DEVELOPMENT OF A PORTABLE PROPANE DRIVEN 300 W SOFC-SYSTEM

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ABSTRACT
Portable power generation is expected to be an early and attractive market for the commercialization of SOFC-systems. The competition in this market is strong at costs per kilowatt, but weak in terms of electrical efficiency and fuel flexibility. Propane is considered as attractive fuel because of its decentralized availability and easy adaptability to other well-established hydrocarbons, such as camping gas, LPG or natural gas.

The Lower Saxony SOFC Research Cluster was initiated as network project to bundle the local industrial and research activities on SOFC technology. Goal is the development of a stand-alone power supply demonstrator on the basis of currently available SOFC stack technology. Electrolyte supported cells are deployed because of their good stability and robustness. Possible application areas are engine-independent power generation for recreational vehicles or off-grid power supply of cabins and boats. Further potential markets are industrial applications with a continuous demand for reliable power, e.g., traffic management, measuring systems and off-grid sensors and surveillance equipment. To cover the technical requirements of those applications, the SOFC system should provide the following features:

- propane as fuel,
- net system electrical power $\geq 300$ We
- net system efficiency $\geq 35\%$
- compact mass and volume,
- time to full load $\leq 4$ hours.

Anode offgas recycle in conjunction with a combined afterburner/reforming-unit in counter flow configuration is used for high efficient fuel gas processing without complex water treatment. All main components are in a planar design and stacked to reduce thermal losses and permit a compact set-up.

INTRODUCTION
The Lower Saxony research cluster consists of 5 institutes from the universities of Braunschweig, Hannover, Clausthal, and the University of Applied Sciences Osnabrück and 9 industrial partners. The project is aimed to introduce innovations in components and system development by an interdisciplinary team of researchers. The consortium adopted the planar design of the SOFC stack to the other high-temperature components like reformer, stack, heat exchanger and afterburner, thus enabling a compact system setup with a high degree of integration. All units are placed on top or below the stack and rigid pipe connections are avoided whenever possible to minimize space requirements and reduce thermal stresses during the heat-up and cool-down phase. In addition, arrangement and connection of the process units has to consider pressure drop and limitations concerning fabrication and system assembly. The high degree of thermal integration in conjunction with the internal recycle of anode offgas promises an electrical system net efficiency above 35\%. That would be remarkable for a small scale system in the power range $< 500$ W.

BASIC BLACK BOX CONSIDERATIONS
Prior to the detailed system design and the component specifications, a simple interactive black box model was used to prove the general feasibility of the system concept approach at different boundary conditions (see Figure 1). The Staxcra Mk200 stack is
equipped with 30 ESC4 cells of H.C. Starck Ceramics and has a rated power output of 700 W, at a fuel utilization (FU) of 75%, if operated with a H₂/N₂ mixture of 40/60 Vol.-%.

For the basic considerations a performance drop of about 5% was estimated if syngas from propane reforming was used instead of the H₂/N₂ mixture, resulting in 665 W gross electrical power output. Assuming an electrical gross system efficiency of 60% the necessary C₂H₆ input is 1,108 W (corresponding 0.022 g/s, HHV). The black box model considers heat losses via the outer system casing by calculating the convective heat flux under the assumption of a flow velocity of the ambient air of v = 0.25 m/s. The heat transfer coefficient α was estimated by

\[ \alpha = 2 + 12 \cdot v^{0.5} \]  \hspace{1cm} (1)

The heat loss is 300 W for a surface temperature of 50 °C and an area of 1.5 m². The system offgas temperature was calculated by closing the energy balance. The offgas temperature is 121 °C for a cathode air flow rate of 1.2931 g/s (corresponding 60 l/h/min).

The cathode air blower causes mainly the parasitic electrical demand of the BoP components. Overall, BoP power demand has to be less than 270 W to obtain the demanded electrical net system efficiency of 35%. For that case, an electrical net power output of 390 W results. Thus, the initial performance goals seem feasible.

Nevertheless, these figures emphasize the impact of heat loss via the surface for small SOFC systems. 27% of the supplied energy are lost in terms of waste heat at 50 °C surface temperature; 480 W is the convective heat loss at 65 °C surface temperature and the overall energy balance no longer agrees. Additional energy supply would be required to maintain a self-sustaining operation. These simple considerations illustrate the basic necessity of a high degree of thermal integration and the need for internal usage of heat fluxes, what has to be taken into account from the beginning of system design.

**Figure 1. Black box model for proof of feasibility (given input data are formatted italic)**

**SYSTEM CONCEPT**

Figure 2 shows a simplified process flow diagram of the proposed system. Propane and anode offgas (AOG) are fed to the reformer. The AOG contains H₂O, CO₂ and heat from the electrochemical oxidation of the H₂ and CO on the SOFC anode. That is used for endothermic steam- and dry-reforming of the propane. The reformer provides the fuel gas for the SOFC stack. The remaining part of the AOG is fed together with the cathode exhaust air
to the afterburner for additional heat generation. Heat is used to maintain the endothermic reformer reactions and for cathode air preheating to about 650 °C, before entering the stack.

Hot anode offgas recycling is a challenging task and no commercial hardware solution is currently available for the desired flow range. Thus, a piston pump was proposed and developed for AOG recycle. Intercooling of the anode offgas cannot be fully avoided due to the temperature limitations of the compressor bearings and seals. Thus, reheating the compressed AOG with the hot AOG from the anode exit using a tube-in-tube heat exchanger seems to be a reasonable compromise.

![Diagram of the propane SOFC system with anode offgas recycle](Image)

Figure 2. Process flow diagram of the propane SOFC system with anode offgas recycle

The lower limit of the AOG recycle rate is determined by the carbon formation boundary in the reformer for the given temperature. It is shown in previous tests that the oxygen to carbon ratio at the reformer inlet \((O/C)_{\text{ref}}\) (Equation 2) is the key figure with respect to carbon formation:

\[
\left( \frac{O}{C} \right)_{\text{ref}} = \frac{\dot{n}_{\text{H}_2} + \dot{n}_{\text{CO}}}{3 \cdot \dot{n}_{\text{H}_2}}
\]

(2)

The parameter \((O/C)_{\text{ref}}\) corresponds to the steam to carbon ratio \(S/C\), well known for steam reforming reactions. Equation 3 and 4 show the strong endothermy of the reforming reactions taken place:

- **Dry-reforming:** \( \text{C}_3\text{H}_8 + 3 \text{CO}_2 \rightarrow 6 \text{CO} + 4 \text{H}_2 \quad \Delta_R^{\text{298}} \text{H} = 622.1 \text{kJ/mol} \) (3)
- **Steam-reforming:** \( \text{C}_3\text{H}_8 + 3 \text{H}_2\text{O} \rightarrow 3 \text{CO} + 7 \text{H}_2 \quad \Delta_R^{\text{298}} \text{H} = 498.6 \text{kJ/mol} \) (4)

Figure 3 shows the equilibrium reformate composition at different temperatures based on stationary process flow sheet simulations (ChemCAD®). The flow rate ratio of AOG to propane has been kept constant at a calculated \((O/C)_{\text{ref}}\) of 1.82. Hydrocarbon conversion is almost complete for reforming temperatures above 700 °C. The fraction of \(\text{H}_2\) and \(\text{CO}\) is greater than 60 Vol.-%. Soot formation is inhibited above 720 °C for the distinct operation conditions.

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Figure 3. Reformate composition for combined steam-/dry-reforming of propane at different equilibrium temperatures, (O/C)_REF = 1.82

While the reformer and burner can be considered as Gibbs reactors (delivering thermodynamic equilibrium values), the flow sheet simulation of the overall process requires the implementation of a confirmed stack characteristic. Key figures for the stack are power output, fuel utilization and electrochemical efficiency at the desired operation point. Thus, a Staxera Mk200/ESC4 stack was evaluated in a stack-test-bench with different fuel gas compositions and throughputs. Figure 4 shows the measured U/I-curves, Table 1 summarizes the stack performance data for the different operation points.

Figure 4. Single cell U/I-curves, Mk200/ESC4 stack, 850 °C stack temperature
Table 1. Experimentally validated stack performance data, Mk200/ESC4 stack, 850 °C stack temperature

<table>
<thead>
<tr>
<th>Fuel gas</th>
<th>Composition [Vol-%]</th>
<th>Flowrate [l/h/min]</th>
<th>Results</th>
</tr>
</thead>
<tbody>
<tr>
<td>H₂/N₂</td>
<td>H₂ 58.2 N₂ 41.8</td>
<td>23.77</td>
<td>FU 65 ηₑ 40 Pₑ 719</td>
</tr>
<tr>
<td>H₂/N₂+steam</td>
<td>H₂ 57.8 N₂ 38.4</td>
<td>24.67</td>
<td>70 41 709</td>
</tr>
<tr>
<td>Reformate</td>
<td>H₂ 47 N₂ 0 CO 35</td>
<td>10.5</td>
<td>71 37 676</td>
</tr>
<tr>
<td></td>
<td>CO₂ 0 CH₄ 10</td>
<td>10.34</td>
<td>77 40 660</td>
</tr>
<tr>
<td></td>
<td></td>
<td>9.59</td>
<td>79 41 626</td>
</tr>
<tr>
<td></td>
<td></td>
<td>9.06</td>
<td>80 41.5 596</td>
</tr>
</tbody>
</table>

*FU, ηₑ and P are maximum values for each test series

As expected, a fuel gas consisting of H₂ in N₂ gains maximum stack power. A voltage drop was observed throughout the complete current range if the fuel is additionally diluted with steam. Synthetic fuel gas mixtures with a composition derived from thermodynamic simulations of propane reforming with AOG were used to determine the stack performance at the suggested operation point. The stack performance estimations that were used in the black-box model are verified by the experiment. 660 Wₑ power output was achieved at a fuel utilization of 77 %, resulting in an electrical stack efficiency of ηₑ = 40 % (for reformate operation, case b, see Table 1). The expected 5 % drop in stack performance with propane reformate syngas was confirmed.

The system concept was investigated in steady-state operation with ChemCAD™ flow sheet simulations. Figure 5 shows the results in terms of an energy flow chart for one operation point.

Figure 5: Energy flow chart for the stationary design-point
Propane with an energy content of 1,128 W is fed to the system. Recycled AOG is added, supplying 402 W of heat and chemical energy of the unused syngas components H₂ and CO to the endothermic reformer. Due to the maximum inlet temperature of the AOG-compressor the AOG will be cooled down to 350 °C and successively re-heated as far as possible in real operation. For the sake of simplicity, the respective heat losses are neglected for the energy flow chart. Additional heat flux of 295 W is needed to maintain the steam- and dry-reforming reactions and is supplied by the combustion of the remaining AOG in the afterburner that is in direct thermal contact to the reformer. Thus, chemical energy content of the syngas leaving the reformer is higher than the power of the initial propane feed. The energy content of the syngas of 1,825 W consists of 1,640 W chemical power and 185 W thermal energy. Assuming a stack fuel utilization (FU) of 75 % and an electrochemical efficiency of 54 % the gross power output is 665 Wₑ. The internal power consumption of the BoP components has to be less than 270 Wₑ for ηₑ = 35 % net efficiency, resulting in 395 Wₑ net system power.

Figure 6 shows a sketch of the assembly concept and a scheme of the gas flows. The main components are enclosed in an inner thermal insulation. Propane enters the system, is mixed with the respective amount of anode offgas delivered by the AOG compressor and fed to the reformer layer. Reformate gas passes the SOFC anode; part of the anode offgas stream is recycled to the reformer inlet, remaining AOG is fed to the catalytic burner that encloses the planar reformer catalyst. The burner exhaust gas enters the heat exchanger for cathode air preheating. Cold cathode inlet air purges the outer casing prior to the entry in the heat exchanger to assure low surface temperatures and thus reduce heat losses through the outer enclosure. The system has only two supply connections, one for the air and propane. Exhaust gas is released by an opening in the outer casing.

Figure 6. Schematic design of component alignment and simplified gas flows
REFORMER DEVELOPMENT

Supplying sufficient heat for the endothermic reforming reactions is a main task to assure a self-sustaining and soot free reformer operation. As shown in Figure 5, a heat flux of about 300 W has to be transferred from the burner section to the reformer to maintain a reforming temperature above 720 °C. A commercial metallic foil substrate is used as support for the reformer catalyst because of its good heat conductivity, what assures good heat transfer through- and in-plane. A catalyst with an activity for both, steam- and dry-reforming of propane is required. For the proof of proper catalyst choice, a catalytic coated foil package was mounted in a simple housing. For the preliminary catalyst tests, a five-layer substrate was used as support for the catalytic coating. The assembly was equipped with thermocouples (see Figure 7) and placed inside a furnace, heated up to 850 °C and fed with a mixture of propane and a synthetic AOG. The furnace simulates the heat supplied by the anode offgas burner. Reformer gas composition and reformer temperatures are determined for different feed compositions and flow rates. Figure 8 shows the reformer outlet temperature and the methane fraction for different input compositions.

![Figure 7. Test set-up for preliminary test of catalytic activity, detail: cross-sectional area of the 5-layer metallic support](image)

The methane fraction can be used as indicator for the degree of conversion. The catalyst is well capable of converting propane if sufficient heat is supplied. For propane flow rates above 0.4 lb/min the reformer temperature falls short of the soot formation boundary. To avoid carbon formation, the tests were terminated at propane flow rates of 0.5 lb/min.
Figure 8: Temperature and CH₄ concentration (dry basis) at reformer exit with different C₃H₈ flow rates, furnace temperature: 850 °C

Good conversion rates at low propane throughput and high reforming temperatures show, that the catalyst is suited for a combined steam- and dry-reforming of propane. For further improved heat transfer through-plane a catalyst substrate with only one layer was chosen for the final reformer set-up and coated with the examined catalyst. The substrate foil has a sheet thickness of 50 μm, resulting in a porosity of 400 cpsi (see Figure 9). The foil package has an overall thickness of 1.41 mm.

Figure 9: Single-layer metallic support for the reformer catalyst

The reformer unit was designed as a joint-part coupling the catalytic afterburner and the reformer layer in a sandwich-design, with the catalytic coated reformer substrate located between two burner plates (Figure 10). A commercial oxidation catalyst was used as burner catalyst, arranged as randomly packed bed above and below the reformer unit.
SYSTEM SETUP

Figure 11 shows a 3D-sketch of the complete system setup. Part of the insulation, the stack compression system and the outer casing are removed for a better view. The AOG-compressor together with the tube-in-tube heat exchanger for the AOG is separated from the hot components by a thermal insulation. The reformer-burner-unit is located on top of the SOFC stack with an adapter plate in between to direct the gas streams in the right manner. Another insulation plate separates the stack from the heat exchanger compartment. The plate heat exchanger is designed as two-stage unit, with a low temperature zone made from alumina and a high temperature unit from Crofer22APU. High temperature steel tubes with bellow compensators connects the burner outlet and the heat exchanger inlet. All other components are connected directly or via the adapter plate.
CONCLUSION AND OUTLOOK

A concept for a SOFC portable power unit in the power range ≥300 W_s,net using current stack technology has been presented. A simple black box model proves the general feasibility of the approach and indicates, that an electrical net efficiency ≥35% is possible using anode offgas recycling and a high level of thermal integration by planar design of the main components promises. The Mk200 stack is well capable of meeting the system requirements and the planar reformer design with a single-layer metal foil as catalyst support and the chosen catalyst has been proven to work under the ambitious conditions of steam- and dry-reforming of propane with AOG. Next step will be the commissioning of the system and the detailed characterisation at different operation points.

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