Control of oxygen-to-carbon ratio and fuel utilization with regard to solid oxide fuel cell systems with anode-offgas recirculation: A review

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Abstract

One of the possible SOFC system-configurations providing the highest potential of electrical DC-efficiency of up to 65% is a SOFC-system with anode exhaust gas recirculation (AEGR), where part of the depleted anode exhaust gas is recirculated and mixed with fresh natural gas upstream of the reformer. For safe and durable operation of a SOFC-system, the oxygen-to-carbon-ratio and the fuel utilization as characteristic parameters must not exceed stack- and reformer-specific thresholds. The determination and control of the characteristic parameters are therefore of crucial importance. However, this poses especially for SOFC-systems with AEGR due to enhanced system complexity a challenging task. In this paper, the authors present an overview on representative control strategies as well as different approaches to determine or diagnose characteristic parameters with emphasis on SOFC-systems with AEGR. Some conclusions are discussed based on the provided overview and outlines recommendations for future research work.

Keywords: SOFC, control, fuel utilization, oxygen-to-carbon-ratio, recirculation ratio, system operation

1 Introduction

Limited (fossil) resources and their consumption, growth of worldwide energy demand and progressive global warming due to the emission of greenhouse gases pose one of the major challenges of the 21st century. As a key to address and overcome this challenge, enhanced use and integration of renewable energies as well as measures to achieve higher energy efficiency and reduction of primary energy consumption are needed [1]. One of the major technologies that has the potential to overcome these challenges is the fuel cell. In general, fuel cells are electrochemical devices converting chemical energy directly to electrical energy by oxidizing a fuel gas and evade the thermodynamic limitations of a Carnot cycle. Fuel cells possess therefore a higher potential of energy conversion efficiency than conventional energy converters such as a gas engine [2–4]. This also leads to low emissions, which can be further reduced to zero emissions by the use of renewable fuels [3]. One of the main fuel cell technologies attracting a lot of interest and increasing attention lately is the high temperature solid oxide fuel cell (SOFC) technology, thus being currently intensively investigated.

In a SOFC, an ion-conducting ceramic is used as an electrolyte, simplifying the construction of a cell and eliminating a complex electrolyte management in comparison to low-temperature fuel cell types [5]. In addition, due to the high operating temperature (600 – 1000°C) needed for the ion-conduction, nonprecious metals can be used as catalyst and natural gas can be reformed internally within the cell, offering a great fuel flexibility in the operation of a SOFC and a reduced complexity for fuel pre-processing [3,5–7]. Moreover, SOFC-technology is very promising, because it is a decentralized power generator offering a high modularity paired with scalability as well as a high electrical efficiency of up to 65% concerning direct current (DC) output [8]. It is enabling a high energy utilization which is comparable to efficiencies reached by large power plants such as gas-steam combined cycle power plants [5,6,9–11].

An overall efficiency of around 90% can be achieved, if the SOFC-system is operated as a combined heat and power generator (CHP) [5,10,12,13]. This is done by utilizing the

available heat in the exhaust gases at a relative high temperature of around 200°C for domestic heating or even as process heat in industrial applications [1,3–5,7,11,14].

However, main challenges for the SOFC-technology are cost reduction, reduced start-up time, improvement of durability with less degradation caused by the high temperatures and enhanced efficiency of a SOFC-system [5,10,15–20]. Another challenge is the shift of the overall energy system to a high share of renewable energies. SOFC-systems have so far mainly been optimized for operation under constant load. In an energy system with a high share of renewable energies, transient operation and shortened start-up time are going to be increasingly important. These challenges need to be addressed in future.

The system configuration with the highest potential regarding efficiency is a SOFC-system with anode exhaust-gas recirculation (AEGR) [21]. In such a device the depleted anode exhaust gas is partially recirculated to increase the electrical efficiency and to supply the reforming process with the needed water vapor [14,21].

For an optimal operation regarding efficiency, durability and to prevent degradation of the stack and cells as core of the SOFC-system, the characteristic parameters fuel utilization (*FU*) and oxygen-to-carbon-ratio (*O/C*) are of key importance. These parameters have to be kept within certain boundaries defined by stack- and reformer-specific limits in regard to the operating point [22]. Moreover, these two parameters need to be controlled closely to its limit values to ensure an efficient, but durable operation of a SOFC-system.

In principle, installing concentration measurement devices at the anode before and after the stack is a possible and easy way to determine the characteristic parameters. Gas chromatography systems or mass spectrometers are in general highly accurate methods for measuring gas compositions, which are used in SOFC application according to the current state-of-the-art. However, these are associated with a high metrological effort (calibration, provision of carrier gas) and comes along with not acceptable additional costs as well as adds significantly to the size of a SOFC-system. In addition, accurate measurements of gas concentrations at high temperatures imply long measurement times of several minutes to half an hour, which is partly due to the extractive measuring methods, making it impossible for dynamic control with a needed high temporal resolution. Accordingly, these methods are primarily suitable for use in laboratories and not for commercial series application.

Several sensors could be also used to measure the concentrations of the combustible gases (CH_4 , H_2 and CO) needed for determination of FU . Alternatively, a so-called combustible gas sensor can be used to measure the combustible gas components as a summarized equivalent. The CO_2 and H_2O concentrations at the anode inlet are further needed for the O/C -ratio. A wide variety of common measuring principles can be used including tunable diode lasers, resistance temperature detector, non-dispersive infrared sensors and electrochemical sensors. The stabilized zirconium-based electrochemical sensor is the most common sensor type for industrial use under demanding conditions, according to Liu et al. [23]. However, to the best of the authors knowledge, no commercially available sensor is suitable for SOFC use. The sensors are either designed as gas warning detectors in the ppm range and exhibit saturation behavior above it, which disables its use at higher concentrations present in a SOFC. Or the sensors meet the concentration range requirements but cannot be used at temperatures of up to 800°C . According to Liu et al. [23], electrochemical sensors for measuring H_2 , CO or hydrocarbons are indeed part of intensive research work, but have not yet reached a sufficient level of maturity and are not commercially available. Standard commercial concentration sensors are thus not suited for concentration measurements in a SOFC fuel gas according to the current state-of-the-art. Different concepts and novel methods are needed to control or determine these parameters in real-time [24].

This review summarizes typical control strategies by giving an overview of some representative control strategies and discusses approaches found in literature to determine characteristic parameters online in a SOFC-system with the purpose of control-integration. In addition, recommendations for future research to enhance performance and commercialization of the SOFC-technology are given. The remainder is structured as follows. First faulty operation states and characteristic parameters of SOFCs are outlined. The following part gives a brief overview about SOFC-systems with special focus on systems with AEGR. The section thereafter provides a review on representative control strategies, followed by a comprehensive overview on different approaches to determine characteristic parameters as main part of this paper. As a result of the conducted literature review, recommendations for future work regarding the determination of characteristic

parameters and control of SOFC-systems with AEGR are derived, followed by a summary.

2 SOFC fundamentals

In this section a brief overview on fundamentals of characteristic parameters and on main relevant aspects of SOFC-systems, especially on systems with AEGR, are given.

2.1 Faulty operation states and characteristic parameters

Fuel depletion as well as carbon formation and deposition ('coking') are the two major faulty operation states, which have to be limited to minimize cell degradation and enhance durability [25–28]. Fuel depletion leads to a shortage of fuel at the anode resulting in a reduction in cell voltage and in re-oxidation of the anode, the latter leading to enhanced cell degradation and irreversible cell damage [8,16,19,27,29–32]. To avoid the occurrence of fuel and air depletion an overstoichiometrical feeding with fuel and air to the anode and the cathode, respectively, is conducted. Whereas the oversupply of air does not strongly influence the efficiency, the supply of additional fuel gas, which is not taking part in the electrochemical conversion, represents a loss of efficiency. The amount of overstoichiometrical feeding of fuel gas to the anode should be therefore minimized to the point, where the occurrence of fuel depletion can be safely prevented.

The fuel utilization FU describes the relation between the electrochemically converted part of supplied fuel gas to total supplied fuel gas as stated in equation (1) and represents one of the major characteristic parameters for SOFC control [29,33,34]. Typical values for FU are in the range of 50-80% [3,8].

$$FU = \frac{\dot{n}_{\text{gas,conv}}}{\dot{n}_{\text{gas,total}}} \quad (1)$$

Analogous to FU for the anode side, the air utilization (AU) can be defined for the cathode side by equation (2) [26,35]. Since a SOFC-system is typically fed with ambient air with known O_2 -concentration ($\sim 21\%$) and air flow rate as well as the stack current I_{Sta} represent easily measurable parameters, AU can be determined without much effort.

$$AU = \frac{I_{\text{Sta}} \cdot N_{\text{cell}}}{4 \cdot F \cdot \dot{n}_{\text{air,in}} \cdot x_{O_2,\text{air,in}}} \quad (2)$$

In dependency of the local temperature, the lack of oxygen in a carbon containing fuel gas can lead to carbon formation at the anode [17,28]. Typical carbon formation processes are the Boudouard-reaction, reverse coal gasification and especially cracking of long-chain hydrocarbons [8,16,29,33,36,37]. This can lead to a disconnection of the

paths for the conduction of the electrons, increased pressure losses and blockage of the reaction zone in the triple phase boundary [8,15,17,29,34]. To prevent carbon formation it is necessary to supply either enough water vapor or oxygen in the fuel gas in dependency of the upstream reforming process [28,29]. For example, water vapor enhances the water-gas shift-reaction leading to a reduced amount of carbon monoxide. This is described by the temperature-dependent parameters oxygen-to-carbon-ratio (O/C) or steam-to-carbon-ratio (S/C), which are stated below in equation (3) and (4) in dependence of a general alkane molecule C_nH_m ¹ [28,38,39]. They represent one of the major characteristic parameters for operation of a SOFC-stack or system.

$$\frac{O}{C} = \frac{\dot{n}_O}{\dot{n}_C} = \frac{x_{H_2O} + x_{CO} + 2x_{CO_2}}{\sum_n n \cdot x_{C_nH_m} + x_{CO} + x_{CO_2}} \quad (3)$$

$$\frac{S}{C} = \frac{\dot{n}_{H_2O}}{\dot{n}_C} = \frac{x_{H_2O}}{\sum_n n \cdot x_{C_nH_m} + x_{CO} + x_{CO_2}} \quad (4)$$

The O/C-ratio describes the ratio of the molar rates of oxygen-bearing to carbon-bearing species in the fuel gas, which are weighted by the related number of atoms in the molecule [1,21]. Too much of oxygen or water vapor results in a dilution of the fuel gas and a drop in the cell-voltage resulting in a loss in efficiency [8,37,40]. Accordingly, these parameters should be kept at a minimal value, where carbon formation is avoided.

2.2 SOFC-Systems

A complete SOFC-system (see figure 1) comprises the three essential process steps: gas treatment, electrochemical conversion and afterburning [5,10,12,36]. Gas treatment consists of gas reforming and gas cleaning (desulfurization) [5,29,36]. The electrochemical conversion takes place in the stack, which represent roughly 25% of the system size [5,29,36]. Additionally, a SOFC-system consists of some balance of plants components such as pumps, blowers and heat exchangers (HEX). For the sake of brevity only a short introduction on the main reforming processes as relevant differentiation is given and the reader is referred to Kendall [36] for further details on system build-ups. Possible reforming processes of natural gas to hydrogen-rich gas are steam reforming (SR), catalytic partial oxidation (CPOx) or autothermal steam reforming (ATR), where SR

¹ Methane: n = 1, m = 4; ethane: n = 2, m = 6; propane: n = 3, m = 8; butane: n = 4, m = 10

shows the highest hydrogen-yield [4,12,16,36,41,42]. ATR is SR, which is combined with CPOx for start-up of the SOFC-system to supply the needed heat for SR-reforming, but will be not explained further in detail for the sake of brevity [16,29,36]. The corresponding equations and reaction enthalpies are shown in table 1.

Figure 1: Basic structure of a SOFC-system with different reforming procedures (based on [8,25])

Table 1: Reforming Processes [12,16,29,36]

Name	Chemical equation	Enthalpy of reaction	Equation
SR	$C_nH_m + n H_2O \leftrightarrow n CO + \left(n + \frac{1}{2}m\right) H_2$	$\Delta^R h_0 > 0$	(5)
CPOx	$C_nH_m + \frac{n}{2} O_2 \leftrightarrow n CO + \frac{m}{2} H_2$	$\Delta^R h_0 < 0$	(6)
ATR	$C_nH_m + k H_2O + \frac{n-k}{2} O_2 \leftrightarrow n CO + \left(k + \frac{m}{2}\right) H_2$	$\Delta^R h_0 \approx 0$	(7)

A system with CPOx-reforming is rather simple, but has a relative low electrical efficiency of about 35% due to a lower hydrogen-generation and a higher risk of carbon formation due to carbon monoxide generation [16,25,29,33]. SR can be realized with an external water supply (EWS) or anode exhaust-gas recirculation (AEGR) [28]. Anode exhaust gas is partly depleted considering oxidable gas components and contains a lot of water vapor due to the electrochemical conversion of H_2 [12,16,43]. In the SR-process the water-gas

shift reaction takes also place, reducing the amount of carbon monoxide and therefore the risk of carbon formation. In case of EWS an additional evaporator is needed, which is driven with heat by the internal heat recovery system or external supply [36]. For AEGR no additional heat exchanger is needed (at nominal load) and a system with AEGR shows a more robust behavior, but has a higher system complexity and challenging start-up [21,33]. SOFC-systems with EWS or CPOx are not as complex as systems with AEGR, since the air or water vapor can be accurately controlled and supplied.

A control strategy enabling to keep the O/C -ratio (or S/C -ratio, as it is preferred) and FU as characteristic parameters in the desired ranges in a SOFC-system with AEGR can be therefore adapted to other SOFC-systems without much effort. In comparison to SOFC-systems with EWS or CPOx, a system with AEGR shows also the highest potential in electrical efficiency of up to 60% with alternating current (AC) as output [14,21,25,42]. Accordingly, this paper focuses on the determination of the characteristic parameters and control strategies intended for SOFC-systems with AEGR.

2.3 SOFC-System with AEGR

AU , FU , O/C -ratio and recirculation ratio are characteristic parameters which need to be controlled and regulated to keep the SOFC-stack in an optimal operation state without risking enhanced degradation or irreversible damage of the cells [1]. As mentioned earlier, AU can be determined directly without much effort. Temperature control of the cell is very often done by the airflow and can be realized by simple temperature measurements at stack inlet and outlet combined with variation of airflow rates. Control of the cathode side is therefore not a major issue and rather simple compared to the determination and control of FU and O/C -ratio, where this paper is focused on.

As already stated, in a SOFC-system with AEGR a part of the depleted fuel gas leaving the anode of the stack is recirculated and mixed with natural gas. The recirculation ratio r is defined as the ratio of recirculated to total anode exhaust gas molar flow as stated in equation (8) [1,21,44]. The recirculation has two purposes. First, since operation of a SOFC-stack is typically limited to a FU in the range of 60 – 80% to avoid fuel depletion, part of the unused fuel is recirculated leading to a higher FU on system level and higher electrical efficiency [1,20,31,34,36,45,46]. Second, the needed water vapor for steam reforming is supplied internally without an external device [33]. On the other hand a too

high recirculation rate leads to a dilution of the fuel gas resulting in a drop of Nernst voltage and output power of the system [8,34,37]. An overview regarding different configurations of SOFC-systems with AEGR can be found in Peters et al. [47].

$$r = \frac{\dot{n}_{\text{rec}}}{\dot{n}_{\text{an,out}}} \quad (8)$$

The minimal and critical *O/C*-ratio by equation (3) occurs in the reformer and at the anode inlet, since the electrochemical conversion of fuel gas leads to a rise in the *O/C*-ratio along the anode [8]. In a SOFC-system with AEGR *FU* can be defined with respect to the molar flow rate of the fuel gas at the anode inlet as stack-*FU* or the natural gas at the system inlet as system-*FU*, see equation (9) and (10) [1,7,8,16,21,25,26,44].

$$FU_{\text{sta}} = \frac{\dot{n}_{\text{gas,conv}}}{\dot{n}_{\text{gas,total,an,in}}} = \frac{I_{\text{Sta}} \cdot N_{\text{cell}}}{F \cdot \dot{n}_{\text{gas,total,an,in}} \cdot K_{e^-, \text{an,in}}} \quad (9)$$

$$FU_{\text{sys}} = \frac{\dot{n}_{\text{gas,conv}}}{\dot{n}_{\text{NG,total,in}}} = \frac{I_{\text{Sta}} \cdot N_{\text{cell}}}{F \cdot \dot{n}_{\text{NG,total,in}} \cdot K_{e^-, \text{NG}}} \quad (10)$$

I_{Sta} is the stack current and K_{e^-} is the number of potentially releasable electrons of the gas mixture at the anode inlet or system inlet. At the anode inlet K_{e^-} follows equation (11):

$$K_{e^-} = \sum_i x_{\text{gas},i} \cdot N_{e^-,i} \xrightarrow{\text{an,in}} K_{e^-, \text{an,in}} = 8 \cdot x_{\text{NG}, \text{CH}_4} + 2 \cdot x_{\text{NG}, \text{CO}} + 2 \cdot x_{\text{NG}, \text{H}_2} \quad (11)$$

Without AEGR ($r = 0$) both system-*FU* and stack-*FU* are equal, as stated in equation (12), showing its relation via recirculation-ratio r [8,21,26,44].

$$FU_{\text{sta}} = \frac{FU_{\text{sys}} \cdot (1 - r)}{1 - r \cdot FU_{\text{sys}}} \quad (12)$$

In addition to equation (12), equation (13) defines the correlation of the recirculation-ratio r , *O/C*-ratio and stack-*FU* or system-*FU* and is visualized in figure 2 for pure methane ($K_{e^-} = 8$, $K_c = 1$, $K_o = 0$) [21]. K_c and K_o are additional gas coefficients and represent the carbon and oxygen coefficients in a gas mixture by the respective weighted sum of the contained species as stated in equation (14) and (15). The shaded area in figure 2 represents the allowable operation range for pure methane as supplied fuel gas, which is defined by typical thresholds for stack-*FU* and *O/C*-ratio [8]. For a given *O/C*-ratio as wells as stack-*FU* and a specific composition of natural gas, system-*FU* and recirculation ratio r are hence uniquely set.

$$r = \left(FU_{sta} \left(\frac{K_{e^-,NG}}{2 \left(\frac{O}{C} \cdot K_{C,NG} - K_{O,NG} \right)} - 1 \right) + 1 \right)^{-1} = \frac{2 \left(\frac{O}{C} \cdot K_{C,NG} - K_{O,NG} \right)}{FU_{sys} \cdot K_{e^-,NG}} \quad (13)$$

$$K_{C,NG} = \sum_i x_{NG,i} \cdot N_{C,i} = x_{NG,CH_4} + 2x_{NG,C_2H_6} + 3x_{NG,C_3H_8} + 4x_{NG,C_4H_{10}} + x_{NG,CO_2} \quad (14)$$

$$K_{O,NG} = \sum_i x_{NG,i} \cdot N_{O,i} = 2x_{NG,CO_2} \quad (15)$$

It can be concluded that for the determination of the characteristic parameters the exact composition in the form of the individual concentrations of the species in a gas mixture must be known. However, the measurement equipment for concentration measurements is aligned with high costs as well as long measuring periods, being an unsuitable method for commercial use and in particular for online measurement with the purpose of dynamic control of a SOFC-system [17,24,26,30,48].

Figure 2: Correlation of characteristic parameters for pure methane as supplied fuel gas (based on Wahl [8])

3 Review on control strategies

The characteristic parameters FU and O/C -ratio represent essential control variables of a SOFC-system. Representative control strategies of SOFC-systems with special focus on systems with AEGR are discussed in the following in relation to these characteristic parameters. For a more general and comprehensive overview on control strategies for SOFC-systems, the reader is referred to corresponding overviews in literature outlined in the following. General overviews on different types of control strategies can be found in Leung et al. [7], Kirubakaran et al. [49] and Hering [25]. Bavarian et al. [50] deal with stationary and dynamic behavior of PEMFC and SOFC, present and evaluate different control strategies. Huang et al. [51,52] focus on dynamic modelling of SOFC-systems and in particular on the concept of model-predictive control. Buonomano et al. [53] and others [54–56] give overviews on control strategies of hybrid systems with gas turbines.

In general, the control strategy is intended to achieve high electrical efficiency and the provision of the required electrical power, as well as long and durable operation with minimized degradation [7,26,57]. Depending on system configuration and operating strategy, the consequence of the latter are restrictions on different system parameters, which must be met [26]:

- Maximum temperature and temperature difference across the stack must not be exceeded due to thermally induced stresses [7].
- AU and FU have to be controlled to avoid depletion of fuel gas at the anode and oxygen at the cathode, but also to maintain high utilization for high efficiency [7].
- Minimal value for the O/C -ratio has to be fulfilled to avoid carbon formation [7]. At the same time, O/C -ratio must be kept low to prevent fuel gas dilution with a corresponding drop of cell voltage.
- Enabling a load change being as quick and accurate as possible, as well as partial load operation with high efficiency [7,57].
- Ensuring a complete combustion of the electrochemically unconverted gas components in the afterburner.
- These demands are to be maintained for a fuel gas supply by the natural gas grid with varying gas quality for commercial SOFC application.

Table 2 and 3 give an overview on some representative control strategies for SOFC-systems. The tables contain the main author, the source number, the plant type, the controller, the fuel gas used and the indication, marked "Proof-of-Concept", whether the postulated control strategy is tested and validated with experiments on a real system. The manipulated and control variables are given, and it is specified whether the characteristic parameters are determined during operation in the respective control strategy.

It can be seen from table 2 and 3 that proportional-integral-derivative-controllers (PID) or model predictive control systems (MPC) are primarily used for implementation of control strategies as superior groups. Whereas an MPC requires a higher computing power than a PID controller, it allows better control of dynamic, more complex and non-linear behavior [58,59]. In addition to high computing times, a complex model as needed for MPC requires a rather high implementation effort for different system configurations and does not represent an easily adaptable and general approach. MPC control strategies can be divided in MPC with linear and nonlinear models, where the latter is called NMPC. An introduction on PID and MPC control is given as supplementary material in the appendix to point out the general differences for the interested reader.

Since the predictive model of the SOFC-system represents an important part in MPC, some representative studies regarding the setups and models will be briefly outlined below. Wu et al. [60] propose an artificial neural network (ANN) as NMPC-controller for the stack voltage during load changes to indirectly keep the FU within a safe range. Han et al. [58] present a MPC-strategy, where an adaption to model deviations is performed with a specific algorithm to counteract the nonlinearities and disturbances with little computational effort. Das and Weisman [30], as well as Madani and Das [61] use an analytical formulation of stack- FU at stationary conditions for its control at load changes. To minimize the deviation from the stack- FU setpoint, the current density is continuously controlled feedforward as a function of the time-delayed change in the gas flow rate. Spivey and Edgar [62] also use a linear model with multiple inputs and multiple outputs for dynamic control of a SOFC-system with AEGR. The model required for control in the work by Wang et al. [2] is a time-discrete state-space model, which is based on simulated input-output data by means of a physical SOFC-system model. Sartipizadeh and Vincent [63] present also a state-space model for an autothermal methane reformer, where the

H₂/CO-ratio and the energy content of the reformat are control variables. The latter is used to ensure sufficient fuel gas supply to the stack and is assumed to be reflected in the stack signal. A Kalman filter is applied in the studies of Murshed [64] and Kupilik [65] to estimate unknown states. The supplied and converted hydrogen at the anode required for stack-*FU* calculation is estimated based on the Kalman filter in the NMPC-strategy by Murshed et al. [64]. A linear model in combination with a Kalman filter is used to estimate the unknown, not measurable quantities of hydrogen concentration and distance d_{CDB} (CDB: Carbon deposition barrier) in the MPC-strategy by Kupilik and Vincent to avoid fuel depletion and carbon formation [65]. d_{CDB} is the distance of a gas mixture in the ternary diagram with the atomic components O, C and H to the limit value of carbon formation. The focus in the following is on the treatment of characteristic parameters as control variable and on respective control strategies, whereby a subdivision according to the supplied fuel gas from H₂ via CH₄ to natural gas is done. Further details on PID or MPC control design will therefore not be explicitly discussed in the following.

Common manipulated variables for SOFC-system control are the current or voltage of the stack, the temperatures at stack or system inlet and the flow rates of fuel gas and air at the system or stack inlet. In a system with AEGR, the recirculation ratio also represents a manipulated variable. Volume flow measurements, which are needed for accurate determination of the recirculation ratio in SOFC-systems with AEGR, are subject to deviations or measurement errors. Therefore, an exact measurement and control of the recirculation flow with its high temperature and the supplied natural gas flow cannot be achieved as it is assumed in the studies [30,57,61] with fixed or known recirculation ratio. The overview of representative control strategies for SOFC-systems in table 2 and 3 verifies also that the characteristic parameters *FU* and *O/C*-ratio are essential control variables as it has been postulated section 2. For a close-loop control of stack-*FU* or *O/C*-ratio by means of a PID control concept, its determination is required first though. As for the PID control strategies, there are several studies in literature concerning MPC, which deal with the control of stack-*FU*. Therein a determination of stack-*FU* is also advantageous for feed back to the MPC controller.

It shall be mentioned in this regard that some studies [2,9,60,62,64,66] are based on very high stack-*FU* values of up to 95%, which exceed the expected limits of maximum

65-70% that can be tolerated in a SOFC-system in the long term. Values for the stack-*FU* above 70% are very likely associated with at least local fuel depletion and associated irreversible damage to the cell. It represents a critical operating condition and should be avoided. Since such high *FU* values can be regarded as beyond stack-specific values and most likely not endurable, a transfer of the control strategy on a real SOFC-system in field application is not given and cannot be recommended by the authors.

Especially in SOFC-systems with AEGR, the characteristic parameters cannot be measured directly during operation or can be quite complex to determine. In principle, *FU* and the *O/C*-ratio could be physically modelled, but physically-based models are currently not suitable for use in real operation due to their complexity and the associated computing time [65]. Accordingly, none of the control strategies presented in table 2 and 3 is based on a closed-loop for the characteristic parameters. The characteristic parameters can solely be calculated with the help of simplifying assumptions such as the restriction to a known composition of the supplied fuel gas. A common restriction in literature is a supply with pure hydrogen or methane. Due to this simplification or the assumption of known gas concentrations, stack-*FU* can be calculated directly by means of the stack-current by equation (9), such as in [3,20,67–69] for PID control strategies and [58,60,62,64,65,70,71] for MPC strategies. This applies to control strategies, where only the SOFC-stack is considered, or SOFC-systems without AEGR, whereby the supplied fuel gas does not undergo any change in atomic composition until stack entry. This is different for control strategies intended for application on SOFC-systems with AEGR.

Stiller et al. [57] calculate the concentrations of the fuel gas species at the stack inlet assuming chemical equilibrium with known temperature, recirculation ratio and pure methane as fuel gas. Stack-*FU* can thus be determined with the aid of the stack current as measured variable by equation (9). Similar to that, Aguiar et al. [72], Zhang et al. [73] and Sartipizadeh and Vincent [63] assume the species concentrations of the fuel gas such as H₂, H₂O, CO, CO₂ and CH₄ at stack inlet to be known or measured by some kind of gas sensors without going into further details of such a measuring system. In conjunction with the respective flow rate and the stack current, stack-*FU* can be easily calculated and controlled with a simple PID-controller.

Kandepu et al. [74] define FU as a function of hydrogen conversion, using CH_4 as supplied fuel gas of the SOFC-system. It is assumed that the respective FU can be determined based on the measured variables in the system. No further detailed information about the practical implementation is given in this respect though. In other studies [58,60,64,66,70,73] stack- FU is also given as a function of the hydrogen flow rate at the anode inlet, which is atypical for SOFC-systems. While neglecting CO and CH_4 as fuel gas species for FU definition is not common, it is nonetheless a useful variant. H_2 is the primarily electrochemically converted fuel gas species and its amount present at the anode is a main factor concerning fuel depletion, which can be used for its monitoring. It also reduces number of species, which need to be known for FU calculation from three (CO , CH_4 , H_2) to one. Monitoring fuel depletion with respect to present hydrogen at the anode is also a rather conservative concept, since CO and CH_4 are also present. A sensor for dynamic measurement of hydrogen concentration at the anode inlet is still needed. To the best of the authors knowledge, there is no commercial sensor, which is intended for direct use in the fuel gas path of a SOFC-system and the respective harsh conditions.

As already mentioned, reforming is needed when pure methane is used as fuel gas, which in general needs to be monitored to avoid carbon formation. A common assumption in this regard is the external water vapor supply to attain a correspondingly O/C - or S/C -ratio, such as in Vrecko et al. [20,67], Mueller et al. [68] and Gaynor et al. [69] for PID control strategies and Wu and Gao [71] or Murshed et al. [64] for MPC control strategies. In case of a reforming process with O_2 or for an autothermal reformer, a respective supply is considered as it is done in the work of Sartipizadeh and Vincent [63].

While CH_4 supply represents a more realistic scenario, as it is the hydrocarbon mainly found in natural gas and requires the use of a reformer, it still does not meet the requirements for commercial use of a SOFC-system. A significant advantage is especially its operation with natural gas from the grid. The limitation of H_2 or CH_4 as supplied fuel gas does not represent a sufficiently practicable approach in this regard. The presented control strategies are not transferable in this form to a typical and commercial SOFC system, which is driven by natural gas with unknown and varying gas quality depending on location and time. Besides from the gas species (CO , CO_2 , CH_4 , H_2 , H_2O) that are

likely to occur as well as their rough concentration ranges, natural gas operation results in a unknown gas mixture at the anode inlet, so that FU cannot be calculated.

Natural gas operation has been considered in PID control strategies by Darjat et al. [9], Carré [75] as well as others [21,25,26] and MPC strategies by Zhang et al. [73], Wang et al. [2] as well as Lawrynczuk [66]. These control strategies will be discussed shortly and presented in more detail.

Zhang et al. [73] use a NMPC concept with moving horizon estimation as optimization approach to estimate state variables for a planar SOFC-cell. It is driven with a gas mixture containing H_2 , H_2O , CO , CO_2 and CH_4 , which is representative for anode inlet conditions of a SOFC-system based on natural gas supply. Due to the limitation to a single SOFC-cell in the studies by Zhang et al. [73] the composition of the gas mixture is known, which is similar to the aforementioned studies by Aguiar et al. [72], Zhang et al. [73] as well as Sartipizadeh and Vincent [63]. A specific knowledge on the exact fuel gas composition at the anode inlet does not represent a tenable assumption for general SOFC-system operation, where anode inlet fuel gas composition is affected by a various effects such as natural gas composition or other disturbances within the SOFC-system.

Since the characteristic parameters are functions of the fuel gas composition, the characteristic parameters can be calculated in principle on the basis of concentration measurements of the gas components in the respective locations of the SOFC-system [76]. Due to the time required for the measurements needed for exact quantification of fuel gas composition, this method cannot be used to set-up a closed-loop control with a sufficient high temporal resolution though [26]. In addition, to the best of the authors knowledge the measurement technology required for concentration measurements, which is currently available as commercial product and intended for use in SOFC-systems such as a high precision gas chromatography system as used in the work by Carré [75] or Hering [25], is rather complex as well as associated with high costs and maintenance [17,26,48]. Therefore, this does not represent a feasible solution for commercial serial application in terms of cost reduction for the competitiveness of SOFC-technology if no cost-effective sensor for the fast analysis of SOFC-relevant gas compositions is available. Darjat et al. [9] develop a control concept for the SOFC-stack within a SOFC-system, which uses adaptive PID-controllers and the stack voltage as control variable. The PID-

controllers are supposed to be particularly suitable for nonlinear systems with slow dynamic response such as SOFC-systems and to be able to adapt to disturbances during operation by the current value of three process parameters. The supplied fuel gas flow rate is the manipulated variable and the prescribed current is regarded as an externally applied disturbance variable. The proposed PID-controllers show a longer rise time during a load change compared to simple PID-controllers, which is balanced by an improved control behavior with respect to lower oscillations of the stack-*FU*. Since an increase of stack-*FU* above the respective limit value poses the risk of fuel depletion with the effect of irreversible cell damage, a smoothing of the fluctuations in the stack-*FU* is a significant advantage. It is assumed though that only hydrogen is converted at the anode due to complete reforming in an upstream reformer. This implies a rather complex and cost-intensive reforming system. It also neglects one of the key benefits of an SOFC compared to a PEM fuel cell that it can run on an "impure" hydrogen-rich fuel gas containing CO and CH₄ as well as its internal reforming capability, which implies the need of a rather simple reformer system.

A complete reforming of the supplied natural gas to hydrogen is also assumed in the work of Lawrynczuk [66]. A NMPC is used to control the voltage of an SOFC-system and to maintain stack-*FU* within thresholds. To minimize computational effort, the predicted trajectories for voltage and stack-*FU* are linearized during operation. The algorithm presented shows an improved behavior in terms of less overshooting and faster adjustment at load changes in comparison to other MPC approaches.

Wang et al. [2] present a data-driven MPC strategy for a SOFC-system. Based on the supplied natural gas and the oxygen flow rate at the system inlet as input variables, the data-driven model determines the control variables stack voltage, stack-*FU*, hydrogen-to-oxygen-ratio in the stack and the pressure difference between anode and cathode. The stack voltage is the only measurable control variable that is used for the realization of a closed-loop control though. Stack-*FU* as an elementary control variable is not measured and is only estimated based on the predictive model without further verification.

A rather different concept for natural gas operation is given by Carré [75] and others [21,25,26]. The control strategy is intended for a SOFC-system with AEGR and is based on the feed-forward control of stack-*FU* and O/C-ratio by means of supplied fuel gas flow

rate and recirculated flow rate as manipulated variables, as can be seen in figure 3. The stack current is a prescribed variable and not considered as manipulated variable. Basis for this control strategy is the assumption that the composition of the supplied natural gas is known, hence being measured or constant reference composition. The characteristic parameters can be calculated based on equation (9) and (13). This approach has been validated by an application on a real SOFC-system with AEGR and accompanying extensive concentration measurements using a gas chromatograph with a measuring rate of ~0.5 hours per measurement at stationary operating points. Accordingly, the values for the characteristic parameters are not to be checked online during operation, so that no close-loop control is implemented. The effect of small malfunctions by degradation or inaccuracies, such as setting of the recirculation blower, on stack-*FU* or *O/C*-ratio are therefore not detectable. Main drawback of this control strategy is the dependence on a natural gas analyzer system though. The proposed feedforward control concept is also critical for dynamic operation phases such as a load change, where the dynamic effects need to be accounted for by a safety buffer and a respective drop in system efficiency.

Figure 3: Schematic control principle by Carré [75] and others [21,25,26] for a SOFC-system with AEGR

In contrast to the work by Carré [75], many control strategies such as in [2,3,9,30,60–62,64–66,73] are also limited to application on a simulation model and a respective verification of the proposed control strategy. Application on a simulation is advantageous and gives the ability to perform many runs without any risk to improve the control strategy

until a final robust version. It also enables the evaluation of an operation of the SOFC-system and its behavior exceeding its limit. The effects of the control strategy on stack-*FU* and the *O/C*-ratio as well as adhering of the respective limit values can be determined by means of comparison with the values from the simulation model. As a final step, especially for a complex system such as a SOFC-system, an experimental evaluation of the control strategy is rather important. It comprises the interaction with its balance of plant as well as its robustness against disturbances such as varying natural gas quality and the effect of small malfunctions, which can have some effects on control.

Based on the review of control strategies and the outlined representative control strategies it can be finally concluded:

- An open-loop control strategy is implemented in the most cases in current SOFC-systems with AEGR, which are based on simplifying assumptions and thus do not pursue a generally valid approach. Setpoints for characteristic parameters are specified feedforward by analytical correlations to control variables.
- If natural gas composition is not measured continuously, a representative gas composition is defined as reference. However, the characteristic parameters are not checked during operation regarding the accuracy of the feedforward control.
- Fluctuating natural gas composition leads to deviations of the system behavior in comparison to a reference [8]. Characteristic parameters must be thus assigned with buffers to avoid critical operating conditions. This results in efficiency loss.
- To ensure safe operation and high system efficiency at the same time, it is indispensable to determine the characteristic parameters.
- During a load change, knowledge about the characteristic parameters is essential. Particularly *FU* may briefly exceed its limit value. Without online determination characteristic parameter with sufficient temporal resolution, exceeding these limit values cannot be detected and influenced by a closed-loop control.
- The basic structure of a PID control as well as MPC implies the determination of characteristic parameters for its direct and safe closed-loop control. Consequently, respective concepts are presented in the following.
- If the most preferable solution of direct determination of characteristic parameters cannot be fulfilled, other available control variables must be used for control. It is

important to know the relation to characteristic parameters in this case and disturbances are considered thoroughly. It applies to sophisticated MPC strategies, where it is conceivable to incorporate characteristic parameters or the H/C -ratio as parameter for the fluctuating natural gas quality as internal state variables of a Kalman filter.

Table 2: Overview on representative SOFC-control strategies based on PID controller

Main author	Source number	Unit	Controller ²	Fuel	Proof-of-Concept	Manipulated variables	Control variables	Online Determination of characteristic parameters ³		
								FU_{sta}	FU_{sys}	O/C, S/C
Qin	[3]	SOFC-stack	PID	H ₂	No	$\dot{n}_{H2,in}, p_{ca}$	FU_{sta}, U_{sta}	Yes	No	No
Chaisantikulwat	[77]	SOFC-cell	PI	H ₂	No	$x_{H2,in}$	U_{cell}	No	No	No
Stiller	[57]	Hybrid (SOFC + gas turbine)	PID	CH ₄	No	$I_{sta}, P_{el}, \dot{n}_{CH4,in}, \dot{n}_{air,in}$	$P_{el}, T_{an,out}, FU_{sta},$	Yes	No	No
Kandepu	[74]	Hybrid (SOFC + gas turbine)	PI	CH ₄	No	$\dot{n}_{NG,in}, \dot{n}_{air,in}$	FU_{sta}, T_{sta}	Yes	No	No
Mueller	[68]	SOFC-stack	PI	CH ₄	No	$I, \dot{n}_{air,in}, \dot{n}_{air,bypass}, \dot{n}_{CH4,in}, \dot{n}_{H2O,in}$	$T_{sta}, P_{el}, FU_{sta}, U_{sta,min}, T_{ca,in}, T_{br}, O/C$	Yes	No	Yes
Gaynor	[69]	SOFC-system	PID	CH ₄	No	$\dot{n}_{CH4,in}, \dot{n}_{air,in}, I, \dot{n}_{H2O,in}$	$FU_{sta}, P_{el}, T_{sta}, O/C$	Yes	No	Yes

² PID: Proportional-integral-derivative controller

³ Note on (Yes): Characteristic parameter is not directly determined, but correlates very strongly with the determined value, or could be derived from it.

Vrecko	[20]	SOFC-system	PI	CH ₄	No	$\dot{n}_{\text{CH}_4,\text{ref}}, \dot{n}_{\text{air},\text{sta}}, \dot{n}_{\text{H}_2\text{O},\text{ref}}, \dot{n}_{\text{CH}_4,\text{br}}, \dot{n}_{\text{air},\text{br}}$	$T_{\text{br}}, T_{\text{ca},\text{out}}, \lambda_{\text{br}}$	Yes	Yes	No
Vrecko	[67]	SOFC-system	PI	CH ₄	No	$\dot{n}_{\text{CH}_4,\text{sta}}, \dot{n}_{\text{air},\text{sta}}, \dot{n}_{\text{CH}_4,\text{br}}, \dot{n}_{\text{air},\text{br}}, \dot{n}_{\text{H}_2\text{O},\text{ref}}$	$T_{\text{ca},\text{in}}, T_{\text{ca},\text{out}}, \Delta T_{\text{ca}}, T_{\text{br}}, \lambda_{\text{br}}$	Yes	Yes	No
Sorrentino	[78]	SOFC-cell	PI	CH ₄	No	$\dot{n}_{\text{air},\text{in}}$	ΔT_{cell}	No	No	No
Inui	[79]	SOFC-cell	Not specified, but likely to be in the PID-group	CH ₄	No	$T_{\text{an},\text{in}}, I, T_{\text{ca},\text{ein}}, \dot{n}_{\text{ca},\text{in}}$	AU, T_{cell}	No	No	No
Carré	[75]	SOFC-system	PI	CH ₄	Yes	$\dot{n}_{\text{NG},\text{in}}, \dot{n}_{\text{air},\text{in}}, \dot{n}_{\text{rec}}$	$FU_{\text{sta}}, AU, \lambda_{\text{br}}, O/C, T_{\text{sta}}, T_{\text{br}}$	Yes	Yes	Yes
Kaneko	[80]	Hybrid (SOFC + gas turbine)	PID	Biogas	No	$\dot{n}_{\text{exhaust gas,bypass}}, \dot{n}_{\text{biogas},\text{in}}, I_{\text{sta}}$	$T_{\text{sta}}, P_{\text{el}}, U_{\text{sta}}$	No	No	No
Aguiar	[72]	SOFC-stack	PID	CH ₄ , H ₂ O, CO, H ₂ , CO ₂	No	$\dot{n}_{\text{NG},\text{in}}, \dot{n}_{\text{air},\text{in}}$	$FU_{\text{sta}}, AU, T_{\text{an},\text{out}}$	Yes	No	No
Carré	[21]	SOFC-system	PI	NG	Yes	$\dot{n}_{\text{NG},\text{in}}, \dot{n}_{\text{air},\text{in}}, \dot{n}_{\text{rec}}$	$O/C, FU_{\text{sta}}, AU, \lambda_{\text{br}},$	Yes	Yes	Yes

Hering	[25]	SOFC-system	PID	NG	Yes	$\dot{n}_{NG,in}$, $\dot{n}_{air,in}$, $\dot{n}_{rec}$, $\dot{n}_{air,bypass}$, $\dot{n}_{blow,air}$, $\dot{n}_{blow,NG}$	FU_{sta} , AU_{max} , O/C , $T_{sta,max}$, $T_{ca,in}$, T_{rec} , ρ_{air} , ρ_{NG}	Yes	Yes	Yes
Gallet Segarra	[26]	SOFC-system	PI	NG	Yes	$\dot{n}_{NG,in}$, $\dot{n}_{air,in}$, $\dot{n}_{rec}$	FU_{sta} , AU , λ_{br} , O/C , T_{sta} , T_{br}	Yes	Yes	Yes
Darjat	[9]	SOFC-stack	PID	NG	No	$\dot{n}_{NG,in}$	U_{cell} , FU_{sta}	No	No	No

Table 3: Overview on representative SOFC-control strategies based on MPC controller

Main author	Source number	Unit	Controller ⁴	Fuel	Proof-of-Concept	Manipulated variables	Control variables	Online Determination of characteristic parameters ⁵		
								FU_{sta}	FU_{sys}	O/C , S/C
Han	[58]	SOFC-stack	MPC	H ₂	No	$\dot{n}_{H2,in}$, $\dot{n}_{air,in}$	U_{sta} , FU_{sta}	Yes	No	No
Wu	[70]	SOFC-stack	MPC	H ₂	No	$\dot{n}_{air,in}$, $\dot{n}_{H2,in}$	U_{sta} , FU_{sta}	Yes	No	No
Wu	[60]	SOFC-System	NMPC	H ₂	No	$\dot{m}_{H2,in}$, l_{sta}	(FU_{sta}) , U_{sta}	Yes	No	No
Yang	[81]	SOFC-stack	NMPC	H ₂	No	$\dot{n}_{H2,in}$, $\dot{n}_{air,in}$	T_{sta}	No	No	No

⁴ MPC: Linear model-predictive controller; NMPC: Nonlinear model-predictive controller

⁵ Note on (Yes): Characteristic parameter is not directly determined, but correlates very strongly with the determined value, or could be derived from it.

Wu	[71]	SOFC-system	NMPC	CH ₄	No	$\dot{n}_{CH_4,in}, \dot{n}_{air,in}, \dot{n}_{air,bypass}$	FU_{sta}, T_{sta}, AU	Yes	No	No
Das	[30]	SOFC-system	MPC	CH ₄	No	$\dot{n}_{CH_4,in}, I$	FU_{sta}	Yes	No	No
Madani	[61]	SOFC-system	MPC	CH ₄	No	$\dot{n}_{CH_4,in}, I$	FU_{sta}	Yes	No	No
Murshed	[64]	SOFC-system	NMPC	CH ₄	No	$\dot{n}_{NG,in}, \dot{n}_{air,in}, \dot{n}_{H_2O,in}$	U_{sta}, FU, T_{sta}	No	No	No
Kupilik	[65]	SOFC-system	MPC	CH ₄	No	$\dot{n}_{CH_4,in}, \dot{n}_{air,in}, U_{sta}$	$T_{ref}, T_{sta}, X_{H_2}, d_{CDB}, I_{sta}$	(Yes)	No	(Yes)
Sartipizadeh	[63]	Reformer	MPC	CH ₄	No	$\dot{m}_{CH_4,in}, \dot{m}_{O_2,in}, \dot{m}_{H_2O,in}$	$T_{ref}, H_2/CO, H_{i,ref,out}, d_{CDB}$	No	No	(Yes)
Spivey	[62]	SOFC-system	MPC	CH ₄ , CO ₂	No	$\dot{m}_{air,in}, p_{gas,in}, T_{gas,in}, U_{cell}, T_{air,in}, p_{sys}$	$AU, FU_{sta}, S/C, P_{sta}, T_{cell,min}, \Delta T_{rad,max}, \eta_{fc,el}$	No	No	No
Zhang	[73]	SOFC-stack	NMPC	CH ₄ , H ₂ , H ₂ O, CO, CO ₂	No	$I, \dot{n}_{air,in}, \dot{n}_{CH_4,in}$	$T_{sta}, P_{el}, FU_{sta}$	Yes	No	No
Sanandaji	[82]	SOFC-stack	MPC	CH ₄ , H ₂ , H ₂ O, CO, CO ₂ , N ₂	No	$U_{sta}, \dot{n}_{NG,in}, \dot{n}_{air,in}$	$I, x_{H_2,an,out}, T_{ca,out}$	No	No	No
Wang	[2]	SOFC-system	MPC	NG	No	$\dot{n}_{NG,in}, \dot{n}_{O_2,in}$	$U_{cell}, FU_{sta}, H_2/O_2, \Delta p_{an,ca}$	No	No	No
Lawrynczuk	[66]	SOFC-System	NMPC	NG	No	$\dot{n}_{NG,in}$	$(FU_{sta}), U_{sta}$	No	No	No
Pohjoranta	[59]	SOFC-system	MPC	NG	No	$\dot{n}_{NG,in}, \dot{n}_{air,in}, T_{air,in}$	$T_{sta,max}, T_{sta,min}$	No	No	No

4 Review on approaches for determination of characteristic parameters

The following section and the overview in table 4 deals with different concepts and approaches for the determination of characteristic parameters. Table 4 lists the main author, reference number, plant type, respective approach and used fuel gas. It is indicated whether the respective approach is examined simulatively or experimentally and which of the characteristic parameters are addressed. Table 4 also specifies whether a determination of the characteristic parameters is considered for feedback control.

Table 4: Overview on approaches for determination of characteristic parameters

Main author	Source number	Unit	Method	Fuel	Simulation	Proof-of-Concept	Evaluated Parameter ⁶			Online Determination of characteristic parameters for control-feedback
							O/C	FU_{Sta}	FU_{Sys}	
Gorgun	[41]	CPOX-reformer	Observer model	CH ₄	Yes	No	(Yes)	No	No	No
Gorgun	[43]	CPOX-reformer	Observer model	CH ₄	Yes	No	(Yes)	No	No	No
Pukrushpan	[83]	CPOX-reformer	Observer model	CH ₄	Yes	No	(Yes)	No	No	No
Das	[45]	SOFC-system	Observer model	CH ₄	Yes	No	(Yes)	(Yes)	No	No
Das	[84]	SOFC-system	Observer model	CH ₄	Yes	No	(Yes)	(Yes)	No	No
Abey Siriwardena	[76]	SOFC-system	Observer model	CH ₄	Yes	No	No	Yes	No	No
Dolenc	[48]	SOFC-system	System model	CH ₄	No	Yes	(Yes)	(Yes)	No	No
Slippey	[85]	SOFC-system	System model	CH ₄	Yes	No	(Yes)	Yes	No	No

⁶ Note on (Yes): Characteristic parameter is not directly determined, but correlates very strongly with the determined value, or could be derived from it.

Dolenc	[17]	SOFC-stack	Soft sensor	CH ₄	No	Yes	Yes	No	No	Yes
Vrecko	[86]	SOFC-stack	Soft sensor	CH ₄	No	Yes	Yes	No	No	Yes
Chen	[22]	SOFC-system	Soft sensor	n. s. ⁷	Yes	No	No	Yes	No	No
Subotic	[87]	SOFC-cell	EIS	CO, CO ₂ , H ₂ , CH ₄ , H ₂ O, N ₂	No	Yes	(Yes)	(Yes)	No	No
Subotic	[88]	SOFC-cell	EIS	CO, CO ₂ , H ₂ , CH ₄ , H ₂ O, N ₂	No	Yes	(Yes)	(Yes)	No	No
Subotic	[89]	SOFC-cell	EIS	CO, CO ₂ , H ₂ , CH ₄ , H ₂ O, N ₂	No	Yes	(Yes)	(Yes)	No	No
Subotic	[90]	SOFC-cell	EIS	CO, CO ₂ , H ₂ , CH ₄ , H ₂ O, N ₂	No	Yes	(Yes)	(Yes)	No	No
Montinaro	[91]	SOFC-cell	EIS	CH ₄ , H ₂ O, H ₂	No	Yes	Yes	No	No	No
Stam	[28]	SOFC-cell	EIS	CH ₄ , H ₂	No	Yes	(Yes)	No	No	No
Ikeda	[92]	PEM-stack	EIS	H ₂	No	No	No	(Yes)	No	Yes

⁷ n. s.: Not specified

Tallgren	[93]	SOFC-stack	EIS	H ₂	No	Yes	No	Yes	No	No
Morel	[94]	SOFC-cell	THD	H ₂	No	Yes	No	Yes	No	No
Mougin	[18]	SOFC-stack	EIS, THD	H ₂	No	Yes	No	(Yes)	No	No
Nusev	[95]	SOFC-stack	EIS	H ₂	No	Yes	No	(Yes)	No	No
Subotic	[96]	SOFC-cell	THD	CH ₄	No	Yes	(Yes)	No	No	No
Subotic	[97]	SOFC-cell	THD	CH ₄	No	Yes	(Yes)	No	No	No
Subotic	[98]	SOFC-cell	THD	CH ₄	No	Yes	(Yes)	No	No	No
Malafronte	[44]	SOFC-system	THD	H ₂ , natural gas	No	Yes	No	Yes	No	No
Pahon	[99]	SOFC-stack	WT	H ₂	No	Yes	No	(Yes)	No	No
Esposito	[100]	SOFC-cell, - system	WT	H ₂ , natural gas	No	Yes	No	Yes	Yes	No
Potyrailo	[24]	SOFC-system	Multiple gas-sensor	H ₂ , CO	No	Yes	No	No	No	No
Haag	[101]	Not specified	Carbon sensor	CH ₄ , C ₃ H ₈	No	Yes	(Yes)	No	No	No

van Herle	[102]	SOFC-system	Lambda sensor	n. s.	No	No	No	No	No	Yes
Ruka	[103]	SOFC-system	Lambda sensor	CH ₄ , natural gas	No	Yes	Yes	No	No	Yes
Ballantine	[27]	SOFC-system	Lambda sensor	n. s.	No	No	Yes	Yes	Yes	Yes
Keegan	[104]	SOFC-system	SOFC-cell	H ₂	No	No	No	No	(Yes)	No
Schäfer	[105]	SOFC-system	Lambda sensor	Natural Gas	Yes	Yes	Yes	(Yes)	Yes	No

For a sensor or a sensor concept determining the characteristic parameters, rough specifications and basic requirements are listed in table 5. These should serve as reference values to achieve a reasonable and expedient implementation of a sensor concept in a control strategy of a SOFC-system, in particular with AEGR. The values for accuracy and response time are rough estimates, since it depends on the respective operating conditions as well as the specific system configuration and no concrete values can be found in literature to the best of the authors knowledge. These values are chosen in a range that ensures the ability to detect and control a change of the characteristic parameters in time with sufficient accuracy. Due to the higher sensitivity of a SOFC to the stack-*FU* and a higher inertia of the *O/C*-ratio, higher accuracy and a shorter response time for *FU* are desirable. Regarding a cost reduction of SOFC-systems, an acceptable cost should also be considered.

Table 5: Requirements on concepts or approaches for determination of characteristic parameters

Charact. parameter	Measurement range [8,75,106]	Accuracy	Response time
Stack- <i>FU</i>	50 – 75%	$\pm 1 - 3\%$	< 60 s (stationary); < 1 – 5 s (dynamic)
System- <i>FU</i>	70 – 90%	$\pm 1 - 3\%$	
<i>O/C</i> -ratio	2 - 3	$\pm 5 - 10\%$	< 60 s (stationary); < 30 s (dynamic)

Based on the literature research, the following statements and conclusions can be made on approaches for determining the characteristic parameters. The different concepts and approaches for the determination of the characteristic can be divided into sensor-based and model-based approaches. This superior categorization in model-based and sensor-based concepts is used in the following to group the different concepts for better structure and readability as visualized in figure 4. Model-based approaches are concepts in which models for a SOFC-system or individual components are set up for. These sensor concepts are therefore solely based on algorithms or software. This differs from the

sensor-based concepts, where a physical component is used as a sensor. It can be an additional sensor or the use of an existing component as sensor such as the SOFC-stack. Further differentiation of model-based or sensor-based concepts is done into observer concepts and data-driven soft sensors or the application of diagnostic fuel cell methods and sensors placed directly in the fuel gas, respectively.

Figure 4: Classification of concepts for determination of characteristic parameters

4.1 Model-based concepts

Model-based concepts include the use of an observer for the reformer or the whole SOFC-system and the establishment of correlations via analytical equations, which are also referred to as data-driven soft sensors. Whereas data-driven system models can be assigned to the latter, physical-based system models are rather part of observer concepts regarding the proposed categorization in this work.

In a data-driven soft sensor, a correlation between a variable of interest and one or more dependent variables is established based on data. The data can be based on simulations or experiments. A data-driven soft sensor therefore represents a purely empirical approach. Consequently, it is a relatively simple but also inflexible modeling approach. It must be kept in mind that data-driven models are only valid in the data range in which

they are trained. A (subsequent) extrapolation beyond the underlying data range used for model generation is therefore not possible without further ado and must be strictly avoided. This means that the data area of interest must be completely known before and included in model generation to achieve reliable and safe predictions. Flexibility in this regard during model application on an operating SOFC-system is restricted and no understanding of complex data-driven soft-sensors regarding the way of correlation between input and output is possible. This is a clear contrast to physically based models, which in general have universal validity and provide physical interpretability, but can come along with a high complexity. All data-driven soft sensors essentially represent more or less a simplification of the real, nonlinear SOFC-system with its associated complexity and should be handled with caution.

Just as for the control strategies a common restriction for model-based concepts is a fuel gas supply with known composition or with pure methane such as in Das and Mukherjee [45], Das [84], Abey Siriwardena and Das [76], Dolenc et al. [48] as well as others [17,41,43,83,85], which enables a simple calculation of the characteristic parameters. Due to this limitation the concepts lack the potential to be transferred to a typical and commercial SOFC system, which is driven by natural gas with varying quality. The prerequisite of stationary operating conditions in [76,85] or a constant and known recirculation ratio is used in some studies [17,45,48,85], which is a simplification of the complexity in real SOFC-systems. The validation of the concepts is also often limited to simulations, such as in Das and Mukherjee [45] or Chen et al. [22]. The abandonment of experimental studies and evaluation of the robustness of a concept has the drawbacks outlined extensively above.

The achievement of a thermodynamic equilibrium at the outlet of the reformer or the anode represents an assumption in physical-based models as in the work of Pukrushpan et al. [83] or Dolenc et al. [48], which is a common and reliable assumption for SOFC-systems in stationary state [107]. This holds also true for the neglect of methane at the anode outlet, which has been used in the work by Dolenc et al. [48] .

An observer is basically characterized as a model for a component or a system, where unknown parameters of interest are estimated based on measured input variables. Gorgun et. al [41,43] and Pukrushpan et al. [83] present observer concepts for CH₄-driven

reformer systems based on the CPOx-principle with regard to the hydrogen supply for a PEMFC. In principle, such a reformer system can also be applied to a SOFC-system to monitor the O/C -ratio or gas composition at the anode inlet, where the latter could be used for calculation of stack-FU. The reformer system by Gorgun et. al [41,43] comprises subsequent reactors for water-gas-shift-reaction and selective oxidation intended for CO-removal. Due to CO compatibility the latter process could be removed for SOFC application, which results in a less complex observer. With the help of measured temperatures, fixed CH_4 supply and pressures the gas concentrations can be determined. Similarly, Pukrushpan et al. [83] use a dynamic, physics-based model, where the O_2 -to- CH_4 -ratio at the reformer inlet is an important influencing factor. Since methane and oxygen supply can be precisely adjusted, the O_2 -to- CH_4 -ratio is also known.

To the best of the authors knowledge there is no relevant work concerning a sole reformer observer, which can be applied to a SOFC-System with AEGR or is based on natural gas supply as being relevant for commercial SOFC-systems. This basically represents a scientific gap for future work. However, the lack of such an observer can probably be attributed to the complex boundary conditions. For an observer of a reformer in a SOFC-system with AEGR, the reformer inlet conditions are coupled to the anode outlet and the related gas composition in addition to a varying natural gas quality. Due to this complexity, no observer can be developed for the reformer alone. It is to be expected that an observer describing the entire SOFC-system will be required.

A correspondingly more comprehensive observer can be found for instance in the work of Das and Mukherjee [45], which is used in subsequent works by Das [84] and Abeyisiriwardena and Das [76]. Das and Mukherjee [45] develop a model-based observer for estimating gas concentrations in a CH_4 -driven SOFC-system with AEGR and steam reforming with constant recirculation ratio. The estimation of the gas concentrations is used to determine characteristic parameters (FU , S/C -ratio) and intended for control of the SOFC-system with a reduced number of gas sensors, which is quite close to an MPC control. The approach assumes that in addition to pressure and temperature, the concentrations of any two gas components in the reformer and the anode can be measured using some kind of gas sensors without giving any further specification. In a subsequent work Das [84] integrates the cell voltage as a measurable quantity into the

model-based observer for the estimation of the gas concentrations. It's still assumed that hydrogen and water vapor concentrations at the reformer outlet and water vapor concentration at the anode outlet are available as measured parameters. However, according to the current state of the art, no simple low-cost and commercial sensor is available for this application, which has been proven to fulfill this requirement. This observer is used by Abeysiriwardena and Das [76] to derive a reduced-order observer for the estimation of stack-*FU* in a CH₄-driven SOFC-system. The observer estimates the hydrogen potential of the fuel gas at the reformer outlet for the determination of stack-*FU*. The hydrogen potential describes the possible hydrogen content in the fuel gas assuming a complete reforming of the fuel gas components to hydrogen. Based on this, a control of the stack current as a function of the estimated hydrogen potential and the set point of the stack-*FU* is proposed for minimizing the deviation of the stack-*FU* during load changes. It's stated that this approach is based on sensors for the measurement of flow rates at the anode inlet and outlet, as well as the supplied fuel gas at the system inlet instead of concentrations measurements. However, cost-effective solutions for determining flow rates, such as common thermal mass flow meters, require knowledge of the gas mixture composition. Such flow meters are also not suited for use under the hot operating conditions at the anode inlet and outlet considering its thermal endurance as well as its technical principle of operation. Since this type of observer concept for SOFC-systems is based on the concentration measurement of one or more gas components, sensor concepts have to be outlined or specified in future work, which are able to fulfill this prerequisite or an adaption of the observer concept to rather available measured quantities should be considered.

Another concept, where a SOFC-system is taken into consideration and modeled, is given in the work by Dolenc et al. [48]. Dolenc et al. [48] propose three different approaches for the determination of the gas composition in a CH₄-driven SOFC-system with AEGR and test them on a 10 kW SOFC-system. In the first approach, a physics-based model is defined, which describes the reforming reactions as well as the electrochemical conversion. The model is solved iteratively with a numerical solution process by predefining an arbitrary gas composition at the reformer inlet as a starting point. To overcome numerical problems such as convergence associated with the complex model

and its implementation on a commercial programmable logic controller (PLC), the second approach simplifies the model from the first approach and is adapted for a simpler PLC implementation. Assuming a constant inlet temperature of the reformer, a linear correlation between the gas compositions at the reformer outlet and inlet is established in the sense of a black-box model. The third approach is a data-driven concept. A linear equation for the description of the different gas compositions as function of the recirculation flow, the supplied methane flow and the stack current as error-free input variables is established based on measurement data. It is shown that the data-based approach has the highest accuracy in predicting the gas composition. This is since the data-driven model is based directly on real data and obviously no assumptions are made for system description. Finally, the authors adapt the simplified physics-based model with few training data sets and achieve an even higher accuracy than with the data-driven model. The enhancement of universally valid physics-based model with a data-driven model to a joined hybrid model poses a great potential to compensate for errors of the physics-based model and adapt to the specific behavior of a real SOFC-system. It still has to be kept in mind, that the empirical part of such hybrid models already represents a minor data-based modelling.

Slippey et al. [85] define analytical equations as data-driven soft-sensor for stack-*FU* and *S/C*-balance to ensure safe and optimum operation of a SOFC-system with AEGR. Stack-*FU* is defined as a function of the maximum hydrogen potential by complete reforming of the supplied fuel gas. The *S/C*-balance describes the excess of water vapor at the reformer inlet, which is not needed for stoichiometric steam reforming. Stack-*FU* and *S/C*-balance are described by means of the stack current, the recirculation ratio, gas composition and mass flow at the system inlet as known system variables. The proposed concept is limited to the use of one hydrocarbon species as fuel gas though.

Another data-driven soft-sensor is set-up by Dolenc et al. [17] for the calculation of the *O/C*-ratio at the anode inlet for a SOFC-system with AEGR, which is intended for implementation in a control strategy [86]. Based on available measurement data and the method of the least square error, an equation for the *O/C*-ratio is defined, where it shows a linear dependence to the stack current, the recirculation flow and the supplied CH₄ flow.

In order to predict the *FU* of a SOFC-system with AEGR in a hybrid system of SOFC and gas turbine, Chen et al. [22] apply a data-driven dynamic model based on an artificial neural network (ANN) as machine learning algorithm in conjunction with the long short-term memory (LSTM) method. By the application of the LSTM method, historical data of the input variables from previous time steps is included as ANN input in the current prediction. The input variables for *FU* prediction are the supplied fuel gas flow and air flow, as well as the stack voltage. For the mapping of input to output data the ANN is trained beforehand with respective data to learn the correlation, which is known as supervised machine learning. For the additional extraction or generation of meaningful features from the mentioned input data a further ANN is applied, which is based on a specific unsupervised machine learning algorithm known as deep auto-encoder (DAE). In the DAE-ANN algorithm only the input parameters are given and the algorithm determines itself relevant output parameters equivalent to a categorization procedure. The output parameters of the DAE-ANN are then fed to the LSTM-ANN as further input variables, which is supposed to reduce the risk of overfitting. It achieves a predictive accuracy with a maximum absolute error of 0.529% regarding *FU* at load changes. The large training data of 4000 data points is based on simulative data. A rather large data set for training of the ANN models in conjunction with a sufficient variation of respective parameters to cover the whole operation range of the system of interest is always required for an accurate prediction potential. The availability of such large data set is not always given and rather limited concerning real data of SOFC-system, which is not just operated at a fixed and safe setpoint but covers a wide-range in part and full load.

While the general concept of a data-driven soft-sensor offers a simple and straightforward possibility to define and determine a correlation describing one or more characteristic parameter, the specific equation or algorithm must be determined for each SOFC-system due to individual system behavior. It also has to be kept in mind that a data-driven soft-sensor represents merely a feedforward concept without direct feedback of the current real conditions in the system. It can be rather see as close to a feedforward control strategy as proposed by Carré [75] and offers the potential of its integration in a respective adapted control strategy. Since feedback of the current conditions in a SOFC-system are beneficial for safe and reliable control, the next section deals with sensor-based concepts.

4.2 Sensor-based Concepts

Sensor-based concepts can be differentiated in diagnostic fuel cell methods and fuel gas sensors. Diagnostic fuel cell methods are commonly known for evaluation of electrochemical processes within a fuel cell and are namely electrochemical impedance spectroscopy (EIS), total harmonic distortion (THD) and wavelet transformation (WT). A general introduction on EIS, THD and WT is given in the appendix for the interested reader. Since these methods are directly applied on the SOFC-stack as it is the case for EIS and THD or analyze the already available stack voltage (WT), no additional sensor must be installed in the SOFC system. This is different for fuel gas sensor concepts, where an additional sensor needs to be installed directly in the fuel gas before or after the anode. The fuel gas sensor concepts found in literature comprise a carbon sensor, photonic multi-gas sensor and statements on the use of a lambda sensor as well as a single SOFC cell.

4.2.1 Diagnostic fuel cell methods

The studies on diagnostic methods are mostly limited to single-cells or short-stacks with few cells and not applied at system level as can be seen in [18,28,87–90,93,94,96,97,99,108]. In addition, simplifying limitations to a known and simplified gas composition such as pure hydrogen or methane as well as a mixture with a defined amount of water vapor are used, see [92–94,96,97,99,108]. In EIS the high measuring effort and the long measuring time to record a complete impedance spectrum is disadvantageous for application in SOFC-systems. The required measuring time of up to 20-30 minutes for EIS is too long for continuous and dynamic control integration, which is needed or at least beneficial for *FU* control [109]. THD has a significantly shorter measurement time due to the limitation to one or a few data points at individual frequencies and is more beneficial in this regard. Despite these drawbacks, there are some EIS-based studies in literature that address the relevant detection of fuel depletion and carbon formation intended for (system) monitoring purposes. These will be briefly outlined, before studies on THD and WT method are discussed.

Subotic et al. [87–90] investigate the influence of different fuel gas compositions, carbon formation and fuel depletion on the impedance spectrum by means of EIS. Similarly, Montinaro et al. [91] investigate the influence of the S/C-ratio on the impedance spectrum.

Stam and Aravind [28] are examining the detection of carbon formation using EIS on button-cells. As a result, the individual contributing factors can be assigned and detected under given conditions. However, fuel depletion and carbon formation represent undesired operating conditions, which lead to degradation and should be avoided in real system operation. A detection of these faulty operating states by means of EIS is thus not regarded as expedient in the sense of a precise and safe control of the correlated characteristic parameters FU and O/C -ratio. Differences in EIS for different gas compositions can also be shown. It is not possible though to derive a general causal relationship to certain gas components and its respective share. Ikeda et al. [92] describe a related EIS-method for a PEMFC-system in which the hydrogen concentration can be determined by comparing measured impedance values with reference values. In theory, this could be used to determine a hydrogen-based FU as outlined section 4.1.

Tallgren et al. [93] present a concept based on the work of Dekker et al. [64], where FU distribution on individual cells of a SOFC-stack can be determined by means of EIS. The components of an equivalent circuit model are adapted and compared with a reference. EIS recording for each single cell requires a high measuring effort, which is not reasonable in a SOFC-system with many 100 cells. Morel and Chatroux [94] show similarly that EIS and THD can be used to detect and distinguish different FU in single cells. Neither for EIS nor THD an exact quantification of the FU values is given though. EIS and THD are also used as monitoring tool for SOFC operation by Mougin et al. [18]. Instead of using the common continuous sinusoidal excitation signal for EIS with a discrete spectrum, a so-called binary PRBS excitation signal (pseudorandom binary sequence) is used to obtain an almost continuous spectrum, which has been introduced to fuel cell diagnostics by Debenjak [110]. In PRBS a randomly switch-type excitation with varying amplitude length is used, which essentially results in a shorter measurement time by an order of magnitude [110,111]. Using THD, a correlation between fuel depletion and an increase in the THD value can be shown qualitatively, but not quantitatively. A similar EIS approach is addressed by Nusev et al. [108] for monitoring of a short-stack by wiring each cell during a 3600-hour test with temporary fuel depletion. A discrete random binary sequence excitation signal with discrete switching times is used instead of PRBS, which is based on Boskoski et al. [95]. Features such as increased ASR can be identified to

detect increased *FU* by an analysis of the associated equivalent circuit parameters. *FU* change by a change of natural gas quality as not easily available measurand in a commercial SOFC-system and its causal differentiation to other causes for fuel depletion such as insufficient volume flow rate is not considered within the studies.

Subotic et al. [96–98] investigate the use of THD for monitoring carbon formation. Individual frequencies can be identified and used for fast detection. An increase in the THD value in the low frequency range can only be detected after a long period of time though, whereby the cell is exposed to critical operating conditions with the risk of irreversible cell degradation. Since no application at stack level is concerned, it shall be questioned if a stack application results in an even later detection of faulty operating states due to some signal noise caused by coupling of the cells to a stack.

In the context of system application, Malafronte et al. [44] comprehensively investigate the use of THD for monitoring the stack-*FU* at values of 60 to 90% in a SOFC-system with AEGR. Based on studies performed on single cells fueled with H_2 frequencies between 0.01 and 1 Hz of the alternating current and amplitudes between 5 and 10% of the direct current value are obtained as operating parameters. The first 3 to 4 harmonic frequencies provide the essential contributions and higher frequencies can be neglected. An exponential correlation between stack-*FU* and THD value with the direct current as further dependent variable is derived and a basic suitability for the detection of (high) stack-*FU* is demonstrated. The THD value also increases considerably during load changes, which is partly attributed to the latency for the calculation of the THD-value though. Greater measurement noise and inexplicable outliers occur, too. The high alternating current amplitudes lead to the linearity condition in the U-i-curve required for THD being violated. Instead, non-linearities are artificially generated, which can be detected. If the linear condition is fulfilled by using an alternating current with small amplitudes, no detectable non-linearities in the form of harmonic distortions should occur at typical stack-*FU* of 50 to 70%. A quantitative exact determination of the stack-*FU* by THD does therefore not seem applicable. It has also to be kept in mind that high amplitudes can lead to short-term fuel depletion during the positive half-wave.

A different concept of fuel cell diagnosis is WT, where the stack voltage itself is analyzed. Pahon et al. [99] investigate WT application on the voltage of a short stack to diagnose

critical operating conditions as well as its state-of-health by variation of operating temperature, FU (40 – 60%), current density and AU . The voltage signal is divided into partial signals using a 5-step discrete WT (DWT) and special features (wavelet energy and entropy) are defined. The features can be used to diagnose a faulty operating state, but no causal and quantitative correlation can be derived. Instead of application on a short stack, it would be interesting to evaluate the relevant application of DWT on the overall stack voltage in a SOFC-system and the respective signal quality with the associated signal noise caused by the coupling of individual cells to a stack in future studies.

Esposito et al. [100] investigate the use of continuous WT (CWT) for the diagnosis of anode re-oxidation at high FU . The experimental studies are conducted on single cells as well as on a natural gas-powered SOFC-system. By comparing the graphic representations of the CWT coefficients, 76.2% of the critical operating conditions with elevated $FU > 75\%$ can be detected. Although considering further parameters such as the standard deviation of the cell voltage, the authors do not derive a causal correlation for the FU -values. Compared to DWT with its discrete values and the respective studies by Pahon et al. [99], CWT is quite complex and not as straightforward. Regarding the automated use in monitoring or control of a SOFC-system, the DWT approach should be attributed a higher potential. The general WT method is a promising tool though because it does not require an additional sensor and does not interfere with normal system operation due to the subsequent analysis of the measured stack voltage.

As final evaluation, the diagnostic methods EIS, WT and THD are primary intended for application in SOFC-systems for the detection and diagnosis of unwanted, critical operating conditions such as carbon formation or fuel depletion. Individual influencing factors in this regard can be assigned and detected. The occurrence of these critical operating conditions should always be avoided by determining and controlling the characteristic parameters at first. A respective quantitative determination of the characteristic parameters with sufficient accuracy does not seem to be achievable with these methods concerning the current state-of-the-art.

4.2.2 Fuel gas sensors

In concepts based on fuel gas sensors an additional sensor is installed directly in the fuel gas before or after the anode for fuel gas sensor concepts. The concepts found in

literature comprise a photonic multi-gas sensor, a carbon sensor as well as the use of a lambda sensor or a single SOFC cell.

Potyrailo et al. [24] present a photonic multi-gas sensor for real-time determination of fuel gas species, which is intended for monitoring and control of a SOFC-system. The functional principle is based on the different interaction of individual species with spatially distributed lamella and ridges of a photonic 3D-nanostructure in the form of different reflectance colors and spectra in comparison to a reference gas (N_2). The illumination with white light as well as the detection of the resulting reflectance spectrum is done with a fiber optic. Various machine learning algorithms are applied for evaluation of the measured reflectance spectrum by means of classification and to derive a correlation to the fuel gas species, which is rather complex. As a result, different gas compositions of pure H_2 and CO as well as a mixture of both at $300^\circ C$ can be detected and distinguished. However, no quantitative correlation regarding their concentrations is given. The effects of elevated temperatures of around $600^\circ C$ regarding durability and the presence of other gas components such as CO_2 or H_2O in the fuel gas of an SOFC are not considered. It would be rather important to evaluate in further studies whether these related gas species can be differentiated from H_2 and CO or an interference is present that makes it difficult to make a clear distinction. It shall be mentioned that a clear distinction into individual gas species is not necessarily required for use in a SOFC-system. It is even advantageous, if a subdivision could be made in the total amount of combustible gas components such as H_2 , CO and CH_4 and the total amount of non-combustible gas components such as H_2O , CO_2 and N_2 . The total amount of combustible gas components such as H_2 , CO and CH_4 can be used to define an equivalent to stack-*FU* concerning fuel depletion.

Figure 5: Schematic principle of a carbon sensor (based on [101,112,113])

Another prototype sensor concept is the in-situ carbon sensor by Haag [101] intended for determination of carbon formation in the reformer of a SOFC-system with external steam supply. Since it is a stand-alone sensor, an application in an SOFC system with AEGR is also feasible. An essential element of the carbon sensor is a catalyst pellet whose electrical resistance is determined by application of a voltage and current measurement. A drawing of the functional principle for the carbon sensor is given in figure 5, where the three-dimensional catalyst-pellet is replaced by an abstract interdigital structure.

Carbon formation occurs on the catalyst pellet forming interconnections, which can be detected by a resistance drop. After carbon deposition has occurred, the catalyst pellet must be regenerated or replaced. Whereas the replacement is not a practicable solution in the sense of continuous, automated operation and monitoring of a SOFC-system, the effect of particles released during regeneration needs to be evaluated in further studies. According to the author, carbon formation can be detected at an early stage. However, the O/C-ratio as relevant parameter for SOFC-system control is not directly determined or at least not addressed in the presented studies. The carbon sensor has some potential for beneficial use in a SOFC-system though since it can be directly used in the fuel gas of a SOFC with the respective harsh conditions. It can detect carbon formation as a stand-alone sensor without other influences in the measurement signal, which is not yet possible according to the current state-of-the-art to the best of the authors knowledge. It allows at least its use as a back-up monitoring tool for safety reasons by placing it in specific locations of a SOFC-system, where it is known to be vulnerable for carbon formation to

enhance safety and durability. It offers the possibility to operate the SOFC-system safely at the limit value for carbon formation, without determining its descriptive variable O/C -ratio. This in turn reduces the dilution of fuel gas and results in a higher system efficiency. A sensor, which structure is similar to a SOFC and can be attributed a good potential for use in a SOFC-system, is the lambda sensor. Lambda sensors, which are also called oxygen sensor in literature, give a statement about the relative amount of oxygen in a gas mixture in form of the lambda value and are used to diagnose oxygen shortage or excess. Oxygen shortage ($\lambda < 1$) can be regarded as equivalent to the amount of unconverted fuel gas making the lambda sensor to a combustible gas sensor. Electrochemical conversion of fuel gas results in an increase of the lambda value by equation (16) over the stack due to the transferred oxygen ions with a higher share of the products CO_2 and H_2O at the anode outlet [105]. By the respective change of the gas composition, different chemical potentials are generated and can be measured against an oxygen reference as signal of a lambda sensor. The signal difference at the anode inlet and outlet characterizes therefore fuel conversion, whereas the oxygen amount in a fuel gas is expected to correlate with the O/C -ratio.

$$\lambda = \frac{\text{available oxygen}}{\text{stoichiometric needed oxygen}} \quad (16)$$

There are essentially two different (commercial) types of lambda sensors, as can be seen in figure 6. The potentiometric or switch-type lambda sensor measures the Nernst voltage of the fuel gas in reference to ambient air. It shows a switch-like signal behavior with low slope at $\lambda < 1$. The amperometric or wide-range lambda sensor pumps oxygen ions by a pumping current in a measuring chamber to achieve stoichiometric conditions therein with $\lambda = 1$. It is separated from the fuel gas by a diffusion barrier and results in a linear signal curve with steady slope of the pumping current as measurand.

Figure 6: Different types of lambda sensors and respective measurements signals (based on [105])

Whereas the remarks of some authors [27,102–104] are restricted to a theoretical consideration of a lambda sensor, simulative and experimental studies are only presented by Schäfer et al. [105] to the best of the authors knowledge.

Van Herle [102] solely mentions the use of a lambda sensor positioned before and after the reformer as a possibility to determine the O/C -ratio to avoid carbon formation and to detect further degradation processes. Further detail is given by Ruka and Basel [103] describing the use of a potentiometric lambda sensor for O/C -ratio monitoring to avoid carbon formation. An integrated heating element is used to maintain the sensor temperature constant to minimize temperature influence and noise. The oxygen partial pressure in the fuel gas is determined by the measured Nernst voltage, from which the O/C -ratio is correlated. However, this only applies under the assumption of known H/C -

ratio of the supplied fuel gas and chemical equilibrium of the fuel gas with known temperature. Further studies regarding the influence of the H/C -ratio and its variation on the determination of the O/C -ratio should be pursued in further work. An additional heated reforming chamber is integrated in the proposed sensor since the fuel gas is to be branched off and monitored before entering the reformer of the SOFC-system. This is complex and associated with great integration effort in commercial SOFC-systems. It would be rather promising to consider a placement of the lambda sensor after the reformer without heated reforming chamber.

Ballantine et al. [27] consider the general use of a lambda sensor to determine FU and S/C -ratio. While considering lambda sensors in amperometric and potentiometric mode to determine FU , only the potentiometric mode is considered for determination of the S/C -ratio. The sensors can be placed anywhere in the fuel gas path of the SOFC-system or be integrated directly into a SOFC-cell to determine stack- FU .

A concept, which is fairly similar to the use of a lambda sensor is proposed by Keegan [104]. Keegan [104] describes the use of one or more SOFC-cells as hydrogen sensor. The SOFC-cell of the sensor has the same structure and materials as the actual SOFC. This makes the sensor suitable for the demanding operating conditions in a SOFC-system with high hydrogen concentrations and high operating temperatures. The principle of the sensor is based on the measurement of the open circuit voltage of the corresponding SOFC-cell, which is similar to a potentiometric lambda sensor. Since it is assumed that only hydrogen is converted at the anode, the measured cell voltage correlates with the hydrogen content in the supplied fuel gas. Other influences such as the operating temperature on the cell voltage or interference with other fuel gas species such as CH_4 or CO are not considered. The proposed concept is especially promising regarding durability of the sensor in accordance to the lifetime of the respective SOFC-system.

While the approaches listed above are considered promising for use in an SOFC-system, some comprehensive studies are given in the work by Schäfer et al. [105] substantiating this hypothesis. Schäfer et al. [105] propose a concept for control of a SOFC-system with AEGR, which is based on the determination of characteristic parameters by commercial lambda sensors. The lambda sensors are placed at the anode inlet and outlet, where the lambda value at the anode outlet correlates with system- FU . The concept is evaluated

with simulative and experimental studies based on a fixed reference natural gas composition, whereby no influence of its H/C -ratio on the lambda value is given. The experimental studies are done in a test rig emulating fuel gas conditions of a SOFC-system with AEGR regarding temperature and gas mixture composition under the prerequisite of chemical equilibrium. The studies show a good potential for lambda sensor application, since the O/C -ratio at the anode inlet can be measured with the potentiometric lambda sensor with the fuel gas temperature as further dependent variable, whereas the amperometric lambda sensor shows good accuracy for determination of system- FU at the anode outlet. Both sensors can be used to determine the electron gas coefficient K_e . A control concept is postulated, where a minimal K_e is used to avoid fuel depletion as substitute to stack- FU . The potentiometric lambda sensor can be applied at the anode inlet and outlet, but the amperometric lambda sensor can solely be used at the anode outlet due to the greater oxygen shortage in the gas mixture at the anode inlet and respective hardware limitations of its control unit. Since a higher potential can be attributed to the amperometric lambda sensor due to its linear signal curve with sufficient slope for precise lambda quantification, further studies are supposed to address the hardware adaption. The work by Schäfer et al. [105] gives a well-founded statement about the general suitability and potential of lambda sensors in SOFC-systems though, which should be pursued in future works. This concerns especially the use of commercial automotive lambda sensors, which have proven reliability and maturity.

5 Discussion and recommendations for future work

It has been shown section 3 that a PID and MPC control implies the determination of characteristic parameters for its direct and safe control. Consequently, concepts for the determination of characteristics parameters regarding implementation in a closed-loop control strategy are needed.

Based on the review on the identified relevant literature and the respective studies, none of the approaches fulfill the need for a sensor determining the characteristic parameters with general validity and sufficient accuracy. As a result, the following recommendations for future works regarding the determination and control of characteristic parameters in SOFC-systems, especially with AEGR, can be derived:

1. Online determination of characteristic parameters requires a sensor for the implementation of a closed-loop control strategy. It should be ideally independent of a respective gas composition. It should also represent a simplification of the sensor technology in a SOFC-system in comparison to costly and time-consuming concentration measurements used currently. A fast measurement method would be beneficial for dynamic control of the characteristic parameters. Regarding the needed cost reduction of SOFC-systems the sensor is also needed at an acceptable cost and commercially available.
2. A respective sensor concept allows an optimal control of the characteristic parameters in steady state with associated higher system efficiency and reliability. In dynamic operation during load changes, compliance with the respective limit values can also be monitored and controlled. Damage can be avoided and durability of the SOFC can be increased. Such a sensor concept can balance the volatility of natural gas composition and allows to achieve maximum system efficiency by setting stack-*FU* to the allowed maximum. While in the stationary case the *O/C*-ratio as well as stack-*FU* and system-*FU* for a SOFC-system with AEGR are important, the dynamic case primarily concerns stack-*FU*. It is due to the low dynamics of the *O/C*-ratio and system-*FU* being merely an efficiency parameter, whereas stack-*FU* characterizes the risk of fuel depletion. In addition, a non-uniform gas distribution of parallel stacks in a SOFC-system can be detected, allowing appropriate countermeasures to be taken.

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3. Therefore, one or more methods for determination of the characteristic parameters shall be developed in future work, where table 5 lists the respective requirements. It can be seen from equation (3), (9) and (10) that gas concentrations or the relevant molar flow rates are required for a direct determination of the characteristic parameters. Alternatively, methods can be developed, which determine the characteristic parameters indirectly with the aid of correlations to other variables such as voltage, temperature and pressure.
 4. The methods can be based on a real hardware sensor or a model-based soft-sensor as well as the combination of both. The method should be an approach, where no or less simplifications are made. A generally valid method is to be aimed at, which can be applied without restriction to different system configurations.
 5. From the authors point of view, it is inevitable to derive any information on gas composition within the SOFC-system. It can be either information on natural gas composition such as the higher heating value, Wobbe index or H/C -ratio as relevant parameters or about the fuel gas compositions at the anode.

Based on the findings outlined above, further statements on more detailed concepts and its respective potential for use in SOFC-systems will be given in the following.

Further studies on the application of THD- and EIS-measurements as well as WT should be pursued. Integrating a frequency generator and an analyzing tool for THD- and EIS-measurements in the power inverter of the SOFC-system, as it has been postulated by Herrmann et al. [114], would be beneficial. For WT, an integrated high-resolution voltage measurement is required. Integration in the inverter is beneficial, since with cost reduction of SOFC-systems being currently one of the major challenges, additional costs for the measuring device and the needed space for integration in a housing is limited. While these methods can probably not be used to determine any of the characteristic parameter with a sufficient accuracy, it can be applied as an additional safety unit or to describe long-term degradation for maintenance planning and other use cases.

For the application as monitoring tools in a real SOFC system, some progress still must be made. First, studies on EIS, THD and WT must be extended from laboratory tests on single cells and short stacks to larger stacks and direct application in SOFC-systems. This comprises the influence of signal noise due to the coupling of individual cells with

varying electrochemical behavior to a stack and the influence of a complex fuel gas mixture typically present at the anode of a SOFC on the signal quality of the stack voltage. A good signal quality is needed to ensure a safe and reliable differentiation of faulty operating states. For control of a SOFC-system, a clear quantitative statement or correlation is desirable in this regard. In THD analysis it could be for instance a maximum THD value, where exceeding results in a safety shutdown of the SOFC-system.

When using EIS and especially THD on a stack, an evaluation of the required amplitude, which is needed to exclude influences by signal noise (due to the individual cells) and to reliably detect process characteristics, must be done. This should be considered critically regarding the maximum current at the high wave and the associated high FU regarding a short-time and local fuel depletion. Another current drawback is the measurement times needed for EIS and THD. The conventional EIS-method has a measurement duration of several minutes and even the THD-method has measuring times of a few seconds to a few minutes at the low frequencies of the exciting alternating current, which are relevant for fuel depletion or carbon formation. Focus should be therefore given to advanced approaches, where the necessary measurement time is reduced. A promising approach in this regard seems to be a change of the exciting alternating current signal, as can be seen by the works outlined above by Debenjak [110] and Mougins [18] or Nusev et al. [108] and Boskoski et al. [95], where a binary sequence excitation signal is used. A similar approach was proposed by Mertens et al. [115], where an excitation of an lithium-ion batteries was performed with square-wave pulse currents. The presented EIS variant is of particular interest for the low frequency components and represents a significant time saving of several orders of magnitude compared to conventional EIS. A promising concept is also the conjunction of EIS with an associated equivalent circuit diagram, as proposed by Tallgren et al. [93] and Nusev et al. [108]. Analysis of the respective parameters can be used in an automated monitoring tool by comparison to a reference. WT has in general the advantages that it requires little computational effort, modelling is less complex than EIS, uses no additional cost-intensive sensor technology, and is highly accurate and reliable [100]. WT can be applied to the stack voltage or current, which means to use the inevitably existing stack as sensor. This is advantageous, since stack voltage and current as key parameters of a fuel cell system are already measured

quantities and WT analysis does not interfere with normal system operation due to its subsequent analysis. WT can be also applied to other measured quantities such as pressures or temperatures in the fuel cell system, but stack voltage is the rather preferred variable. Since electrical and mass transport processes have time constants in the range of milliseconds and tenths of seconds, a change in gas composition at the anode affects the stack voltage with a sufficient high dynamic in comparison to the rather slow influence on temperature measurements due to thermal inertia of the system [62]. The cell process of interest (fuel gas transport, coking) can be selected for WT analysis similar to EIS and THD using a suitable low frequency or frequency range. High frequency parts like noise, which are not interesting for signal analysis, can be explicitly eliminated by this method. For a comprehensive evaluation of the potential of WT, a sufficiently large amount of real measured data of a SOFC system must be available at first, too. This should be based on a large and wide-ranging variation of the relevant manipulated variables such as stack-*FU* or *O/C*-ratio as well as a sufficient temporal resolution of the stack voltage. A greater potential as automated monitoring tool is attributed to DWT, since CWT analysis is complex, which makes it not suitable to implement on PLC. Corresponding to the approaches shown by Pahon et al. [99], parameters can be defined based on discrete values by DWT, which can be used for SOFC-system control and monitoring.

Whereas the most preferable solution of direct determination of characteristic parameters with an independent hardware sensor should be favored, a soft sensor can also be considered. For the general case of a soft sensor the authors propose the concept of a hybrid model as most promising. A hybrid model represents a combination of both physics-based and data-based models as it has been done by Dolenc et al. [48], achieving simplified structure with a reduced number of equations as well as good accuracy and validity [18,116]. The basic structure of a hybrid model is given by a physics-based model, which is extended by a data-based model. In the proposed concept suggested for future work by the authors the physics-based model shall represent a universal valid model of a SOFC-system. An adaption and calibration of the general SOFC-system to a specific layout of an SOFC-system is pursued by the conjunction with a data-based model. The data-based models can range from simple linear regression to complex methods such as ANN for mapping of the relationship between input to output

parameters. In addition to the commonly used ANN, the authors propose a Gaussian process model. It has the advantage to give a statement on the reliability of the estimated value, which can be used for safety concerns.

For a soft sensor application, it would also be advisable to have a correlation to the characteristic parameters for safety reasons, where disturbances are considered. This applies to a sophisticated MPC strategy, where it is conceivable to incorporate characteristic parameters or the fluctuating natural gas quality by the H/C -ratio as internal state variables. In this regard, a Kalman filter with a state-space model of the SOFC-system should be included, where internal state variables get updated by comparison of variables, where the estimation can be compared to a measured value. The predestined variable would be again the stack voltage because it is an already measured system quantity and has the benefit of a quick response time to changing fuel gas quality.

As it has been mentioned above though, the most preferable solution represents a direct determination of characteristic parameters with an additional and independent hardware sensor placed at the anode inlet and outlet to directly get information about the gas compositions. This would open up the possibility to calculate or correlate the characteristic parameters stack- FU and O/C -ratio straightaway. A sensor which is to be placed directly in the fuel gas at the anode inlet and outlet has to fulfill and withstand harsh conditions such as high temperatures in conjunction with high concentrations of H_2 , H_2O , CH_4 , CO_2 and CO though. A fast response time and accuracy as indicated in table 5 is also challenging to fulfill, but beneficial. Gas chromatography systems or mass spectrometers are in general highly accurate concentration measurement devices but are not suited for commercial application in a SOFC-system, as outlined above. Standard commercial sensors measuring the individual gas species or the combustible gas components as a summarized equivalent are also not suited for direct application in a SOFC fuel gas and can be excluded according to the current state-of-art.

A sensor family rather made for similar harsh conditions as present at the anode are automotive exhaust gas sensors. Automotive exhaust gas sensors are mature components extensively used as well as optimized and therefore very attractive for integration in SOFC-systems. The risk of a new development of a sensor prototype can therefore be avoided. Automotive exhaust gas sensors, being placed at the anode in- and

outlet, have the advantage to provide information directly from the fuel gas as well as being a cost-effective technology. Potential suitable sensors are the lambda sensor and the particle sensor, where the latter is in principle an equivalent to the carbon sensor by Haag [101]. The individual positions of the different sensors are depicted in figure 7 and shows the outline of a control system, which can be implemented in a SOFC-system with AEGR for control of characteristic parameters based on the strategy by Carré [75]. Recirculation ratio and fuel gas supply at system inlet are manipulated variables.

Figure 7: Integration of carbon or particle and lambda sensor in a SOFC-system

In the view of the authors, lambda sensors have great potential for use in SOFC-systems, which has been proven in the work by Schäfer et al. [105]. As outlined above, lambda sensors diagnose oxygen excess or shortage in a fuel gas, where the latter is equivalent to the total amount of unconverted fuel gas species. The difference of the measurement signal at the anode inlet and outlet characterizes therefore the fuel conversion in a SOFC-system, which is characterized with stack-FU. The oxygen amount in a fuel gas can be expected to correlate with the O/C-ratio. This information can be used to determine characteristic parameters or related gas properties and finally to control the manipulated values. However, it should be noted that no direct and explicit quantification or differentiation of single gas components can be expected with lambda sensors. An overall statement about the proportion of fuel gas species or the reversed proportion of oxygen-bearing species in a gas mixture can be expected making it a reasonable alternative to the currently used expensive gas measurement technology.

As mentioned above, a higher potential can be attributed to the amperometric lambda sensor due to its continuous linear signal curve with sufficient slope, which is needed for precise quantification of characteristic parameters. The work by Schäfer et al. [105] should be persecuted with subsequent comprehensive simulative and experimental studies as essential content of future work. It comprises the application of the amperometric lambda sensor in anode inlet conditions, the direct integration of lambda sensors in the fuel gas of a SOFC-system to assess the respective interactions and the derivation of control concepts adapted to lambda sensors as well as its robustness.

A great potential has the combination of the information gained from a lambda sensor with observer concepts. This concerns in particular observer concepts, which are based on the imprecise assumption of a measured or known gas species concentration such as Das [84] and others [45,76]. The respective quantity would need to be replaced by the lambda value as overall information of the gas mixture composition. WT application on the measurement signal of the lambda sensor to achieve a deeper analysis of the signal could be also addressed in future work. It is beneficial since the basic structure and functional principle of a lambda sensor is close to a fuel cell with related electrochemical behavior. It should be especially considered, if no correlation can be established based on the sole measurement signal of the lambda sensor. By application of DWT, further parameters can be used to establish a correlation. A combination would be also conceivable, if the open circuit voltage is measured with a single SOFC-cell as sensor, which is based on the cell structure used in the SOFC-system itself as proposed by Keegan [104]. Due to the equivalence and the current-free operation, it can also be used for degradation monitoring of the stack.

While lambda sensors can be considered for the characteristic parameters FU and O/C -ratio, it only applies to the latter in the case of automotive particle sensors, which are intended to measure the soot emissions after the particle filter [112]. In comparison to the carbon sensor by Haag [101] the catalyst pellet is replaced by an interdigital structure with two pectinated electrodes. Although lambda sensors are expected to have the higher potential to be used in SOFC-systems to determine the O/C -ratio quantifiable next to FU , the particle sensor has still some potential for beneficial use as outlined above for the carbon sensor by Haag [101]. Its study should be preferred to the carbon sensor by Haag

[101], since it is not a prototype sensor but a mature and cost-effective sensor technology with proven reliability.

A modified operation mode of the particle sensor and more advanced concepts should be considered. This comprises operation of the particle sensor at a slightly lower temperature than the fuel gas mixture at the anode inlet and the evaluation of the resistance change rate during carbon formation on the sensor surface. Due to a slightly lower temperature, initiation of carbon formation occurs first on the sensor surface. An equivalent process could be implemented for the reformer inlet with the present longer-chain hydrocarbons. The resistance change rate could be also correlated in conjunction with the sensor temperature to the O/C -ratio.

The effect of regeneration and the particles released in its process needs to be evaluated, though. This concerns the operation of the SOFC-system as well as the impacts on the exposed anode. It has led under similar conditions in the development of automotive lambda sensors to the exhaust gas electrode being coated with a porous protective layer to protect it from particles. Since this cannot be considered feasible for the anode in an SOFC-system, the effect of particles during regeneration should be considered in future work. Placing regeneration in stationary operating phases, where no sudden carbon formation can be expected, should be also evaluated.

As a final analysis, the authors expect that none of the presented approaches alone will be sufficient to set up a fully comprehensive control or monitoring concept. The natural gas composition and its variation for instance is expected to have some influence on the signal quality of the proposed hardware sensors. Based on a thermodynamic equilibrium consideration the H/C -ratio influences the equilibrium composition of a fuel gas while all other parameters such as temperatures are kept fixed. Since the hardware sensors such as a particle or a lambda sensor are intended for direct use in the fuel gas, it is expected to have a respective influence, which needs further studies.

It will also come down to a combination of methods, which depends on the specific SOFC-system configuration and application. If the focus in operation of a SOFC-system is on electric power generation, high FU is needed for high system efficiency, which requires precise FU determination. If heat in the exhaust gas is also used, it would be conceivable to lower the stack- FU and thus hedge reasons such as fluctuating natural gas quality with

a safety buffer without the need of exact *FU* quantification. Other available variables can be used for control and EIS or THD can be used as monitoring tool.

6 Conclusions

The authors present an overview on representative control strategies with special focus on SOFC-systems with anode exhaust gas recirculation (AEGR) and different approaches to determine characteristic parameters. The characteristic parameters of SOFC-systems are the O/C -ratio as well as stack- and system- FU .

The overview of control strategies for SOFC-systems shows that the characteristic parameters FU and O/C -ratio are essential variables for the control of a SOFC-system. However, especially in SOFC-systems with AEGR, the characteristic parameters cannot be measured directly during operation. Accordingly, none of the control strategies is based on a respective closed-loop control.

The presented approaches to determine characteristic parameters can be divided into sensor-based and model-based approaches. The sensor-based approaches include the use of a lambda-sensor or a single SOFC-cell. In addition, the use of a carbon sensor, photonic multi-gas sensor and the diagnostic methods EIS, THD and wavelet transformation of the stack voltage are discussed. Model-based approaches include the use of an observer for a SOFC-system or the reformer and the establishment of correlations, which are also referred to as data-driven soft sensors. Furthermore, physical-based and data-driven system models are used. However, none of the approaches found in literature fulfill the need for a sensor determining the relevant information necessary for SOFC system control with general validity and sufficient absolute accuracy.

Therefore, it can be concluded that one or more methods for the determination of the characteristic parameters are still needed and shall be developed in future work. This method should be an approach where no or few simplifying assumptions are made. In addition, a generally valid method is to be aimed at, which can be applied without restriction to different system configurations or layouts. Based on the essential gap regarding the determination of characteristic parameters, comments and statements on more detailed concepts and its respective potential for use in SOFC-systems are given. The online determination of characteristic parameters would enable an enhanced operation of a SOFC-system with higher efficiency, safety, durability and independence

of natural gas composition. This holds true even for dynamic control if the measurement method is sufficiently fast.

7 Appendix

7.1 Control strategies based on PID control

Figure S1 shows a schematic structure of a simple PID control loop, which is used to briefly explain its principle for a better understanding of the reader for respective control strategies. The interested reader is referred to the relevant literature such as Johnson and Moradi [117] or Vilanova and Visioli [118] for its (mathematical) theory, differences as well as application areas.

Figure S1: Schematic principle of a simple PID control loop (based on [119–122]). Step responses (red lines) are shown for the individual elements in the case of a stepwise control deviation e .

A control loop is composed of the controller, the controlled process and a feedback of the controlled variable y [119–121]. In the context of an SOFC application, the SOFC system represents the controlled process and the air inlet temperature of the stack is a possible variable to be controlled. The current value x of a controlled variable y is compared as feedback with the setpoint variable w [119–121]. The difference is the control deviation e , which is due to disturbances on the process [122]. A disturbance can be an intentional change of the setpoint value for the stack current in a SOFC system or the unintentional change due to heat losses, varying natural gas quality or measurement inaccuracies, such as for the recirculation rate. The control deviation e is fed to the controller, which is composed of a combination of P, I and D elements [119–122]. The P element represents

the proportionally acting part with a proportional amplification of the control deviation e [119–122]. The I element is an integrally acting component, whereby the control deviation e is integrated in time to the controlled variable y [119–122]. In comparison, the I element has a slower control behavior than the P element, but can eliminate the control deviation completely [119,122]. The D element represents the differentiating component, which reacts to the change rate of the control deviation e and can only be used in combination with another control element [119–122]. The calculated value for controlled variable y is finally fed to the controlled system, which can be the setpoint of the air flow rate at stack inlet.

7.1 Control strategies based on MPC systems

Figure S2 shows the schematic principle of a MPC control approach. Equivalent to the introduction of PID controllers, this is only intended to contribute to a general understanding of the reader for MPC control strategies. For a detailed description of MPC control the reader is referred to the corresponding literature [123–126].

An MPC control concept is used for the predictive control of multivariable and therefore complex processes [59,127–129]. An MPC control concept essentially consists of the controlled process and the MPC controller, whereby in the context of an SOFC application the SOFC system represents the controlled process [127]. The MPC controller is composed of a discrete-time dynamic simulation model of the controlled process and an optimization part [127–130]. The simulation model needs to replicate the respective process with sufficient accuracy to ensure reliable predictions [127,128].

The simulation models can generally be divided into linear and nonlinear as well as adaptive approaches [58,127,129]. Linear process models are used if the process behavior can be linearized without major deviations or a process being operated at a fixed setpoint, where just disturbances need to be eliminated [129]. Nonlinear models are used for processes with nonlinear behavior and are called nonlinear model predictive control (NMPC) [129]. NMPC can be realized for example with machine learning methods such as artificial neural networks [128,129,131]. In the adaptive approach, the controller or its parameters are adapted to changing process behavior [58].

Based on predicted future input variables (future input prediction), the simulation model predicts the future process behavior for some time steps (state prediction), which is known as prediction horizon [127–130,132]. In this way, the effects of the manipulated variables $y(t)$ on the process can be predicted and optimized for a better and expedient control before feeding it to the real process [129,130]. For optimization, constraints are considered and an objective or cost function needs to be minimized, which requires a controlled process with sufficiently slow system dynamics so that optimization can be performed for each time step [127–130,132]. In addition to the setpoint of the controlled variables $w(t)$, the current state $x(t)$ of the system serves as input for the MPC controller, whereby disturbances on the process can be taken into account [58,129,130]. Predicting and optimizing the setpoints $y(t)$ results in a higher computational effort compared to a PID control approach but enables better control of dynamic and nonlinear processes/behavior in terms of higher control performance [58]. In addition to high computing times, a complex model as needed for MPC control requires a rather high implementation effort for different system configurations and therefore does not represent an easily adaptable and general approach for a control system.

Figure S2: Schematic principle of a MPC system (based on [127,128,130,132])

An improvement of system models, where unknown quantities are present or internal, non-measurable variables are needed for definition of the system model, can be achieved by applying a so-called Kalman filter [133]. For application of a Kalman filter, the system

of interest is described with a mathematical model, which contains the unknown, non-measurable system variables [17,133–135]. The input and output variables of the system model represent measurable variables. The output variables of the system are estimated with the model and compared with the measured values of the output variable. Any difference is then minimized using a Kalman filter as a state estimator by adjusting internal states. If the values match, then the model reflects the real system and the unknown system variables can be calculated using the model. Based on the combination of a model with available measured data by a Kalman filter, the accuracy of calculated output values can thus be increased [133].

7.2 Basics on EIS and THD

In EIS an alternating current is imposed on the SOFC and the responding alternating voltage is measured, so that the impedance can be calculated. This is done for a broad range of frequencies leading to an impedance spectrum, often visualized in a Nyquist diagram. For characterization and analysis of the individual contributions to the measured impedance spectrum of an EIS, equivalent circuit models (ECM) are applied. The different areas in the Nyquist diagram and thus corresponding cell processes are described with elements such as inductors, capacitors, resistors, constant phase elements and Warburg-elements. The parameters of the individual elements can be used for fault detection and isolation using reference values and a fault detection matrix. For the fault analysis, DRT-analysis (DRT: distribution of relaxation times) is also used.

In the THD-method a sinusoidal alternating current is superimposed on the stack operating direct current at a single certain frequency. The resulting alternating voltage is analyzed as a system response regarding harmonic distortions. This is shown in figure S3 schematically on the typical U/I-curve of a fuel cell with an error-free state (blue dotted line) in comparison to a faulty state (red solid line), which can be caused for instance by fuel depletion. In general, the use of THD and EIS should allow the detection of faulty operating conditions in a SOFC such as carbon formation or fuel depletion at an early stage and suitable countermeasures to be taken in time [18].

Figure S3: Schematic principle of a THD analysis (based on [136–138])

7.3 Basics on WT

As can be seen in figure S4, a signal is to be described in WT by means of a wavelet, which generally represents a rapidly decaying, wave-like oscillation with a mean value of zero and, in contrast to sinusoidal waves, has a finite time duration [100,139–143]. There are different mother wavelets (e.g. Morlet, Daubechies, Mexican-Hat, etc.) with different sizes and shapes, which have to be selected depending on the particular application [100,139,140,143–145]. In WT, scaling $a > 0$ by stretching or compressing and shifting b along the time line represent the core concepts and generate wavelet variants based on a basic wavelet [100,139–143].

The main methods in WT are the discrete (DWT) and continuous WT (CWT), which differ in scaling and shifting [139,145]. For CWT, continuous scaling factors can be selected for signal analysis, allowing fine scale analysis. The number of coefficients in DWT is significantly lower than in CWT, resulting in lower memory requirements and shorter computation times [142,146,147]. The basic scaling factor is two and is multiplied by integer values to obtain further scaling factors [141,143,144,146]. Conceptually DWT can be understood as the application of a filter bank to the input signal, in which the signal is divided in multi-stage steps by a high and low pass filter into two sub-frequency bands

with high and low frequency ranges cutting the frequency into half with each step [99,143,144,146,148].

Figure S4: Schematic principle of a wavelet transformation (based [99,100,146])

Glossary

Latin characters

Symbol	Meaning	Unit
d	distance	-
F	Faraday-constant	C/mol
h	enthalpy	kJ
H_i	Lower heating value	kJ/kg
I	Current	A
k	Number of water molecules in ATR-reaction	-
K	Gas coefficients	-
$\dot{m}$	mass flow rate	kg/s
N	Number	-
$\dot{n}$	Molar flow rate of gas	Mol/s
P	pressure	bar
r	Recirculation rate	%
T	Temperature	°C
U	voltage	V
V	Volt	V
x	concentration	%

Greek characters

Symbol	Meaning	Unit
$\Delta^R h_0$	enthalpy of reaction	kJ/kg
Δp	pressure difference	bar
ΔT	temperature difference	K
λ	fuel-air-ratio	-

Abbreviations

Abbreviation	Meaning
AC	Alternating current
AEGR	Anode exhaust-gas recirculation
ANN	Artificial neural network
ASC	Anode supported cell
ASR	Area specific resistance
ATR	autothermal steam reforming
<i>AU</i>	Air utilization
CDB	Carbon deposition barrier
CHP	Combined heat and power system
CH ₄	Methane
CO	Carbon monoxide
CPOx	Catalytic partial oxidation
CWT	Continuous wavelet transformation
DAE	Deep auto encoder
DB	Data-based
DRT	Distribution of relaxation time
DWT	Discrete wavelet transformation
ECM	Equivalent circuit models
EIS	Electrochemical impedance spectroscopy
ESC	Electrolyte supported cell
EWS	External Water supply
<i>FU</i>	Fuel utilization
GPC	Generalized predictive control
HEX	Heat exchangers
<i>H/C</i>	Hydrogen-to-carbon-ratio
LHV	Lower heating value
LSTM	Long short term memory
MIMO	Multiple input multiple output
MPC	Model predictive control

NMPC	Nonlinear model predictive control
PB	Physically-based
PEMFC	Polymer electrolyte membrane fuel cell
PID	Proportional integral derivative
PLC	Programmable logic controller
PRBS	Pseudo-random binary sequence
ppm	Parts per million
O ₂	Oxygen
O/C	Oxygen-to-carbon-ratio
R	Reaction
RBF	Radial basis function
S/C	Steam-to-carbon-ratio
SOFC	Solid oxide fuel cell
SR	Steam reforming
THD	Total harmonic distortion
WT	Wavelet Transformation

Subscripts

Subscript	Meaning
air	Supplied air to fuel cell
an	Anode
biogas	Biogas
blow	Blower
br	Burner
bypass	Bypass
c	Carbon
ca	Cathode
cell	Cell of a fuel cell
CH ₄	Methane
conv	Converted

DC	Direct current
e-	Electrons
El	Elctrical
exhaust	Exhaust
fc	Fuel cell
gas	Fuel gas in the fuel cell system
H ₂	Hydrogen
H ₂ O	Water
in	Inlet
m	Number of H-atoms in a molecule
max	Maximum
min	Minimum
n	Number of C-atoms in a molecule
NG	Natural gas
o	Oxygen
out	Outlet
rad	Radial
rec	Recirculation
ref	Reformer
sta	Fuel cell stack
sys	Fuel cell system
Total	Total amount

8 References

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