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AN INNOVATIVE BURNER CONCEPT FOR THE CONVERSION OF ANODE OFF-GASES FROM HIGH TEMPERATURE FUEL CELL SYSTEMS

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Abstract Because of the limited fuel utilization in high temperature fuel cell systems, the exhaust gases from the anode of the stack contain some concentration of combustibile species (mainly H_2 and CO). An innovative burner concept for converting efficiently such type of gases is presented in this work. The development of the burner is part of the development process of a complete SOFC based micro-CHP system and there, a challenging combination of technical requirements needs to be fulfilled. The burner has to convert stably the very low-calorific value gases during steady state operation of the system and the high-calorific value reformate gas during start-up and shut-down. Furthermore, both types of gases have a very high temperature when exiting the anode, ranging from 650°C to 850°C. The presented burner design utilizes as combustion air the exhaust from the cathode of the stack and involves a single stage of combustion in the case of low-calorific value gases while in the case of high-calorific value gases, the complete process is carried out via two combustion stages.

Keywords Two-stage combustion, high temperature fuel cells

INTRODUCTION

In recent years, fuel flexible combustion systems seem to gain increased attention due to their potential economic and environmental benefits. Most often, the driving force for research and practical development in this field is the need to utilize energy sources which would otherwise be wasted like the exhaust gases from other primary conventional or “less” conventional energy production systems. One promising option for efficient residential power supply is the cogeneration of heat and power by means of a fuel cell based CHP (Combined Heat and Power) system because of reducing the consumption of primary energy and also reducing carbon dioxide emissions while increasing the overall system efficiency by generating electrical energy and heat simultaneously. Among such fuel cell systems, high temperature fuel cells, such as the Solid Oxide Fuel Cell

(SOFC) and the Molten Carbonate Fuel Cell (MCFC) can achieve high efficiency in practice, mainly because the thermal energy of the exhaust gas can be recovered [1, 2, 3].

The interest in developing CHP systems using the technology of high temperature fuel cells has increased the relevance of the study on combustion technologies, able to convert the exhaust gases from the anode of the fuel cell stack. In order to achieve a reasonable cell voltage and to protect the anode from oxidation, there is always a certain minimum partial pressure of hydrogen required, meaning that some concentration of combustible species (mainly H_2 and CO), determined by the fuel utilization, is present in the anode exhaust gas, which can vary, depending on the type of the cell itself and on the operational conditions [4].

Especially in the case of high temperature fuel cells, which have operation temperatures in the range of 650°C to 1000°C, the after-burner has to fulfill a challenging combination of technical requirements; it should be able to convert stably the very low-calorific value gases during steady state operation of the system and the high-calorific value reformate gas during start-up and shut-down. In addition, both types of gases have a very high temperature when exiting the anode ranging from 650°C to 850°C.

Different post-combustion options have been developed and tested over the years on the basis of both, non-premixed and premixed combustion processes. Voss et al [5] and Yen et al [2] investigated the premixed combustion of ultra-lean H_2/CO mixtures, highly diluted in inert gases and developed inert porous media based burners. The numerical and experimental studies of the premixed burners showed that a complete conversion of gas mixtures over a wide calorific value range was possible. Drawbacks concerning this approach are related to the complexity of such systems and to the strict requirements for process controlling so as to avoid excessively high temperatures or temperature gradients which could affect the stability of the porous materials.

A study on a catalytic burner for a high- temperature stationary fuel cell power generating system was investigated by Yu et al [4]. Three commercially available catalysts were used to study flow and combustion characteristics of this system in terms of performance and stability over various operating parameters. Catalytic combustion is a promising alternative to homogenous combustion regarding its total oxidation capability of low calorific value gases at low temperatures [6]. However, catalysts have a limited lifetime due to degradation and they are cost-intensive.

Schloss et al [7] developed a non-catalytic, diffusion type burner for post-combustion of the off-gases of SOFC stacks, fed with Diesel CPOX reformate. In comparison to a catalytic system and combustion in porous media, this burner design has several advantages. There is no need for sensitive, cost-intensive catalysts and porous media that reduce the lifetime of the system and therefore result in a higher system maintenance effort. Based on these deliberations a diffusion flame burner has been chosen in this study for complete conversion of the anode off-gases of an SOFC based system with a natural gas CPOX reformer. It provides a good compromise of flame stability, emissions and lifetime towards cost and installation space. Furthermore the integration and the control- and measurement effort of such systems is less than e.g. in premixed combustion systems.

In the current paper, the concept and first prototype design for a new burner suitable for post-combustion of the anode off-gases of an SOFC stack are being presented. Calculations that support the development of the burner as well as some first experimental results are presented.

CONFIGURATION OF A HIGH TEMPERATURE SOFC-CHP SYSTEM

The presented burner is being developed in the framework of the EU-Collaborative Project “FC-DISTRICT”, which is financially supported by the European Commission in the 7th framework program. The overall objective of the FC-DISTRICT project is to optimize and implement an innovative energy production and distribution concept for sustainable and energy efficient districts. For this purpose, decentralized co-generation of heat and electricity should be coupled with optimized building and district heat storage and distribution network. A small-scale SOFC based micro-CHP (Cogeneration of Heat and Power) unit is being developed for the needs of the project. A schematic diagram of this system is shown in Figure 1. The system is operating with natural gas that is converted via Catalytic Partial Oxidation (CPOX) to a hydrogen-rich gas. A CPOX based system has the advantage of a simple layout with lowest initial and operational costs. The nominal electrical power output of the system is 1.5 kWel and is achieved by using an advanced planar, compact SOFC stack.

The application of the SOFC technology allows the use of synthesis gas mixtures with H2 and CO as fuel. An advantage of the SOFC technology is a higher tolerance against fuel impurities [8]. The targeted efficiencies of the micro-CHP system are η=0.9 for total efficiency and η>0.3 for the electrical efficiency. The fuel cell system is able to modulate in the range of 1:3.

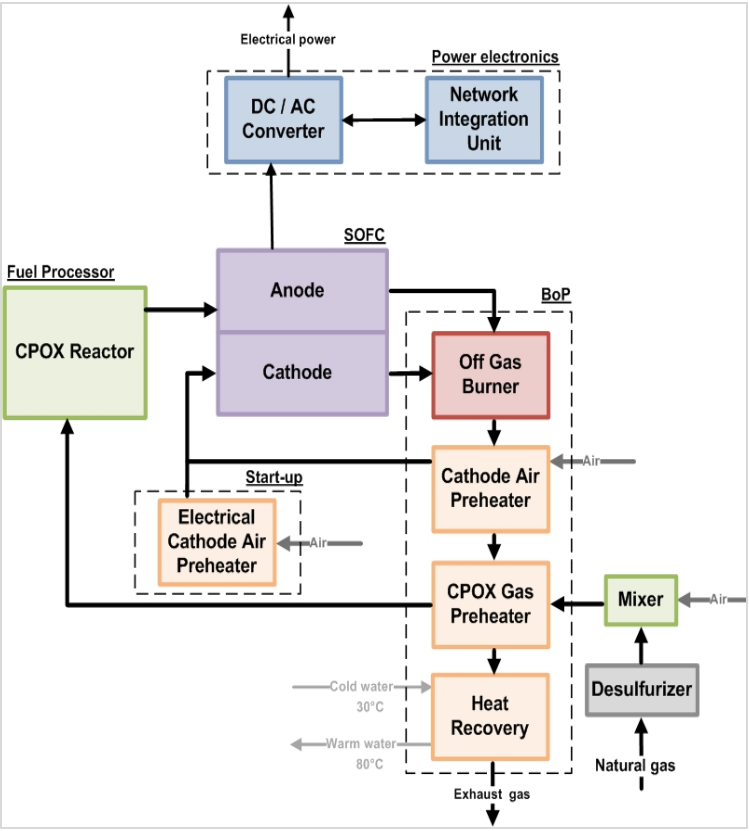

Figure 1. Schematic Diagram of FC-DISTRICT micro-CHP system

A range of innovative balance of plants components, like high temperature heat exchangers and a post combustion system are being developed for an efficient integration of the system. A new burner concept is being studied for the conversion of the anode off-gases from the SOFC stack, taking always into consideration additional requirements that arise from the integration of the burner into a larger system. The anode off-gases of a high temperature fuel cell contain chemically bound enthalpy in form of unconverted H_2 and CO and a high amount of the inert components of CO_2, N_2 and H_2O, that results in a limited chemical conversion of the combustible components when utilizing conventional combustion technology [9].

BURNER DEVELOPMENT

Process Requirements

The described burner was designed with the aim to operate over a wide range of gas compositions with different calorific values and for high preheating temperatures. Moreover, requirements for very low pressure losses over the burner had to be fulfilled as well as “near zero” pollutant emissions in all operational modes of the micro-CHP unit.

The major technical challenge for the burner development results from the different operation modes starting from combustion of weak gases like the anode off-gas during normal operation and ending with reformate gases showing high hydrogen content and calorific value. In Table 1 the composition, temperature and LHV (Lower Heating Value) of the anode off-gas and the CPOX reformate are summarized for the case of system operation with the maximum electrical stack power of 1.5 kW_{el} and the minimum possible electrical stack power of 0.5 kW_{el}, what results from the system modulation range of 1:3.

Table 1. Compositions of anode off-gas and reformate for different electrical stack powers and stack fuel utilizations

		anode off-gas				**reformate**
electical stack power [kW]		1.5	1.5	0.5	0.5	0.5 - 1.5
stack fuel utilization [%]		70	80	75	85	70 - 85
operation mode		steady-state operation				start-up/shut-down
temperature [°C]		780°C				830°C
gas composition	x_{H2} [Vol.-%]	10.7	7.2	8.9	5.4	34.3
	x_{CO} [Vol.-%]	4.7	3.1	3.9	2.3	16.7
	x_{CO2} [Vol.-%]	13.9	15.5	14.7	16.3	1.8
	x_{N2} [Vol.-%]	42.8	42.8	42.8	42.8	42.8
	x_{H2O} [Vol.-%]	27.4	31.0	29.2	32.7	3.8
LHV [kJ/Nm³]		1748	1168	1452	873	5808

Additionally, the composition of the anode off-gas is shown for different stack fuel utilizations, which are in the range of 70% to 80% and for lower electrical stack power even higher (85%). At low fuel utilizations, the concentrations of H_2 and CO in the anode off-gas increase and therefore, the heating value of the gas becomes higher. From Table 1 it is obvious, that the concentration of the combustible components, H_2 and CO, in the gaseous fuel varies in the range of 5.4 to 34.3 Vol.-% and 2.3 to 16.7 Vol.-% respectively. This results in a LHV of the gaseous fuel in the range of 873 to 5808 kJ/Nm^3 and a thermal power of the developed burner in the range of 0.2 to 1.4 kW. The temperature of the anode off-gas is approximately 780°C in steady state operation, however the temperature of the reformate, coming from the anode of the fuel cell in transient operational modes (start-up and shut-down) may vary in the range of 500°C to 850°C.

Furthermore, the cathode off-gas contains a high amount of oxygen not consumed at the cathode. Excess oxygen is present in the cathode off-gas because the percentage of oxygen fed to the fuel cell's cathode is higher than the stoichiometric amount for complete reaction with H_2 and CO so as to enhance oxygen kinetics at the cathode. Excess oxygen is also used to compensate for the effect of aging and external contaminants on the cathode. [10]

With the aim of having simple and compact overall system architecture and reducing the control and measurement effort, the design of the developed burner is based on a diffusion type process where the anode off-gases are directly combusted with exhaust gases from the cathode of the stack, which is oxygen depleted air. Thus, no additional air stream is required for this process and consequently no additional air blower, which of course acts in favor of the system simplification.

Two-stage Burner Concept

In Figure 2 the adiabatic flame temperatures are shown for different equivalence ratios φ and for representative mixture compositions of anode off-gas and reformate (see Table 1). The adiabatic flame temperatures were computed with the software package *ChemKin Reaction Design 4.1.1*, using the chemical and phase equilibrium calculation. In the case of the reformate different preheating temperatures of the gas were also examined (500°C, 700°C and 850°C). As it can be seen, for decreasing fuel utilization the adiabatic flame temperatures of the anode off-gas become higher and the same behavior is observed for higher preheating temperatures of the reformate.

The cathode air flow varies depending on the operation of the fuel cell system and is 204 Nl/min for a nominal electrical stack power of 1.5 kW$_{el}$ and a stack fuel utilization of 80%. Utilizing the total amount of cathode air for the combustion of the respective low-calorific value anode off-gas would result in an equivalence ratio of φ=0.04. In Figure 2, the grey bar indicates the range of equivalence ratios (φ=0.5-1) where complete combustion of the anode off-gas with low emissions is possible. Therefore, the required amount of air is less than 10% of the available amount. The computed maximum temperatures for these equivalence rations range from 1200°C to 1550°C.

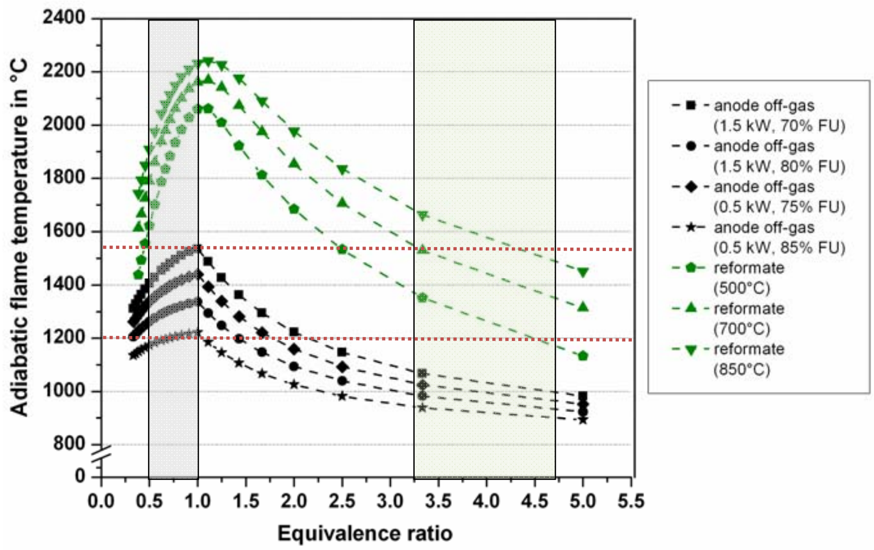

Figure 2. Adiabatic flame temperatures dependent on equivalence ratio for anode off-gas for different electrical stack powers (0.5 kW$_{el}$ and 1.5 kW$_{el}$) and stack fuel utilizations (70%-85%) and for reformate for different preheating temperatures (500°C, 700°C, 850°C)

In case of the high-calorific value reformate the same amount of air corresponds to an equivalence ratio in the range of φ=3.2 to 5, indicated with the green bar in Figure 2. This would mean under-stoichiometric combustion of the reformate with maximum temperatures ranging again from 1200°C to 1550°C. This is an interesting finding since it allows a two-staged process (see Figure 3), where in the first stage the temperature conditions would remain more or less the same regardless of the type of gas which is being combusted. Concerning the reformate, conversion can be completed in the second stage using the rest of the cathode air flow as combustion air. Of course a proper and stable air flow splitting under all operational conditions is the big technical challenge for realizing such a concept.

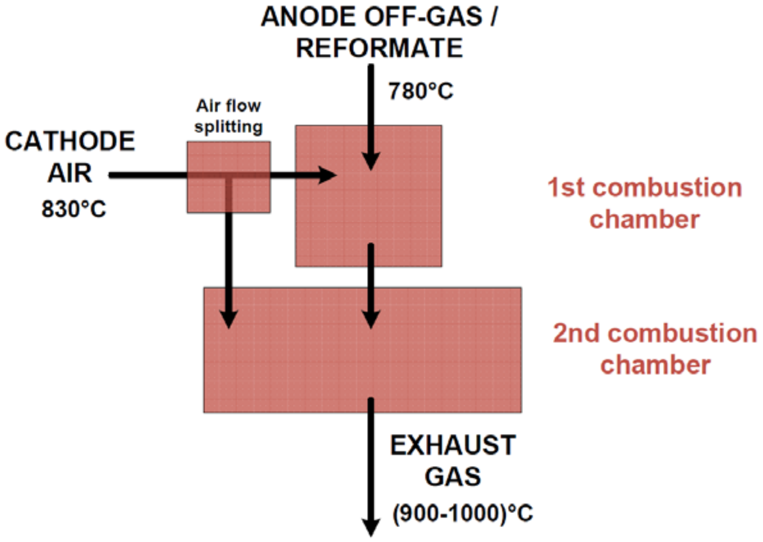

Figure 3. Schematic diagram of the two-stage burner concept

First Burner Prototype

Basic engineering of the burner prototype was performed with the support of plug flow reactor calculations with the software package *ChemKin Reaction Design 4.1.1*, using the reaction mechanism *GriMech 3.0* of Smith et al [11], in order to estimate CO and NO_x emissions.

In Figure 4 the calculated species concentration of H_2, H_2O, CO, CO_2 and NO over the burner residence time are presented for anode off-gas combustion in the first chamber (for an electrical stack power of 1.5 kW_{el} and a stack fuel utilization of 80%) and an equivalence ratio of $\varphi=0.9$, where an adiabatic flame temperature of 1330°C is reached. It can be observed that a residence time of 3.5 ms is enough for complete conversion however for longer residence times NO formation increases.

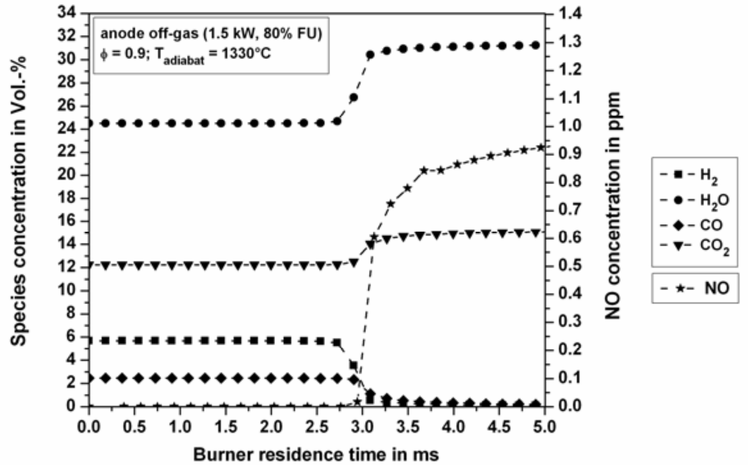

Figure 4. Calculated conversion of species (CO, H_2, H_2O, CO_2 in Vol.-% and NO in ppm) in the first combustion chamber over burner residence time for anode off-gas (780°C) with an electrical stack power of 1.5 kW_{el} and a stack fuel utilization of 80%

The concentrations of the species for the reformate gas (for an electrical stack power of 1.5 kW_{el} and a stack fuel utilization of 80%) are presented in Figure 5 and Figure 6. In Figure 5 the results for the under-stoichiometric combustion (φ=3.3) in the first combustion stage of the burner are shown. Results for the second stage of combustion are shown in Figure 6, where the conversion of major species is being completed.

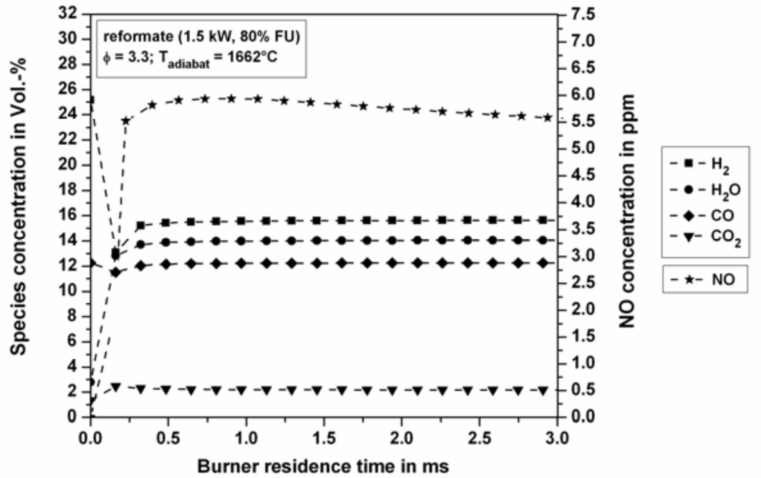

Figure 5. Calculated conversion of species (CO, H_2, H_2O, CO_2 in Vol.-% and NO in ppm) in the first combustion chamber over burner residence time for reformate (850°C) with an electrical stack power of 1.5 kW_{el} and a stack fuel utilization of 80%

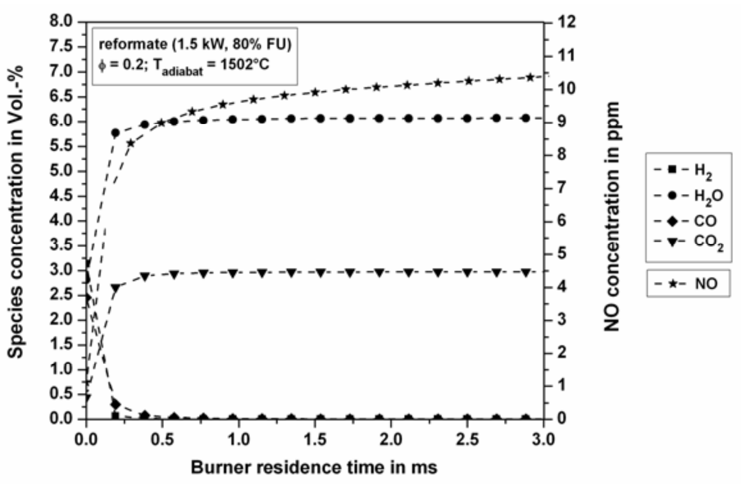

Figure 6. Calculated conversion of species (CO, H_2, H_2O, CO_2 in Vol.-% and NO in ppm) in the second combustion chamber over burner residence time for reformate (850°C) with an electrical stack power of 1.5 kW_{el} and a stack fuel utilization of 80%

Taking into consideration these results the first burner prototype was designed (Figure 7). The two-stage burner consists of two cylindrical, concentric metal pipes with inner diameters of 70 mm and 26 mm, respectively. The gaseous fuel enters the inner pipe, which forms the first combustion chamber, in axial direction from the top of the burner through a pipe with an inner diameter of 21 mm. The total cathode air enters the outer pipe in radial direction through a tangential placed pipe with an inner diameter of 26 mm. The proper splitting of the air is realized via 12 holes with a diameter of 4 mm in the upper part of the inner pipe and in the radial direction. When the air enters the burner through the tangential inlet, part of it passes directly through these holes into the inner pipe, the first combustion chamber. In order to increase this effect, a metallic plate with 10 holes (diameter 8 mm), which are arranged in a circle, was placed between the inner and the outer burner pipes at the end of the first combustion chamber, as shown in Figure 7.

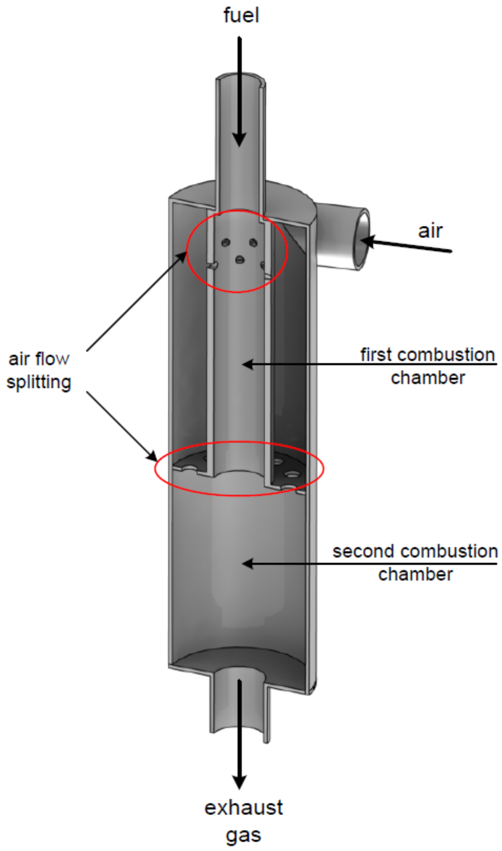

Figure 7. Cross section of the first burner prototype

In the case of the low-calorific value anode off-gas the complete combustion takes place in the first chamber, which has a length of 140 mm, and the exhaust gas is mixed with the rest of the cathode air flow before leaving the burner in axial direction. In the case of the high-calorific value reformate under-stoichiometric combustion takes place in the first chamber, since the ratio between fuel and combustion air remains the same. This means that the exhaust of the first chamber has still a calorific value and is completely converted with the rest of the cathode air flow in the second chamber, which has a length of 106 mm.

Some preliminary experimental characterization of the presented two-stage burner concept was performed with temperatures in the first combustion chamber reaching a maximum 1045°C in case of the anode off-gas and 1560°C in case of the reformate, for an electrical stack power of 1.5 kW_{el} and a stack fuel utilization of 75%. This behaviour indicates, that more than the targeted 10% of the total cathode air flow entered the first chamber. The exhaust gas analysis and the combustion temperatures show that major species were completely converted for all tested conditions and a pressure drop between 8 mbar and 9 mbar on the air side was recorded.

CONCLUSIONS AND OUTLOOK

In order to achieve high overall efficiencies and to avoid emissions of combustible or toxic gases a fuel cell system needs an off-gas treatment unit with heat regeneration. The off-gas treatment unit should be a compact and robust system with low control and maintenance effort. For this reason a two-stage diffusion type burner is under development for direct conversion of the anode off-gas with the cathode exhaust as oxidant stream. Based on numerical investigations the need of splitting the total cathode air stream was identified and a two-stage burner concept was elaborated. The first experimental results showed that a safe and stable operation of the presented burner is possible for the whole operational range of the SOFC system. However design optimization is necessary especially concerning the splitting of the cathode air flow. Minor adaptations are also required concerning the length of the burner which can be decreased further as well as the pressure losses in the component.

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NOMENCLATURE

CHP	Combined Heat and Power
FU	Fuel utilization
LHV	Lower Heating Value
MCFC	Molten Carbonate Fuel Cell
SOFC	Solid Oxide Fuel Cell
φ	Equivalence ratio

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