A novel concept for an atmospheric solid oxide fuel cell (SOFC) hybrid system applying a sub-atmospheric gas turbine is proposed. Based on a developed process model suitable operating conditions are studied. The resulting performance is compared to different state-of-the-art SOFC system concepts. The comparison shows that pressurized SOFC/GT systems offer the highest electric efficiency. The novel concept as well as a two-stage SOFC system offer lower efficiencies, but still well above 60%. However, avoiding the complex system requirements of pressurized SOFC/GT systems, these concepts are promising for highly efficient electricity generation.

Keywords: Anode Off-gas Recirculation, Electric Efficiency, SOFC/GT, Solid Oxide Fuel Cell, Sub-atmospheric Gas Turbine

1 Introduction

According to the International Energy Agency, a continuing increase in energy demand is expected in the next decades, coupled with a further increase of renewable energy plants [1]. Even though the amount of energy generated from renewable sources has inclined over recent years, it is not expected to cover the global energy demand in a mid-term future and society will therefore still be dependent on fossil energy sources [2]. In the light of global climate change, an efficient use of these fossil resources is of utmost importance in order to limit the environmental impact [3]. SOFC-based systems are of special interest in this context due to the potential to achieve a high system efficiency for electricity generation [4]. Additionally, SOFC systems might become an important component in a future renewable energy system, where energy carriers are produced via power-to-gas concepts [5–7]. Due to the existing storage and distribution infrastructure, methane is a promising product for such a concept and therefore considered as fuel in this study.

Several concepts for electricity generation via SOFC have been proposed in literature. Simple SOFC concepts based on once-through setups combining reforming and SOFC are already being applied for residential applications [8,9]. The utilization of anode off-gas recirculation offers the potential to increase the efficiency of such systems considerably, an increase in electric efficiency by five to nine percentage points (pp) was reported in literature [10–12]. Even higher efficiencies can be reached with concepts applying two-stage conversion [13–15] or via coupling of SOFC with gas turbines [16]. Especially SOFC/GT hybrid systems have gained increasing interest as electric system efficiencies of $\eta_{el,sys} > 60\%$ can be reached for small- as well as large-scale applications without the need for a water management system, as necessary in concepts based on steam reforming [17,18]. However, while gas turbines and fuel cells are commercially available, most of the SOFC/GT hybrid system concepts remain in the demonstration phase [4,19–21]. At the beginning of 2018, Mitsubishi Hitachi Power Systems, Ltd. received the first order for its pressurized SOFC/GT hybrid power generation system, developed for the commercial and industrial market [22]. The full-scale operation was scheduled to start in February of 2019 [23]. Even though pressurized SOFC systems offer a high potential for efficient electricity production, the necessity for the implementation of a high-pressure vessel allowing for the operation of the SOFC stacks at temperatures in the range of $\theta = 800^\circ$ C makes these systems complex and costly. An alternative allowing for the utilization of advantages like high effi-
ciency, but at reduced system complexity, might be atmospheric SOFC/GT systems. However, as for most of the system concepts published so far, only pressurization and expansion of the necessary air stream was considered, the efficiency gain in comparison to pressurized hybrid systems was rather low [4, 24]. Deeper integration of the gas turbine is possible for the coupling of an atmospheric SOFC with a sub-atmospheric expansion. In this paper, such a hybrid system concept for small and medium commercial and industrial applications is proposed. In this concept, an atmospheric SOFC is arranged with a gas turbine operated at sub-atmospheric pressure to form an inverted Brayton cycle. By means of process modeling and simulation tools, the novel proposed concept is developed and compared to state-of-the-art SOFC systems including the pressurized SOFC/GT hybrid system concept.

2 Studied System Concepts

Within this study, different means of SOFC system concepts were modeled in order to allow for a comparison to the proposed novel atmospheric SOFC/GT system. In this section, the systems studied as well as the assumptions applied for modeling are presented.

2.1 SOFC Systems

The basic approach of improving the efficiency of simple once-through SOFC systems is the increase of the global fuel utilization \( FU' \) while maintaining a suitable local fuel utilization \( FU \) at stack level. Two different pathways have been discussed in literature: (i) single-stage SOFC module with anode off-gas recirculation [25–27], and (ii) two-stage SOFC module with a fuel regenerator between the modules [13–15, 28].

2.1.1 SOFC/AGR

An SOFC with anode off-gas recirculation (SOFC/AGR) allows for the utilization of unconverted fuel as well as for providing steam for the reforming process. When an AGR is implemented in the system, the global fuel utilization \( FU' \) depends on the anode off-gas recirculation ratio

\[
AGRR = \frac{\dot{n}_{\text{recycle}}}{\dot{n}_{\text{m, out}}} \tag{1}
\]

as well as on the fuel utilization in the stack \( FU \) and can be calculated by:

\[
FU' = \frac{FU}{1 - AGRR(1 - FU)} \tag{2}
\]

The AGRR is used to quantify the ratio between the molar flow recirculated \( \dot{n}_{\text{recycle}} \) and the entire anode off-gas molar flow \( \dot{n}_{\text{m, out}} \).

A simplified flowsheet of an SOFC/AGR system is depicted in Figure 1. The anode off-gas can be recirculated by means of an ejector or blower. The utilization of ejectors allows for a higher reliability, as they have few – if any – moving parts, as well as for the possibility to recirculate at high temperatures [12]. However, the fluid pressure must be boosted to drive the ejector. Moreover, the outlet flow rate and pressure cannot be controlled for a common ejector [29]. Meanwhile, when a blower is used to recirculate the gas, the flow rate can be easily controlled [12]. However, as high-temperature recirculation with a blower is still technologically challenging, in the present work a low-temperature recirculation is considered. The pressure drop in the SOFC is expected to be well below 5% [30–32]. Thus, the power demand is very small compared to the power output of the SOFC. An exemplary calculation of the energy demand of the blowers for the system configuration given in Table 4 shows that a pressure drop of 50 mbar over the SOFC would lead to a reduction in efficiency by just 0.1%. Therefore, the electricity demand of the blower was not included in the calculations. Furthermore, the pressure drop within the system is neglected entirely in order to reduce system complexity and retain the degree of freedom in order to be able to carry out parameter variations.

Different means of heat integration are included in the studied system. Besides gas/gas heat exchangers allowing for heat recovery inside the system, a gas/liquid heat exchanger for the generation of hot water at \( \theta = 60^\circ \text{C} \) is implemented.

Methane is supplied as fuel to the system and mixed with the recirculated anode off-gas, which is pre-heated in heat exchanger AGR-Hx. After mixing, the fuel stream is fed to the external reformer, where the reforming reactions take place. Due to the recirculated anode off-gas containing electrochemically produced steam, the system does not depend on an external water supply [26]. However, because of the endothermicity of the reforming process, thermal energy has to be provided, which is realized via purge gas combustion. The established industrial design for this kind of reformer would be a multi-tubular top-fired type with combustion within the shell [33].

On the cathode side, the incoming fresh air stream is fed to the heat exchanger Air-Hx1 to cool down the AGR flow to \( \theta = 160^\circ \text{C} \) while supplying thermal energy to the fresh air stream. After that, the air stream is heated up by the hot
exhaust gases of the combustion side of the external reformer using heat exchanger Air-Hx2, and is then fed to the SOFC cathode side. The cathode off-gas and the residual anode off-gas downstream of the AGR splitter are mixed and used to provide heat to the reformer as already stated.

2.1.2 Two-Stage SOFC System

The two-stage system model allows for highly efficient power generation by utilizing the anode off-gas of the first stage of stacks by regeneration of the process gas and feeding it to a second stage [15]. Therefore, the system can be operated with a high global fuel utilization \(FU\) while keeping a lower local \(FU\) in each of the two stages. The global \(FU\) in the two-stage SOFC system is given according to the following equation [13]:

\[FU' = FU_{S1} + (1 - FU_{S1})FU_{S2}\] (3)

where \(FU_{S1}\) and \(FU_{S2}\) represent the fuel utilization in the first and in the second stage, respectively.

Based on various concepts proposed in literature [13, 14], a two-stage SOFC system, also referred to as ‘cascade system’ was derived (Figure 2). As mentioned, the system consists of two fuel cell stages placed in series and a steam condenser used as fuel regenerator to remove product water from the anode-off-gas of the first stage (SOFC-1). Nakamura et al. [15] found that in order to provide an appropriate fuel inlet composition for the second stage (SOFC-2), a ratio between the cell area of the first stage to the second stage of 3:1 is necessary.

On the anode side, the fresh methane and water are supplied to the system. The methane stream is heated up to \(\vartheta = 100^\circ C\) and the water is vaporized in the heat exchangers Fuel-Hx1 and Wtr-Hx, respectively. The pre-heated methane and steam are mixed and subsequently fed to the external reformer to carry out the endothermic reforming process. The proposed external reformer is similar to the design utilized for the SOFC/AGR system, but in this case, the residual off-gas of the second stage is supplied to the combustion-side of the reformer. The anode off-gas from SOFC-1 is cooled by the regenerated anode off-gas and the fresh airflow by the heat exchangers Fuel-Hx2 and Air-Hx1, respectively. The water content in the anode off-gas of the stage SOFC-1 is removed by the steam condenser with a condensation temperature of \(\vartheta = 60^\circ C\) and a \(H_2O\) removal rate of 90%, as stated in [15]. Then, the regenerated anode off-gas is heated up to \(\vartheta = 650^\circ C\) and supplied the second stage.

The cathode side of the SOFC cascade concept is set up similarly to the SOFC/AGR model. The incoming fresh air stream is fed to the heat exchanger Air-Hx1 to cool down the SOFC-1 anode off-gas to \(\vartheta = 160^\circ C\) in order to support the subsequent condensation of water. Subsequently, the air stream is heated up by the hot exhaust gases of the combustion side of the external reformer Air-Hx2, and is then fed to the cathode side of the two SOFC stages [14].

2.2 Hybrid Systems

The hybrid systems are based on coupling of SOFC and gas turbine. The concept aims for an efficient SOFC off-gas utilization by combustion in a burner unit and expansion of the resulting hot tail gas in a gas turbine, in order to generate additional electric energy [34]. Solid oxide fuel cells are particularly beneficial for such a process concept, due to the high operating temperature of the stacks in the range of \(\vartheta = 600–1,000^\circ C\).

Process concepts available from literature can be classified into two categories: atmospheric and pressurized processes. In case of the latter one, as the name implies, the fuel cell is operated at pressurized conditions by being placed downstream of the compressor and upstream of the turbine [35]. Advantage of this concept is that efficiency is not only increased by the additional gas turbine but also due to an increase in cell voltage at elevated pressures [36, 37]. However, the necessity for a pressure chamber makes the system costly and challenging in terms of process safety. Therefore, atmospheric SOFC/GT systems were proposed as alternative. So far, in these concepts most often only the air stream is compressed, heated in a recuperator, and expanded in a turbine. While in this concept no pressure chamber is necessary, it suffers from the disadvantage that the thermal energy included in the off-gas streams can only be transferred to the air stream via a heat exchanger. Besides the need for this additional heat exchanger with high transfer area, the gases are “pumped” through the system. In case of leakage, critical gases like hydrogen and carbon monoxide can leave the system, which could be a safety issue.

Another pathway for atmospheric operation of the SOFC in a hybrid system is the sub-atmospheric operation of the gas turbine. So far, only one process concept has been proposed. Facchinetti et al. [38, 39] developed a concept which incorporates a 5 kW SOFC module operating at atmospheric pressure with a gas turbine driven in an inverted Brayton cycle mode. In their concept, the cathode and anode off-gases are expanded in separate gas turbines in order to utilize the oxygen-conducting electrolyte to produce pure carbon dioxide on the anode side by condensing water upstream the compressor.
However, if no CO₂ separation from the system off-gas is necessary, a less complex system concept might be advantageous. In the present model, only one turbine is used because the SOFC off-gases are mixed upstream of the burner-side of the reformer. Besides the resulting decrease in complexity and therefore in capital expenditures, the proposed concept avoids the injection of additional air and steam into the burner for combustion and cooling, respectively, as it is necessary in the concept of Facchinetti et al. [38, 39]. Additionally, in order to improve the performance of the proposed system model, an anode off-gas recirculation is considered. This allows to avoid water co-feeding, which was required in the concept of Facchinetti et al. Generally, the concept proposed here aims for a more simple technical realization of an atmospheric SOFC/GT system with a sub-atmospheric gas turbine.

The different hybrid systems studied in this publication are explained in more detail in the following sections.

2.2.1 Pressurized SOFC/GT

Pressurized hybrid systems based on a conventional Brayton cycle have been extensively studied [4, 16, 29, 34, 40–42]. As mentioned above, the SOFC stack is located between the compressor outlet and the turbine inlet and effectively functions as a combustion chamber. Often, AGR is also employed in such systems as a very efficient means of reforming.

The investigated model was adapted from Azizi et al. [43] and is depicted in Figure 3. In the implemented model just as in the literature process concept, the methane feed is pressurized and subsequently reformed with the anode off-gas at operating pressure. The resulting syngas is fed to the anode side of the SOFC where it is partially converted. A part of the anode off-gas is recycled while the other is mixed with the cathode off-gas and combusted in order to achieve a high temperature. The hot exhaust gas, which is also used to provide heat for the reformer, is fed to the turbine where the gas is expanded, in order to generate electricity.

On the cathode side, the incoming fresh air is supplied to the heat exchanger Air-Hx1 to cool down the AGR flow and is then fed to the compressor to be pressurized to operating pressure. The compressed air is fed to the cathode side being previously heated in heat exchanger Air-Hx2. The cathode off-gas and the residual gas downstream of the AGR splitter are mixed and combusted in the reformer shell. The combustion off-gas is expanded in the turbine, and the exhaust gas is fed to the hot-side of the heat exchanger Air-Hx2, and then leaves the process subsequent to the generation of hot water in the heat exchanger HWG.

2.2.2 Atmospheric SOFC/GT with Sub-Atmospheric Expansion

In this work, an atmospheric hybrid system is proposed (Figure 4). The feed methane is mixed with recycled anode off-gas and fed to the reformer. The resulting syngas is partially converted in the SOFC and part of the anode off-gas is recycled to the reformer while the other is mixed with the cathode off-gas, combusted and utilized to supply thermal energy to the endothermal reforming reactions. The off-gas of the hot side of the reformer is expanded in a turbine, cooled and again pressurized to ambient conditions.

On the cathode side, the fresh air is heated in the heat exchangers Air-Hx1 and Air-Hx2, and then fed to the cathode side. The cathode off-gas and the residual gas downstream of the AGR splitter are mixed and fed to the reformer hot-side. Then, the reformer off-gas is expanded in the turbine to sub-atmospheric pressure and fed into the hot-side of the heat exchanger Air-Hx2.

Upstream of the compressor, a cooler is placed. The idea of installing a cooler is to remove heat and depending on the operating conditions also material (water) from the system, which has a positive effect on the performance of the system due to a smaller gas stream being compressed at a lower inlet temperature. The exhaust gas is pressurized up to atmospheric pressure and then leaves the process.
3 Process Modeling and Simulation

An SOFC system consists of two main process steps: reformer and SOFC. In the case of hybrid systems, a gas turbine is coupled with the SOFC system in order to utilize the tail gas for generating additional electric energy [34]. The modeling approaches for the main process steps are presented in the following section.

3.1 Modeling

The modeling and simulation of the proposed SOFC and hybrid systems introduced in Section 2 were performed using the flowsheet-based simulation package Aspen Plus. The Predictive Soave-Redlich-Kwong (PSRK) property method was used for the calculation of thermodynamic properties in order to achieve reliable results for the high pressure, high-temperature processes. The material streams are specified by choosing the MIXCISLD class, so as to check whether solid carbon is formed in the process. The databank Aspen Property Pure32 was used to obtain the property data for the components present in the process.

The aim of this work is to compare several modes of interconnecting the main process steps. Due to the number of possibilities for process integration, the models are developed in a hierarchical structure, which means using subsystems (‘hierarchy blocks’). Besides offering a well-organized system, the utilization of ‘hierarchy blocks’ provides flexibility when modeling and simulating process chains. It allows for placing and arranging blocks in different layouts, limiting the modeling time and computational overhead.

3.1.1 SOFC Model

In this study, the considered SOFC systems are assumed to be fed with pure methane (CH4). On the anode side of the cell, the oxidation reactions for hydrogen

\[ H_2 + \frac{1}{2}O_2 \rightarrow H_2O \quad \Delta_{\text{f}}h^0 = -241.8 \text{ kJ mol}^{-1} \quad (4) \]

carbon monoxide

\[ CO + \frac{1}{2}O_2 \rightarrow CO_2 \quad \Delta_{\text{f}}h^0 = -283.2 \text{ kJ mol}^{-1} \quad (5) \]

the water-gas shift reaction

\[ CO + H_2O\rightleftharpoons CO_2 + H_2 \quad \Delta_{\text{f}}h^0 = -41.2 \text{ kJ mol}^{-1} \quad (6) \]

as well as the steam reforming reaction may occur:

\[ CH_4 + H_2O\rightleftharpoons CO + 3H_2 \quad \Delta_{\text{f}}h^0 = 206.3 \text{ kJ mol}^{-1} \quad (7) \]

In accordance with literature it is assumed that chemical equilibrium is reached in the fuel cell [44–46]. Therefore, in the applied SOFC model (Figure 5), the anode side is modeled by an ‘RGibbs block’. The necessary oxygen for the oxidation reactions (Eqs. (4) and (5)) is transferred from the cathode side with a ‘Sep block’.

The electrochemical and thermal properties of the SOFC are modeled in Aspen Plus using a ‘Calculator block’ that allows for the implementation of an Excel spreadsheet into the SOFC flowsheet. Within this spreadsheet, the electric current resulting from the conversion of CO and H2 is calculated by:

\[ I = 2n_{H_2+CO\text{, react}}F \quad (8) \]

where \( n_{H_2+CO\text{, react}} \) is the molar flow of H2 and CO that reacted within the cell and \( F \) is the Faraday constant. With the maximum electric current

\[ I_{\text{max}} = 2n_{H_2+CO\text{, SOFC\text{-in}}F} \quad (9) \]

the fuel utilization can be derived:

\[ \text{FU} = \frac{I}{I_{\text{max}}} \quad (10) \]

As Eqs. (4)–(6) are coupled, the Nernst’s potential can be calculated considering only the hydrogen oxidation reaction [11]:

\[ E_N = E_0 - \frac{R_m T}{2F} \ln \left[ \frac{x_{H_2\text{, SOFC\text{-out}}/C_18/C_19}}{x_{H_2\text{, SOFC\text{-in}}/C_18/C_19}} \left( \frac{p_0}{p} \right)^{1/2} \right] \quad (11) \]

In Eq. (11), \( E_0 \) is the reversible electrochemical cell voltage, \( R_m \) is the universal gas constant, \( T \) is the temperature and \( p_0 \) is the standard pressure. For the calculation of \( E_N \), the molar fractions of H2, CO and O2 at the SOFC outlet were applied [11]. The difference between the Nernst’s potential and the occurring cell voltage of a fuel cell is due to irreversible losses, which are generally referred to as overpotential. There are several reasons for the voltage drop with the three most important being activation, ohmic and concentration overpotentials.

The polarization is influenced by numerous mechanisms, which depend on operational parameters such as current density, cell voltage, temperature, pressure, gas composition and material properties. In order to limit the time needed for the calculation to a reasonable degree, a shortcut method proposed by Gandiglio et al. [47] was implemented. In this case
the loss mechanisms are summarized in a constant area specific resistance \( ASR \). With the \( ASR \), the cell voltage can be calculated applying the current density \( J \) by:

\[
V_{\text{cell}} = E_N - ASR \cdot J \quad (12)
\]

The advantage of using the \( ASR \) is that it simplifies the expression of the overall cell performance, as it gives a simple correlation between applied current density and the resulting voltage. In addition to the literature model, the temperature dependence of the \( ASR \) was considered according to an approach by Saarinen et al. [48]:

\[
ASR = ASR_0 \exp \left( \frac{E_a}{R_m} \left( \frac{1}{T} - \frac{1}{T_0} \right) \right) \quad (13)
\]

where the constant \( ASR_0 \) is the area specific resistance at the reference temperature \( T_0 = 1,123.15 \text{ K} \), and \( E_a \) is the activation energy of the electrochemical reactions. In Table 1 the values for \( ASR_0 \), \( T_0 \), and \( E_a \) applied in this study are given. They were determined from experimental data for the electrolyte-supported fuel cell stacks developed at Fraunhofer IKTS [49]. As operation temperature, the SOFC outlet temperature is used. The applied simple model is not as accurate as a complete model, but because the overpotential in electrolyte-supported cells is governed by ohmic resistances in the considered range of operating conditions, the implementation should be sufficient for a first approach to the system modeling while shortening the simulation time significantly.

The reaction enthalpy of the oxidation of hydrogen and carbon monoxide is not only obtained in the desired form of electric energy but also released in the form of heat. Therefore, a heat balance for the SOFC is crucial in order to obtain feasible simulation results. The heat streams occurring are represented by the dashed lines in Figure 5. The stream of thermal energy from the oxidation reaction is derived from the aforementioned ‘Calculator block’ for thermochemical calculations. Subtracting the heat necessary to heat the eucts to cell temperature as well as losses to the ambience, the remaining heat is applied to the product streams. Derived from this calculation is the temperature of the product streams, which is considered to be the cell temperature. Due to the thermal energy released by the oxidation reaction being temperature dependent this causes a calculation loop. Due to heat conduction, a part of the heat from the oxidation reaction is transferred from the SOFC to the reformer (\( Q_{\text{ref}} \)).

| Parameter | Unit    | Value   |
|-----------|---------|---------|
| \( ASR_0 \) | \( \Omega \text{ cm}^2 \) | 0.236   |
| \( E_a \)  | J mol\(^{-1} \) | \( 8.64 \times 10^4 \) |
| \( T_0 \)   | K       | 1,123.15|

### 3.1.2 Gas Turbine Model

In case of the hybrid systems, for both compressor and turbine, an isentropic model is used. Isentropic, mechanical and generator efficiencies of \( \eta_{\text{isen}} = 72\% \), \( \eta_{\text{mech}} = 100\% \) and \( \eta_{\text{gen}} = 98\% \) were assumed, respectively [50, 51]. For both hybrid systems (pressurized and atmospheric), the pressure ratio defined with

\[
\beta = \frac{P_{\text{comp.out}}}{P_{\text{comp.in}}} \quad (14)
\]

is used to study the influence of gas turbine operation on system efficiency.

### 3.1.3 Reformer Model

For the studied SOFC systems applying endothermal reforming most likely a catalytic reformer heated by burners would be applied [33]. This means that the reforming step is comprised of two major thermally linked steps, which are both defined by a chemical equilibrium: the endothermal catalytic reforming and the exothermal combustion. Therefore, in Aspen Plus, the reformer and burner are both modeled using the equilibrium reactor ‘RGibbs’ and are linked by a heat stream. The reformer side is considered to be isothermal with the heat being provided by the combustion side. For the combustion side the heat duty was defined to be equal to the heat demand of the reforming block and the exhaust temperature was subsequently calculated. In order to ensure a sufficient heat flux and a reasonable size of the reformer, an appropriate temperature difference between the combustion-side outlet temperature and the reforming-side is necessary. In this work, a minimum temperature difference of \( \Delta T > 50 \text{ K} \) was considered to be sufficient.

### 3.1.4 Heat Integration

The efficiency of SOFC systems is highly dependent on heat integration, as the process is operated at high temperature. Therefore, several heat exchangers are included in the different process concepts studied. In the developed model, unless otherwise specified, the minimum difference between the hot-side inlet and cold-side outlet was set to \( \Delta T = 50 \text{ K} \).

As low heat losses are crucial for SOFC system efficiency, the hot system components (SOFC, reformer) will be arranged in close proximity to each other. With the reformer being operated in endothermal mode and the SOFC in exothermal mode, a heat transfer between the two units is expected to occur due to conduction. In order to include this influence, a heat transfer of 10% of the thermal power generated in the fuel cell from the SOFC to the reformer is assumed. The heat losses to the ambient were assumed as 3% of the thermal power generated in the fuel cell. For all heat exchangers thermal losses of 10% of the transferred heat were assumed [52].

In case of the two-stage cascade system (Section 2.1.2), a steam condenser is modeled, using a separator unit ('Sep') that allows for the separation of components based on a specified split fraction. For the investigated model, the \( \text{H}_2\text{O} \) removal
rate is set to 90%, according to [15]. On the other hand, for the model of the proposed atmospheric SOFC/GT system, the condenser is modeled using a two-outlet flash unit (‘Flash2’), in which water is condensed depending on the dew point at the given operating conditions.

### 3.2 Simulation

The simulation of SOFC systems is very complex, due to various modes of integration of the processes, which cause recycles and information feedbacks resulting in calculation loops. All calculation loops in Aspen Plus were implemented with their own convergence block to get better control over the nesting and convergence criteria. It is recommended to reduce the convergence criterion by an order of magnitude with each level of nesting in order to ensure accuracy on the convergence of the outermost calculation loop. In the following, the calculation loops are listed from the innermost to the outermost nested.

#### 3.2.1 Tear Streams

When Aspen Plus is running in sequential modular mode, any recycle streams will be solved iteratively. By default, the stream to be teared is determined by the software and a corresponding convergence block is generated. Also the location is determined by a flowsheet analysis algorithm, which proved to be unreliable in certain cases. Therefore, a manual approach was favored. The ‘tear streams’ of the proposed atmospheric SOFC/GT hybrid system are listed in Table 2.

#### 3.2.2 Design Specifications

In the process of modeling the SOFC systems, the Aspen Plus native function ‘design specification’ was used extensively. It allows for the automatic adjustment of specified variables in order to achieve a constant target value, performing similarly to a feedback controller in a chemical plant. Guesses are made for the so-called ‘manipulated variables’ [52, 53]. A ‘design specification’ also causes iteration loops, as Aspen Plus repeatedly tries values for an input parameter until the target value of the specified variable is reached. A termination criterion must be specified for the target value. The ‘design specifications’ used in the base case simulations are listed in Table 3.

#### 3.3 Performance Definitions

Elaborate material and heat integration schemes of the different process steps are necessary to allow for a high SOFC system efficiency. Furthermore, different process concepts are compared in this work. This results in a high degree of complexity when evaluating the different SOFC and hybrid system concepts. Therefore, the net electric efficiency of the system [42]

\[
\eta_{\text{el.sys}} = \frac{P_{\text{el.sys, out}}}{\dot{n}_{\text{CH}_4, in} \cdot LHV_{\text{CH}_4}}
\]

(15)

with \(\dot{n}_{\text{CH}_4, in}\) as the inlet molar flow rate of methane and \(LHV_{\text{CH}_4}\) as the methane lower heating value was chosen as main indicator for system performance. The output power of the system

\[
P_{\text{el.sys, out}} = P_{\text{el,SOFC,out}} + P_{\text{el,GT,out}}
\]

(16)

is calculated from the AC net power generated by the SOFC \((P_{\text{el,SOFC,out}})\) and GT \((P_{\text{el,GT,out}})\). For the two-stage SOFC system,
$P_{d,SOFC,out}$ includes the net AC power generated by both stages.

As part of a further analysis, the combined heat and power (CHP) efficiency of the system is calculated. This indicator includes the heat $Q_{HWG}$ that can be utilized for hot water generation in the heat exchanger HWG:

$$\eta_{CHP} = \frac{P_{d,sys,out} + Q_{HWG}}{n_{CH,in}LHV_{CH_i}} \quad (17)$$

3.4 Sensitivity Analysis

In most of the published works on SOFC systems, the evaluation is focused on the performance at a standard operating point. This paper aims for the comparison of different SOFC system in terms of efficiency. Therefore, in order to evaluate the performance of the introduced concepts, sensitivity analyses of selected main design parameters were carried out using Aspen Plus. The ‘design specifications’ described in Table 3 were utilized in order to ensure reasonable operating conditions throughout the sensitivity analyses.

As the models are based on a number of parameters that have significant influence on system performance, multiple sensitivity analyses were conducted. Two parameters per analysis were chosen to be treated as independent design parameters and were varied so as to determine and discuss feasible operating conditions for each system. When conducting a sensitivity analysis all other parameters were kept constant. The baseline parameters that were used in this study to conduct the analysis and evaluation of the systems are given in Table 4.

As discussed in Section 3.1.3, for the reformer, a minimum temperature difference between the hot-side and cold-side outlet stream of $\Delta t = 50 \text{ K}$ should be ensured to allow for a sufficiently efficient heat transfer within the reformer. In Table 4, it is noted that the reformer cold-side outlet temperature for the SOFC systems was set to $\theta_{ref,out,CS} = 750 \text{ °C}$, resulting in a target temperature of hot-side outlet of $\theta_{ref,out,HS} > 800 \text{ °C}$. In case of the hybrid systems, the reformer cold-side outlet temperature was set to $\theta_{ref,out,CS} = 800 \text{ °C}$, resulting in a target temperature of $\theta_{ref,out,HS} > 850 \text{ °C}$ for the hot-side outlet. The higher temperature was chosen, in order to allow for a sufficient minimum turbine inlet temperature (TIT).

In order to define a baseline for system comparison purposes, the methane molar flow rate $n_{CH_4,in}$ for all systems was set to 1 kmol h$^{-1}$ (223 kW based on LHV). For the fuel cells developed at Fraunhofer IKTS, long-term stable operation at $J = 400 \text{ mA cm}^{-2}$ was demonstrated [49]. Therefore, this current density was used for the calculations in this study. In case of the two-stage system, the current density in the second-stage is calculated for the given stack ratio of 3:1 by:

$$I_{2S} = 3I_{1S}\frac{I_{2S}}{I_{1S}} \quad (18)$$

where $I_{1S}$ and $I_{2S}$ are the current densities of the first and second stage, respectively; and $I_{1S}$ and $I_{2S}$ the current generated by each stage.

| Parameter | Unit | SOFC AGR | Two-stage | Pressurized | Atmospheric |
|-----------|------|-----------|-----------|-------------|-------------|
| Gas Turbine | Isentropic efficiency $\eta_{i,es}$ | % | 72 | 72 | 
| | Pressure ratio $\beta$ | | 3 | 2 | 
| | Generator efficiency $\eta_{g,es}$ | % | 98 | 98 | 
| SOFC | Outlet temperature $\theta_{SOFC,out}$ | °C | 830 | - / 830 | 830 | 830 |
| | Pressure $p_{SOFC}$ | bar | 1 | 1 | 3 | 1 |
| | Area specific resistance $ASR$ | Ω cm$^2$ | 0.28 | - / 0.28 | 0.28 | 0.28 |
| | Reactant utilization $FU$ | % | 75 | - | 75 | 75 |
| | Heat transfer to reformer | % | 10 | 10 | 10 | 10 |
| | Heat losses to ambience | % | 3 | 3 | 3 | 3 |
| | Inverter efficiency $\eta_{i,es}$ | % | 95 | 95 | 95 | 95 |
| Reformer | Cold-side outlet temperature $\theta_{ref,out,CS}$ | °C | 750 | 750 | 800 | 800 |
| | Anode off-gas recirculation ratio $AGRR$ | % | - | - | 30 | 30 |
| | Methane conversion in the reformer $X_{CH_4,ref}$ | % | - | 50 | 50 | 50 |
| System | Global fuel utilization $FU'$ | % | - | 90 | - | - |
4 Results and Discussion

In the following section, sensitivity analyses for the different studied system concepts are presented. The results allow for the identification of advantageous operating conditions for the system concepts and a subsequent comparison between them (Section 4.4). Specific focus is on the evaluation of the proposed atmospheric SOFC/GT system (Section 4.3).

4.1 Sensitivity Analyses for the SOFC System Models

4.1.1 SOFC/AGR

For the SOFC/AGR system, as the name implies, the anode off-gas recirculation ratio $\text{AGRR}$ is a parameter of interest for the sensitivity analysis. On the other hand, as the SOFC module is an exothermic process, the internal methane reforming within the stack could represent a sink for thermal energy in order to limit the air-fuel equivalence ratio $\lambda$

$$\lambda = \frac{n_{\text{O}_2, \text{SOFC,in}}}{n_{\text{O}_2, \text{SOFC,stoich}}} \quad (19)$$

and therefore cathode air flow rate. Ideally, a major amount of methane could be reformed in the SOFC avoiding the usage of an external reformer. However, internal reforming can lead to considerable temperature gradients within the stacks causing thermal stress and increasing the risk of carbon formation [54–56]. In Figure 6, the dependence of the system electric efficiency upon anode off-gas recirculation ratio (AGRR) and methane conversion in the external reformer $X_{\text{CH}_4, \text{ref}}$ is plotted. Due to the mentioned drawbacks of internal reforming, a fraction of at least 50% of the fed methane is considered to be converted in the reformer.

As mentioned in Section 3, the reformer hot-side outlet temperature should be at least $T_{\text{ref,out,HS}} = 800{^\circ C}$, in order to satisfy the requirements for efficient heat transfer. The operating points which do not fulfill this constraint are marked in Figure 6 as hatched. The blacked out area represents operating points where carbon formation in the reformer or in the SOFC occurs in thermodynamic equilibrium. For an increasing share of internal reforming, the tendency toward carbon formation within the system decreases, as the partial pressure of oxygenated species is higher in the SOFC due to the occurring oxidation reactions. Figure 6 shows that the AGR has a considerably higher influence on system efficiency than the fraction of conversion in the reformer. This is due to the global fuel utilization increasing significantly with higher AGR, as can be seen in Figure 7. The highest efficiency of $\eta_{\text{el,sys}} = 59.7\%$ is reached with an external methane conversion fraction of $X_{\text{CH}_4, \text{ref}} = 50\%$ and an anode off-gas recirculation ratio of about $\text{AGRR} = 62\%$. As mentioned, a further increase of AGR would be possible. However, this would lead to a more challenging design of the reformer unit.

As the fraction of methane converted in the reformer $X_{\text{CH}_4, \text{ref}}$ does not have a significant impact on the system performance, in Figure 8 the dependence of the electric voltage and power generation by the SOFC system upon AGR is plotted at constant $X_{\text{CH}_4, \text{ref}}$.

4.1.2 Two-Stage SOFC System

The aim behind the two-stage layout – similar to anode off-gas recirculation systems – is the increase of global fuel utilization $FU^*$ in order to reach higher efficiencies. The fuel utilization within the two stages has a considerable influence on the system. As Figure 9 shows, there is only a slight influence on the system efficiency. However, the current density of the...
second stage as well as the number of stacks necessary is influenced (Figure 10). This is due to the fixed ratio between first stage and second stage being 3:1 [15]. As a result, in order to detect an optimum, a multi-variable optimization needs to be carried out. An optimization of a two-stage SOFC system is beyond the scope of this work and was therefore not implemented. The boundary conditions of the literature source [15] were assumed instead.

4.2 Sensitivity Analysis for the Pressurized SOFC/GT System Model

4.2.1 Reforming

Analogously to the SOFC/AGR concept, the AGR ratio and the methane conversion in the external reformer were varied. The results for the overall system efficiency are plotted in Figure 11. For the analysis, the parameters listed in Table 4 were used, except for the parameters varied in the plot. It can be seen that the possible operating conditions are limited. As outlined, reformer hot-side outlet temperatures of at least \( T_{\text{ref,out,HS}} = 850 \, ^\circ\text{C} \) were considered, in order to ensure an appropriate heat transfer in the reformer and feasible operating conditions for the gas turbine. Infeasible operating points with regards to this restriction are marked as hatched areas. The resulting low range of feasible operating conditions is due to the large air-fuel equivalence ratio \( \lambda \) that is needed to cool the stack at increasing global fuel utilization with increasing AGR. A comparison to Figure 6 highlights the increase in efficiency which can be achieved for hybrid systems with AGR.
4.2.2 SOFC Module

The operating conditions of the SOFC module itself has a considerable influence on the efficiency of the pressurized hybrid system. In Figure 12, the dependence of the system efficiency on SOFC operating pressure \( p_{SOFC} \) and \( FU \) is plotted. The results show that the highest efficiencies are reached at high fuel utilizations and intermediate pressures. Due to the increasing energy demand for air compression, the maximum efficiency of the studied pressurized system is in the range of \( p_{SOFC} \approx 3 \) bar.

The dependence of the electric power generated by the SOFC and the gas turbine upon \( p_{SOFC} \) and \( FU \) is given in Figures 13 and 14. It can be seen that the highest power output of the gas turbine can be reached at low fuel utilizations providing more fuel for the turbine. However, due to the lower electric efficiency of the gas turbine in comparison to the SOFC, it is advantageous to operate the SOFC close to the technical upper boundary.

4.3 Sensitivity Analysis for the Atmospheric SOFC/GT System Model

4.3.1 Reforming

Similar to the SOFC and pressurized hybrid system, the AGR ratio and the methane conversion in the external reformer were varied. In Figure 15 the overall system efficiency is depicted. It can be seen that the range of possible operating conditions is even lower than in case of the pressurized system. This is due to the large air-fuel equivalence ratio \( \lambda \) that is needed to cool the stack when recirculation is increased. The large amount of air limits the temperature increase in the exhaust gas combustion and subsequently results in a low temperature level at the GT inlet. Assuming a stack outlet temperature of \( \theta_{SOFC, out} = 830 \) °C, the highest electric system efficiency that can be achieved is \( \eta_{el,sys} = 63.6\% \).
With the aim of analyzing the influence of the AGR ratio and $X_{CH_4,ref}$ on the system performance in a larger range of operating conditions, the system efficiency for different SOFC outlet temperatures $\theta_{SOFC,out}$ is depicted in Figure 16. It is shown that higher outlet temperatures lead to a larger range of feasible operating conditions. This is due to less air being necessary for stack cooling, allowing for a higher combustion inlet temperature, which subsequently results in a higher temperature at the gas turbine inlet. For higher stack temperatures, an increase in system efficiency results, e.g., for $\theta_{SOFC,out} = 900^\circ$C a maximum system efficiency in the studied range of operating conditions of $\eta_{el,sys} = 66.4\%$ results. Higher temperatures would lead to a further increase in efficiency. However, the operating temperature of the SOFC is limited due to material constraints.

### 4.3.2 SOFC Module

As mentioned earlier, it was observed that for higher SOFC outlet temperatures higher system efficiencies can be reached. Furthermore, the fuel utilization was found to be of major importance for the efficiency of an SOFC system. Therefore, the influence of both parameters on efficiency are studied in Figure 17. The upper limit for the fuel utilization was set to $FU = 80\%$, as for higher values an increase in overpotential and in degradation rate can occur [57]. The highest efficiency, as expected, can be reached at the highest fuel utilization and SOFC outlet temperature considered. For these conditions the electricity generation in the SOFC as well as in the gas turbine are optimized. As discussed in Section 4.3.1, a further increase in efficiency is possible by increasing the AGRR.

That the efficiency of the system is mainly governed by the stack performance can be deduced from Figure 18. However, as the contour lines are angled slightly steeper in Figure 18, compared to Figure 17, an additional influence of the sub-atmospheric gas turbine is implied (Section 4.3.3).

In Figure 19, the dependence of the electric voltage generated by the SOFC upon $FU$ and $\eta_{el,sys}$ is plotted. The highest cell voltage of $V_{cell} = 0.83$ V is reached at $FU = 60\%$ and $\theta_{SOFC,out} = 900^\circ$C. This is because the hydrogen fractions at the anode outlet are higher at lower fuel utilization. Therefore, the Nernst’s potential is higher (see Eq. (11)). As the ASR decreases with increasing temperature, higher cell voltages can be reached at elevated temperatures.

### 4.3.3 Gas Turbine

The pressure ratio $\beta$ of the gas turbine has considerable influence on hybrid system performance. It was therefore studied, how the sub-atmospheric operation of the gas turbine affects the efficiency of the process. As the expanded gas contains a high fraction of steam, the influence of a cooling device downstream of the turbine was also evaluated by varying the temperature of this process step. The results are
depicted in Figure 20. The outlet pressure was varied in range $p_{turb,out} = 0.3$–1 bar. The outlet temperature of the condenser $\theta_{\text{cooler,out}}$ was varied from 10 to 50°C. It is worth noting that for operation below $\theta_{\text{cooler,out}} = 25$°C additional cooling equipment would be necessary. However, for the variation presented here, a single-step condensation was assumed, as this simplification does not influence heat and mass balance. Figure 20 shows that an optimum for the turbine outlet pressure in the range of $p_{turb,out} > 0.4$ exists. Cooling of the gas stream also leads to an increase in efficiency as a lower volume flow has to be compressed subsequently. Below $\theta_{\text{cooler,out}} = 25$°C condensation occurs, leading to a steeper increase in efficiency due a further reduction in volume flow.

In order to further analyze the effect of the cooler, a hybrid system without cooler was modeled. This would allow for a higher temperature in the hot water generator HWG (Figure 4) and thus a higher CHP efficiency. The dependence of the electric system efficiency and compressor inlet temperature upon the $p_{turb,out}$ is plotted in Figure 21. It can be observed that omitting the cooler leads to a significant decrease in efficiency even in comparison to high cooler temperatures like $\theta_{\text{cooler,out}} = 50$°C. For a technical realization, a cooler downstream the turbine should therefore be included in the system if the aim is on maximization of the electric system efficiency.

### 4.4 Process Efficiency Comparison

In Sections 4.1 to 4.3, feasible operating conditions for the different SOFC systems were identified with help of sensitivity analyses. In order to allow for a comparison of the concepts, optimized operating conditions were selected. They are listed in Table 5. For the sake of comparison, the SOFC temperature was set to $\theta_{\text{SOFC,out}} = 830$°C for all studied systems even though it was already stressed that especially in case of the hybrid systems a higher temperature can yield higher efficiencies. All other process parameters remain like they are given in Table 4.

The simulation results at optimized conditions are summarized in Table 6. Based on the results, a higher electric efficiency is achieved by the SOFC/GT systems compared to the SOFC systems. The highest electric and CHP efficiency values are reached by the pressurized system with an efficiency of

| Parameter | Unit | SOFC | SOFC/GT |
|-----------|------|------|---------|
| **Gas Turbine** | | | |
| Pressure ratio $\beta$ | | 3 | 2 |
| Cooler outlet temperature $\theta_{\text{cooler,out}}$ | °C | | 35 |
| **SOFC** | | | |
| Outlet temperature $\theta_{\text{SOFC,out}}$ | °C | 830 | – / 830 | 830 | 830 |
| Operating pressure $p_{\text{SOFC}}$ | bar | 1 | 1 | 3 | 1 |
| Fuel utilization $FU$ | % | 75 | 70 / 67 | 75 | 75 |
| **Reformer** | | | |
| Anode off-gas recirculation ratio AGRR | % | 60 | 33 | 30 |
| Methane conversion in the reformer $X_{\text{CH}_4,\text{ref}}$ | % | 50 | 50 | 50 |
| **System** | | | |
| Global fuel utilization $FU^*$ | % | 88(a) | 90 | 82(a) | 81(a) |

(a) Calculated from Eq. (2)
The lowest values are reached by the AGR system with an efficiency of \( \eta_{\text{el,sys}} = 59.5\% \) and \( \eta_{\text{CHP}} = 68.4\% \), respectively; the performance gain by utilizing a gas turbine amounts to 6.5 pp and 16.4 pp, respectively. In the case of the SOFC systems, air forms the bulk flow, as indicated by the given air-fuel equivalence ratio \( \lambda \). Thus, significant heat transfer occurs in the air heat exchangers, which is relevant in terms of economic feasibility. The two-stage and the novel atmospheric system reach a comparable electric efficiency of \( \eta_{\text{el,sys}} \approx 63.5\% \). However, it has to be pointed out that in case of the two-stage system a higher total stack number is necessary. On the other side, the proposed atmospheric SOFC/GT system concept of course necessitates a gas turbine instead. So both concepts offer an interesting potential for efficient electricity generation avoiding the drawbacks of pressurized SOFC operation, namely the need for large pressure vessels.

It is worth noting that in case of the novel atmospheric system, the heat removed by the cooler was not considered in the calculation of the CHP efficiency, because the cooler is only operated at \( \theta_{\text{cooler, out}} = 35^\circ \text{C} \). However, when the cooler outlet temperature is set to \( \theta_{\text{cooler, out}} = 60^\circ \text{C} \) and is therefore used as a HWG, the CHP efficiency can reach a value of \( \eta_{\text{CHP}} = 80.4\% \) with the electric system efficiency decreasing to \( \eta_{\text{el,sys}} = 62.6\% \). So the novel hybrid system offers flexibility regarding the supply of electricity or heat, depending on the demand.

5 Conclusions

A novel atmospheric SOFC hybrid system applying an inverted Brayton cycle was proposed. In order to allow for a suitable comparison, the novel process as well as several concepts proposed in literature were modeled with help of the software Aspen Plus. Sensitivity analyses were conducted, in order to identify the most suitable operating conditions for the studied systems. A comparison of the novel concept with an SOFC/AGR system, a two-stage SOFC system as well as a pressurized hybrid system was conducted. The electric efficiencies for the studied systems were calculated to be: pressurized SOFC/GT (\( \eta_{\text{el,sys}} = 66.0\% \)) > novel atmospheric SOFC/GT (\( \eta_{\text{el,sys}} = 63.5\% \)) = two-stage SOFC (\( \eta_{\text{el,sys}} = 63.4\% \)) > SOFC/AGR (\( \eta_{\text{el,sys}} = 59.5\% \)). Even though the efficiency for the novel atmospheric system is slightly lower than in case of a pressurized hybrid system, it offers the advantage that no pressure vessel is necessary. This makes the concept considerably cheaper and more easily scalable. The proposed atmospheric SOFC/GT system with sub-atmospheric expansion therefore offers an interesting perspective for the application in efficient electricity generation.

List of Symbols

| Symbol | Description |
|--------|-------------|
| A      | Area / m²   |
| AGRR   | Anode off-gas recirculation ratio |
| \( E_a \) | Activation energy / kJ mol⁻¹ |
| \( E_N \) | Nernst potential / V |
| \( F \) | Faraday constant / 96485.3 A s mol⁻¹ |
| FU     | Fuel utilization |

Table 6 Simulation results at optimized conditions for the studied SOFC systems.

| Parameter               | Unit       | SOFC                      | SOFC/GT   |
|-------------------------|------------|---------------------------|-----------|
|                         |            | AGR                       | Two-stage | Pressurized | Atmospheric |
| Electric power \( P_{\text{el,GT,ouz}} \) | kW         |                           | 16.6      | 15.7       |            |
| SOFC                    |            |                           | first / second stage |           |            |
| Air-fuel equivalence ratio \( \lambda \) | 5.46       | 5.54 / 16.1   | 2.63      | 3.36       |            |
| Number of stacks        | 125        | 99 / 33                   | 116       | 115        |            |
| Electric power \( P_{\text{el,SOFC,ouz}} \) | kW         | 139.6                     | 115.8 / 32.8 | 137.8      | 132.9     |
| System                  |            |                           |           |            |            |
| Electric efficiency \( \eta_{\text{el,sys}} \) | %          | 59.5                      | 63.4      | 66.0       | 63.5       |
| CHP efficiency \( \eta_{\text{CHP}} \) | %          | 68.4                      | 74.8      | 84.8       | 71.4       |
**Global fuel utilization**

| Symbol | Description       |
|--------|-------------------|
| $I$    | Current / A       |
| $J$    | Current density / A m⁻² |
| $i$    | Molar flow / mol s⁻¹ |
| $P$    | Power / W         |
| $p$    | Pressure / Pa      |
| $Q$    | Heat flux / W      |
| $R_u$  | Universal gas constant / 8.314 J mol⁻¹ K⁻¹ |
| $t$    | Time / s          |
| $T$    | Temperature / K    |
| $V$    | Voltage / V        |
| $x$    | Molar fraction     |
| $X$    | Conversion         |

**Greek Symbols**

| Symbol | Description       |
|--------|-------------------|
| $\beta$| Pressure ratio    |
| $\eta$ | Efficiency        |
| $\theta$| Temperature / °C  |
| $\lambda$ | Air-fuel equivalence ratio |

**Subscripts and Superscripts**

| Subscript | Description            |
|-----------|------------------------|
| 0         | Standard, reference    |
| an        | Anode                  |
| CHP       | Combined heat and power |
| comp      | Compressor             |
| CS        | Cold side              |
| el        | Electric               |
| gen       | Generator              |
| HS        | Hot side               |
| HWG       | Hot water generator    |
| in        | Inlet                  |
| inv       | Inverter               |
| isen      | Isentropic             |
| max       | Maximum                |
| mech      | Mechanical             |
| min       | Minimum                |
| out       | Outlet                 |
| react     | Reacted                |
| ref       | Reformer               |
| rev       | Reversible             |
| S1        | Stage 1                |
| S2        | Stage 2                |
| stoich    | Stoichiometric         |
| sys       | System                 |
| t         | Technical              |
| th        | Thermal                |
| turb      | Turbine                |

**Abbreviations**

| Abbreviation | Description       |
|--------------|-------------------|
| AGR          | Anode off-gas recirculation |
| ASR          | Area-specific resistance |
| GT           | Gas turbine        |
| pp           | percentage points  |
| SOFC         | Solid oxide fuel cell |
| TIT          | Turbine inlet temperature |

**References**

[1] International Energy Agency, *World Energy Outlook 2018*.
[2] C. Zou, Q. Zhao, G. Zhang, B. Xiong, *Natural Gas Industry B* 2016, 3, 1.
[3] A. Mazza, E. Bompard, G. Chicco, *Renewable Sustainable Energy Rev.* 2018, 92, 794.
[4] L. van Bier, T. Woudstra, M. Godjevac, K. Visser, P. V. Aravind, *J. Power Sources* 2018, 397, 382.
[5] M. Bailera, P. Lisbona, L. M. Romeo, S. Espatolero, *Renewable Sustainable Energy Rev.* 2018, 92, 794.
[6] A. Varone, M. Ferrari, *Renewable Sustainable Energy Rev.* 2015, 45, 207.
[7] G. Gahleitner, *Int. J. Hydrogen Energy* 2013, 38, 2039.
[8] H. R. Ellamla, I. Staffell, P. Bujlo, B. G. Pollet, S. Pasupathi, *J. Power Sources* 2015, 293, 312.
[9] T. Pfeifer, A. Chakradeo, N. Ahire, J. Baade, M. Barthel, C. Dosch, R. Näke, M. Hartmann, *Fuel Cells* 2017, 17, 550.
[10] R.-U. Dietrich, J. Oelze, A. Lindermeir, C. Spitta, M. Steffen, T. Küster, S. Chen, C. Schlitzberger, R. Leitner, *J. Power Sources* 2011, 196, 7152.
[11] M. P. Heddrich, M. Jahn, A. Michaelis, R. Näke, A. Weder, *Fuel Cells* 2013, 13, 612.
[12] R. Torii, Y. Tachikawa, K. Sasaki, K. Ito, *J. Power Sources* 2016, 325, 229.
[13] K. Nakamura, T. Ide, S. Taku, T. Nakajima, M. Shirai, T. Dohkoh, T. Kume, Y. Ikeda, T. Somekawa, T. Kushi, K. Ogasawara, K. Fujita, *Fuel Cells* 2017, 17, 535.
[14] T. Nakajima, K. Nakamura, T. Ide, M. Shirai, T. Dohkoh, S. Taku, K. Ishihika, K. Ogasawara, K. Fujita, *ECS Trans.* 2017, 78, 181.
[15] K. Nakamura, T. Ide, K. Ishihika, S. Taku, T. Nakajima, T. Dohkoh, M. Shirai, S. Akabane, T. Hatae, K. Ogasawara, *Proceedings 13th European SOFC & SOE Forum* 2018, Lucerne, Switzerland, 2018, A1205.
[16] A. Buonomano, F. Calise, M. D. d’Accadia, A. Palombo, M. Vicedomini, *Appl. Energy* 2015, 156, 32.
[17] R. Payne, J. Love, M. Kah, *ECS Transactions* 2009, 231.
[18] T. Elmer, M. Wóral, S. Wu, S. Riffat, *Energy Convers. Manage.* 2016, 124, 29.
[19] M. Steilen, C. Saletti, M. P. Heddrich, K. A. Friedrich, *ECS Trans.* 2015, 68, 479.
[20] F. Leutcht, W. G. Bessler, J. Kallo, K. A. Friedrich, *Appl. Energy* 2018, 211, 479.
[21] M. Henke, M. Steilen, C. Schnegelberger, M. Riedel, M. Hohlöch, S. Bucheler, M. Herbst, A. Huber, J. Kallo, K. A. Friedrich, *ECS Trans.* 2015, 68, 85.
[22] *Fuel Cells Bulletin* 2018, 2018, 6.
[23] *Fuel Cells Bulletin* 2018, 2018, 7.
[24] S. K. Park, T. S. Kim, *J. Power Sources* 2006, 163, 490.
[25] R. Peters, M. Engelbracht, W. Tiedemann, I. Hoven, R. Deja, V. N. Nguyen, L. Blum, D. Stolten, ECS Trans. 2017, 78, 2489.
[26] M. Engelbracht, R. Peters, L. Blum, D. Stolten, ECS Trans. 2015, 68, 283.
[27] L. Blum, Q. Fang, L. G. J. de Haart, J. Malzbender, N. Margaritis, N. H. Menzler, R. Peters, ECS Trans. 2017, 78, 1791.
[28] T. Somekawa, K. Nakamura, T. Kushi, T. Kume, K. Fujita, H. Yakabe, Appl. Therm. Eng. 2017, 114, 1387.
[29] J. Chen, Y. Chen, H. Zhang, S. Weng, Energy 2018, 163, 1.
[30] E. Riensche, J. Meusinger, U. Stimming, G. Unverzagt, J. Power Sources 1998, 71, 306.
[31] P. Sarmah, T. K. Gogoi, Energy Convers. Manage. 2017, 132, 91.
[32] M. Shamoushaki, M. A. Ehyaei, F. Ghanatir, Energy 2017, 134, 315.
[33] R. Reimert, F. Marschner, H.-J. Renner, W. Boll, E. Supp, M. Brejc, W. Liebner, G. Schaub, Ullmann’s Encyclopedia of Industrial Chemistry, Wiley-VCH, Weinheim, Germany, 2012.
[34] M. L. Ferrari, A. Turan, D. Sanchez, U. M. Damo, Hybrid Systems Based on Solid Oxide Fuel Cells: Modeling and Design, 1st ed., John Wiley & Sons Inc, Hoboken, New Jersey, USA, 2017.
[35] M. L. Ferrari, J. Power Sources 2011, 196, 2682.
[36] S. H. Chan, H. K. Ho, Y. Tian, J. Power Sources 2002, 109, 111.
[37] S. H. Jensen, X. Sun, S. D. Ebbesen, M. Chen, Fuel Cells 2016, 16, 205.
[38] E. Facchinetti, D. Favrat, F. Marechal, Fuel Cells 2011, 11, 565.
[39] E. Facchinetti, D. Favrat, F. Marechal, Fuel Cells 2014, 14, 595.
[40] M. Henke, C. Willrich, M. Steilen, J. Kallo, K. A. Friedrich, ECS Trans. 2013, 57, 67.
[41] S. Seidler, M. Henke, J. Kallo, W. G. Bessler, U. Maier, K. A. Friedrich, J. Power Sources 2011, 196, 7195.
[42] S. H. Park, Y. D. Lee, K. Y. Ahn, Int. J. Hydrogen Energy 2014, 39, 1799.
[43] M. A. Azizi, J. Brouwer, Appl. Energy 2018, 215, 237.
[44] B. Eisavi, A. Chitsaz, J. Hosseinpour, F. Ranjbar, Energy Convers. Manage. 2018, 168, 343.
[45] V. Liso, A. C. Olesen, M. P. Nielsen, S. K. Kær, Energy 2011, 36, 4216.
[46] J. Jia, Q. Li, M. Luo, L. Wei, A. Abudula, Energy 2011, 36, 1068.
[47] M. Gandiglio, A. Lanzini, P. Leone, M. Santarelli, R. Borchiellini, Energy 2013, 55, 142.
[48] J. Saarinen, M. Halinen, J. Ylijoki, M. Nononen, S. Pekka, J. Kiviaho, J. Fuel Cell Sci. Technol. 2007, 4, 397.
[49] S. Megel, M. Kusnezoff, W. Beckert, N. Trofimenko, C. Dorsch, A. Weder, M. Jahn, A. Michaelis, C. Bienert, M. Brandner, S. Skrabs, W. Schulmeyer, L. S. Sigl, Proceedings 12th European SOFC & SOE Forum, Lucerne, Switzerland, 2016, A0908.
[50] M. Samavati, M. Santarelli, A. Martin, V. Nemanova, Energy 2017, 122, 37.
[51] F. J. Gutiérrez Ortiz, P. Ollero, A. Serrera, S. Galera, Chem. Eng. J. 2013, 218, 309.
[52] G. Herz, E. Reichelt, M. Jahn, Appl. Energy 2018, 215, 309.
[53] W. D. Seider, J. D. Seader, D. R. Lewin, S. Widagdo, Product and Process Design Principles: Synthesis, Analysis and Design, 3rd ed., John Wiley, Chichester, United Kingdom, 2010.
[54] M. A. Abdelkareem, W. H. Tanveer, E. T. Sayed, M. E. H. Assad, A. Allagui, S. W. Cha, Renewable Sustainable Energy Rev. 2019, 101, 361.
[55] K. Nikooyeh, A. A. Jeje, J. M. Hill, J. Power Sources 2007, 171, 601.
[56] S. Wongchanapai, H. Iwai, M. Saito, H. Yoshida, J. Power Sources 2012, 204, 14.
[57] Q. Fang, L. Blum, R. Peters, M. Peksen, P. Batfalsky, D. Stolten, Int. J. Hydrogen Energy 2015, 40, 1128.