Results of the measurement of SOFC fuel cell stacks under pressure conditions

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Abstract
Results from the measurement of SOFC fuel cell stacks under pressure conditions are presented. As part of a measurement campaign, the operation of a stack system is investigated, particularly under the operating conditions of a recuperated micro gas turbine. Above all, the performance and effectiveness of selected stack types at various operating pressures and operating temperatures are measured. With the test facility set up for such investigations, cell systems could be examined under atmospheric conditions and with pressures of up to 5 bar.

It is shown that in operating conditions under pressure, the output of the fuel cell systems are improving. From a gauge pressure of 4 bar, the performance curve is flattened and higher pressures only produced a marginal increase in performance. Furthermore, the cells tested at overpressure show a steady-state behavior more quickly under load change requirements than in atmospheric operation. This means that more flexible operating modes with faster response behavior can be realized. By choosing a suitable operating temperature, the efficiency of the system is further increasing. Care was taken to select the operating conditions of the cell systems so that coking can be prevented.

Finally, a statement can be made about the pressure dependence of the fuel conversion rate. Parasitic reactions at the anode can be related to the power output. The tests carried out have shown that the high-temperature fuel cell is a promising service provider of the future. A combination of SOFC high-temperature fuel cells and micro gas turbines in one machine could, in addition to the internal provision of the required pressure, also lead to better dynamics of the entire system and increase the energy yield from the primary energy source.

Keywords: Fuel Cell, MGT-SOFC, Pressurized System, Measurement data

1. Introduction
In a fuel cell, the chemical energy of the fuel is not converted into electrical energy by releasing heat, but directly into electricity through an electrochemical process. This so-called cold combustion has the
advantage that losses are significantly lower, as the diversion of energy conversion via heat generation and subsequent work output to generate electrical energy is replaced by such a direct process with lower heat loss. Highly efficient fuel cells are e.g., SOFC (solid oxide fuel cells). These are high-temperature fuel cells which use membranes made of yttrium-stabilised zirconium oxide as a solid electrolyte. This type of fuel cells shows an electrical efficiency of up to more than 50% at operating temperatures around 700°C under atmospheric conditions.

Simulations show that an increase in the operating pressure of SOFC fuel cells can increase cell performance. As a result, hybrid energy converters can be developed which make better use of primary energy on the basis of micro gas turbines, whose combustion chamber has been replaced by an SOFC fuel cell stack grape. By reusing the fuel cell exhaust air with an afterburner chamber for the conversion of residual fuel in a turbine, the electrical yield and the electrical efficiency of the entire system can be further increased [1-5].

Such an increase in efficiency can be used in hybrid systems consisting of micro gas turbines and high temperature fuel cells (MGT-SOFC) to develop small, decentralized energy converters with high electrical efficiency [6]. In this context, electrical efficiencies in continuous operation of 65% and more can be achieved in optimal utilization of the primary energy input. At the BTU Cottbus-Senftenberg, Germany, a research project on the development of such efficient energy converters is currently being completed under the development name TurboFuelCell [1, 6].

In order to demonstrate the increase in efficiency, a test facility has been developed at the LS VFA to investigate SOFC fuel cells under pressure conditions. Individual fuel cell stacks can be supplied with fuel and the load behavior at different operating temperatures and pressure ranges can be investigated. In this investigation, measured values and analyses up to a maximum operating pressure of 5 bar absolute are presented on the response behavior, power output and efficiency of the fuel cell conversion. The investigated systems are operated with pure hydrogen.

2. Determination of performance and efficiency of fuel cells
In an SOFC fuel cell, the chemical energy of the fuel is directly converted into electricity by electrochemical processes. Such a process, called "cold combustion", offers the advantage that losses are significantly lower. For their electrochemical reactions, such fuel cells use hydrogen as fuel, which is supplied to them either directly or as a fossil pre-product. In addition to hydrogen, air is supplied as a second operating medium (oxidizer), from which the oxygen is to react with the hydrogen. Both reaction partners are separated from each other by means of an ion-conducting material, the electrolyte. Yttrium-stabilized zirconium oxide in the form of a solid oxide membrane is used in SOFC systems. This membrane is permeable to oxygen ions O²⁻. In the process, the O²⁻ ions migrate through the membrane after absorbing electrons on the cathode side and react on a catalyst layer of nickel with electron-donating H⁺ ions on the anode side to form water. The electron migration can be used as electric current via the connected electrodes [7-9].

\[
\text{Anode:} \quad 2 \text{H}_2 + 2 \text{O}_2 \Leftrightarrow 2 \text{H}_2\text{O} + 4 \text{e}^- \quad E_0=0 \text{ V} \tag{1}
\]

\[
\text{Cathode:} \quad \text{O}_2 + 4 \text{e}^- \Leftrightarrow 2 \text{O}_2 \quad E_0=1.23 \text{ V} \tag{2}
\]
In the process, the $\text{O}_2^-$ ions migrate through the membrane after absorbing electrons on the cathode side and react on a catalyst layer of nickel with electron-donating $\text{H}^+$ ions on the anode side to form water. The electron migration can be used as electric current via the connected electrodes [7-9].

These normal potentials are only valid for standard conditions (T=25 °C and p=101325 Pa). The difference between the normal potentials of both electrodes is called reversible cell voltage or open-circuit voltage.

$$
\Delta E_0 = E_{0,\text{Cathode}} - E_{0,\text{Anode}}
$$

The reversible cell voltage can be used to determine the GIBBS free reaction enthalpy ($\Delta G_0$). This indicates how large the useful energy is related to the mole of fuel gas.

$$
\Delta G_0 = -z \cdot F \cdot \Delta E_0
$$

The reaction determines the number of electrons involved ($z$). For the reaction in fuel cells, this has a value of 2, resulting in the following free enthalpy

$$
\Delta G_0 = -237.13 \text{kJ/mol}
$$

Conversely, this equation can also be used to calculate the reversible cell voltage $E_0$ using the Nernst equation [7].

$$
E_0 = \frac{-\Delta G_0}{zF}
$$

With the Faraday constant $F$. Since the electrical voltage of a single cell with $\Delta E_0=1.23$ V is very low and not sufficient for technical applications, the operating voltage can be increased by connecting several fuel cells in series. In the stacks investigated here, 30 cells are connected in series.

An ideal voltage is only present in no-load operation. The theoretical value is never reached, since voltage losses occur, e.g., in case of reaction inhibitions and insufficient gas diffusion. Therefore, values between 0.9V and 1V are more likely to be measured under no-load conditions. Furthermore, voltage losses are caused by activation losses, whereby $\text{H}_2$ molecules give off electrons at the catalyst and form protons. Similarly, ohmic losses are caused by the voltage drop at the membrane or the electrodes on the fuel cell. In the diffusion-controlled area, a further voltage loss is caused by the drop in concentration shortly before the gas escapes from the cell area.

2.1 Fuel cell efficiency [7]

The efficiency of the fuel cell $\eta_{FC}$ shows the ratio of supplied chemical energy and generated electrical energy, which summarizes the losses inside the fuel cell.

$$
\eta_{FC} = \frac{\text{electrical power}}{\text{chemical performance in used hydrogen}}
$$

This can be represented in an efficiency chain, so that the internal losses can be more specifically attributed to their origins and the Fuel Utilization of Hydrogen in the Fuel Cell $F_U$:

$$
\eta_{FC} = \eta_{\text{SOCFC}} \cdot \eta_u \cdot \eta_{H} \cdot \eta_{F_U}
$$

A theoretical efficiency $\eta_{th}$ can be defined by analysing the fuel cell reaction:

$$
2\text{H}_2(g) + \text{O}_2(g) \Rightarrow 2\text{H}_2\text{O}(l)
$$

Since a free enthalpy of the reaction of $\Delta G_0 = -237.13 \text{kJ/mol}$ and an enthalpy under standard conditions of $\Delta H = 285.83 \text{kJ/mol}$ can be determined, regarding the theoretical efficiency follows:

$$
\eta_{th} = \frac{\Delta G_0}{\Delta H} = \frac{237.13 \text{kJ/mol}}{285.83 \text{kJ/mol}} = 82.96\%
$$
These 82.96% represent the theoretical maximum which can be achieved with the reacting substances $\text{H}_2$ and $\text{O}_2$. By means of the voltage efficiency $\eta_U$, the internal losses due to electro catalysis, electrolyte and cell design are shown. This can be calculated by means of the ratio of clamp voltage to open-circuit voltage:

$$\eta_U = \frac{U_{\text{cell}}}{E_0}$$  \hspace{1cm} (12)

$U_{\text{cell}}$ is the voltage applied to the cell at the current operating point and always follows the model of the characteristic curve. $E_0$ is the electromotive force, which depends, among other things, on the temperature and the activity of the oxidized and reduced form. This can be represented, among other things, with its dependence on pressure, numbers of electrons transferred during the reaction $z$ and the Faraday constant $F$:

$$E_0(p) = E_0(p_0) - \frac{\Delta \nu \cdot R \cdot T}{z \cdot F} \cdot \ln \left( \frac{p}{p_0} \right)$$  \hspace{1cm} (13)

Here, $\Delta \nu$ refers to the molten change of the gas molecules ($\Delta \nu = -1.5$). Higher partial pressures thus lead to higher stresses and cause an increase in performance. The dwell time in the cells required for improved diffusion is also increased for the same mass flow.

The current efficiency or also FARADAY efficiency ($\eta_I$) indicates the ratio of the theoretical maximum gas volume $V_{\text{theo}}$, which can be determined from the measured current, to the actually consumed gas volume $V_{\text{transformed}}$:

$$\eta_I = \frac{V_{\text{theo}}}{V_{\text{transformed}}}$$  \hspace{1cm} (14)

If one now sets the theoretically possible current in relation to the actual current efficiency, one obtains the current efficiency $\eta_I$:

$$\eta_I = \frac{I}{I_{\text{theo}}} = \frac{I}{E \cdot F \cdot n_{\text{H}_2}}$$  \hspace{1cm} (15)

Voltage efficiency ($\eta_U$) and current efficiency ($\eta_I$) depend on the cells used and indicate how efficiently they convert the chemical energy supplied to them into electrical energy. Thus they form the internal or also electrical efficiency ($\eta_{el}$):

$$\eta_{el} = \eta_I \cdot \eta_U$$  \hspace{1cm} (16)

The heating value efficiency ($\eta_H$) determines the ratio of the heating value of the usable material to the heating value of the supplied mixture:

$$\eta_H = \frac{n_{\text{H}_2} \cdot \sum x_i}{\sum n_{\text{H}_2} \cdot x_i}$$  \hspace{1cm} (17)

Finally, there is the Fuel Utilization ($F_U$). This indicates how high the amount of hydrogen used in electricity generation is compared to the amount of hydrogen supplied and is expressed as the ratio of the hydrogen used to the hydrogen supplied.

$$F_U = \frac{n_{\text{H}_2, \text{use}}}{n_{\text{H}_2, \text{supplied}}}$$  \hspace{1cm} (18)

This is needed to evaluate the practical application in order to prevent depletion at the electrodes. If the fuel utilization is 100%, the oxygen partial pressure rises so high that the cell voltage collapses. Since the anode usually contains part nickel, the risk of nickel oxide formation would increase as a result. This would lead to the formation of inactive areas, which would only become available again with a subsequent reduction of the cell with a sufficiently high hydrogen concentration. As only pure hydrogen was used for the experiments, the efficiency chain can be further shortened:

$$\eta_{FC} = \eta_I \cdot \eta_{el} \cdot F_U$$  \hspace{1cm} (19)

In the investigations, the electricity efficiency was not taken into account, but the converted fuel flow was determined from the electrical power achieved. The hydrogen used for the calculation of the fuel utilization was calculated using the actual electricity flow, so that parasitic reactions are neglected. Therefore, electrical efficiency and fuel utilisation were combined into one factor.
\[ \hat{F}_U = \eta_{el} \cdot F_U \]  

(20)

Thus, the mass flows for the determination of the fuel utilization in a 30 cell-stack are as follows:

\[ \dot{n}_{H_2,use} = \frac{30 \cdot I}{2 \cdot \hat{F}} \]  

(21)

\[ \dot{n}_{H_2, supplied} = \frac{m_{H_2, norm} \cdot p_{norm}}{R_{H_2} \cdot T_{norm} \cdot M_{H_2}} \]  

(22)

### 2.2 Determination of performance

Fuel Utilization is also needed to determine the chemical performance of the hydrogen consumed:

\[ P_{chem, use} = \dot{n}_{H_2, supplied} \cdot H_{S,H_2} \cdot \hat{F}_U \]  

(23)

For a fuel cell, a characteristic curve can be drawn with which the electrical voltage falling across the current and the resulting power can be determined. Since the active area of the fuel cell also has a significant effect on the power, it is common practice to specify the voltage not via the current but via the current density as shown in Figure 1.

Since the electrical power of the fuel cell is defined as the product of current and voltage, the characteristic curve results in a point of maximum power. However, since the efficiency of the fuel cell decreases with increasing current, the areas of highest efficiency are, however, in the partial load area. The power characteristics of the fuel cell can also be determined from the characteristic curve. Thus, the point of intersection of the voltage line with the Y-axis, or voltage axis, results in the open-circuit voltage of the fuel cell. This is also called OCV (open-circuit voltage) and is lower than the theoretically achievable, reversible cell voltage due to losses at the cathode. In fuel cells, the electrical voltage does not occur directly, but takes some time to build up in the cell.

**Figure 1.** Current-Voltage characteristic of a fuel-cell [7]

### 3. Test facility [10]

At the VFA chair of the BTU Cottbus-Senftenberg, a test facility has been set up with which fuel cell stacks can be investigated under various pressure conditions and temperature influences. All operating parameters of the stack could be monitored and the respective test object could be supplied with different operating media.

For the SOFC fuel cells under investigation, an external heat source had to be integrated into a pressure vessel, with which a stack could be heated to an operating temperature of approx. 800°C. For this purpose, a commercially available ceramic furnace was used. The furnace chamber had to be insulated against the pressure vessel to prevent excess temperature on its outer wall. To monitor the temperature, various thermocouples were installed in fuel gas lines, air inlets and outlets and gas chambers. This enabled the heat distribution of the stack to be resolved in three dimensions. The control of the gas flows was realized by means of mass flow controllers, which can realize a minimum flow rate of 0.7 (mlN)/min. Afterwards the gases are led to the test stand by means of a hose system. There, the
gases are transported in scale-free pipes into the pressure chamber and further into the interior of the furnace. There they are preheated to the required inlet temperature in a 2 m long pipe bend inside the furnace and then fed to the fuel cell stack. Since the fuel cell membranes are sensitive to pressure surges, pressure sensors were installed on the anode and cathode sides. This allowed a maximum permitted pressure difference of 50 mbar to be regulated and monitored, so that the stack could be tested under constant conditions without damage. Figure 2 shows the realized test facility for the determination of pressurized SOFC Fuel Cells.

An important component of the test stand is the outer pressure vessel. This enables tests to be carried out at increased pressure. The boiler pressure is built up by blowing in approx. 300 lN/min of air and flushed for heat dissipation. A manometer and pressure sensors are used to monitor the boiler pressure. To monitor the performance development of the stack, all 30 cells of a stack were equipped with voltage sensors. This allowed a good overview of damage to individual cells during the tests. Furthermore, these always provided information about the current performance point of the stack. The regulation and control of the test stand was carried out with a LabView application. SOFC stacks from the Chinese manufacturer SOFCMAN were used for the tests. Their characteristic curves were to be validated and extended for a future characteristic map.

The current density (current intensity divided by active cell area) is often used in the presentation of results. This allows cell efficiencies of different manufacturers to be displayed. In this comparison only the stack type SOFCMAN, 64cm², 30 cells [11] was used. Therefore, the current intensity was used for comparison and classification of the measuring points.

Figure 2. Test Facility to determine pressurized SOFC-Fuel Cells.

3.1 Measurement and data acquisition
In the test facility, the SOFC fuel cell from the company SOFCman [11] was heated to an operating temperature of 700°C and the working pressure in the pressure vessel was set to operating pressure. The stack behaviour was measured in the plant at absolute operating pressures between 1bar and 5bar. After reducing the fuel cell stack with pure hydrogen, the SOFC fuel cell system is operated with the hydrogen flow for power operation on the anode side and a suitable air flow on the cathode side. The pressure in
the flow is thus set to operating pressure. To operate the stack, a current intensity was set depending on the fuel flow and the power output of the stack was recorded. Under these conditions, stack temperatures, pressure difference and stack current voltage were measured. In addition, the cell current voltage was measured for each cell of the used 30-cell stack. The fuel utilisation was calculated from the electrical results and the efficiency of the stack power was determined. The data obtained at different stack loads were analysed and graphically processed. By adjusting the operating temperature and pressure, an attempt was made to optimise the performance of the system. When using pure hydrogen as the fuel system, no further analysis of the exhaust gas composition is required, because the fuel utilisation can be calculated from the power and the heat output of the stack. This will be carried out in subsequent work when the fuel cells are operated with synthesis gas instead of hydrogen.

4. Test results

To compare the performance of the fuel cells, performance data are analyzed in particular. Thus, the increase in stack performance due to the increased operating pressure can be clearly documented. Furthermore, SOFC stacks under pressurized atmosphere show an increased response behaviour after load changes. A stationary operating state under correspondingly maximum power output is set up much faster. This is particularly advantageous when using the systems under load change conditions.

Furthermore, the efficiency of the cell systems must be assessed in particular. Here, an improvement in the utilization of the primary energy input can be seen above all through the improved conversion of the fuel.

Figure 3 now shows the performance curves of the examined fuel cells under different load requirements as a function of the operating overpressure. In relation to a power output of about 620 W maximum power under atmospheric conditions, power increases of more than 60 W/stack from 30 cells can be seen.

![Figure 3. Performance curves for different load requirements](image)

Figure 4a shows the increase in electrical efficiency at maximum stack power. The result is related to the total energy content (calorific value) of the supplied hydrogen fuel (blue). The green curve shows the efficiency related only to the fuel converted with the fuel utilization. The second graph (4b) shows the increase in efficiency when the operating pressure is increased to 3.5 bar (g). Thereby the power increases almost linearly with increase. If the demanded power is reduced, the decrease in stack power is greater than the increase with an increase in power demand. This is due to the fact that the stack has a higher operating temperature at higher power and that a cooling down at load reduction does not succeed completely to the equilibrium temperature at load increase. Thus, a hysteresis loop is formed in the power characteristic curve. This appears to be less pronounced at higher pressure.
Figure 4. a) Efficiency and power increase at increased operating pressure, b) efficiency at increased stack load [10]

Efficiencies also decrease with increasing power demand. An atmospherically operated stack at full power demand (40 A), for example, shows a drop in efficiency to just over 40% compared to an idling load of more than 80%. At higher pressures, on the other hand, an electrical efficiency of almost 98% can be determined with a very low load demand of almost 0 A. At maximum load, this is still measured at just over 50%. The fuel cell efficiency even reaches about 59%. Even when looking at the efficiency, a hysteresis between increasing load decrease and decreasing load demand can be recognized. Here too, this is less pronounced at higher pressures.

Furthermore, the response behaviour of the stacks to load changes will be investigated. When considering the voltage curve after the load requirement has been increased by increasing the stack current intensity, it can be seen that the voltage response reaches a maximum faster at the considered higher operating pressure and thus a stationary operation is ensured. If one compares the temporal tensions with the tensions reached after 15 minutes, it becomes clear that especially with higher load requirements the new operating point needs longer time to reach the maximum possible power, expressed by the highest voltage. Tests with higher operating pressure show that the lower voltage at the beginning of the load requirement does not deviate so much from the final voltage. In addition, the equilibrium voltage is usually reached earlier than 15 minutes after the start of the test. (Figure 5)
Finally, the average increase of the operating voltage at load demand in the form of demanded stack current will be considered. The increase in pressure may well reach an increase of more than 11%. This increase decreases with the pressure increase, so that at a limit pressure no improvement of the stack performance is to be expected. (Figure 6)

**Figure 5.** Comparison of the response behaviour of SOFC fuel cell stacks at different operating pressures [10].

**Figure 6.** Increase of the operating voltage for the same load requirements at increased operating pressure compared to atmospheric test conditions (load point=30A)

### 5. Analysis

Within the scope of the investigations, operating data of SOFC fuel cells at atmospheric operating conditions and at higher pressures were compared. Various partial load cases were also investigated and the influence of the operating pressure on the performance of such systems was shown.

Pressurized fuel cells provide higher performance at the same fuel mass flow compared to the same cells operated only under atmospheric pressure conditions. An increase in stack performance of up to 11% at maximum cell load was observed. This increase is particularly noticeable at high loads. Similarly, higher performance increases can also be achieved at lower load requirements. For commercial applications an optimum between stack performance (cost factor) and pressure charging can thus be found. A significant increase can only be achieved up to operating pressures of about 8 bar. At higher pressure the curve also flattens out noticeably.

The response behaviour and regulation of the fuel cell stack during pressurized charging is still much faster than under atmospheric operating conditions. A stationary operating condition can be reached faster. This is particularly advantageous for applications to compensate for load changes. Likewise, if the load is increased, the initially achieved cell voltage is higher compared to the maximum achievable voltage after a 15-minute settling period. Thus, the difference between the minimum and maximum voltage after a load change is significantly lower. Furthermore, a hysteresis of the power development between power increase and power reduction at the same operating point can be seen. The temperature
dependence of the power output of a cell stack plays a particularly important role here. In the case of a load reduction, the stacks must first cool down to a temperature relevant to the operating point in order to achieve optimum power output. This requires a longer start-up phase. This hysteresis and also the start-up time can be significantly reduced by pressure charging. This reduces the temperature dependency, especially with load changes.

Furthermore, the electrical efficiency for stack yield can be increased by up to 10% over the entire power range. In lower partial load ranges, it is evident that the pressure increase can even result in a higher efficiency increase. Thus, the selection of stack packs also requires consideration of the number of stacks and stack load and the resulting stack wear.

The pressure ranges in which an increase in performance is significant correspond to the efficient operating pressures of recuperated micro gas turbines. This demonstrates the compatibility of the two energy conversion systems, micro gas turbine and SOFC fuel cell. This can be used in hybrid systems to significantly increase the electrical energy yield from primary energy systems. In the reference [1], it was shown that an electrical efficiency of up to 65% can be achieved.

Finally, it should be noted that the tests have not yet allowed any conclusions to be drawn regarding degradation/longevity. Long-term tests are planned to be carried out for this purpose. These will be carried out in subsequent projects after the expansion of the test facility to continuous operation. For this purpose, further tests will be performed to investigate the temperature dependence of the power output.

6. Conclusion
A system for the investigation of pressure charged SOFC fuel cells has been introduced. Results from atmospheric and pressurized fuel cell tests were presented. It was shown that pressure charging can increase the performance of the investigated stacks by up to 11% and increase the efficiency by more than 10%. Furthermore, the improvement of the response behavior of a cell stack to load changes under pressure-charged operating atmosphere was determined and the reduced temperature dependence of the power output was shown. Especially in hybrid systems with recuperated micro gas turbines operating in a similar pressure range, pressure charging can be used to significantly increase the primary energy utilization.

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