which promises high electrical efficiency and low environmental impact. For the first time, the suitability of biogas as a fuel for proton exchange membrane fuel cell systems (PEMFC) has been experimentally confirmed. The advantages of this particular type of fuel cell are its low operating temperatures, modular system design and moderate costs. Measurement results from a 650 W_{el} test plant show a cell efficiency of 58% at a power density of 0.14 W/cm². A particularly problematic component is the steam reformer with a thermal energy efficiency of 38%. A model calculation on the basis of an optimised PEMFC system shows that an electrical efficiency of over 40% can be obtained.

Keywords: biogas, CHP, energy, fuel cell, PEM, steam reforming

1. INTRODUCTION

Fuel cell technology provides a promising alternative to the conventional electrodynamic use of gaseous hydrocarbons in combined heat and power plants (CHP). Fuel cells produce less noise and pollutant emissions while achieving higher electrical efficiency, particularly in partial load operation. Proton exchange membrane fuel cells exhibit a comparatively higher power density and make less material demands as a result of low operating temperatures, thus allowing for lower costs.

PEMFC systems are being developed for stationary use with natural gas as fuel. As this type of fuel cell is only able to utilise hydrogen, the natural gas needs to be reformed to a hydrogen rich gas (reformate). Biogas has similar properties to natural gas and is by far the most cost effective renewable fuel. However, biogas has a lower power density, noticeably higher carbon dioxide content and exhibits various other harmful components such as sulphur compounds and ammonia.

The intention of research at the ATB is to develop and test a PEMFC system as an efficient, cost effective and reliable technology for generating electrical energy from biogas. Primarily, the focus is on experimentally verifying the compatibility of biogas with PEMFC and determining the system parameters needed for optimal operation. In order to estimate the attainable electrical efficiency the most important characteristic energetic values for an optimised system are calculated.

1.1 Processing Biogas and Hydrogen Generation

At present, several fuel cell systems are in development for distributed energy supply based on natural gas. The use of biogas as a fuel requires the systems to be technically adapted and optimised, as biogas differs to natural gas in several aspects (Schmersahl and Scholz, 2004).

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The methane content and thus the energy density is lower and irregular. The high content of associated catalyst poisons, above all sulphur compounds, demands additional gas purification.

The use of natural gas and biogas in fuel cells requires the methane to be converted. State of the art is steam reforming as used by the petrochemical industry on an industrial scale. During steam reforming, a catalytic conversion of methane with steam to hydrogen, carbon monoxide and carbon dioxide occurs. The reforming temperatures are at 650 to 850° C. As well as the steam reforming, desulphurisation, carbon monoxide conversion (CO-Shift) and carbon monoxide elimination are integrated into the fuel processor. The thermal efficiency η_{Ref} of the fuel processor can reach more than 80% (Schmitz, 2002). After the reforming process a CO-shift is carried out in one or two steps. Subsequently, the natural gas reformat exhibits a hydrogen content of 70% to 80% referring to the dry gas. The carbon monoxide content amounts to less than 1% (Krause et al., 2001). Due to the low operating temperature of [90° C, PEMFCs demand additional gas purification to reduce the carbon monoxide build up during reforming to below 10ppm. This is achieved by selective oxidation (selox) of carbon monoxide with atmospheric oxygen requiring a catalyst.

1.2 PEM Fuel Cells

The fuel cell stack consists of several layered single cells. The membrane electrode assemblies (MEA) are separated by the bipolar plates which feed the reactant gases. Around the stack there are peripheral components designed for media supply, cooling, process control, DC regulation and voltage conversion.

First experiences of PEMFC systems fueled by natural gas at the demonstration stage show a net system efficiency of 25% at 5 kW_{el} (Koschowitz, 2003) up to 35% at 200 kW_{el} (Pokojski, 2001)]. The optimisation of a 1 kW_{el} system at the Universität Gesamthochschule Essen delivered a maximum electrical efficiency of 42% (Schmitz, 2002).

1.3 Present Research in Biogas Fuel Cells

Besides those of the ATB, research and development activities for three different biogas fuel cell systems are known of in Europe. One research project aims to adapt the molten carbonate fuel cell (MCFC), namely the Hot Module of the company MTU, for use with biogas (Ott and Tamm, 2003). Another group of scientists at the Federal Research Institute for Agriculture is conducting research into steam reforming of biogas with subsequent CO₂ sequestration. The hydrogen produced is used to generate electricity in a PEMFC. In Switzerland is an adapted solid oxide fuel cell (SOFC) for residential power generation on test operation with biogas (Schuler, 2001).

The above named research projects are all characterised by the fact that they aim to adjust natural gas technology for use with biogas by modified gas processing. Particular emphasis is placed on the removal of the associated harmful gases prevalent in biogas. The purity requirements are largely determined by the reformer catalyst. However, the fuel cell electrodes are also damaged cumulatively and irreversibly by sulphur compounds, for example (DOE, 2000). The higher carbon dioxide load in biogas compared to natural gas calls for a change in fuel processor design. The reformat exhibits a lower hydrogen content causing a decrease in cell voltage. The fuel cell stack can be adapted by changing the flow fields and the catalyst loading of the anode (Beckmann, 2003).

2. MATERIALS AND METHODS

2.1 Supply of Raw Gas

The ATBs fuel cell test stand is supplied with biogas from two solid state fermenters (Linke et al., 2001). The substrate used is a mixture of fermented manure, manure and corn silage. The resulting biogas is stored in a 16 m³ balloon gas-holder. The methane content of 60% is varied by adding manufactured gases (CH₄, CO₂).

2.2 Experimental System

The test stand consists of the fuel processor with integrated desulphurisation and the fuel cell. Both units include a programmable logic controller (PLC). The PLC of the fuel cell assumes higher ranking functions such as initialising the start/stop procedures as well as the load demand. Media supply of the test plant secures the flow of biogas, deionised water and tap water used to feed the cooling circuits.

The fuel cell unit has two independent water-cooled fuel cell stacks manufactured by Schalt- and Regeltechnik GmbH Berlin. A test stack with 4 cells (150 W_{el}) serves to examine the effect of harmful gases, the operating stack with 14 cells (650 W_{el}) is intended to examine system behaviour and determine the energetic parameters.

The fuel is treated by a fuel processor with reformer, two stage shift converter and selective oxidation (figure 1). The reformer load is regulated by fed process gas mass flow in the range of 20% to 100% (1.4 to 5.8 l_N/min). For operation with 60% methane in the biogas, a maximum hydrogen output of 2.3 kW with a hydrogen content of 64% is specified.

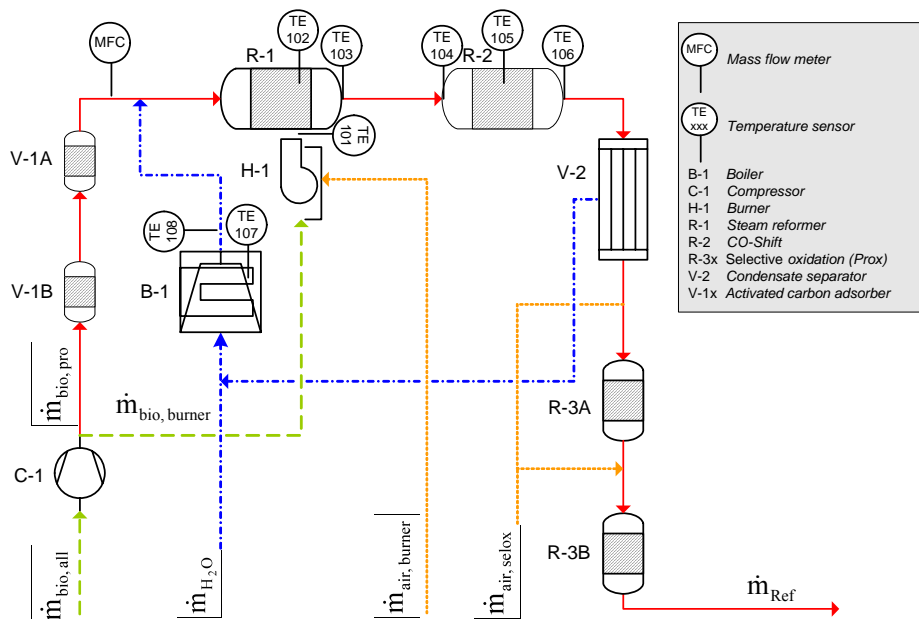


Figure 1. Mass flow scheme of the fuel processor

The fuel processor integrates gas compression and desulphurisation as well as steam generation and has internal heat recovery. The raw gas is compressed to 720 mbar by the compressor C-1. The gas flow is then separated. The combustion gas is fed into the

atmospheric burner H-1 and the process gas is fed into two activated carbon filters that are impregnated with copper and adsorb hydrogen sulphide. The purified gas passes through a mass flow controller (MFC) and, en route, is mixed with steam at a S/C ratio of 3.5 (steam to carbon ratio, carbon from methane). The mixture is preheated in a heat exchanger, induced into the steam reformer R-1 and passes through the ring shaped reforming catalyst (G-90B). Here it is heated up by the burner H-1 and by the counter current flow of reformat which is transported through the inner cylinder. Thereby a temperature of about 720 to 840° C is obtained at the outlet of the catalyst bed (TE-103). The reformat passes over a heat exchanger, is directed through both shift converter stages at a temperature of 250 to 300° C and cooled. Cooling to approximately 30° C results in condensate formation, which is trapped (V-2). Following this, the reformat is fed to the selective oxidation, consisting of the water-cooled catalysts R-3A und R-3B (50 to 60° C).

The reformat with a dew point of 40 to 45 °C is supplied to the fuel cell via a heated hose. Depending on the position of the valve, it is either passed into the anode chamber of one of the two fuel cell stacks or through a bypass directly to the outlet. In the fuel cell stack it is fed through the flow fields of the bipolar plates to the polymer electrolyte membrane (Primea 5621 series, 207 cm²). Operating pressure is 1.3 bar and operating temperature is 45 to 48 °C.

2.3 Gas Analysis

The gas measurement equipment consists of an online gas analyser by Pronova (Berlin) and a drum gas meter to record the amount of supplied gas. Process gases are taken from three sample ports in order to analyse and record the composition of biogas, reformat and anode off-gas (H₂, CH₄, CO₂, CO). The volumetric concentration of CH₄, CO₂ und CO is determined by infrared spectroscopy and the concentration of H₂ is measured by a thermal conductivity detector. The total biogas volume flow is measured with a drum gas meter and the process biogas volume flow is recorded by the thermal mass flow controller of the fuel processor.

2.4 Experimental Procedure

65 experiments in total are conducted in 5 test series to evaluate the reformer, the fuel cell and the entire system (Schmersahl, 2004). To examine the reformer, the influence of selective oxidation on the composition of the reformat and the operational behaviour for biogas with different methane content are analysed. For this, the selox air volume flow and the reformer load (process gas mass flow) are varied. The methane conversion rate u of the fuel processor is calculated as follows:

$$u = \frac{\dot{m}_{\text{CH}_4,\text{c}}}{\dot{m}_{\text{CH}_4,\text{pro}}} \quad (1)$$

where: $\dot{m}_{\text{CH}_4,\text{c}}$ = methane converted
 $\dot{m}_{\text{CH}_4,\text{in}}$ = methane input to process

The fuel processor efficiency η_{ref} can be defined as:

$$\eta_{\text{Ref}} = \frac{\dot{m}_{\text{H}_2,\text{ref}} \cdot (h_{\text{LHV}})_{\text{H}_2}}{\dot{m}_{\text{CH}_4,\text{in}} \cdot (h_{\text{LHV}})_{\text{CH}_4}} \quad (2)$$

where: $\dot{m}_{H_2,ref}$ = hydrogen output
 $\dot{m}_{CH_4,in}$ = methane input
 $(h_{LHV})_i$ = Specific enthalpy of component i

For the fuel cell, polarisation curves for various compositions of biogas are recorded and investigations into the influence of excess hydrogen flow are undertaken. Operational behaviour and performance of the fuel cell are described by polarisation curves, indicating the relationship between voltage and current density. The current density i , relevant for physical size and costs, is defined as the ratio of the current I_{FC} to the membrane surface area A_C . The thermoneutral maximum voltage of 1.25 V refers to the lower heating value of hydrogen:

$$V_{C,rev} = \frac{-(h_{LHV})_{H_2}}{2 \cdot F} = 1.25 \text{ V} \quad (3)$$

where: $V_{C,rev}$ = reversible maximum cell voltage
 $(h_{LHV})_{H_2}$ = specific enthalpy of hydrogen (= -241.8 kJ/mol)
 F = Faraday constant (= 96487 As/mol)

This leads to a practical definition of cell efficiency as a function of the cell voltage V_C :

$$\eta_c = \frac{V_C}{1.25V} \quad (4)$$

The arrangement of individual fuel cells comprises the fuel cell stack. From the point of view of fluid dynamics, the cells are in parallel connection whereas the electric connection is in series, so that the voltage of the single cells is added to V_{FC} . As not all of the supplied fuel is converted by the cells, efficiency loss occurs. Fuel utilisation μ_f can be defined as:

$$\mu_f = \frac{\dot{m}_{H_2,u}}{\dot{m}_{H_2,ref}} \quad (5)$$

where: $\dot{m}_{H_2,u}$ = hydrogen utilised
 $\dot{m}_{H_2,ref}$ = hydrogen input

Equation 6 gives the stack efficiency η_{BZ} for a fuel cell with z individual cells:

$$\eta_{FC} = \mu_f \cdot \frac{V_C}{1.25V} \quad (6)$$

The gross electrical system efficiency η_{Sys} is given by the ratio of effective electrical power P_{el} to fed fuel not accounting for auxiliary energy consumption:

$$\eta_{Sys} = \frac{P_{el}}{\dot{m}_{CH_4,in} \cdot (h_{LHV})_{CH_4}} = \eta_{Ref} \cdot \eta_{FC} \quad (7)$$

3. RESULTS

3.1 Fuel Processor

The PEMFC requires a fuel largely free of carbon monoxide. In order to attain a CO content of < 10 ppm, the reformat is purified by selective oxidation with atmospheric oxygen. Reformat entirely free of carbon monoxide is obtained by adding a volumetric air flow rate of more than 5% of the reformat flow rate. Stochastic CO maximum values of > 250 ppm occur when less than 2.5% air is added. Increasing the air flow rate decreases the hydrogen and carbon monoxide content in the reformat. The slight decrease in CO_2 content can be explained by dilution. The strong reduction in hydrogen concentration is caused by the reaction of hydrogen with surplus atmospheric oxygen. The air flow control of the fuel processor does not give a fixed reformat/air ratio. At lower reformer load, increasing excess air causes a minimally inferior reformat quality. For the further operation of the test stand, an air supply of 5% of the reformat volume flow is adjusted.

A high hydrogen output requires a high methane conversion rate. Measurement results show a decrease in methane conversion rate with increasing reformer load indicating insufficient catalyst activity. The lowest methane conversion rate of 74% for biogas is acquired at full load with 65% methane. At 30% load, the conversion rate increases to about 90% for all compositions of biogas. Reasons for the low conversion rate are assumed to be the relatively low reforming temperatures of $700 - 740$ °C (figure 2).

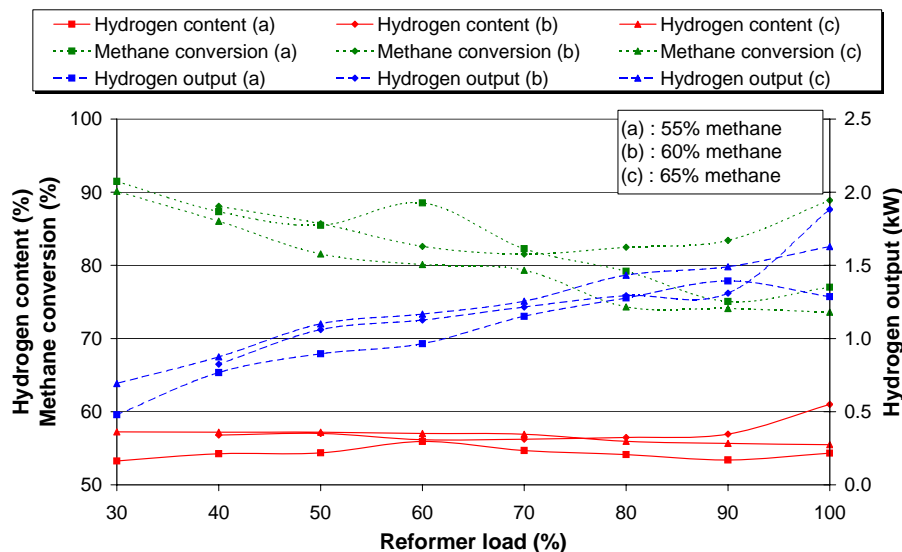


Figure 2. Methane conversion and hydrogen output for different methane content in biogas

Measurement results are represented as an average value over 5 minutes. The reformat composition is not constant. The concentration of hydrogen fluctuates by a maximum of 4.5 percent by volume within 5 minutes.

Reforming results in different concentrations of hydrogen in the reformat depending on the methane content of biogas. Measurements show an average hydrogen content of 53% (55%

CH₄), 55% (60% CH₄) and 56% (65% CH₄). The peak value for biogas with 60% methane was obtained at a higher reforming temperature of 840 °C at full load.

3.2 Fuel Cell

The performance of fuel cells is characterised by polarization curves. Increasing current density lowers the voltage and hence decreases cell efficiency. This leads to difficulties in determining the rated power, since there is a trade-off between high output and efficiency.

The characteristic curves of the entire operating stack for different compositions of raw gas (55% to 65% CH₄ content) show currents barely deviating from each other at current densities of up to 0.25 A/cm² (figure 3). With increasing current densities the different hydrogen concentrations in the reformat, caused by different methane concentrations in the biogas, result in increasing voltage differences. Thus, the test stand attains a maximum power of more than 600 W_{el} with biogas containing 65% methane. At a methane content of 55% only 500 W_{el} is obtained, then the voltage of the weakest individual cell falls to below 450 mV leading to an automatic breakdown of load operation.

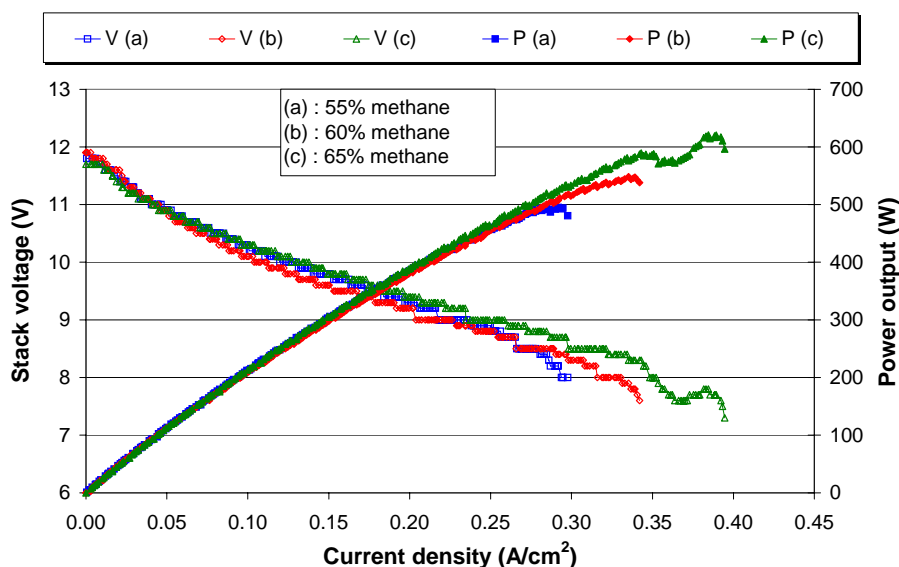


Figure 3. Polarisation curves and power output for different methane content in biogas

The characteristic curves of the single cells display significant differences. A comparison of the most efficient cell 7 with the weakest cell 2 shows a difference in cell voltage increasing over the entire range of performance. At a current density of 0.2 A/cm², the difference amounts to approximately 80 mV. At a current density of 0.35 A/cm² the difference increases to more than 100 mV, corresponding to a difference of more than 8 percent in efficiency. This is assumed to be due to irregular flow conditions (figure 4).

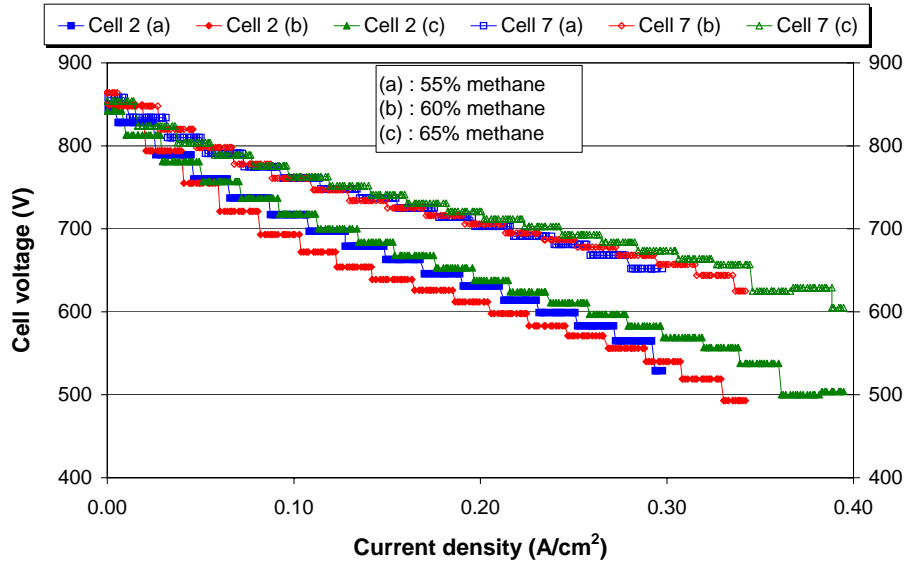


Figure 4. Polarisation curves for individual cells

Only part of the supplied hydrogen is utilised in the fuel cell. The hydrogen overflow ratio α_H is the ratio of the supplied hydrogen to that used; it is the reciprocal value to the fuel utilisation rate μ_f . Its influence on the cell voltage is slight up to a critical limit and depends on the current density. At $\alpha_H < 1.25$ (20 A) or $\alpha_H < 1.4$ (40 A) the cell voltages of single cells collapse, whereas other cell voltages show only slight changes.

A higher methane content in the biogas and thereby a higher hydrogen content in the reformat allows for a smaller hydrogen overflow ratio. Adjusting a constant hydrogen overflow ratio is not possible for the existing test stand, as the reformats pressure and hydrogen content fluctuate. This lowers fuel utilisation and thereby decreases electrical efficiency.

3.3 Power Balance

The energetic analysis requires a model which allows the necessary values to be calculated from the measured volume fractions, volume flow rates and electric quantities (**fig. 5**).

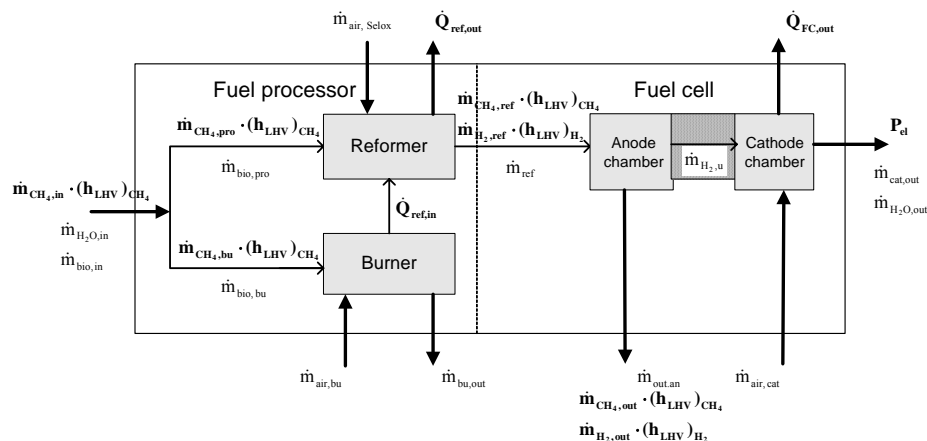


Figure 5. Simplified mass and energy flow scheme

The conversion of volumetric flow rates into mass flow rates needed for this assumes an ideal gas as well as standard temperature and pressure.

The hydrogen mass flow utilised is $\dot{m}_{\text{H}_2,\text{u}}$ and amounts to (Winkler, 2002):

$$\dot{m}_{\text{H}_2,\text{u}} = \frac{I_{\text{FC}} \cdot z \cdot M_{\text{H}_2}}{2 \cdot F} \quad (8)$$

where: I_{FC} = Amperage
 z = Number of cells
 M_{H_2} = Molar mass of hydrogen

Taking into consideration the measured volume fractions the reformat hydrogen mass flow is calculated as follows:

$$\dot{m}_{\text{H}_2,\text{Ref}} = \frac{x_{\text{H}_2,\text{ref}} \cdot (1 - x_{\text{H}_2,\text{an}})}{x_{\text{H}_2,\text{ref}} - x_{\text{H}_2,\text{an}}} \cdot \dot{m}_{\text{H}_2,\text{u}} \quad (9)$$

Where: $x_{\text{H}_2,\text{ref}}$ = Hydrogen fraction of reformat
 $x_{\text{H}_2,\text{an}}$ = Hydrogen fraction of anode off-gas

A power balance is established for different operating points to assess the performance of the test stand. The measurements show a gross electrical system efficiency η_{sys} of 12 % at a current density i of 0.29 A/cm² and a fuel utilisation μ_f of 70 %. At partial load the efficiency is $\eta_{\text{sys}} = 11$ % at $\mu_f = 62$ %.

The reason for the low system efficiency is to be found in the inefficient fuel processor with a thermal efficiency of less than 38% as well as in the process design, as a large part of the chemically bound energy leaves the system unused as anode off-gas.

A particular process design such as the one developed and tested at the Bergakademie TU Freiberg uses the anode off-gas to supply thermal energy to the reformer (Walter, 2001). For this design an estimate was made of the attainable gross system efficiency η_{sys} for the generation of electricity from biogas with PEM-fuel cells based on the experimental results and reliable published data.

Besides the efficiency rates already described, the DC/AC conversion needed to feed into the grid is taken into account assuming an efficiency $\eta_l = 0.95$. Calculations are made for $\alpha_{\text{H}} = 1.2$ (model 2 and 4) and $\alpha_{\text{H}} = 1.4$ (model 1 and 3). This corresponds to a fuel utilisation rate of 71% or 83% respectively. The fuel processor efficiency is estimated at 68% (model 1 and 2) according to manufacturer's specifications and 80% (model 3 and 4) according to published values. Operation points are determined on the basis of the polarisation curve of cell 7 with $U_z = 670$ mV at 0.3 A/cm² and $U_z = 720$ mV at 0.2 A/cm² (cf. figure 4). Assuming a methane conversion rate of 97%, the electrical efficiency is calculated with:

$$\eta_{\text{sys}} = (1,03 + (1 - \mu_f) \cdot \eta_{\text{ref}}) \cdot \eta_{\text{ref}} \cdot \mu_f \cdot \frac{U_c}{1,25\text{V}} \cdot \eta_l \quad (10)$$

The result is a maximum gross electrical system efficiency of 39% to 42% (table 1).

Table 1: Efficiency of an optimised PEMFC system for biogas (simulation)

Model	1	2	3	4
Input values				
η_i (%)	95	95	95	95
η_{ref} (%)	68	68	68	80
μ_f (%)	71	71	83	71
V_C (mV)	670	720	670	720
Results				
η_{sys} (%)	30	32	33	37

4. CONCLUSIONS

The suitability of PEMFC technology for generating electricity from biogas has been experimentally confirmed for the first time. Measurements taken at a 650 W_{el} test stand exhibit a cell efficiency of $\leq 58\%$ at a current density of 0.2 A/cm². However, this value is only achieved by individual cells. Cell voltages vary considerably. Differences increase with increasing power output and lower excess hydrogen overflow. The reasons for this are assumed to be the irregular gas flow and cell humidification and can thereby be regarded as a constructive problem of the fuel cell stack.

The fuel processor proves to be a particularly problematic component with a thermal efficiency of approximately 38% and a methane conversion rate of 75% to 90%, resulting in a low hydrogen yield. The influence of methane content on the energetic efficiency of the fuel cell system is small at the low power densities, which are necessary to obtain a high electrical efficiency. A calculation on the basis of the strongest single cell results in a maximum gross electrical system efficiency of 40% at a power density of 0.14 W/cm² in an optimised system using the anode off-gas to supply thermal energy to the reformer.

In summary, the assessment can be made that the system is not fit for the market. The use of biogas requires an efficient and reliable biogas fuel processor and a basic system optimisation. Once this is achieved, biogas cogeneration on the basis of PEMFCs has very good energetic prospects.

Studies are continuing in cooperation with fuel cell and fuel processor developers. The aims are system optimisation, determining the durability and degradation of components and identifying the harmful components in the biogas in order to ascertain the appropriate tolerance limits of the fuel cell system.

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NOMENCLATURE

Symbols

A_C	Membrane surface area	cm^2
F	Faraday constant (= 96487 As/mol)	
$(h_{LHV})_i$	Specific enthalpy (ref. to lower heating value)	kJ/mol
i	Current density	A/cm^2
I_{FC}	Amperage	A
\dot{m}_i	Mass flow of component i	g/h

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M_i	Molar mass of component i	g/mol	
\dot{Q}	Heat flow	kJ/h	
P_{el}	Electrical power	W	
x_i	Volume fraction of component i	%	
u	Methane conversion rate	%	
V_C	Cell voltage	mV	
$U_{c,rev}$	Reversible cell voltage		V
z	Number of cells		
α_H	Hydrogen overflow ratio		
η_{BZ}	Electrical stack efficiency	%	
η_C	Electrical cell efficiency	%	
η_I	Inverter efficiency	%	
η_{ref}	Thermal fuel processor efficiency	%	
η_{sys}	Gross electrical system efficiency	%	
μ_f	Fuel utilisation rate		

ABBREVIATIONS

CO-Shift	Carbon monoxide shift reactor
CHP	Combined Heat and power production
LHV	Lower heating value
MCFC	Molten carbonate fuel cell
MEA	Membrane electrode assembly
MFC	Mass flow controller
PEMFC	Proton exchange membrane fuel cell
Selox	Selective oxidation
SOFC	Solid oxide fuel cell
PLC	Programmable logic controller
ppm	Parts per million