

CONDITION MONITORING, PROGNOSIS, AND LIFE EXTENDING CONTROL FOR SOLID OXIDE FUEL CELLS

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SPREMLJANJE IN NAPOVEDOVANJE STANJA TER VO-
DENJE TRDNO OKSIDNIH GORIVNIH CELIC S CIL-
JEM PODALJŠANJA ŽIVLJENJSKE DOBE NAPRAVE

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Abstract

Despite their environmental benefits and high energy conversion efficiencies, solid oxide fuel cells (SOFCs) have yet to achieve their full market potential. Currently, the main focus of SOFC research is on extending the lifespan of the fuel cells. One way of achieving higher reliability and durability of the fuel cells is through the development and application of diagnostic, monitoring, and control algorithms.

This dissertation proposes an integrated control of the SOFC system, where longevity of the stack is directly incorporated in the control strategy, which is a major novelty compared to the conventional control systems. Conventional controllers typically manipulate flows and temperatures of the stack to keep the system running in safe operation. Conversely, this thesis develops an integrated control system that has direct impact on the durability of the stack, thus making the system health-aware. The implementation of the proposed solution requires development of several conceptually different, yet interconnected modules.

The controller consists of two layers. The low-level part keeps the system operating in the corresponding set-point, while the supervisory controller (SC) defines the said set-points. The SC is formulated as a nonlinear program which minimises the cost function of two conflicting objectives: (i.) the degradation rate of the SOFC stack, and (ii.) system efficiency.

While the system efficiency can be easily calculated, the degradation rate has to be estimated via structure of Kalman filters (KFs). The so called state of health (SoH) estimator implements a lumped stack model in an unscented Kalman filter (UKF) algorithm to recursively identify the area specific resistance (ASR) of the stack. In the subsequent step, a model describing temporal evolution of the ASR is identified from the semi-past data, and remaining useful life (RUL) predictions are obtained accordingly. The developed approach is invariant to operating conditions which gives it a major advantage over the conventional solutions.

Physical sensors are essential in control and monitoring of any system. However, installing in-situ instrumentation becomes impractical on an industrial scale. Therefore, this thesis develops soft sensors capable of estimating process variables such as stack temperature gradients and fuel gas composition. The temperature soft sensor can be directly exploited by the low-level controller to constrain the stack gradients, while the gas composition estimates are directly employed by the SoH estimator.

Both the soft sensors and SoH estimator are experimentally validated on a 10 kW SOFC system. The integrated control strategy, however, is fully validated on the model of the said 10 kW system. The simulation study shows the capability of the developed control system to minimise the degradation rate of the stack, thus extending the total life span of the system. It is shown that an increase in lifespan comes at the cost of system efficiency.

Furthermore, the control system with an integrated SoH estimator and soft sensors were implemented on standard industrial equipment. All of the algorithms were tuned to the experimental system and run on-line. Due to the issues related to poor condition of the system, only a preliminary experimental validation is discussed in the thesis.

Povzetek

Navkljub številnim okoljevarstvenim prednostim in visokem izkoristku trdno-oksidge gorivne celice (SOFC) še ne dosegajo polnega tržnega potenciala. Trenutno so raziskave usmerjene v podaljšanje življenjske dobe naprave. Med drugim lahko višjo vzdržljivost in zanesljivost gorivnih celic zagotovimo z razvojem diagnostičnih algoritmov, ter algoritmov za vodenje in spremljanje stanja.

Delo naslavlja sintezo integriranega vodenja SOFC sistema, v okviru katerega je zagotavljanje daljše življenjske direktno vključeno v strategijo vodenja. Prav ta novost loči predlagan pristop v primerjavi z običajnimi. Pri tem sistem vodenja poleg klasičnih veličin, kot so pretoki in temperature, vključuje še stanje SOFC sklada. Implementacija integriranega sistema vodenja zahteva razvoj nabora konceptualno različnih, vendar medsebojno povezanih modulov.

Vodenje sistema je realizirano na dveh nivojih. Nižji nivo vodenja sestavljajo klasične regulacijske zanke, ki skrbijo da sistem deluje v ustrezni delovni točki, med tem ko nadzorni regulator (SC) definira prav to delovno točko. Nadzorni regulator je formuliran v obliki nelinearnega programa, ki minimizira kriterijsko funkcijo sestavljeno iz dveh nasprotujočih zahtev: (i.) čim počasnejša degradacija SOFC sklada ter (ii.) čim višji izkoristek sistema.

Medtem, ko je izkoristek sistema moč zlahka izračunati, pa moramo za oceno hitrosti degradacije uporabiti strukturo Kalmanovih filtrov (KF). Najprej s pomočjo nepristranskega Kalmanovega filtra (UKF) in modela SOFC sklada izračunamo značilko, ki opisuje stanje sklada (SoH). V našem primeru je to specifična upornost gorivnih celic (ASR). V naslednjem koraku iz pol-preteklih podatkov identificiramo model, ki opisuje časovni potek omenjene upornosti ASR. Ta model lahko nato s pridom uporabimo za napovedovanje preostale življenjske dobe naprave (RUL). Glavna prednost razvitega pristopa je zanesljiva ocena SoH ter napoved RUL navkljub spremenljivim delovnim pogojem.

Senzorji so bistvenega pomena pri vodenju in spremljanju stanja katerekoli naprave. Namestitev nekaterih senzorjev na komercialni napravi lahko hitro postane nepraktično, oziroma drago. Zatorej bomo v okviru dela predstavili tudi pristop za razvoj mehkih senzorjev. Predstavili bomo dva takšna senzorja, s katerimi je moč oceniti temperaturne gradientne znotraj SOFC sklada, kot tudi sestavo plinov na različnih lokacijah v sistemu.

Oba mehka senzorja, kot tudi algoritem za ocenjevanje SoH in napovedovanje RUL so eksperimentalno preizkušeni na 10 kW SOFC sistemu. Integrirani sistem vodenja pa je v celoti validiran le na modelu omenjenega 10 kW sistema. Simulacijski rezultati kažejo, da je z implementacijo razvitega integriranega sistema vodenja moč zmanjšati hitrost degradacije sklada, s tem pa povečati življenjsko dobo sistema. Rezultati kažejo, da lahko dobo podaljšamo na račun zmanjšanja izkoristka.

Celotni sistem vodenja z integriranimi mehкими senzorji in ocenjevalcem SoH smo implementirali na standardni industrijski opremi. Vsi algoritmi so bili ustrezno nastavljeni in uporabljeni med delovanjem sistema. Zaradi slabega stanja sistem, v delu obravnavamo le preliminarno analizo rezultatov.

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List of Abbreviations

AC	adaptive control
AFC	alkaline fuel cell
ANNC	artificial neural network control
AOGR	anode off-gas recycle
ASR	area specific resistance
BoL	beginning-of-life
BoP	balance of plant
c.i.	confidence interval
CM	condition monitoring
DMFC	direct methanol fuel cell
ECM	equivalent circuit model
EIS	electrochemical impedance spectroscopy
EoL	end-of-life
EU	European Union
FCH JU	fuel cells and hydrogen joint undertaking
FDI	fault detection and isolation
FF	feed-forward
FLC	fuzzy logic control
FSM	fault signature matrix
FTA	fault tree analysis
FTC	fault tolerant control
FU	fuel utilisation
HT-PEM	high temperature polymer electrolyte membrane
KF	Kalman filter
MC	Monte Carlo
MCFC	molten carbonate fuel cell
MPC	model predictive control
OCR	oxygen-to-carbon ratio
P&O	perturbation and observe
PDE	partial differential equation
PDF	probability density function
PEMFC	proton exchange membrane fuel cell
PI	proportional-integral
PID	proportional-integral-derivative
PLC	programmable logic controller

RUL		remaining useful life
SAFC	. . .	sulphuric acid fuel cell
SC		supervisory controller
SOEC	. . .	solid oxide electrolysis cell
SOFC	. . .	solid oxide fuel cell
SoH		state of health
SPFC	. . .	solid polymer fuel cell
TEC		thermal expansion coefficient
UKF		unscented Kalman filter
YSZ		yttria-stabilized zirconia

Chapter 1

Introduction

Ensuring sustainable power generation is becoming a global challenge. The scientific community addresses the issue by developing new, more efficient, and cleaner systems for power generation. With an optimistic outlook on the future, a bundle of promising green technologies are participating in the race. Among others, fuel cell technology is an important competitor.

A fuel cell is an energy conversion device, which converts chemical energy into electrical energy through electrochemical combination of hydrogen and oxygen [1]. The most attractive characteristic of a fuel cell is clean energy production at high-efficiency rates [2]. In ideal situations, the only by-products of the energy conversion are pure water and heat. However, pure hydrogen is rarely found in nature and needs to be acquired elsewhere. Based on the hydrogen source, fuel cell technology distinguishes among solid oxide fuel cells (SOFCs) [3], low-temperature proton exchange membrane fuel cells (PEMFCs) [4], high temperature polymer electrolyte membranes (HT-PEMs) [5], direct methanol fuel cell (DMFC), sulphuric acid fuel cell (SAFC), molten carbonate fuel cell (MCFC), solid polymer fuel cell (SPFC), and alkaline fuel cell (AFC) [2].

There are numerous advantages for SOFCs operating at high temperatures (600°C - 1000°C). Firstly, high operating temperatures of SOFC activate catalytic activity of the fuel cell, hence facilitating internal reforming of the different types of fuel. High fuel flexibility makes the SOFC more versatile as it may be used with most of the hydrocarbon fuels in gaseous or even liquid form. Furthermore, the unspent exhaust energy can be recuperated via e.g. gas turbine, or heat exchangers, thus inducing the overall efficiency of the SOFC system to range up to 90%[6].

Another advantage of the SOFCs is their applicability in reversible, electrolysis mode. Under such operation, the fuel cells can be used for hydrogen production from carbon dioxide and water vapor. The fuel cells employed in electrolysis mode are commonly referred to as solid oxide electrolysis cells (SOECs). Taking into account the capability of the same device operating in SOFC or SOEC mode, one can understand why these types of fuel cells can be coupled with other eco-friendly energy sources as, for example, surplus energy storage devices.

The market position

The SOFC market can be segmented as in Figure 1.1 [7]. The applications are diverse and range from micro-fuel cells with less than 1W power outputs to multi-MW power generation plants. SOFCs are applicable in stationary standalone applications as well as a part of smart grids. Stationary fuel cell applications cover emergency back-up power supply (EPS), remote-area power supply (RAPS), and distributed heat and power generators (CHP). Portable applications of fuel cells include power generators for outdoor personal

uses (such as camping or climbing), power supply for consumer electronic devices, and military equipment and transportation [8], [9].

SOFCs were first successfully used in the Apollo programme in the late 1960s. Since then, there have been ups and downs in regards to investments and research endeavour cycles in the development of the technology. Although viewed as “almost ready to go” a decade ago, we are still not witnessing a massive deployment of the SOFC technology in the market. The commercialization of SOFC technology is stuck in the Technological Valley of Death, which exists between the pilot/demonstration and commercialization phases of the technological development cycle. Significant financial resources are required to introduce fuel cell products on the market. Recently, both Topsoe Fuel Cells (in Denmark) and CFCL (in Australia and Germany) announced to cancel their SOFC related activities.

On the other hand, some market reviews like the report [10] provide an encouraging perspective for the SOFC market. The report defines and segments the global solid oxide fuel cell market with an analysis and forecast of revenue. The SOFC market will grow from an estimated \$2.19 billion in 2013 to \$3.32 billion by 2018. Part of the market activities that are hoped to trigger more massive deployment are supported by public bodies, like the EU Commission through the Ene.field project [11] or the Japanese government through the NEDO project [12].

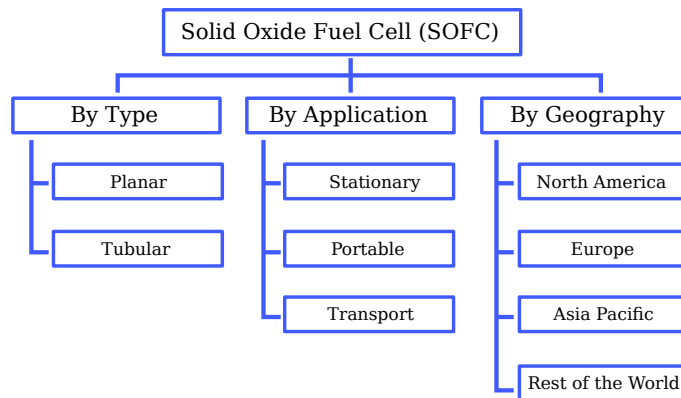


Figure 1.1: Global SOFC market segmentation [7].

Research endeavour

The European Union (EU) supports the development of fuel cells through the fuel cells and hydrogen joint undertaking (FCH JU). A total of over 500 M€ has been devoted to the development of stationary fuel cell systems [13]. Among others, SOFCs hold the leading position for stationary applications. SOFCs receive approximately 47% of the total FCH JU funding. Despite the vast effort devoted to perfecting the technology, broad market adoption of the SOFC systems remains rather modest [13], [14]. The two main drawbacks are the high cost of the fuel cells, and the rapid degradation of the cell materials. Currently, the average stack costs approximately 4,200 €/kW, with an average lifetime of over 30,000 hours. The research and innovation projects focus on all aspects of the fuel cells stack, from basic material research up to the manufacturing processes. FCH JU has given notable attention to the development of diagnostic, monitoring and control of the SOFC systems with the aim of improving efficiency, reduction of degradation, total cost, durability and reliability.

1.1 SOFCs in a Nutshell

While descriptions of the theory and phenomena involved are abundant in the literature, e.g. [3], [15], [16], the following paragraphs cover only the basic working principle of the SOFCs.

The SOFC is an electrochemical device, which converts chemical energy directly into electrical energy. The energy conversion is achieved via electrochemical oxidation of hydrogen with oxygen. Since the energy conversion is direct, i.e. without first converting chemical energy into mechanical, the SOFCs can achieve high efficiencies.

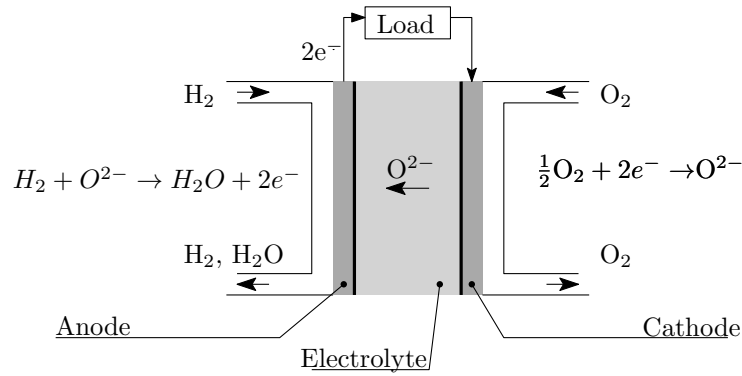


Figure 1.2: The basic principles of a SOFC and its essential components.

Figure 1.2 displays the basic principles and essential components of a single cell. A cell consists of three adjacent layers: (i.) anode, (ii.) cathode, and (iii.) electrolyte. The main purpose of the electrolyte is to transport oxygen ions O^{2-} from the cathode to the anode, while at the same time preventing direct contact between anode and cathode chambers. The ions are formed via oxygen reduction reaction at the cathode, which is continuously fed with oxygen. On the other side, at the anode, the O^{2-} ions react with hydrogen in the process of hydrogen oxidation. In addition to H_2O , two electrons and some heat are released. The electrons travel through the external load to reach the cathode, where they participate in oxygen reduction reaction. In addition, one of the major advantages of the SOFC is its ability of internal reforming of the fuel. This property allows the fuel cells to operate not only on pure hydrogen, but on also other hydrogen-rich fuels, such as methane.

Theoretically, the single SOFC voltage can reach up to 1.2 V, depending on temperature, pressure and gas composition. The power output, however, is heavily conditioned with the active area of the fuel cell. Typically, the larger the active area of the cells, the higher the power output. However, the limiting factor for the size of the individual fuel cells is related to the thermo-mechanical stress induced due to thermal expansion coefficient (TEC) mismatch of the adjacent layers. With larger cells, temperature gradients over the cells become larger, thus causing higher stress on the materials, hence compromising their safe operation. Therefore, to increase power output, a number of fuel cells is connected in parallel to form a fuel cell stack.

Since the SOFCs operate at high temperatures, the stack is usually enclosed in an insulated housing to reduce heat losses. The stack then connects with a balance of plant (BoP) module, which carries out the pre-treatment of the incoming gases. The BoP consists of different interconnected components, such as heat-exchangers, blowers, fuel reformers, pipes, valves, etc.

1.2 Degradation Mechanisms

Some of the degradation mechanisms occurring in the fuel cells can be intuitively correlated with the high temperature at which SOFC stacks operate. Generally, thermal stress caused by a mismatch in TEC represents one of the main factors that shorten the lifespan of the SOFC [17]–[19]. On the other hand, micro-structural degradation is known to actively reduce the total operational time of an SOFC. The phenomena usually occur in the porous material of the anode, cathode, or electrolyte, and cause severe changes in porosity distribution of the material after long periods of operation [20], [21]. Similarly, improper reaction conditions may lead to chromium poisoning of the electrodes [22] and carbon deposition on the anodes [23].

The degradation mechanisms that hinder the overall performance of the SOFC stack can be classified into several groups:

- Thermo-mechanical stress. High operating temperatures make the SOFC prone to thermo-mechanical stress, especially during the start-ups, shutdowns, and rapid load changes. The stack components (anodes, cathodes, electrolytes, seals, interconnect, etc.) are made of different materials, which have different TEC. Therefore, a temperature gradient over the stack that is too high causes severe mechanical stresses, which results in cracks in different components, e.g. sealing. This issue is being intensively studied in e.g. [24]–[26]
- Anode/Cathode delamination. Delamination occurs directly on the interface of adjacent layers [27]. The detachment can be caused by repeated cycling of the SOFC from room to operating temperatures [28], build-up of thermal stresses during manufacturing of the materials [29], and/or heat dissipated due to current load cycling [30]
- Nickel oxidation. Another major degradation phenomenon occurs directly on the porous Ni-doped yttria-stabilized zirconia (YSZ) anode, when the fuel supply to the anode is interrupted either intentionally or due to a fault [31]. In such a case, the oxygen partial pressure at the anode is increased, causing formation of NiO, hence impeding the performance of the fuel cell. Even though the phenomenon is reversible – NiO reduces under hydrogen supply [32] – the redox cycling may cause severe changes in the cermet’s microstructure leading to faster, irreversible performance degradation [33].
- Nickel coarsening. Nickel coarsening is a well known and studied degradation phenomenon in the Ni-YSZ anodes [34]–[36]. The Ni particles in the anode serve dual roles as catalyst for hydrogen oxidation and electrical current conductors, while also facilitating steam reforming of the hydrocarbon fuels [37]. However, long term operation at the elevated temperatures cause the kinetics of the Ni particles. Initially the nickel particles are uniformly spread on the anode surface to facilitate catalytic performance of the fuel cells and agglomerate with time. Coarsening can be influenced mainly by temperature [21], [38], and fuel gas humidity [39].
- Carbon deposition on the anode surface. The fuel flexibility of SOFCs is one of their major advantages. However, the direct use of hydrocarbon fuels leads to a faster degradation of the cells, a phenomenon known as carbon coking, or carbon deposition. When carbon coking occurs, reaction sites are blocked, hence hindering the performance of the fuel cells [40]–[42]. The subject has been extensively studied, and it has been found that coking is mostly affected by the temperature and vapor content within the cell, both of which are directly associated with vapor formation due to hydrogen oxidation [43]–[48].

- Anode poisoning. Special attention is required when fueling the cells with commercially available hydrocarbon based fuels, since these may contain additives and impurities that might cause anode poisoning. Most experimental studies focus on sulphur poisoning of the anodes, either under pure hydrogen operation [49]–[51], or hydrocarbon fuel mixtures [52]–[54]. The effects of poisoning can be observed as performance deterioration [51], [55].
- Interconnect oxidation and chromium poisoning. Interconnects in planar SOFCs are typically made of cheap ferritic stainless steel, which usually has 20-30% chromium content [56]. However, with ongoing operation, the electrical conductivity of the interconnect-electrode interface is degraded due to formation of a chromium oxide scale on the steel. The oxidation rate increases with higher oxygen partial pressure [56]. Furthermore, the chromium oxides formed may be outgassed and deposited to the cathode and lead to reduction of oxygen reduction reactions [57]–[59].

At a given moment, more than one degradation phenomenon can be present within the stack. For instance, higher current densities cause higher thermal stress on the material, generating overall stress on the cell micro-structure. On the other hand, with higher current densities, higher amounts of water vapor are formed at the anode reaction sites, thus in a way protecting the anode from solid carbon formation [44]. Moreover, in the above mentioned references, as well as in others, for instance, in [60], it has been shown that the overall degradation rate of the SOFC is strongly influenced by operating conditions. For instance, Ni oxidation can be avoided by keeping the fuel utilisation (FU) sufficiently low. Or, by keeping the oxygen-to-carbon ratio (OCR) above corresponding level, the carbon coking can be prevented. Furthermore, the temperature is the most influential factor for many degradation mechanisms [61], e.g. Ni coarsening, cracking, delamination. A more thorough review of the degradation mechanisms is available in [62].

The main effect of these degradation phenomena can be observed as an increase of cell losses. As the material ages, the parasitic phenomena take place inside the stack and accumulate over time. Consequently, the polarisation losses gradually increase and result in a gradual decrease in voltage. In some cases, electrochemical impedance spectroscopy (EIS) can be used to quantify this change. Nevertheless, the exact cause of the failure of the fuel cells can only be diagnosed by postmortem analysis.

1.3 Towards Higher Reliability and Efficient Operation

In the last years, more and more interest has been given to raising the reliability of SOFC operation and its life span. Consequently, a great deal of effort has been spent in developing condition monitoring of SOFC systems, particularly in diagnosis. In the most recent literature, prognostics of SOFC systems has been receiving the focus. Most of the research into control and optimal operation has been completed during the last decade or two, with the main focus being on basic control for stabilizing the operating point. A range of mostly model-based methods has been proposed.

At least for the SOFCs, these topics have been treated separately. So far, to the best of the author's knowledge, no attempt to their holistic treatment has been given. That will be presented in Chapter 5 below.

Diagnosis of SOFC systems

There are a plethora of papers dealing with diagnostics of SOFC. The approaches in these papers can be classified as (i.) active methodologies (based on EIS measurements),

and (ii.) passive model-based approaches. The EIS approaches belong to a group called active diagnosis, where system excitation is required in order to extract the relevant information. Alternatively, the passive model-based approaches employ already available operational measurements, i.e. require no additional excitation and instrumentation to infer the diagnostic information.

Usually the approaches that employ EIS measurements observe the impact of various degradation mechanisms on the impedance of the fuel cells. The EIS measurements are performed at a fixed operating point of the system. The current is then superimposed with the corresponding excitation signal, and the voltage response is measured [63]. The amplitude of the excitation signal is selected sufficiently low, so that the highly non-linear nature of the fuel cells can be approximated by simple linear models, e.g. non-parametric impedance spectra, equivalent circuit model (ECM). The changes in spectra or parameters of the ECM are associated with degradation mechanisms occurring in the cells/stack [40].

Conversely to the EIS approaches, the model-based approaches build on simplified physical or data-driven models of the SOFC system [64], [65]. In such a case, the parity relation is used for fault detection and isolation (FDI) [66]. The approaches following this methodology, in its simplest configuration, simulate the model of the system in parallel with the process for consistency check. When the residual between measurement and model output is (ideally) zero-mean white Gaussian noise, the system is considered fault-free. In the opposite case, when the residual departs from its nominal behavior, incipient fault is detected. The approaches that fall within this range mainly deal with detecting incipient faults in auxiliary components installed around the fuel cell stack.

Prognosis

In addition to diagnostic information, on-line predictions of remaining useful life (RUL) of the fuel cell stack is of particular interest for the overall management of the system. The RUL prediction can be efficiently exploited for scheduling of the maintenance works so that either stack can be refurbished or replaced in a timely manner. On the other hand, concurrent information about the ongoing degradation rate can be effectively exploited by the properly designed control system. However, even though significant work has been done on PEMFCs [67], [68], only a few results exist for the SOFCs [60], [69], [70]. The underlying work is aimed to reduce the gap.

Performance optimisation

High performance of complex systems can be achieved by efficient operation of all integrated components, which puts constraints on the overall operation of the system [16], [71].

There have been a plethora of control systems developed for PEMFC systems, see the review paper [72]. These approaches apply different control methods, from classical feedforward-feedback proportional-integral-derivative (PID) controllers, to adaptive control (AC), model predictive control (MPC), artificial neural network control (ANNC) and fuzzy logic control (FLC). Their main control objective is to prevent fuel starvation of the fuel cells, regulate flow rates and temperatures of the input gases, etc. Similar conclusions can be drawn for the control of the SOFC systems. The control systems need to handle the abrupt change in the fuel cell voltage during transient operation, load following, respect the constraints of the components, and improve efficiency [71], [73]–[78]. A more thorough review is given in Chapter 5.

Further maximisation of the reliability and availability of the fuel cell systems is possible through fault tolerant control (FTC). The FTC system is a control system which is able

to accommodate faults among system components while also maintaining system stability and the desired level of system performance [79]. While the issue has been studied to some extent for the PEMFCs [80], literature on FTC on SOFC is scant. To the best of the author's knowledge, only two such control strategies have been developed so far [81], [82].

1.4 Purpose of the Dissertation

The main challenge of this work is to answer the following question:

"How to design a control system that will be capable of exploiting the information on system condition and achieving optimal efficiency while slowing down the degradation processes as much as possible in order to reach higher reliability of operation and longer life of the system?"

This should be done at minimal cost of instrumentation, minimal computational burden and easy maintainability. Since no additional and expensive instrumentation is allowed, knowledge of internal process variables, needed to monitor the operational constraints, have to be *inferred* from available process data.

Soft sensor design

As discussed above, the longevity of a SOFC stack is greatly affected by the imposed operating conditions. However, measuring some of the influential variables, such as inner stack temperatures and gas composition, is too costly for commercial applications. The remedy is to replace physical sensors by *soft sensors*.

Information about the thermal condition inside the stack is of significant practical importance for SOFC operation. Unfortunately, in-situ thermocouples installed on industrial scale stacks are extremely impractical due to issues related to sealing and wiring. Therefore, this thesis investigates whether stack internal temperatures can be reconstructed by employing mathematical models and conventional, more easily accessible, process variables.

OCR is widely accepted parameter to characterize the reforming reactions inside a SOFC under loaded conditions [15]. Improper reforming conditions inside the stack may foster parasitic chemical reactions that accelerate carbon deposition on the active surface. It is practical to have information regarding reforming conditions at hand, i.e. OCR. However, on-line chemical measurements are feasible only in the laboratories. Installation of physical sensors would be too costly in commercial applications. This thesis investigates a possible remedy based on software sensors for the reconstruction of gas composition inside the fuel cell system employing simple structures.

Condition monitoring

Having the information regarding state of health (SoH) of the stack can significantly improve the overall reliability and availability of the SOFC system. Usually, the authors consider voltage as a relevant variable to describe momentary SoH. The degradation rate is usually expressed as the percentage of voltage drop per 1000 hours. However, particularly under non-stationary operation, fluctuations in the voltage hinder this kind of characterisation. Therefore, developing a SoH estimator that is independent of operating conditions is of vital importance. Then, the SoH information and its historical evolution can be easily exploited for RUL predictions.

Health-aware performance optimisation

Including the overall health of the system can greatly benefit the overall performance of the system throughout its life-span. Only very few works have appeared in other areas [83],

[84], but no such attempt has been made on SOFCs. The point is that the slow and gradual degradation of the fuel cells has to be taken into consideration when optimising the operational conditions of the SOFC system.

The optimising performance strategy tries to find a balance between contradictory objectives: (i.) maximisation of the power conversion efficiency, and (ii.) maximising the life-span of the SOFC system. To this aim, a two-tier control structure is proposed, which consists of low-level controllers and the supervisory control module. The main objective of the low-level controllers is to keep the system operating at the desired set-point. The task of the supervisory controller is to take into account information of the stack operation and define the set-points for the low-level controller. By exploiting information on the health of the fuel cell stack as an input to the controller, the SOFC system becomes health-aware.

1.5 Aims

The aim of this dissertation is to come up with a set of algorithms that will improve durability, reliability, and the life span of the SOFC system. At the same time efficiency of the stack should be considered. To do so, we investigate an integrated control strategy which includes several modules. The thesis addresses design and experimental validation of

- the soft sensor capable of estimating SOFC stack internal temperatures,
- the soft sensor of fuel gas composition of a SOFC system,
- the state of health estimator and predictor of the RUL,
- health aware control of a SOFC system.

1.6 Hypotheses

The dissertation builds on the following hypotheses:

- Thermal and chemical variables inside the stack can be reconstructed by employing relatively simple models and easily accessible process variables.
- SoH estimation and RUL predictions of the SOFCs can be achieved under non-stationary operating conditions.
- By utilizing the information on health of the stack in control design, a longer lifespan of the fuel cells can be achieved.

1.7 Scientific Contributions

The research, work, and findings presented within the dissertation mainly contribute to the field of diagnostics, condition monitoring, and control of the SOFC systems. The original scientific contributions are as follows.

A systematic approach for reconstruction of internal stack temperatures.

Published in: B. Dolenc, D. Vrečko, Đ. Juričić, *et al.*, “Online estimation of internal stack temperatures in solid oxide fuel cell power generating units,” *J. Power Sources*, vol. 336, pp. 251–260, 2016

The paper proposes a systematic approach to the development of an estimator capable of reconstructing temperatures inside the stack. First, the approach discusses the causal relations among corresponding process variables and reconstruction of the maximal/minimal stack temperatures. Second, employing the experimental data, the structure, as well as parameters, of the soft sensor is sought directly on the historical recordings employing the sub-space identification methodology. Two sub-models are considered, one that maps process inputs with internal temperatures, and a second that maps the input variables directly with the temperature of the stack outlets. Finally, the two sub-models are then merged into a single soft sensor that employs both process input variables and temperatures of at the stack outlets to reconstruct the internal temperatures. Despite the structural simplicity of the soft sensor, the results demonstrate high precision estimates even under a degrading stack.

A fuel gas composition estimator of a SOFC system.

Published in: B. Dolenc, D. Vrečko, Đ. Juričić, *et al.*, “Online gas composition estimation in solid oxide fuel cell systems with anode off-gas recycle configuration,” *J. Power Sources*, vol. 343, pp. 246–253, 2017

The paper proposes an on-line approach for fuel gas composition in a SOFC system in anode off-gas recycle (AOGR) configuration. For the systems in AOGR, estimating gas composition is a non-trivial task. The classical approach, employing equilibrium calculations, becomes computationally demanding due to partial recirculation of the anode exhaust. Therefore, particularly for on-line implementation, the paper proposes, and justifies, a number of simplifications, which result in an estimator of structural simplicity.

As an alternative, the work also discusses a data-driven approach. In this case, a model for the soft sensor is identified directly from available experimental data. It is shown that higher precision estimates can be achieved in this way, provided a large amount of experimental data is available.

The paper concludes with a hybrid estimator, which combines the simplified equilibrium calculation and the data-driven term, which provides high precision estimates. Compared to a purely data-driven approach, only a small part of experimental data is required.

SoH estimation and RUL prediction.

Published in: B. Dolenc, P. Boškoski, M. Stepančič, *et al.*, “State of health estimation and remaining useful life prediction of solid oxide fuel cell stack,” *Energy Convers. Manag.*, vol. 148, pp. 993–1002, 2017, ISSN: 0196-8904

In this work, SoH estimation and RUL prediction for the SOFC stack is discussed. The paper proposes a model-based approach for on-line SoH estimation. Employing a lumped stack model in an unscented Kalman filter (UKF) algorithm, the estimator recursively identifies the area specific resistance (ASR) of the stack and employs it as a feature for SoH. With ASR as SoH feature, the approach becomes independent of operating conditions. For the RUL prediction, the semi-past data from the SoH estimator is employed to identify its corresponding model of time evolution. With this model at hand, straightforward prediction of temporal SoH evolution is available to predict RUL accordingly.

Health-aware performance optimisation.

In this contribution, the thesis proposes an integrated control system. The so-called health-aware control employs information about the SoH of the system and aims at maximising the life-span of the fuel cell stack. A two-tier control strategy consists of low-level controllers, which keep the system operating in corresponding operating point, while the supervisory controller optimises the said operating point with respect to a set of predefined control objectives. The problem is formulated as a nonlinear optimisation program. The control objectives are condensed in a form of cost function with a set of constraints, where the degradation rate and system efficiency play a major role. The problem is solved by employing extremum seeking model-free optimisation. To the best of the author's knowledge, a similar idea has never been addressed before, at least in the field of SOFCs.

1.8 Structure of the Dissertation

The remainder of the thesis is organised as follows. Chapter 2 describes the approach for fuel gas composition estimation in the system in AOGR configuration. Further, Chapter 3 describes the temperature soft sensor capable of reconstructing stack internal temperatures. Chapter 4 presents the SoH estimator and RUL predictor for the SOFC stack. Finally, Chapter 5 integrates the above mentioned modules in an integrated health-aware control strategy. Furthermore, a set of simulations is performed for validation purposes.

All chapters include experimental validation of what they propose. The fully integrated control system is extensively validated only on a system model, while also supported with a short experimental campaign.

Chapter 2

Soft Sensors for Fuel Gas Composition

One of the most critical operating parameters of the SOFC is the fuel gas composition feed to the anode. Having on-line information about the fuel gas composition can be beneficial in more than one way. First, with an improper fuel feed, the system can be brought to the operating mode where formation of solid carbon can deposit on the anode surface [88]. Second, the information about the molar flows to the stack will be used further in the thesis. For instance, Chapter 4 develops an algorithm, where gas composition at the anode inlet is one of the main variables required for corresponding calculations.

As discussed in the Introduction, one way of avoiding carbon deposition is by keeping the so-called OCR above a certain limit. OCR is simply a ratio of the oxygen and carbon atoms inside the fuel stream [15]. In order to monitor the value of OCR, one needs precise knowledge of the gas molar flows to the stack. While it is entirely feasible to sample and measure the composition of the gas fuel flow, it becomes too costly for application on an industrial scale.

The chemical composition of fuel gas flow to the anode depends on several factors, out of which the fuel flow to the system and load demand are the most influential. However, due to fuel reforming within the pre-reformer, as well as inside the stack, the gas composition changes when passing through the reaction chambers. Moreover, one also needs to take into account partial recycling of the anode exhaust, the configuration commonly known as AOGR. Mathematical models that enable estimation of the gas composition at different points inside the system assume thermodynamic equilibrium. To solve these algebraic models one needs to employ cumbersome numerical optimisation methods.

The soft sensors in our work are able to reconstruct gas composition at the anode inlet, thus providing information regarding reforming and electrochemical conditions inside the stack on the system level. The equilibrium calculation is simplified down to the bare minimum, so to avoid the optimisation tools. The soft sensors that are based solely on 1st principles are found to be inaccurate when neglecting phenomena such as leakage in the stack. Therefore, the work considered in this chapter also develops data-driven and hybrid fuel gas composition estimators.

The developed approaches mainly address the estimation of gas composition of a SOFC system in AOGR configuration. Nevertheless, the soft sensor based on the simplified 1st principle model can easily be adapted for different system configuration, e.g. a system excluding anode recycling. Furthermore, the design of data-driven soft sensor is rather general. Provided the experimental data is available, the approach can be adopted for any stack/system configuration. However, the lack of such data limits the applicability of the developed algorithms to the use of the simplified 1st principle approach.

The chapter includes the following publication:

B. Dolenc, D. Vrečko, Đ. Juričić, *et al.*, “Online gas composition estimation in solid oxide fuel cell systems with anode off-gas recycle configuration,” *J. Power Sources*, vol. 343, pp. 246–253, 2017



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Online gas composition estimation in solid oxide fuel cell systems with anode off-gas recycle configuration

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H I G H L I G H T S

- Gas composition estimators for SOFC systems in AOGR configuration are considered.
- Distinct approaches are developed and compared: data-driven and stoichiometric.
- Simple estimators of high precision are demonstrated.
- The developed estimators are reliable and easy to implement on standard PLCs.
- Implementation is demonstrated on 10 kW SOFC system.

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Degradation and poisoning of solid oxide fuel cell (SOFC) stacks are continuously shortening the lifespan of SOFC systems. Poisoning mechanisms, such as carbon deposition, form a coating layer, hence rapidly decreasing the efficiency of the fuel cells. Gas composition of inlet gases is known to have great impact on the rate of coke formation. Therefore, monitoring of these variables can be of great benefit for overall management of SOFCs. Although measuring the gas composition of the gas stream is feasible, it is too costly for commercial applications. This paper proposes three distinct approaches for the design of gas composition estimators of an SOFC system in anode off-gas recycle configuration which are (i.) accurate, and (ii.) easy to implement on a programmable logic controller. Firstly, a classical approach is briefly revisited and problems related to implementation complexity are discussed. Secondly, the model is simplified and adapted for easy implementation. Further, an alternative data-driven approach for gas composition estimation is developed. Finally, a hybrid estimator employing experimental data and 1st-principles is proposed. Despite the structural simplicity of the estimators, the experimental validation shows a high precision for all of the approaches. Experimental validation is performed on a 10 kW SOFC system.

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1. Introduction

Following the first commercialization successes in Japan in the area of combined heat and power generation [1], solid oxide fuel cells (SOFCs) are showing promising prospects in stationary applications. However, even low CO₂ emissions and high efficiency are insufficient for final market deployment of the technology due

to problems associated with durability, reliability and, finally, the continuous high cost of SOFC systems [2].

During the 2014–2020 time period, the European Union is striving to increase electrical efficiency and the durability of fuel cells [3]. To address these challenges, significant effort has been invested in better understanding degradation mechanisms and strategies for their accommodation.

Anode off-gas recycle (AOGR) configuration in an SOFC system is often employed in order to increase the overall efficiency of power conversion. In such a configuration, a portion of steam rich anode depleted gas is recirculated allowing extra heat to be recuperated

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and used to heat up the stack inlet gases. Benefits of the AOGR on the efficiency of the system are described in Refs. [4–10]. Further, a significant effort is being devoted to the development of control strategies that are also aiming at higher efficiency, while also considering the durability of the stack [11–13]. Control strategies in these papers aim to control stack temperatures in an effort to limit the degradation rate of the stack materials, e.g. temperature dependent nickel agglomeration [14].

Among others, carbon deposition of anode is one of the main degradation phenomena occurring in SOFC stacks [2,15,16]. Reaction sites are blocked by carbon layer on the surface, hence lowering the efficiency of the system. However, in Ref. [17] it is shown that by proper conditioning of the input gases, i.e. temperature, pressure, and oxygen-to-carbon ratio (OCR), carbon deposition can be mitigated.

OCR is a widely accepted variable that characterises reforming reactions inside SOFC under loaded conditions [17]. Improper reforming conditions inside the stack may trigger parasitic chemical reactions that accelerate carbon deposition on the active surface. Moreover, keeping the OCR at the corresponding rate can have indirect positive impacts on the lifespan of SOFC system. For example, a higher methane reforming rate can be achieved at lower temperatures, hence increasing the efficiency of the entire SOFC system, see Ref. [17]. Moreover, lower stack temperatures reduce the rate of nickel agglomeration on the Ni-doped YSZ anodes, hence improving the overall life span of the stack [14].

OCR is defined as the relative ratio of oxygen and carbon atoms inside a fuel stream; therefore, in order to calculate OCR, gas composition of the fuel stream is needed. While it is entirely feasible to measure gas composition by employing appropriate chemical measurements in a laboratory environment, such a solution may become too costly when commissioning SOFC systems on an industrial scale. In order to overcome cost associated problems, software estimators are often exploited. Immeasurable variables are estimated employing easily available process measurements and a physical model.

In this paper, we aim to present and analyse three different approaches for online gas composition estimation. The paper extends the work presented in Ref. [18]. In order to estimate gas composition at any location in the system, 1st-principles and a lumped modelling approach with standard assumptions are considered [19,20]. In such a case, when estimating gas composition of the fuel stream in an SOFC system with AOGR configuration, the computation complexity of the estimators needs to be properly addressed. As a result of modelling assumptions and AOGR, a system of three algebraic relations must be solved. Such an approach has several disadvantages: (i.) the reliability of the estimates can be jeopardised by numerical instabilities, and (ii.) the implementation on the industrial programmable logic controller (PLC) is more demanding.

To overcome numerical issues related with PLC implementation, a simplified estimator for gas composition estimation of the SOFC system in AOGR configuration is proposed. Further, in contrast with 1st-principle estimators, a data-driven approach is also discussed. It is shown that even with a surprisingly simple model structure, estimates of remarkable precision can be achieved. Finally, by combining a data-driven approach with 1st-principles, a hybrid approach is proposed. The main contribution of the presented work are simple, easy to implement and reliable gas composition estimators that can be easily applied on a standard industrial PLC. Moreover, these results show that the complex electrochemical phenomena related to SOFC systems can be described by relatively simple models, suitable for online applications.

The paper is organised as follows. In Section 2, gas composition estimators are discussed. Next, Section 3 shows experimental

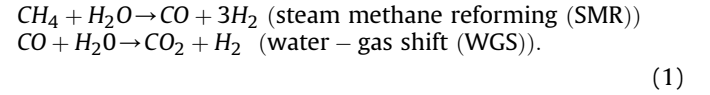
validation of the discussed approaches and comments on results. And, finally, the last section contains concluding remarks.

2. Three-point gas composition estimation

In the present work, we focus on an SOFC system in AOGR configuration. In such a case, a portion of depleted anode gas is recycled and mixed with fresh fuel. The gas mixture is then led through a pre-reformer, where partial reforming of methane takes place before entering the anode. Hence, gas composition differs at three separate locations: (i) pre-reformer inlet, (ii) pre-reformer outlet, and (iii) anode outlet. Fig. 1 shows a causality graph of the corresponding system variables.

2.1. General consideration

Two main reactions describe the steam-methane reforming process occurring in the system, i.e. the pre-reformer and stack itself:



In addition to these, hydrogen oxidation takes place in the stack:



Considering these reactions, gas composition at corresponding outlets, pre-reformer (left) and anode (right) outlets, can be estimated as

$$\begin{aligned} \dot{n}_{\text{H}_2\text{O}}^{\text{ro}} &= \dot{n}_{\text{H}_2\text{O}}^{\text{ri}} - x_r - y_r & \dot{n}_{\text{H}_2\text{O}}^{\text{ao}} &= \dot{n}_{\text{H}_2\text{O}}^{\text{ai}} - x_a - y_a + z \\ \dot{n}_{\text{H}_2}^{\text{ro}} &= \dot{n}_{\text{H}_2}^{\text{ri}} + 3x_r + y_r & \dot{n}_{\text{H}_2}^{\text{ao}} &= \dot{n}_{\text{H}_2}^{\text{ai}} + 3x_a + y_a - z \\ \dot{n}_{\text{CH}_4}^{\text{ro}} &= \dot{n}_{\text{CH}_4}^{\text{ri}} - x_r & \dot{n}_{\text{CH}_4}^{\text{ao}} &= \dot{n}_{\text{CH}_4}^{\text{ai}} - x_a \\ \dot{n}_{\text{CO}}^{\text{ro}} &= \dot{n}_{\text{CO}}^{\text{ri}} + x_r - y_r & \dot{n}_{\text{CO}}^{\text{ao}} &= \dot{n}_{\text{CO}}^{\text{ai}} + x_a - y_a \\ \dot{n}_{\text{CO}_2}^{\text{ro}} &= \dot{n}_{\text{CO}_2}^{\text{ri}} + y_r & \dot{n}_{\text{CO}_2}^{\text{ao}} &= \dot{n}_{\text{CO}_2}^{\text{ai}} + y_a, \end{aligned} \quad (3)$$

where $\dot{n}_i^l$ denotes the molar flow of i^{th} gas component, $i \in \{\text{H}_2\text{O}, \text{H}_2, \text{CH}_4, \text{CO}, \text{CO}_2\}$ at the location l , $l \in \{\text{ri}, \text{ro}, \text{ai}, \text{ao}\}$, i.e. pre-reformer inlet/outlet and anode inlet/outlet. The unknowns x_r (x_a), y_r (y_a), and z are defined as the rates of the corresponding reaction, SMR, WGS, and hydrogen oxidation reactions, see (1) and (2), respectively [21]. The unknown z is directly associated with the hydrogen oxidation reaction (2) and can be directly estimated by employing Faraday's law [22]:

$$z = \frac{n_{\text{cell}} I}{n_e F}, \quad (4)$$

where n_{cell} denotes number of cells in the stack, I is current demand, n_e number of free electrons, and F is Faraday's constant.

Thermodynamic equilibrium is an assumption often employed in SOFC system modeling [19,22–26]. Moreover, in Ref. [5], the

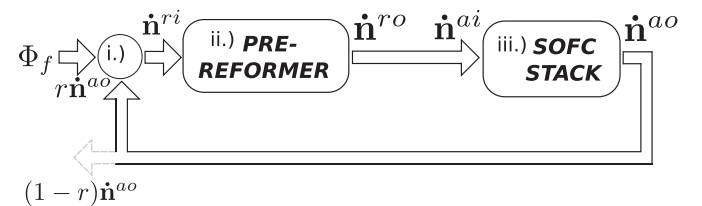


Fig. 1. Causality graph of an SOFC stack in AOGR configuration.

authors address the issue for particular system employed in our study and conclude that equilibrium was reached. Therefore, one can evaluate the unknowns x_r (x_a) and y_r (y_a) by employing SMR and WGS reaction equilibrium constants, i.e. K_{SMR} and K_{WGS} :

$$K_{SMR} = \frac{p_{H_2}^3 \cdot p_{CO}}{p_{CH_4} \cdot p_{H_2O}}, K_{WGS} = \frac{p_{CO_2} \cdot p_{H_2}}{p_{CO} \cdot p_{H_2O}}, \quad (5)$$

where $p_i = \dot{n}_i / (\dot{n}_{tot} + 2x)$ denotes partial pressure of the i^{th} gas component at the corresponding reactor outlet; and, $\dot{n}_{tot}$ is the total molar flow of the gases entering the reactor. Note that partial pressure of the i^{th} species is considered proportional to its molar flow [27]. In addition, the equilibrium constants (5) are directly associated with the change in Gibbs free energy, and can be expressed as follows [28]:

$$\log K_i = A_i T^4 + B_i T^3 + C_i T^2 + D_i T + E_i, \quad (6)$$

where K_i is a corresponding constant in (5), $i \in \{SMR, WGS\}$, and T is the temperature at the outlet of the reactor. The parameters A_i , B_i , C_i , D_i , and E_i are documented in Table 1 [28].

To summarise, putting the expressions from (3) into (5) while considering (6), one obtains a system of two non-linear equations with two unknowns for each individual reactor, i.e. the pre-reformer or stack:

- i.) x_r and y_r for the pre-reformer outlet and
- ii.) x_a and y_a at the anode outlet.

Solving the corresponding system to the corresponding unknowns, one can evaluate gas composition at the outlet of the reactor.

For easier discussion in the following sections, let us rewrite the relations (3) as follows:

$$\begin{aligned} \dot{\mathbf{n}}^l &= \begin{bmatrix} \dot{n}_{H_2O} \\ \dot{n}_{H_2} \\ \dot{n}_{CH_4} \\ \dot{n}_{CO} \\ \dot{n}_{CO_2} \end{bmatrix}, \quad \dot{\mathbf{n}}^{ao} = \dot{\mathbf{n}}^{ai} + \underbrace{\begin{bmatrix} -1 & -1 & 1 \\ 3 & 1 & -1 \\ -1 & 0 & 0 \\ 1 & -1 & 0 \\ 0 & 1 & 0 \end{bmatrix}}_{\mathbf{M}_a} \begin{bmatrix} x_a \\ y_a \\ z \end{bmatrix}, \\ \dot{\mathbf{n}}^{ro} &= \dot{\mathbf{n}}^{ri} + \underbrace{\begin{bmatrix} -1 & -1 \\ 3 & 1 \\ -1 & 0 \\ 1 & -1 \\ 0 & 1 \end{bmatrix}}_{\mathbf{M}_r} \begin{bmatrix} x_r \\ y_r \end{bmatrix}. \end{aligned} \quad (7)$$

2.2. Classical approach

The aim of this section is to briefly describe and highlight the problems related to the computational complexity of gas composition estimation. The targeted estimates are gas composition estimates at three specific locations: pre-reformer inlet $\dot{\mathbf{n}}^{ri}$, pre-reformer outlet $\dot{\mathbf{n}}^{ro}$ /anode inlet $\dot{\mathbf{n}}^{ai}$, and anode outlet $\dot{\mathbf{n}}^{ao}$, as

shown in Fig. 1.

With respect to Fig. 1, the following processes completely define gas composition at any location in the system:

- i.) mixing of portion r of the anode outlet $\dot{\mathbf{n}}^{ao}$ with fresh methane Φ_f supplied by the methane blower,

$$\dot{\mathbf{n}}^{ri} = r \cdot \dot{\mathbf{n}}^{ao} + [0 \ 0 \ 1 \ 0 \ 0]^T \Phi_f, \quad (8a)$$

- ii.) reforming process in the pre-reformer,

$$\dot{\mathbf{n}}^{ro} = \dot{\mathbf{n}}^{ri} + \mathbf{M}_r [x_r \ y_r]^T, \quad (8b)$$

- iii.) reforming process and hydrogen oxidation in the stack,

$$\dot{\mathbf{n}}^{ao} = \dot{\mathbf{n}}^{ai} + \mathbf{M}_a [x_a \ y_a \ z]^T. \quad (8c)$$

Accordingly, the gas composition estimation follows a successive, three step solution. However, although simple at first glance, solving these three processes can be problematic. Firstly, in order to calculate the mixture of the gas composition at pre-reformer inlet $\dot{\mathbf{n}}^{ri}$, gas composition at the anode outlet $\dot{\mathbf{n}}^{ao}$ must be known. Secondly, in order to estimate anode outlet gas composition $\dot{\mathbf{n}}^{ao}$, anode inlet composition $\dot{\mathbf{n}}^{ai}$ must be known. Finally, in order to estimate pre-reformer outlet/anode inlet $\dot{\mathbf{n}}^{ai}$, pre-reformer inlet $\dot{\mathbf{n}}^{ri}$ must be known. Hence, mathematically speaking, the AOGR configuration inherently incorporates an algebraic loop. Moreover, as explained in the previous section, the calculation of the equilibria of the reforming process in the pre-reformer and stack requires a numerical solution. For this purpose, various optimisation techniques can be applied. In this work, the problem was solved applying Newton's method [29].

The entire procedure consists of the following steps:

- i.) initiate arbitrary gas composition vector at the pre-reformer inlet $\dot{\mathbf{n}}^{ri}$;
- ii.) solve the pre-reformer equilibrium employing Newton's method and calculate $\dot{\mathbf{n}}^{ro}$ employing (8b);
- iii.) solve the stack equilibrium employing Newton's method and obtain $\dot{\mathbf{n}}^{ao}$ by employing (8c);
- iv.) calculate the gas composition at the pre-reformer inlet employing (8a);
- v.) iterate steps (ii-iv) until convergence.

The results of this procedure are hereafter denoted by $\mathcal{M}_f$.

Problems related to numerical optimisation.

The main disadvantage of implementing the optimisation algorithm on a standard PLC is related to convergence. For instance, solving the equilibrium in the pre-reformer and stack requires an iterative method, meaning that an "initial guess" of the approximate solution is required and corrected in the next step, until termination criteria are satisfied. In the case that the initial guess is "far away" from the real solution, the iterative sequence may take a longer time than allocated by the PLC. Further, the non-linear objective function may be non-convex. In such a case, the optimisation algorithm may converge to a local minimum, where the termination criteria are not fully accomplished. Improperly taking care of such a situation may cause the optimisation algorithm to get stuck without a solution. From a practical point of view, this means that the safety of the system's operation might come into question.

Table 1
Coefficients for temperature dependent estimation of equilibrium constants in (6) [28].

Const.	Reforming	Shifting
A_i	-2.6312×10^{-11}	5.47×10^{-12}
B_i	1.2406×10^{-7}	-2.574×10^{-8}
C_i	-2.2523×10^{-4}	4.6374×10^{-5}
D_i	1.95027×10^{-1}	-3.91500×10^{-2}
E_i	-66.139488	13.209723

2.3. Model simplifications and adaption for easy implementation

In order to overcome numerical issues related to optimisation for easy implementation on a PLC, a simplified model of the reforming processes in AOGR configuration is proposed hereafter. Several simplifications are employed:

- i.) the pre-reformer model is simplified;
- ii.) an analytic solution of the recycling loop is derived;
- iii.) the reforming model inside the stack is simplified.

2.3.1. Simplification of the pre-reformer model

The main idea is to solve the pre-reformer equilibrium once by employing sophisticated optimisation methods off-line on a standard PC, and then to use the solution in subsequent steps. For this purpose, a simple pre-reformer model, as discussed in Refs. [20,30], was built.

The pre-reformer model $\mathcal{M}_{pr}$ relates pre-reformer input variables $\mathbf{u}_{pr}$ with corresponding outputs $\mathbf{y}_{pr}$,

$$\mathbf{u}_{pr} \mapsto \mathbf{y}_{pr},$$

where the input vector $\mathbf{u}_{pr} = [\dot{\mathbf{n}}^{ri} \ T_{ri}]^T$ consists of $\dot{\mathbf{n}}^{ri}$ from (8b) and the corresponding inlet temperature of the gases T_{ri} . Similarly, the output vector $\mathbf{y}_{pr} = [\dot{\mathbf{n}}^{ro} \ T_{ro}]^T$ consists of $\dot{\mathbf{n}}^{ro}$ from (8b) and the corresponding outlet temperature of the gases T_{ro} .

The model $\mathcal{M}_{pr}$ was then used to simulate outlet gas composition. During the simulation, inlet gas temperature T_{ri} was kept constant. During the experiment, described later in the paper, the reformer inlet was controlled at all times at a certain set-point, thus justifying the choice.

The acquired input/output data was employed to identify a simplified black-box model of the pre-reformer. Since the purpose of this model is to find a simple surrogate model for the reforming process, the outlet temperature is of no interest. Assuming a linear relationship among input/output variables, a simplified pre-reformer model $\hat{\mathcal{M}}_{pr}$ reads

$$\dot{\mathbf{n}}^{ro} = \mathbf{R}\dot{\mathbf{n}}^{ri} + \mathbf{C}, \quad (9)$$

where $\mathbf{R}$ and $\mathbf{C}$ are constant matrix and column vector, respectively. Model parameters were identified from simulated input/output data employing least-squares.

Fig. 2 compares the results of the two models. The example in Fig. 2 shows only H_2O molar flow at the pre-reformer outlet

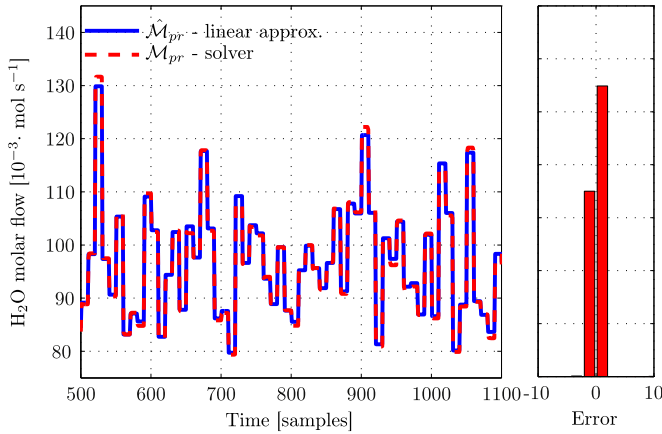


Fig. 2. Comparison of the two models—non-linear optimisation model and corresponding simplified model. The figure shows H_2O molar flow at the pre-reformer outlet only. The rest are omitted for brevity.

employing non-linear optimisation model $\mathcal{M}_{pr}$, while the blue solid plot shows the results from simplified model $\hat{\mathcal{M}}_{pr}$ employing (9). The histogram on the right shows the difference between the optimisation model and the corresponding simplified model. Clearly, the error is low, thus indicating high precision.

The results also show that the reforming model can be easily described with a simple linear structure rather than as a non-linear optimisation problem. However, the proposed simplification is only valid when inlet gas temperature is constant. When inlet temperature is changing, the model should be adapted. Figures of the remaining gas component in the fuel stream are omitted for brevity.

Experimental validation of the proposed simplified model can be found in the Appendix.

2.3.2. Analytic solution of the AOGR

The recycling loop described by (8) can be solved analytically. Considering that $\dot{\mathbf{n}}^{ro} = \dot{\mathbf{n}}^{ai}$, and the results from previous section (9), one can rewrite (8) as follows:

$$\dot{\mathbf{n}}^{ao} = \dot{\mathbf{n}}^{ai} + \mathbf{M}_a[x_a \ y_a \ z]^T \quad (10a)$$

$$\dot{\mathbf{n}}^{ai} = \mathbf{R}\dot{\mathbf{n}}^{ri} + \mathbf{C} \quad (10b)$$

$$\dot{\mathbf{n}}^{ri} = r \cdot \dot{\mathbf{n}}^{ao} + [0 \ 0 \ 1 \ 0 \ 0]^T \Phi_f. \quad (10c)$$

Putting (10c) into (10b), and then the result into (10a), and after some manipulation, one can express $\dot{\mathbf{n}}^{ao}$ as

$$\dot{\mathbf{n}}^{ao} = \underbrace{(\mathbf{I} - r\mathbf{R})^{-1}(\mathbf{R}[0 \ 0 \ 1 \ 0 \ 0]^T \Phi_f + \mathbf{C})}_{\mathbf{A}} + \underbrace{(\mathbf{I} - r\mathbf{R})^{-1}\mathbf{M}_a[x_a \ y_a \ z]^T}_{\mathbf{B}}. \quad (11)$$

In (11), $\mathbf{A}$ and $\mathbf{B}$ are constant matrices parametrised by known process variables. The only unknowns are x_a and y_a from (8c), and z is known from (4).

2.3.3. Analytic solution of the WGS in the stack

Relation (11) unambiguously defines anode outlet gas composition $\dot{\mathbf{n}}^{ao}$ from easily accessible process variables. The only unknowns x_a and y_a can be found by employing equilibria constants (5) and optimisation. However, since the aim is to overcome the optimisation, we apply a common assumption of total reforming of methane feed in stack, hence x_a is known and equals to the amount of methane at the anode inlet. Such a choice is easily justified since the methane conversion rate at low pressures and high temperatures is nearly 100% [17].

Since the entire methane feed is reformed, one of the equilibrium equations, e.g. SMR from (5), can be dropped. In such a case, by putting the relations from (11) into the WGS equilibrium equation from (5), the problem reduces to solving a quadratic equation for the unknown y_a in the form of

$$p_2 y_a^2 + p_1 y_a + p_0 = 0. \quad (12)$$

The unknown x_a can be easily found from (11) since $\dot{\mathbf{n}}_{\text{CH}_4}^{ao} = 0$ holds due to total methane reforming, and y_a is known from (12)¹. Finally, the anode outlet gas composition $\dot{\mathbf{n}}^{ao}$ can be easily calculated employing (11). Likewise, gas composition at the remaining locations can be easily calculated considering (10b) and (10c).

¹ Note that (12) has two solutions, one of which is infeasible and easily recognised.

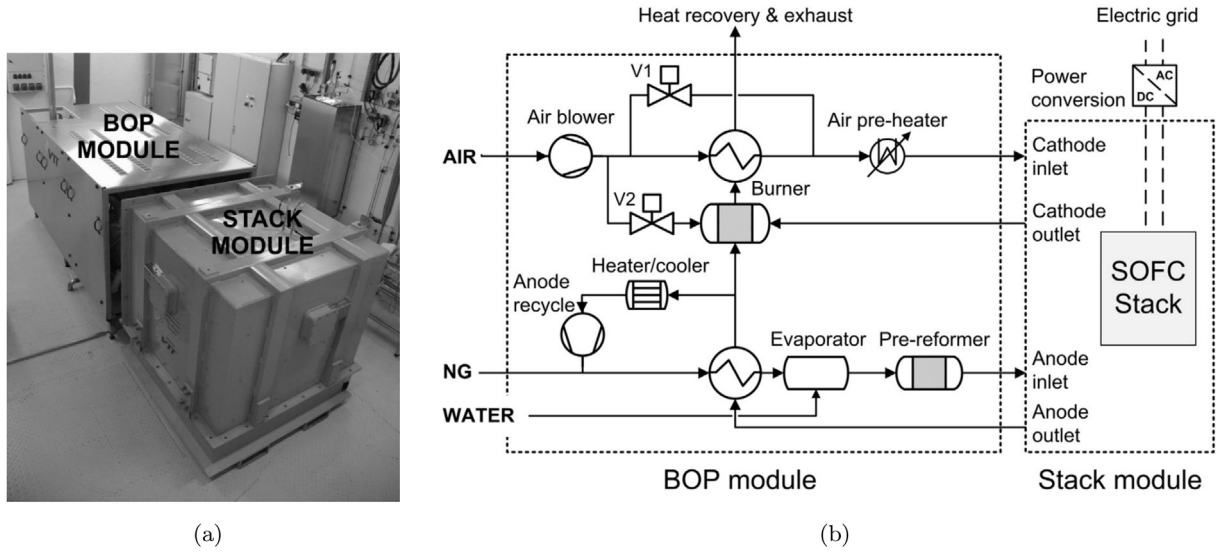


Fig. 3. The SOFC power generating unit installed at VTT (a) and schematic representation (b) [33,34].

To summarise, the simplified model $\mathcal{M}_s$ is fully defined by (4), (10), (11) and (12).

2.4. Data-driven approach

In this section, we explore the potentials of data-driven approaches for online gas composition estimation. Data-driven models are generally more accurate than 1st-principle models as they are built directly from measurements. These measurements inherently incorporate the phenomena neglected by physical models, see Ref. [31]. Contrary to the estimators described in the previous section, the model parameters are identified directly from experimental data.

The inputs of the data-driven model are selected based on the physics as discussed in previous sections and collected in an input vector $\mathbf{u}$,

$$\mathbf{u} = [\Phi_f \Phi_r I]^T, \quad (13)$$

where Φ_f denotes molar flow of the methane feed, Φ_r is the recycle flow which is directly associated with the recycling ratio r from (8), and I is stack current. The targeted outputs are vectors describing gas composition at three separate locations as in (8), stacked in an output matrix $\mathbf{y}$:

$$\mathbf{y} = [\dot{\mathbf{n}}^{ri}]^T [\dot{\mathbf{n}}^{ai}]^T [\dot{\mathbf{n}}^{ao}]^T]^T. \quad (14)$$

As shown in the previous section, the problem of solving the equilibria can be, under certain assumptions, overcome by a simple quadratic equation. Here we go one step further and approximate the solution of the equilibrium. To this aim, a non-linear equation can be linearised and one can write

$$\mathbf{y} = \mathbf{f}(\mathbf{u}) \approx \bar{\mathbf{y}} + \frac{\partial \mathbf{f}}{\partial \mathbf{u}} \bigg|_{\mathbf{u}=\bar{\mathbf{u}}} \Delta \mathbf{u},$$

where vector $\Delta \mathbf{u}$ denotes centred input data, $\bar{\mathbf{u}}$ and $\bar{\mathbf{y}}$ are vectors of mean values of input and output variables respectively, and $\Psi \in \mathbb{R}$ is a matrix of the unknown parameters to be identified. Denoting

$$\Psi = \frac{\partial \mathbf{f}}{\partial \mathbf{u}} \bigg|_{\mathbf{u}=\bar{\mathbf{u}}},$$

the data-driven model for online gas composition estimation $\mathcal{M}_{dd}$ reads

$$\mathbf{y} = \bar{\mathbf{y}} + \Psi(\mathbf{u} - \bar{\mathbf{u}}). \quad (15)$$

The model parameters Ψ are identified via linear least-square method directly from experimental data [32].

3. Experimental validation

The experiments were carried out on an SOFC system. The SOFC stack had 80-cells with a total of 550 cm² of active area and a balance of plant (BoP) module as shown in Fig. 3(a) [33,34]. Owing to the well-instrumented system, an extensive set of different types of data was collected during the experiment. The experimental data contain pressure and temperature measurements, flows of the gases, and electrical variables of the system. Moreover, chemical measurements were conducted on dried samples of gas mixtures from relevant locations, like the pre-reformer inlet, anode inlet, and anode outlet. Therefore, this data is an ideal candidate for evaluation of the discussed approaches. A thorough description of the measurement procedures and data treatment, and the system itself, are available in Refs. [12,35].

3.1. The results

Fig. 4 shows the estimated gas composition at the pre-reformer outlet/anode inlet, i.e. $\dot{\mathbf{n}}^{ro}$ or $\dot{\mathbf{n}}^{ai}$. Individual subfigures show components in the gas composition vector from (7): (a) H₂O, (b) CO, (c) CH₄, (d) H₂, and (e) CO₂. There are four plots in each figure (a–e) belonging to full optimisation estimator $\mathcal{M}_f$, its simplified version $\mathcal{M}_s$, and data-driven one $\mathcal{M}_{dd}$. Additionally, there is an experimental measurement of the corresponding species denoted by “Measurement”.

By closely observing Fig. 4(a) and (b), one notices a similar precision for all of the estimates. There is no significant difference among the estimates of $\mathcal{M}_f$, $\mathcal{M}_s$, and $\mathcal{M}_{dd}$. However, when observing the remaining three estimates (c–e), there is significant off-set

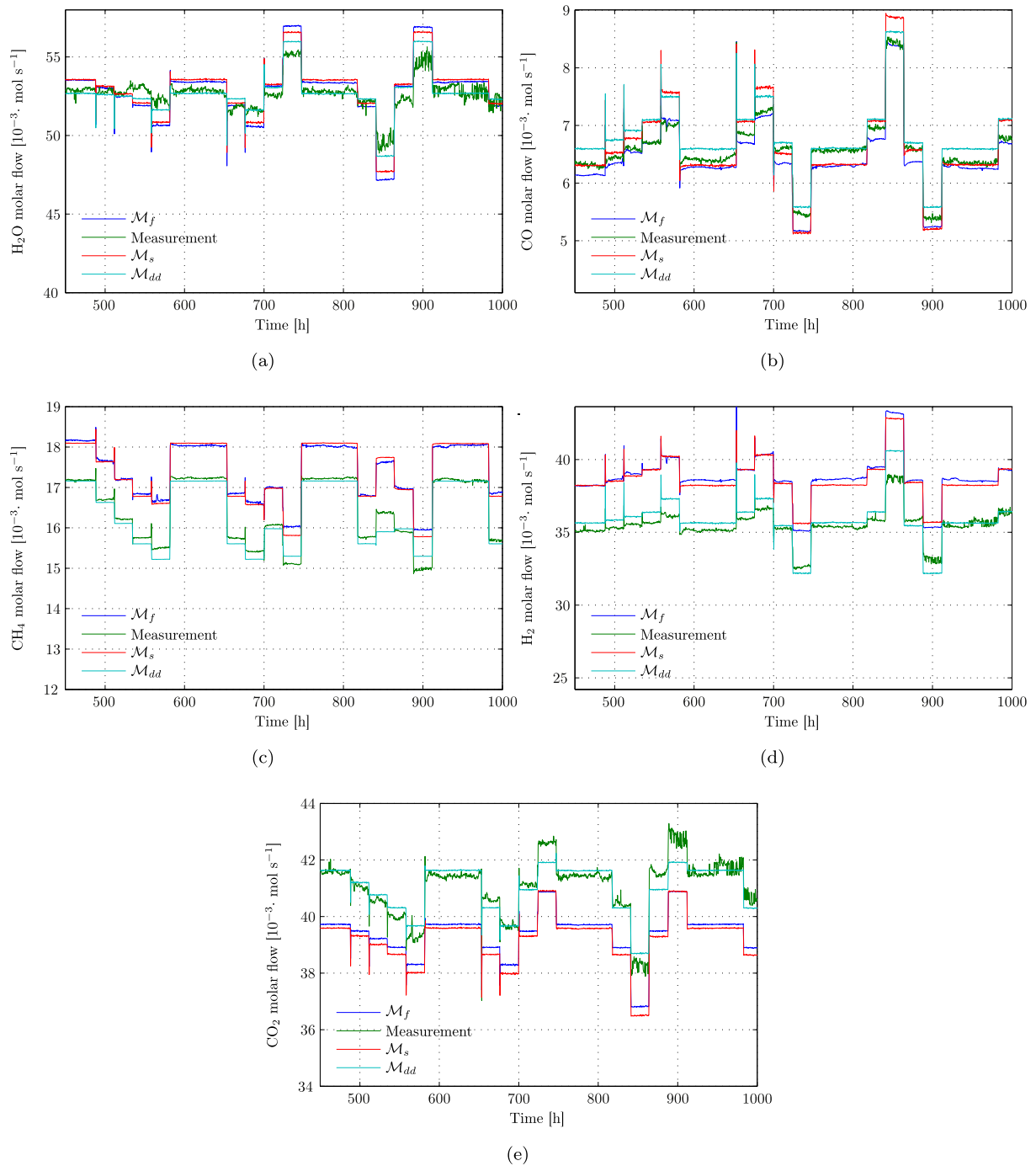


Fig. 4. Estimated pre-reformer outlet gas composition $\dot{n}^o$ for the three described approaches – full optimisation estimator $\mathcal{M}_f$, simplified estimator $\mathcal{M}_s$, and data-driven estimator $\mathcal{M}_{dd}$ – compared to corresponding experimental measurements.

between the stoichiometric estimates $\mathcal{M}_f$ and $\mathcal{M}_s$ and measurements, while the data-driven estimator $\mathcal{M}_{dd}$ accurately estimates all of the variables. As shown in Ref. [36], the stack itself was subjected to external and internal leakages. It is reasonable to expect leakages in an SOFC stack, both between the air and fuel channels as well as out of both of these. In each of these cases, the leakages affect the fuel gas composition in the stack and should thus somehow be accounted for by the OCR estimates. Typically, leakages will lead to the amount of oxygen increasing in the fuel compartment, which means that analytical estimates for OCR will underestimate this ratio. Given that the leakages maintain constant throughout the operation life of the stack, as shown, e.g., in Ref. [36], it is then

possible to incorporate their effect in a data-based estimator for gas composition.

In addition to the leakage effects, the data-based gas composition estimate will also account for errors in, for example, the SMR and WGS reactions' equilibrium constants, the measurement errors in the system temperature, pressure and flow measurements, as well as the kinetic limitations of the electrochemical and chemical conversion processes within the system. The physical models, although more general, are affected by the errors in all of these factors. The data-driven model $\mathcal{M}_{dd}$ also takes into account these non-modelled effects, which is neglected by $\mathcal{M}_f$ and $\mathcal{M}_s$, hence giving estimates of greater accuracy.

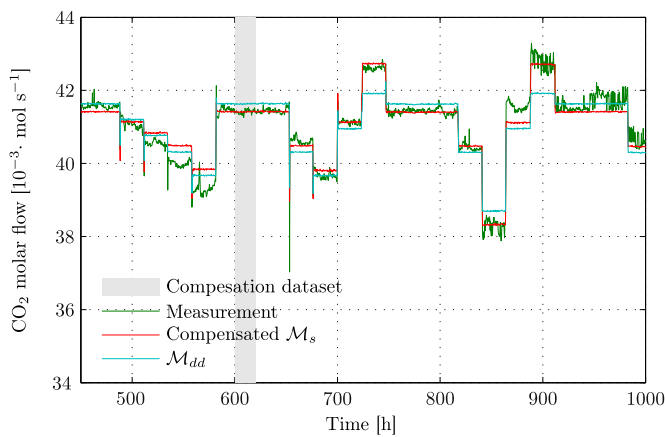


Fig. 5. Compensated estimate of CO₂ molar flow at the anode inlet.

The example shows gas composition estimation only at the pre-reformer outlet/anode inlet, i.e. $\dot{n}^0$. Figures of the estimates at remaining locations are similar and are therefore omitted for brevity. Reformer outlet gas composition was selected because this is the critical point for estimation, i.e. directly at the anode inlet.

3.2. Off-set compensation

The main disadvantage of data-driven models is that they are specifically designed for a system on which the experiment was performed. For example, the considered stack is affected by leakage that is specific for this unit only. Moreover, in order to build such an estimator, a rich experimental data set is required, which may prove to be too expensive for application on an industrial scale. Alternatively, the more that the physical models are generalized, the more they can be adapted for any system. With this approach, however, there is no guarantee that the estimates will be accurate due to possible effects of non-modelled phenomena. A reasonable trade-off is to have a short tuning data set. In such a case, the off-sets of the stoichiometric models can be compensated.

The result of combining the stoichiometric and data-driven approaches is illustrated in Fig. 5. Note that the off-set in Fig. 4(e) was compensated by identifying the correction term from experimental data, marked by the shaded area. The compensation data only included 60 data-points. Surprisingly, the estimates from the compensated simplified model, denoted by “Compensated $\mathcal{M}_s$ ” in Fig. 5 shows an even greater accuracy than the data-driven estimator $\mathcal{M}_{dd}$. Higher accuracy is probably due to the non-linear structure of $\mathcal{M}_s$, which is neglected by $\mathcal{M}_{dd}$.

4. Conclusion

The paper discusses three approaches for online gas composition estimation from easily measurable process variables, such as flows, electric measurements, and temperatures. The three presented approaches are based on (i) classical approach $\mathcal{M}_f$, (ii) model simplification and adaption for easy implementation $\mathcal{M}_s$, and (iii) data-driven approach $\mathcal{M}_{dd}$. It is shown that the most accurate and straightforward estimates are obtained when employing $\mathcal{M}_{dd}$. However, in order to build such estimator, a rich data set is required. Alternatively, it is shown that the stoichiometric model can be simplified so that its implementation on a PLC is as easy as a linear data-driven one. However, such estimators neglect some phenomena and the resulting estimates are less accurate.

In the last section of the paper, a hybrid approach for gas composition estimation is proposed. By combining a short

experimental data set from which offsets are identified together with physical knowledge, estimates of high precision are obtained. Surprisingly, the estimator $\mathcal{M}_s$ shows an even higher accuracy than data-driven estimator $\mathcal{M}_{dd}$. The only requirement to derive such an estimator is a short tuning data set. In doing so, a compromise among requirements of data-driven and 1st-principle approaches is achieved in terms of data requirements, accuracy of the estimates, and implementation simplicity.

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Appendix

Experimental validation of the simplified pre-reformer model (9) was achieved using the following parameters:

$$\mathbf{R} = \begin{bmatrix} 0.8186 & 0.2486 & -0.0619 & -0.4899 & 0.0758 \\ 0.2851 & 0.6326 & 0.1027 & 0.4923 & -0.0512 \\ -0.0479 & 0.0588 & 0.9707 & -0.0128 & -0.0105 \\ -0.0795 & 0.1319 & -0.0196 & 0.5231 & 0.0993 \\ 0.1344 & -0.1896 & 0.0338 & 0.4707 & 0.9129 \end{bmatrix},$$

$$\mathbf{C} = 10^{-4} \times \begin{bmatrix} -0.5224 \\ 0.4570 \\ -0.0548 \\ -0.5015 \\ 0.4239 \end{bmatrix}.$$

These parameters are valid for a specific temperature of the inlet gases only. During our experiment, the temperature of the inlet gases is kept constant throughout the operation, hence the choice. In the opposite case, when the pre-reformer gas inlet temperature is fluctuating, the simplified model needs to be properly adapted.

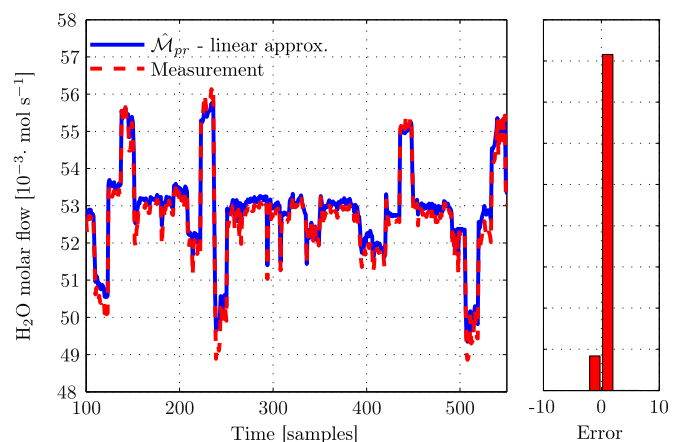


Fig. A.1. Experimental validation of the simplified pre-reformer model (9). The figure shows prediction of the H₂O molar flow at the pre-reformer outlet and its corresponding measurement. The error histogram on the right is narrow and centred around zero, signifying high precision estimates. The remaining gas components are omitted for brevity.

Figure A.1 shows an excerpt from the experimental validation data set. The considered experiment is the same as shown in Section 3. More specifically, the figure shows H_2O molar flow at the pre-reformer outlet. The plot on the right shows the prediction error histogram, which is narrow and centred around zero, thus indicating high precision. It is clearly evident that the simplified linear model is capable of simulating the outlet gas composition.

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Chapter 3

Soft Sensors for Internal Stack Temperatures

The durability of a SOFC stack is considerably influenced by the operating conditions imposed on the stack. Amongst others, the temperature distribution is one of the most influential. Generally, higher operating temperatures are a dominating factor responsible for the majority of degradation phenomena [61]. However, there exist certain operating conditions, where the degradation rate is reduced for a set of phenomena at elevated temperatures [89], and conversely increased for others [21]. Moreover, the temperature distribution over the stack directly influences the mechanical stress imposed on the fuel cells. Thus, having direct control over thermal conditions inside the stack can be of great benefit with respect to the total life-span of the system [77].

Commissioning of a new system with thermocouples that would facilitate direct measurement of temperature on the anode/cathode surface comes with two main drawbacks. First, installation of several thermocouples brings difficulties with the integrity of the system. Placing the sensors inside the anode/cathode chambers requires corresponding wiring, thus requires additional incursions in the seals, which could lead to the problems related to anode/cathode leaks. Moreover, materials of which thermocouples are made of, are unrelated with the operation of the SOFC and may provoke poisoning of the electrodes, thus hindering the total life-span of the system. Furthermore, installing a network of thermocouples on the industrial scale can become costly. Usually, the manufacturers of all kinds of assets aim at minimising the total cost of the production; thus, they tend to limit the physical sensors to the bare minimum. If and when possible, the physical sensors are replaced by software sensors, which employ mathematical models and information from already available measurements to estimate required process variables.

There are a number of models describing the phenomena occurring inside the stack [6], [90], [91]. Depending on the application, these models describe behavior of the system with respect to fluid dynamics, thermodynamics, mass transport, and electrochemistry. The models include a set of interconnected partial differential equation (PDE), usually solved employing commercially available multiphysics software. Even though the models are capable of performing numerous simulations, which are a valuable source of the information for the design of the fuel cell systems and its components, the cost of obtaining such detailed information becomes unreasonable for on-line applications. The sophisticated numerical methods that compute the solution of the PDEs may require significant computing power, i.e. expensive hardware, and may take too much time.

The work in this chapter develops a data-driven approach for estimation of stack internal temperatures and advances the approaches of the works described in [92], [93]. Similarly to the above work, measurements on a reference system are needed in order to derive the

soft sensor. First, the paper discusses the causality of the input/output variables on the system level. The most influential variables that impact the thermal condition inside the stack are sought based on analysis of the system and physical knowledge of the underlying processes. Then, the structure of the model is sought directly from carefully designed experimental data. Two dynamic sub-models of the system, one describing the thermal conditions inside the stack and one which maps input of the system to the output gas temperatures, are merged into a single model, which is able to reconstruct the minimal and maximal stack temperatures. Moreover, the developed software sensor is able to reconstruct these temperatures regardless of the undergoing degradation mechanisms in the fuel cells, which cause gradual and slow drift in the temperatures.

Like in the case of gas composition soft sensor, the developed temperature soft sensor can be easily adapted for a different stack technology provided that the experimental data is available. Alternatively, the temperature soft sensor could be designed in a different manner. One of the possible solutions is to employ a more or less complex 1st principle model in a state estimation algorithm, such as UKF, or particle filter. This way, not only the stack temperature profiles could be estimated, but also other process variables, such as current densities over the fuel cells, could be reconstructed. However, such an approach comes at a cost of higher computational loads, and more difficult parametrisation of the employed model – more model parameters need to be taken apriori.

Choosing to employ soft sensors instead physical sensors always comes as a compromise between cost/feasibility and accuracy. For instance, if during the design stage of the soft sensor some of the crucial phenomena are neglected, the estimates become inaccurate should the mentioned phenomena occur in the system after the commissioning. Therefore, clear specification of the soft sensor functionality and requirements are of vital importance for the designer. In the case of the data-driven soft sensor design, this information should be carefully incorporated into the design of the experiment. On the other hand, modeling assumptions and simplifications need to be properly addressed and justified when employing a more physical model for soft sensor design. For instance, neglecting the degradation phenomena in the design stage can cause a drifting error in the estimates.

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Online estimation of internal stack temperatures in solid oxide fuel cell power generating units



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HIGHLIGHTS

- A systematic data-driven approach for SOFC temperature estimator design is proposed.
- The estimator relates available sensor readings with stack inner temperatures.
- Remarkably simple model structure demonstrates high precision estimates.
- Precision of the estimates remains unaffected under degradation phenomena.
- Implementation is demonstrated on 10 kW SOFC system.

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ABSTRACT

Thermal stress is one of the main factors affecting the degradation rate of solid oxide fuel cell (SOFC) stacks. In order to mitigate the possibility of fatal thermal stress, stack temperatures and the corresponding thermal gradients need to be continuously controlled during operation. Due to the fact that in future commercial applications the use of temperature sensors embedded within the stack is impractical, the use of estimators appears to be a viable option.

In this paper we present an efficient and consistent approach to data-driven design of the estimator for maximum and minimum stack temperatures intended (i) to be of high precision, (ii) to be simple to implement on conventional platforms like programmable logic controllers, and (iii) to maintain reliability in spite of degradation processes. By careful application of subspace identification, supported by physical arguments, we derive a simple estimator structure capable of producing estimates with 3% error irrespective of the evolving stack degradation. The degradation drift is handled without any explicit modelling.

The approach is experimentally validated on a 10 kW SOFC system.

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1. Introduction

Solid oxide fuel cells (SOFCs) are becoming a promising and competitive technology for CO₂ reduction and the realisation of sustainable power sources. The first commercialisation successes in Japan in the area of combined heat and power, recently reported in Ref. [1], indicate encouraging prospects for stationary applications in the near future. However, the market deployment of the

technology is still unsatisfactory due to the problems associated with the durability, reliability and, finally, still too high cost of SOFC systems [2].

According to Atanasiu [3], one of the main objectives during the 2014–2020 period in the European Union is to increase the electrical efficiency and durability of fuel cells. In order to tackle these challenges, significant effort has been invested in better understanding the degradation mechanisms and strategies for their accommodation.

A range of degradation and failure modes affect the durability and reliability of SOFC and associated systems. The most notable examples refer to the ageing of electrode materials [4], thermal

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stresses causing electrode delamination and electrolyte cracking [5], phenomena originating from thermo-chemical processes such as electrode poisoning, carbon deposition, oxide layer growth, etc. [6]. Ageing and degradation also result in a reduction in the active area in the cells, thus decreasing the efficiency and the life span of SOFC power generating systems. If the stack is suddenly subjected to an excessive thermal stress, it may lead to cracks in cell components. Moreover, even thermal cycling in nominal operating conditions increases the overall stack degradation [7].

A number of new solutions have been proposed in the search of materials resistant to various degradation mechanisms, for example [8,9]. The most recent advances in electrode materials are presented in Ref. [10], where ageing and understanding the degradation mechanisms have been recognised as two of the main issues targeted by the scientific community. However, even the most advanced SOFCs are prone to degradation and failure if subjected to improper operating conditions. For instance, excessive temperature gradients through the stack result in unrestrained thermal stress on the stack components, thus increasing the risk of components cracking. Furthermore, allowing too high operating temperatures generally decreases the lifespan of the SOFC [11]. Also, by neglecting the conditions required for chemical reactions, a stack can become exposed to various poisoning hazards, such as carbon deposition [12]. Hence, overall stack degradation can be alleviated by restricting stack operation to the safe region [13].

In order to mitigate the possibility of fatal thermal stress, stack temperatures and the corresponding thermal gradients need to be controlled throughout operation. An approach where the thermal conditions inside an SOFC system are considered as the control objective was recently proposed in Refs. [14,15]. The authors present a control strategy for maintaining a constant stack gradient and the keeping exhaust temperature at a desired set-point. Such an approach builds on precise temperature measurements inside the stack. In spite of a rich set of available temperature sensors, the in-situ implementation of thermocouples inside the stack is rather costly for massive deployment and impractical due to wiring through the sealing materials. The aim of this paper, therefore, is to propose an estimator of the internal stack temperatures, which will replace physical sensors and hence eliminate difficulties associated with in-situ installations. It is mandatory for the estimator to (i) be precise, (ii) to require low computational effort, and (iii) to be easy to implement on conventional low-cost programmable logic controllers.

In order to evaluate temperatures inside the stack, various models have been developed, ranging from 1st-principle to data-driven models [16,17]. Some data-driven models use neural

networks [18] or linear regression [19]. Although 1st-principle models are very desirable in the design stage, they are rather impractical for online temperature estimation due to the significant numerical effort. Grey-box models, however, are easier to develop and implement, provided enough reference data are available [20]. The idea is to start from physical causal relationships and then use information from experimental data to refine the model structure search and the estimation of the parameters.

The work below revisits the problem discussed in Refs. [19,21–23] and suggests an alternative approach to that described in the mentioned references. The notable advances of the approach could be summarised as follows:

- i.) In accordance with the principle of parsimony, the simplest model structure is sought directly from experimental data by analysing singular values of stacked input and output recordings;
- ii.) It is shown that a simple linear model can achieve remarkably high precision estimates;
- iii.) Drifts caused by degradation are inherently incorporated in the model, hence explicit characterisation of the degradation processes is unnecessary.

The rest of the paper is organised as follows. First, the system on which the experiments for data acquisition were performed is described in Section 2. The structural and parametric identification of the process model is described in Sections 3 and 4. The derivation of the temperature estimator and experimental validation is presented in Section 5. Finally, last Section comprises the concluding remarks.

2. System description

The experiments were conducted on an SOFC power generating unit, comprised of an SOFC stack and a balance of plant (BoP) module, as shown in Fig. 1(a) [21,24]. The stack under consideration is comprised of 80 fuel cells with a total active area of 550 cm², sealed in an insulated housing. In such a manner, it can provide power for a maximum 12 kW electric load. The feasible operating range for the unit in question is defined by the following variables: an air flow rate in the range of 700–1600 l/min and a natural gas flow rate up to 35 l/min. The recycle flow rate can reach up to 200 l/min, while the maximum operating temperature is 850 °C at the exhausts and 800 °C at the inlets.

The BoP module consists of two blowers, a methane pre-reformer, an afterburner, a water evaporator, and heat

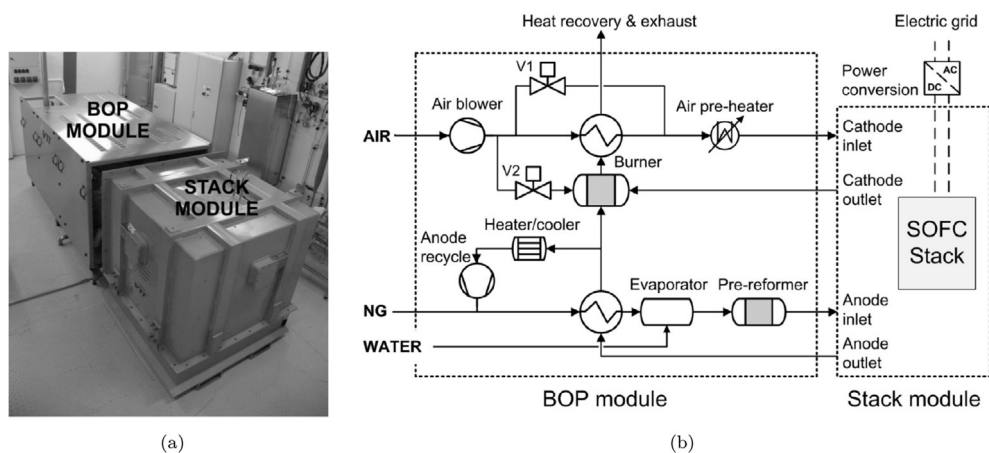


Fig. 1. The SOFC power generating unit installed at VTT (a), and a schematic representation (b).

exchangers. The air blower supplies ambient air to the cathode inlet, while the anode recycle blower drains a portion of the depleted anode exhaust and supplies it back to the fuel stream. The set-up is commonly known as anode off-gas recycling (AOGR). The AOGR configuration provides water required for steam reforming of the natural gas as well as heat supporting the reactions. The remaining anode exhaust is led through the afterburner, where all the remaining unused gases are burnt. The excess heat is then used to pre-heat the cathode inlet gases. The main role of the water evaporator, however, is to provide enough water to the reactor during the start-up of the system and is switched off during normal operation. Fig. 1(b) shows the schematic representation of the entire system.

The study below is performed on operational data gathered during an experiment that lasted approximately 1500 h. During this time, the system was operated in nominal operating conditions. More details on the experiments can be found in Refs. [19,21].

3. Selection of the model structure

Selection of the model structure is central to data-driven modelling. Out of a large set of candidate model structures, including linear and non-linear models, it is reasonable first to try the simplest possible candidate and then seek alternatives if the results achieved are unsatisfactory. Therefore, we focused on a class of linear state-space dynamic models to describe the physical relationships between system inputs and outputs. The modelling procedure consists of three steps:

1. Determining the causal model structure and establishing which independent inputs and process outputs appear in the mathematical model;
2. Structural identification of the model, i.e. selection of the model order based on singular values of data matrix compound of input and output measurements;
3. Estimation of model parameters from the data.

In the first step, it is important to take into account all the influential inputs and to check for the presence of process and control loops. The former imply correlated inputs, which can result in reduced model identifiability and bias.

The last two steps require sufficient system excitation in order to excite all the dominant system eigenmodes. Insufficient excitation of some of the systems eigenmodes has the consequence that particular aspects of the system dynamics remain unmodelled.

3.1. Causality analysis

The aim of a causality analysis is to find which process variables appear as independent inputs (causes) required to describe the outputs (consequences). Failure to overlook control and process loops can lead to biased and misleading estimates of the model parameters. Let us first carry out a brief analysis of causality.

Referring to Fig. 1(b), ambient air is pumped into the system by an air blower producing the flow rate Φ_a . Prior to entering the cathode, the air temperature $T_{cath,in}$ is controlled at a pre-defined value $T_{cath,in}^*$, which is used as a manipulated variable by the higher-level control system. Since the dynamics of the control loop is fast enough compared to the dominant system dynamics, we can assume $T_{cath,in} = T_{cath,in}^*$ and take it as an independent system input.

Similarly, the fuel is delivered to the system by means of a controlled flow rate Φ_f . The fuel is then mixed with a constant portion of steam-rich anode exhaust. The temperature of the gas mixture is controlled at a constant value prior to entering the pre-

reformer. Therefore, the reformer outlet temperature (which at the same time is the fuel temperature $T_{anode,in}$ at the anode inlet) is a result of the chemical processes in the pre-reformer. The chemical processes depend on the anode inlet gas composition, which, in turn, depends on the methane feed rate Φ_f and the demanded load current I . Therefore, $T_{anode,in}$ is a dependent variable input and therefore eliminated from model identification.

Finally, the demanded load current I is also considered as an independent input variable. It defines the hydrogen demand for power generation, and also influences heat production inside the stack due to ohmic losses in materials.

Another phenomenon that has an impact on the behaviour of an SOFC are parasitic processes taking place inside the stack, for instance stack leakages. In fact, in Ref. [25] it was shown that the system under consideration is affected by such leakage phenomena. Evidently, these processes have a direct impact on the output variables. However, assuming there is no substantial change in these processes during normal operation of the stack, their specific characterisation becomes unnecessary.

To sum up, the thermal processes within the stack depend on the demanded load current I , the fuel flow rate Φ_f , air flow rate Φ_a , and air temperature $T_{cath,in}$, stacked in an input vector $\mathbf{u}$:

$$\mathbf{u} = [\Phi_f \ \Phi_a \ T_{cath,in} \ I]^T. \quad (1)$$

The process inputs $\mathbf{u}$ result in process outputs $\mathbf{y}$, which can be divided into two parts:

- Inner stack temperatures of interest, i.e. the maximum and minimum temperatures, T_{min} and T_{max} , respectively; and
- Output temperatures on the anode and cathode sides $T_{anode,out}$ and $T_{cath,out}$.

The stack outputs are hereafter denoted by the output vectors $\mathbf{y}_1$ and $\mathbf{y}_2$:

$$\begin{aligned} \mathbf{y}_1 &= [T_{min} \ T_{max}]^T \\ \mathbf{y}_2 &= [T_{anode,out} \ T_{cath,out}]^T. \end{aligned} \quad (2)$$

The causal representation is shown in Fig. 2.

3.2. State-space representation of stack temperature models

Physical processes within the stack run on three different time scales: (i) fast electro-chemical processes, (ii) fluid transport, and (iii) slow thermal processes. Compared to the electro-chemistry and fluid dynamics, the temperatures exhibit the slowest dynamics. Hence, from the aspect of thermal processes, electro-chemistry and fluid dynamics are almost instantaneous.

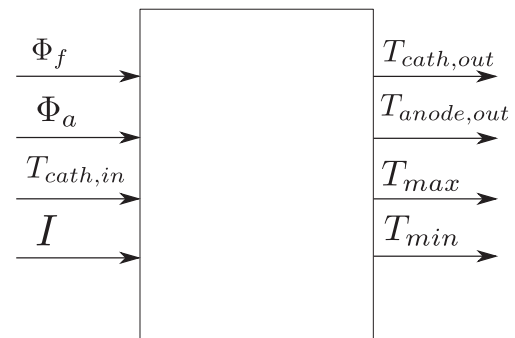


Fig. 2. The causal structure of the thermal processes in the stack.

An appropriate candidate for the temperature model is the linear time-invariant state space model:

$$\begin{aligned}\mathbf{x}(k+1) &= \mathbf{A}\mathbf{x}(k) + \mathbf{B}\mathbf{u}(k) \\ \mathbf{y}(k) &= \mathbf{C}\mathbf{x}(k) + \mathbf{D}\mathbf{u}(k) + \mathbf{v}(k)\end{aligned}\quad (3)$$

where $\mathbf{u} \in \mathbb{R}^p$ is the vector of inputs, $\mathbf{x} \in \mathbb{R}^n$ is the state vector and $\mathbf{y} \in \mathbb{R}^q$ is the vector of measured outputs, while matrices $\mathbf{A} \in \mathbb{R}^{n \times n}$, $\mathbf{B} \in \mathbb{R}^{n \times p}$, $\mathbf{C} \in \mathbb{R}^{q \times n}$, and $\mathbf{D} \in \mathbb{R}^{q \times p}$ are system matrix, input, output, and feed-through matrices, respectively. Noise vector $\mathbf{v}(k)$ is zero-mean white Gaussian noise mimicking random effects in sensors and unmodelled effects, and k indicates the sampling instance kT_s , where T_s is the sampling time.

Each output vector in (2) can be associated with a state space model

$$\mathcal{M}_i : \begin{cases} \mathbf{x}_i(k+1) = \mathbf{A}_i \mathbf{x}_i(k) + \mathbf{B}_i \mathbf{u}_i(k) \\ \mathbf{y}_i(k) = \mathbf{C}_i \mathbf{x}_i(k) + \mathbf{D}_i \mathbf{u}_i(k) + \mathbf{v}_i(k) \end{cases} \quad i \in \{1, 2\} \quad (4)$$

where the system matrices are of appropriate dimensions.

4. Subspace identification of the partial models $\mathcal{M}_1$ and $\mathcal{M}_2$

The subspace identification methods build on geometrical projections in the high dimensional row or column spaces. They rely on the fact that if input and output data are properly stacked into structured block Hankel matrices, it is possible to retrieve certain subspaces that are related to the system matrices associated with the state-space model. Detailed theory can be found in Refs. [26,27]. An essential inherent property of the method is its ability to adjust the structure of the dynamic model to the information contained in experimental data records. The essentials of 4SID methods are highlighted in the Appendix.

4.1. Signal pre-processing

Prior to the model identification, the data needs to be properly conditioned. Fig. 3(a) shows the time plots of the model inputs. From the Figure it is clearly evident that the inputs are stationary signals fluctuating around their mean values. Therefore, the input signals are centered as follows:

$$\Delta u_i(k) = u_i(k) - \bar{u}_i, \quad (5)$$

where $u_i(k)$ denotes the i^{th} element of the input vector, see Eqs. (1) and (3), and $\bar{u}_i$ denotes the corresponding mean value.

Process outputs are depicted in Fig. 3(b). In contrast to the input signals, the output signals are no longer stationary but show a drift, which is due to the degradation of the stack during the experiment. Such a drift can be directly associated with various degradation phenomena such as nickel agglomeration in the Ni-based composite anodes [28]. The phenomenon manifests itself in an increase in the ohmic and/or polarisation resistances of the stack, which lead to higher heat losses inside the stack and an increase in stack temperatures.

In order to capture the dynamics of the thermal processes, while ignoring the process of degradation, the drift needs to be removed. Hence the signal $y_i(k)$ is assumed to be composed of a stationary noise $n(k)$ and the linear drift $\bar{y}_i(k)$

$$y_i(k) = \bar{y}_i(k) + n(k) = \alpha_i k T_s + \beta_i + n_i(k) \quad (6)$$

where $y_i(k)$ is the i^{th} true output signal, e.g. the stack maximum temperature during normal operation, α_i represents the increase rate of the temperature, β_i is the offset, and $n_i(k)$ is additive noise.

The optimal parameters $\hat{\alpha}_i$ and $\hat{\beta}_i$ are estimated from the data in the training set by using the least squares criterion

$$\begin{pmatrix} \hat{\alpha}_i \\ \hat{\beta}_i \end{pmatrix} = \arg \min_{\{\alpha_i, \beta_i\}} \frac{1}{N} \sum_{k=1}^N (y_i(k) - \alpha_i k T_s - \beta_i)^2. \quad (7)$$

Fig. 4 shows an excerpt from the recorded stack maximum temperature and the identified drift.

With estimated $\hat{\alpha}_i$ and $\hat{\beta}_i$ we proceed to identification with the de-trended output data $\Delta y_i(k)$

$$\Delta y_i(k) = y_i(k) - \hat{\alpha}_i k T_s - \hat{\beta}_i. \quad (8)$$

4.2. Model order selection

A crucial step in sub-space identification is the selection of the model order n in the state space model. The key idea in our approach is to analyse two dynamic models. Model $\mathcal{M}_1$ relates the vector of system inputs $\mathbf{u}$ to the minimum and maximum temperatures, represented in the output vector $\mathbf{y}_1$, i.e. $\mathcal{M}_1 : \mathbf{u} \mapsto \mathbf{y}_1$. The second model, $\mathcal{M}_2$, relates $\mathbf{u}$ to the output temperatures $\mathbf{y}_2$, i.e. $\mathcal{M}_2 : \mathbf{u} \mapsto \mathbf{y}_2$.

From result ii.) in the Appendix, see Eq. (A.10), it follows that the model order can be derived from singular values (SV) of the projection matrix constructed from measured data. Moreover, according to [29], each column of the resulting matrix can be viewed as a response of the system to the non-zero initial state without persistent excitation, thus unambiguously characterising the dynamic response of the system. Therefore, the dominant dynamics of the system can be captured by observing the eigenvalues of the corresponding matrix. In the ideal noise-free case, as long as the model order n is smaller than the true system order n^* , the n singular values are non-zero. If $n > n^*$, the first n^* singular values are different from zero while the remaining $n^* - n$ ones drop to zero. This is a clear indication of the associated model order. The estimation of the model parameters is then based on results (A.3) and (A.4) in the Appendix. However, in the case of noise contaminating the measurements, all of the singular values are usually non-zero. Therefore, only the first few, which are relatively significant with respect to the highest singular value, should be considered. Consequently, to enhance the decision-making regarding the model order, we will apply additional model quality criterion, i.e. the mean squared error (MSE):

$$MSE = \frac{1}{N} \sum_{k=1}^N \mathbf{e}(k)^T \mathbf{Q} \mathbf{e}(k) \quad (9)$$

where N denotes the number of samples in the data set, $\mathbf{e}(k) = \mathbf{y}(k) - \hat{\mathbf{y}}(k)$ is the vector of prediction errors, and $\mathbf{Q}$ is the weighting matrix. In our case, $\mathbf{Q} = \mathbf{I}_2$.

In order to ascertain evidence supporting the modelling procedure, the analysis was performed on two data sets labelled DS1 and DS2. Firstly, the model order of $\mathcal{M}_1$ is identified. Six candidate model orders were analysed: $n \in \{1, 2, \dots, 6\}$. Fig. 5(a–b) provides model quality indicators with respect to the model order. From the plots showing the MSE as a function of the model order, an insignificant reduction in the MSE can be observed for model orders above $n > 2$. By analysing the singular values of the projected data matrix, it is clearly evident that the dominant dynamics can be described by only considering the first two singular values. Therefore, the selected model order is $n = 2$.

A similar analysis was performed on model $\mathcal{M}_2$, which resulted

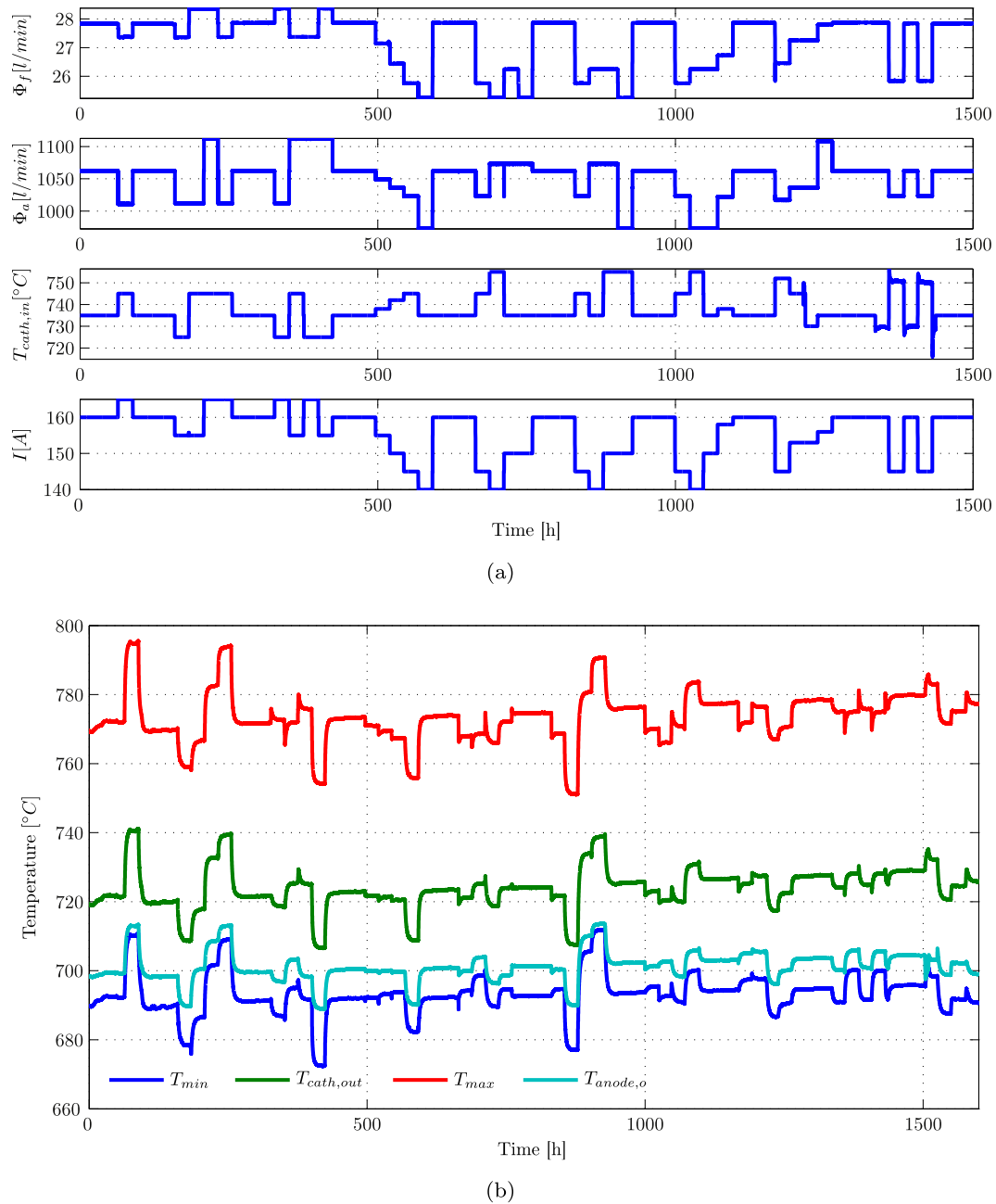


Fig. 3. Process inputs(a) and outputs (b) for the model in Fig. 2. The inputs are stationary signals fluctuating around the mean value. The inner stack temperatures exhibit about 10 °C linear drift over 1500 h of operation due to the stack degradation. Consequently, drift can also be observed in exhaust temperatures.

in an almost identical pattern of indicators in terms of candidate model order n , see Fig. 5(c–d). The second order turns out to be a suitable option again. This sounds surprising at the first glance. Namely, a stack is a distributed parameter system, which can, in principle, be described by a (high-dimensional) state space model. The two spots with maximum and minimum temperatures can be viewed as just two states of such a high-dimensional model. That implies that between T_{min} and T_{max} , on one hand, and output temperatures, on the other, there are some extra dynamic states. Consequently, the order of model $\mathcal{M}_2$ should be higher than the order of $\mathcal{M}_1$. However, evidence obtained from the data suggests equality, meaning that the expected extra dynamics are simply negligible. A plausible physical explanation of such an outcome might be in the fact that the heat capacity of the stack is much

greater than that of the flowing gases. Furthermore, the physical dimensions of the stack are rather small, hence the measurements inside the stack are relatively close to those from the exhausts.

The quality of models $\mathcal{M}_1$ and $\mathcal{M}_2$ is judged from the validation data set,¹ see Fig. 6. Notice that model outputs are compared with the de-trended measured data in order to eliminate the influence of the degradation effects. A comparison of nominal model and de-trended measured data shows the satisfactory precision of the prediction model. This in turn explains why $\mathcal{M}_1$ itself cannot be

¹ These models only hold for the operating envelope depicted in Fig. 3. However, in Ref. [30] we show that similar models describe system behaviour in a wider operating range.

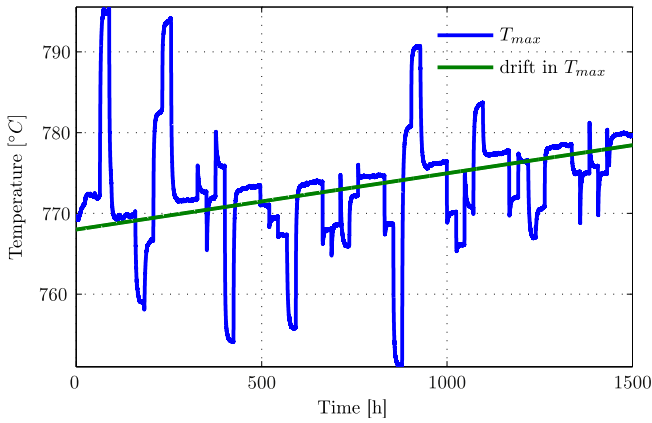


Fig. 4. An excerpt from the recorded maximum temperature within the stack (T_{max}) and the indicated trend.

used as an estimator of stack minimum and maximum temperature. Indeed, even if we start a simulation from initial conditions equal to the initial conditions of the true system, the stack degradation will cause a gradual increase in the prediction error, which renders the estimator model $\mathcal{M}_1$ useless.

The effect of non-modelled drift can be observed in Fig. 7. Due to stack degradation, a large prediction error, especially towards the

end of the experiment, can easily be seen. Stack temperatures and, consequently, exhaust temperatures entail non-modelled drift caused by stack degradation. Its slow evolution is difficult to model, especially by taking into account only the available identification data set. At the end of the experiment, at $t = 1500h$, the drift leads to an error of approximately 10°C .

However, a closer analysis of the process outputs in Fig. 3(b) that the drift in stack inner temperatures is nearly identical to the one of exhaust temperatures. Therefore, the problem of designing the estimator for inner stack temperatures, that will be unaffected by degradation, is solved by fusing the two models $\mathcal{M}_1$ and $\mathcal{M}_2$ into a single predictor model.

5. Derivation of a simple predictor model by fusing models

Drifts in stack and output temperatures preclude using model $\mathcal{M}_1$ as an estimator. We will obtain such an estimator by fusing $\mathcal{M}_1$ and $\mathcal{M}_2$.

From model order analysis above it follows that the two state-space models have the same order $n = 2$. In this case there exists a similarity transform $\mathbf{x}_2 = \mathbf{T}\mathbf{x}_1$, where $\mathbf{T}$ is an appropriate non-singular matrix. Since $\mathbf{C}_2$ is quadratic and non-singular, we can express $\mathbf{x}_2$ as follows

$$\mathbf{x}_2(k) = \mathbf{C}_2^{-1}\mathbf{y}_2(k) - \mathbf{C}_2^{-1}\mathbf{D}_2\mathbf{u}(k) - \mathbf{C}_2^{-1}\mathbf{v}_2(k).$$

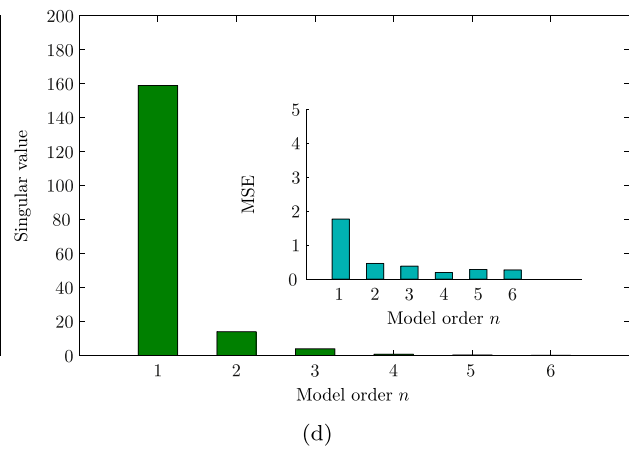
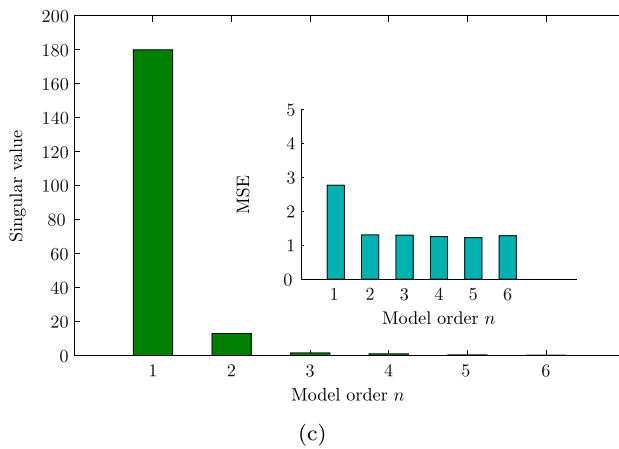
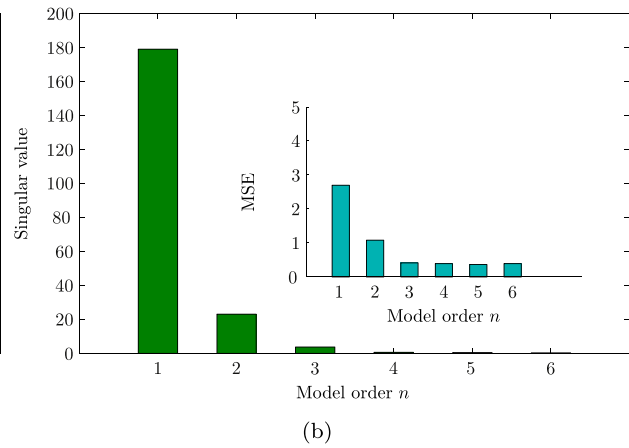
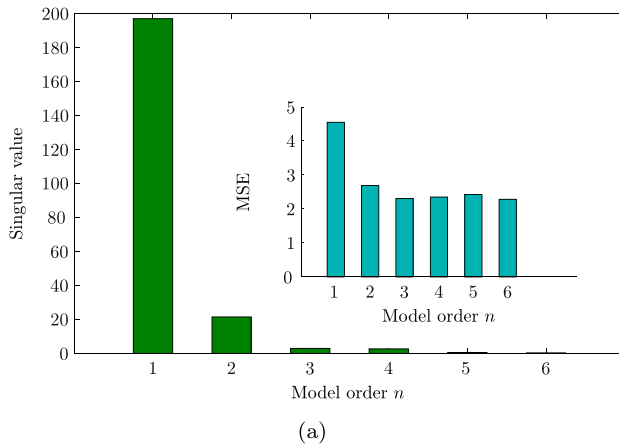


Fig. 5. The quality criteria of the identified models: $\mathcal{M}_1$ on (a) DS1 and (b) DS2, and $\mathcal{M}_2$ on (c) DS1 and (d) DS2. From the eigenvalues plots, one can notice that third and higher eigenvalues do not change much, which indicates that the model order $n = 2$ seems an appropriate selection. A similar indication comes from the MSE plots. Even if we raise the system order higher than 2, the criterion function barely decreases. That means the minimum MSE of the prediction error can be obtained almost at system order $n = 2$. Therefore, the selected model order is two. Fig. 6 shows the corresponding simulation.

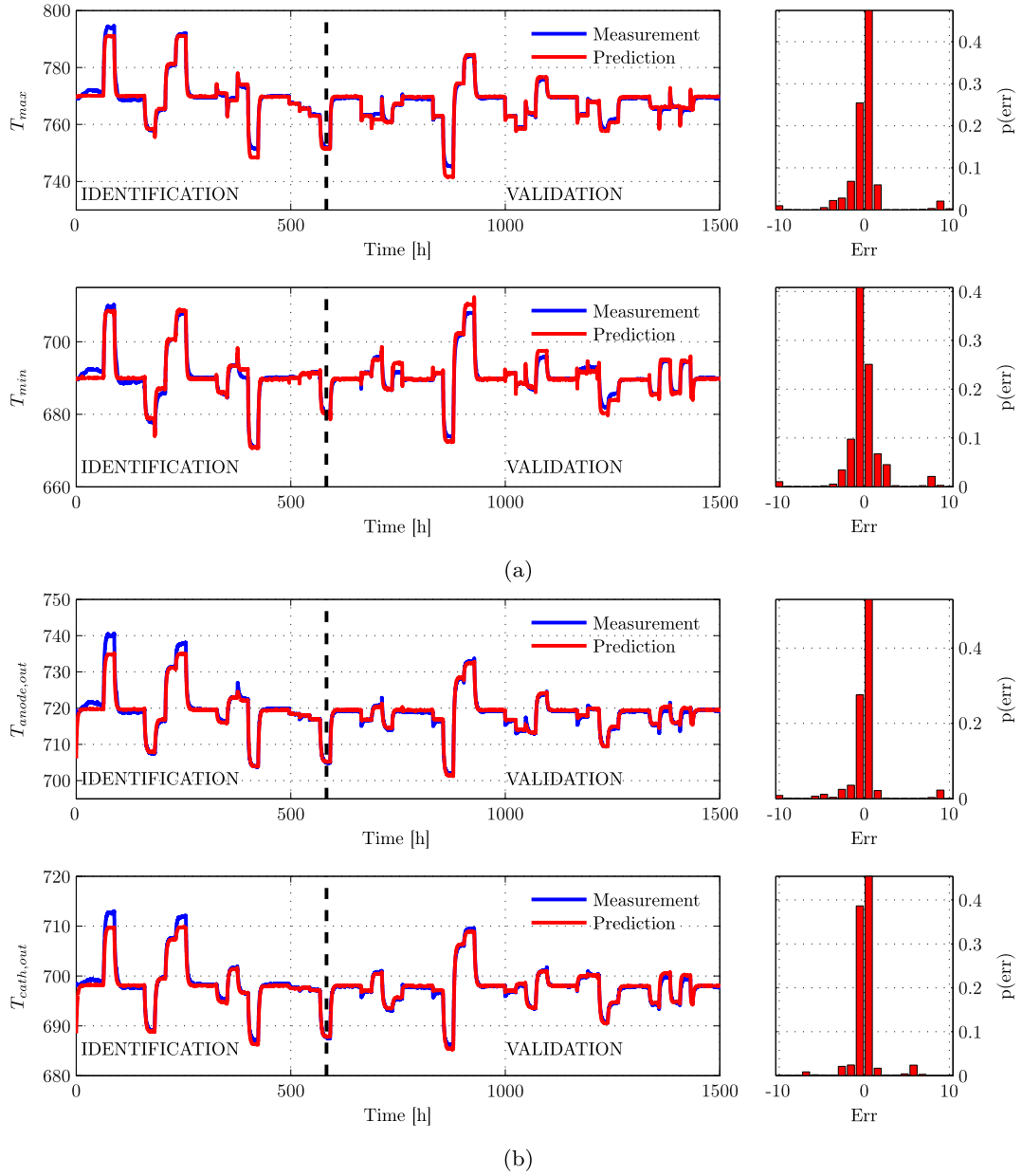


Fig. 6. Prediction results for $\mathcal{M}_1: \mathbf{y}_1 = [T_{max} \ T_{min}]^T$, i.e. stack temperatures, and $\mathcal{M}_2: \mathbf{y}_2 = [T_{anode,out} \ T_{cath,out}]^T$, i.e. exhaust temperatures. Plots on the right-hand side show histograms of the corresponding prediction error, which remain low for the entire data set. The vertical dashed line splits the time series into identification and validation data sets.

We put this expression into the corresponding equation in (4) and get

$$\mathbf{y}_1(k) = \underbrace{\mathbf{C}_1 \mathbf{T} \mathbf{C}_2^{-1}}_{\Psi_{y_2}} \mathbf{y}_2(k) + \underbrace{(\mathbf{D}_1 - \mathbf{C}_1 \mathbf{T} \mathbf{C}_2^{-1} \mathbf{D}_2)}_{\Psi_u} \mathbf{u}(k) + \underbrace{\mathbf{v}_1(k) - \mathbf{C}_1 \mathbf{T} \mathbf{C}_2^{-1} \mathbf{v}_2(k)}_{\xi(k)}. \quad (10)$$

Eq. (10) can be put into a compact regression form, which is represented by the following static relation

$$\mathcal{M}_3\{\mathbf{u}, \mathbf{y}_2\} \mapsto \mathbf{y}_1$$

i.e.

$$\mathcal{M}_3: \mathbf{y}_1(k) = \Psi_{y_2} \mathbf{y}_2(k) + \Psi_u \mathbf{u}(k) + \xi(k). \quad (11)$$

Note that $\xi(k)$ is zero mean white Gaussian noise.

The unknown parameters Ψ_{y_2} and Ψ_u are evaluated by the least squares method. In our case, this results in

$$\begin{aligned} \Psi_{y_2} &= \begin{bmatrix} 0.5259 & 0.5294 \\ -0.2648 & 1.2730 \end{bmatrix}, \Psi_u \\ &= \begin{bmatrix} -2.6182 & -0.0041 & 0.0561 & 0.1875 \\ 0.3745 & -0.0370 & -0.0489 & 0.2208 \end{bmatrix}. \end{aligned} \quad (12)$$

Fig. 8 shows the reconstructed inner stack temperatures employing model $\mathcal{M}_3$ (11) and parameters (12). Note that the precision of the temperature estimates is within $2\sigma \approx 1^\circ\text{C}$ and $2\sigma \approx 1.8^\circ\text{C}$ for maximum and minimum temperatures, respectively.

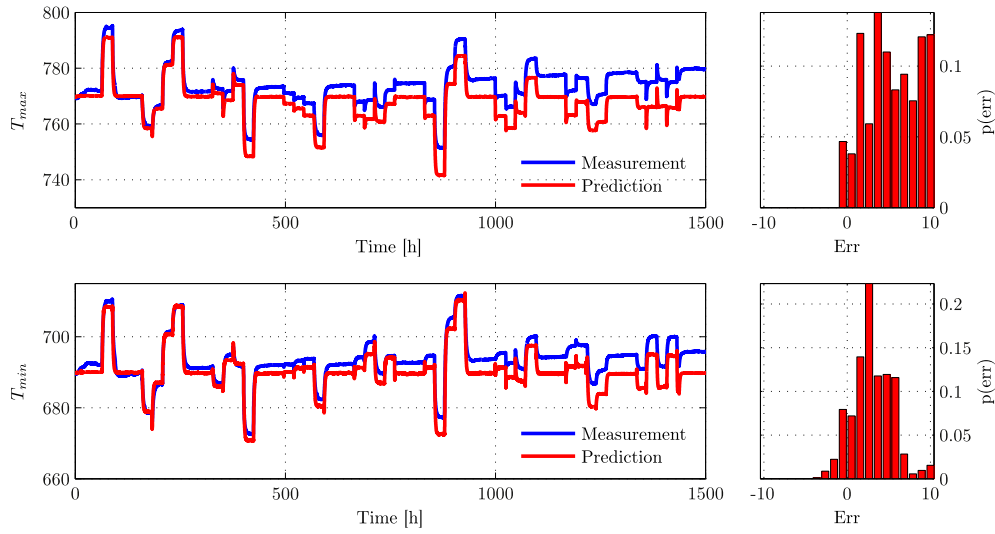


Fig. 7. Prediction results of the stack temperatures obtained from $\mathcal{M}_1$. Due to degradation mechanisms, stack temperature slowly drifts. Such a drift is difficult to predict, hence the prediction error increases over time. The histograms on the right of each of the plots show the error histograms of the corresponding time series.

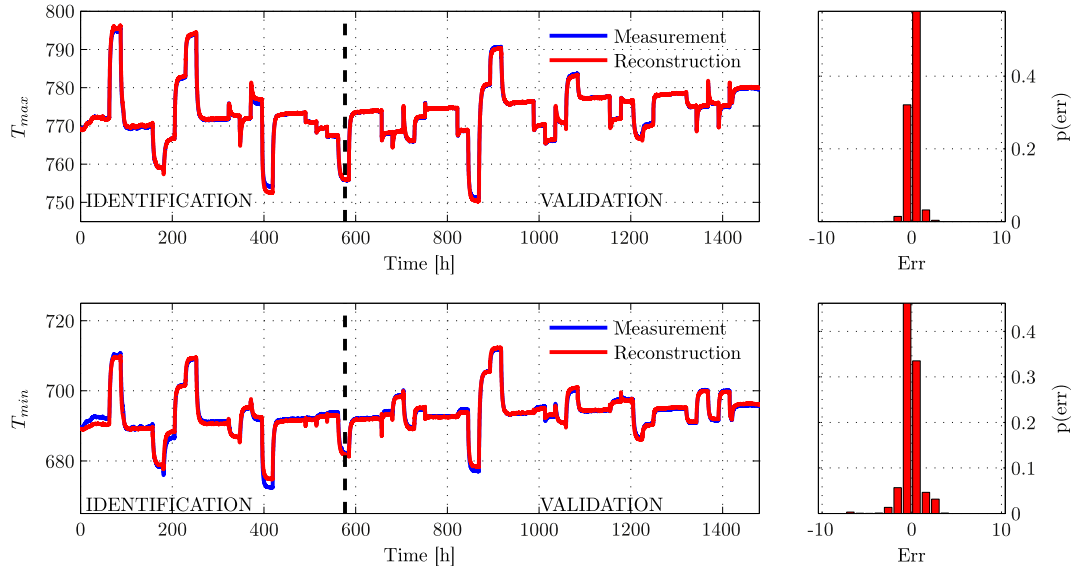


Fig. 8. The reconstruction results of model $\mathcal{M}_3$: $\mathbf{y}_1 = [T_{max} \ T_{min}]^T$, i.e. stack temperatures. Plots on right hand-side show histogram the corresponding reconstruction error, which are significantly narrower than the one from Fig. 7, thus indicating higher precision.

Eq. (11) shows the simple relationship between inner stack temperatures, and system input and system output temperatures. This result is important as it shows that during operation of a stack that is not instrumented internally with thermocouples (as is always the case in commercial applications) the maximum and minimum inner temperatures can be reconstructed by a linear combination of measurable system inputs and measurable system outputs.

It is important to stress also that model $\mathcal{M}_3$ successfully handles the problem of drift in stack temperatures. The drift in exhaust temperatures compensates the one in the stack, hence there is no need for explicit characterisation of the drift. The explanation arises from the fact that degradation affects states of the models $\mathcal{M}_1$ and $\mathcal{M}_2$ equally so the similarity transform always exist and brings to Eq. (11). Predictions shown in Fig. 8 are obtained directly from centered raw data. Trends in temperatures inside and outside the stack cancel almost ideally on both sides of (11). Furthermore, the

error histograms are narrower than the ones from Fig. 7, indicating higher estimation precision.

It should be stressed, however, that the relationship (11) cannot be claimed to generally apply for all stacks, i.e. it holds in the particular case study owing to the specific circumstances related to the system construction and system excitation. In the case of larger stacks, or a different configuration of the system, it is likely that expression (11) should be replaced by a dynamic relation.

6. Conclusion

The main result of the paper is an efficient and consistent data-driven approach to the design of on-line estimators of maximum and minimum stack internal temperatures. By working with experimental data, we show that subtle use of a subspace identification approach results in a simple estimator model $\mathcal{M}_3$, which is extremely convenient for use in real applications on inexpensive

process hardware.

The derivation of the causal relationship of the estimators relies on physical reasoning. Model order, as well as model parameters, are derived directly from high quality experimental data recorded during normal operation of an SOFC power generating unit. The resulting estimator builds on the linear relationship between standard measured variables such as flows, temperatures, and electric quantities. Experimental verification shows that the accuracy of the estimated minimum and maximum internal stack temperatures is below 3%, irrespective of drifts caused by stack degradation. Moreover, it must be stressed that no additional modelling or characterisation of the degradation is required, hence making the approach ideal for commercial implementation.

Acknowledgement

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Appendix

The system model

$$\mathbf{x}(k+1) = \mathbf{A}\mathbf{x}(k) + \mathbf{B}\mathbf{u}(k) \quad (\text{A.1})$$

$$\mathbf{y}(k) = \mathbf{C}\mathbf{x}(k) + \mathbf{D}\mathbf{u}(k) + \mathbf{v}(k) \quad (\text{A.2})$$

where $\mathbf{u} \in \mathbb{R}^p$, $\mathbf{x} \in \mathbb{R}^n$, $\mathbf{y} \in \mathbb{R}^q$, $\mathbf{A} \in \mathbb{R}^{n \times n}$, $\mathbf{B} \in \mathbb{R}^{n \times p}$, $\mathbf{C} \in \mathbb{R}^{q \times n}$, and $\mathbf{D} \in \mathbb{R}^{q \times p}$, and $\mathbf{v}(k) \sim \mathcal{N}(0, \Sigma_v)$ is zero-mean white Gaussian noise mimicking random effects in sensors and unmodelled effects.

Given the state $\mathbf{x}(i)$ at time sample i , the system response at time sample $k+i$ equals

$$\mathbf{x}(k+i) = \mathbf{A}^k \mathbf{x}(i) + \sum_{j=0}^{k-1} \mathbf{A}^{k-j-1} \mathbf{B} \mathbf{u}(i+j) \quad (\text{A.3})$$

By combining (A.3) and (A.2) the relationship between the recorded input and output data sequences $\{\mathbf{u}(i), \dots, \mathbf{u}(i+r-1)\}$ and $\{\mathbf{y}(i), \dots, \mathbf{y}(i+r-1)\}$ can be represented in the following stacked form

$$\begin{pmatrix} \mathbf{y}(i) \\ \vdots \\ \mathbf{y}(i+r-1) \end{pmatrix} = \underbrace{\begin{pmatrix} \mathbf{C} \\ \vdots \\ \mathbf{C}\mathbf{A}^{r-1} \end{pmatrix}}_{\mathcal{O}_r} \mathbf{x}(i) + \underbrace{\begin{pmatrix} \mathbf{D} & 0 & 0 & \dots & 0 \\ \mathbf{C}\mathbf{B} & \mathbf{D} & 0 & \dots & 0 \\ \vdots & \vdots & \vdots & \ddots & \vdots \\ \mathbf{C}\mathbf{A}^{r-2}\mathbf{B} & \mathbf{C}\mathbf{A}^{r-3}\mathbf{B} & \dots & \mathbf{C}\mathbf{B} & \mathbf{D} \end{pmatrix}}_{\mathcal{F}_r} \underbrace{\begin{pmatrix} \mathbf{u}(i) \\ \vdots \\ \mathbf{u}(i+r-1) \end{pmatrix}}_{\mathbf{u}_{i,r}} + \underbrace{\begin{pmatrix} \mathbf{v}(i) \\ \vdots \\ \mathbf{v}(i+r-1) \end{pmatrix}}_{\mathbf{v}_{i,r}} \quad (\text{A.4})$$

Note that $\mathcal{O}_r$ is the extended observability matrix. It furthermore must be noted that the matrices $\mathcal{O}_r$ and $\mathcal{F}_r$ depend on the system matrices $\{\mathbf{A}, \mathbf{B}, \mathbf{C}, \mathbf{D}\}$.

By repeating Eq. (A.4) for different time instances, we obtain the following data structures

$$\mathbf{Y}_{0,r,N} = (\mathbf{y}_{0,r} \mathbf{y}_{1,r+1} \dots \mathbf{y}_{N-1,N+r-2})$$

$$\mathbf{X}_{0,N} = (\mathbf{x}(0) \dots \mathbf{x}(N-1))$$

$$\mathbf{U}_{0,r,N} = (\mathbf{u}_{0,r} \mathbf{u}_{1,r+1} \dots \mathbf{u}_{N-1,N+r-2})$$

$$\mathbf{V}_{0,r,N} = (\mathbf{v}_{0,r} \mathbf{v}_{1,r+1} \dots \mathbf{v}_{N-1,N+r-2})$$

resulting in the following expression

$$\mathbf{Y}_{0,r,N} = \mathcal{O}_r \mathbf{X}_{0,N} + \mathcal{F}_r \mathbf{U}_{0,r,N} + \mathbf{V}_{0,r,N} \quad (\text{A.5})$$

We make the following assumptions:

- i.) *Assumption 1.* The sequences $\mathbf{u}(k)$, $\mathbf{x}(k)$ and $\mathbf{v}(k)$ are ergodic stochastic processes and $\mathbf{v}(k)$ is uncorrelated with $\mathbf{u}(k)$ and $\mathbf{x}(k)$.
- ii.) *Assumption 2.* The input is persistently exciting, meaning that all the system eigenvalues are excited. This can be expressed with the following rank condition

$$\text{rank} \left(\lim_{N \rightarrow \infty} \frac{1}{N} \begin{pmatrix} \mathbf{X}_{i,N} \\ \mathbf{U}_{i,r,N} \end{pmatrix} \begin{pmatrix} \mathbf{X}_{i,N}^T & \mathbf{U}_{i,r,N}^T \end{pmatrix} \right) = n + rm \quad (\text{A.6})$$

From Assumption 1, we can readily see the covariance:

$$\mathcal{V}_r = \lim_{N \rightarrow \infty} \frac{1}{N} \mathbf{V}_{i,r,N} \mathbf{V}_{i,r,N}^T = \underbrace{\begin{pmatrix} \Sigma_v & 0 & \dots & 0 \\ 0 & \Sigma_v & \dots & 0 \\ \vdots & \vdots & \ddots & \vdots \\ 0 & 0 & \dots & \Sigma_v \end{pmatrix}}_{r \text{ times}}$$

We follow the MOESP (Multivariable Output-Error State-Space) approach of [27], which relies on the RQ factorisation of data matrices. In particular

$$\begin{pmatrix} \mathbf{U}_{0,r,N} \\ \mathbf{Y}_{0,r,N} \end{pmatrix} = \begin{pmatrix} \mathbf{R}_{11} & 0 \\ \mathbf{R}_{21} & \mathbf{R}_{22} \end{pmatrix} \begin{pmatrix} \mathbf{Q}_1 \\ \mathbf{Q}_2 \end{pmatrix} \quad (\text{A.7})$$

where $\mathbf{Q}_i$ are orthonormal matrices.

Now we introduce the projection matrix

$$\Pi_{0,r,N}^\perp = \mathbf{I}_N - \mathbf{U}_{0,r,N}^T (\mathbf{U}_{0,r,N} \mathbf{U}_{0,r,N}^T)^{-1} \mathbf{U}_{0,r,N} \quad (\text{A.8})$$

By combining (A.5), (A.7) and (A.8) and taking into account the conditions above, two important results can be derived [27].

- i.) *Result 1.* The range of the extended observability matrix is

$$\text{range}(\mathcal{O}_r) = \text{range}(\mathbf{R}_{22} \mathbf{Q}_{22}) \quad (\text{A.9})$$

- ii.) *Result 2.* The SVD decomposition of matrix $\mathbf{R}_{22}$ obeys the following relation

$$\lim_{N \rightarrow \infty} \frac{1}{\sqrt{N}} \mathbf{R}_{22} = (\Phi_1 \ \Phi_2) \underbrace{\begin{pmatrix} \sqrt{\Sigma_n^2 + \mathcal{V}_{(1)}} & 0 \\ 0 & \sqrt{\mathcal{V}_{(2)}} \end{pmatrix}}_{\Sigma} \begin{pmatrix} \Psi_1^T \\ \Psi_2^T \end{pmatrix} \quad (\text{A.10})$$

where Φ_i and $\Psi_i, i \in \{1, 2\}$ are unitary matrices. Matrix Σ_n^2 contains the non-zero singular values of the matrix $\mathcal{O}_r \mathbf{M}_X \mathcal{O}_r^T$ where

$$\mathbf{M}_X = \lim_{N \rightarrow \infty} \frac{1}{N} \mathbf{X}_{i,N} \Pi_{i,r,N}^\perp \mathbf{X}_{i,N}^T$$

The other matrices in the expression are $\mathcal{V}_{(1)} = \mathcal{V}_r(1 : n, 1 : n)$ and $\mathcal{V}_{(2)} = \mathcal{V}_r(n+1 : rq, n+1 : rq)$.

The expression (A.10) is essential for system order determination. The matrix of singular values Σ consists of two parts, one with "small" singular values $\sqrt{\mathcal{V}_{(2)}}$ and the other with "large" singular values $\sqrt{\Sigma_n^2 + \mathcal{V}_{(1)}}$. The large singular values mainly arise from the excited system eigenstructure. If there is no noise in the outputs $\Sigma_v = \mathbf{0}$, then it is easy to identify n non-zero eigenvalues and hence determine system order. The task becomes less obvious if the signal to noise ratio is not high enough.

Without going into technical details, which can be found in Ref. [27], two more results need to be presented.

- iii.) *Result 3.* Eq. (A.9) states that the column space of matrix $\mathcal{O}_r$ and $\mathbf{R}_{22} \mathbf{Q}_{22}$ are the same. From linear algebra [31] and the results of (A.9) and (A.10), it follows that

$$\text{range}(\mathcal{O}_r) = \text{range}(\Phi_1).$$

From this, one can calculate the system matrices $\mathbf{A}$ and $\mathbf{C}$ up to a similarity matrix $\mathbf{T}$, i.e.

$$\Phi_1 = \mathcal{O}_r \mathbf{T} = \begin{pmatrix} \mathbf{C} \mathbf{T} \\ \mathbf{C} \mathbf{T}^2 \\ \vdots \\ \mathbf{C} \mathbf{T}^{r-1} \end{pmatrix} \quad (\text{A.11})$$

- iv.) *Result 4.* The system matrices $\mathbf{B}$ and $\mathbf{D}$ and the initial condition $\mathbf{x}(0)$ can be found by solving the least squares problem

$$\begin{aligned} \hat{\mathbf{T}}_r &= \arg \min_{\mathbf{T}_r} \left\| \mathbf{Y}_{0,r,N} - \mathbf{T}_r \mathbf{Y}_{0,r,N} \right\|^2 \\ &= \mathbf{Y}_{0,r,N} \mathbf{U}_{0,r,N}^T \left(\mathbf{Y}_{0,r,N} \mathbf{Y}_{0,r,N}^T \right)^{-1} \end{aligned} \quad (\text{A.12})$$

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Chapter 4

Condition Monitoring of SOFC Stack

Condition monitoring (CM) is a process of monitoring some system parameters, which carry information about its condition. The task is to detect, isolate, and identify an incipient fault in the system. Fault detection and isolation (FDI) approaches have been vastly applied from mechanical systems [94], to electrochemical ones [95], in particular to PEMFCs [67]. The FDI information is then exploited in condition-based maintenance, which is vital for modern complex systems in industry, vehicles, military and commercial vessels, etc. [96]. Its main objective is to (i.) maximise the availability of the asset, while at the same time (ii.) minimising the maintenance and operating costs.

The tools that facilitate CM can be effectively exploited in a proactive manner. The health-aware control employs information about the health to actively increase overall reliability of the system. The SOFC community has focused mostly on the development of algorithms, aiming at characterizing microstructural degradation of the fuel cells [40] where EIS measurements, e.g. [68], [97], [98], are employed. To measure EIS of the fuel cells, or stacks, the system is perturbed with sinusoid-like signals. These signals are superimposed at specific direct current (DC), and the voltage response of the system is measured. Then, employing different methodologies, the frequency response of the fuel cells is usually employed to characterize the behavior of the system. The frequency response is usually obtained via Fourier transform. Recently, an advanced approach for impedance estimation employing wavelet transform has been developed as an alternative [63]. The frequency response of the system is then used in different ways with the aim of detecting the onset of a fault. For instance, the information of the Nyquist plot is aggregated into a single condition monitoring indicator [99], an equivalent circuit model is fitted to experimental data and its corresponding parameters are monitored [100], or simple polynomial models are employed to describe impedance [101].

The monitoring of the different components in the BoP has also received a fair amount of attention. These FDI approaches usually build on model-based diagnosis [102]. Model-based diagnostics employs the model of a system under consideration to generate the so-called residuals. In the simplest methodologies, the model is simulated in parallel with the process and the residuals are generated simply by subtracting the model predictions and measurements on the process. Ideally, the residual is zero when the system is fault-free and deviates away when fault occurs. To this aim, a plethora of models of SOFC systems exists in the literature [103]. The models are then employed to perform the so called fault tree analysis (FTA) [104] and develop the corresponding fault signature matrix (FSM). In more detail, Arsie, Filippi, Marra, *et al.* [18] introduced the FTA to the SOFC community and demonstrated the applicability of the approach for on-line fault detection and isolation. The approaches deal with detecting different faults, e.g. air and fuel leakage, stack and reformer degradation [64], other authors deal with reformer faults [42], faults in blowers,

and controllers [65]. The FDI systems developed in the mentioned work has been recently expanded and adapted for the SOFC system in AOGR configuration [105].

Apart from diagnosis, prognostics of the SOFC received only limited attention. The following chapter briefly reviews the state-of-the art and proposes an approach for SoH estimation and RUL prediction for the SOFCs, thus filling the gap in the literature. The approach was developed and validated in [87]. Additional experimental validation, performed on a different SOFC stack, is described in [106].

RUL predictions are achieved by employing a model which describes the ongoing degradation. Monte Carlo (MC) simulation are then employed to simulate how the system will degrade in future. The simulations are stopped when the ASR reaches the end-of-life (EoL) criterion. Consequently, the EoL criterion becomes one of the most important parameters in the algorithm and its selection needs to be addressed properly. The assumption here is that the EoL specification is provided by the end-user of the SOFC system. For instance, the EoL can be calculated from minimal allowed voltage output of the system, or from maximal allowed drop in efficiency. Nevertheless, the ASR can be regarded as a direct measure of losses in the SOFC stack.

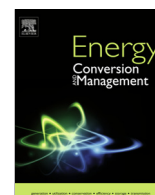
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State of health estimation and remaining useful life prediction of solid oxide fuel cell stack

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ABSTRACT

Having concurrent information regarding the state of health (SoH) of an operating solid oxide fuel cell (SOFC) stack can actively improve its overall management. Firstly, operating the SOFC by taking into account current health conditions, and its anticipated trend, can be beneficial to the total life span of the stack. Furthermore, such information can be of great importance to the maintenance staff, e.g. unplanned shutdowns can be avoided.

Relatively little work has been done in the field of remaining useful life (RUL) prediction of SOFCs. The majority of work employs stack/cell voltage as a direct link for RUL predictions. This paper proposes an integrated approach for SoH estimation based on stack's Ohmic area specific resistance (ASR). Subsequently, a drift model that describes the ASR increase over time enables accurate RUL prediction. The approach consists of three steps. Firstly, an Unscented Kalman filter, based on a lumped stack model, estimates the current ASR value. Secondly, a drift model for describing the temporal evolution in ASR is recursively identified employing the linear Kalman filter. Finally, employing the identified drift model, Monte Carlo simulation is performed to predict future time evolution in ASR and so to obtain RUL. The developed approach is validated with experimental data from a 10 kW SOFC power system. The results confirm that ASR is a viable SoH indicator for the SOFC stack.

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1. Introduction

High electrical efficiency and low level of emissions are two of the important advantages promoting solid oxide fuel cell systems (SOFCs) as a clean, efficient and sustainable power source of the future. In the last few years, approximately 20k SOFC installations are reported worldwide [1,2]. However, due to issues related with durability and reliability with respect to high cost of SOFC systems, deployment to the market is still modest [3]. In order to reach the goal, the bulk of research has been conducted in the area of material science with the aim to improve electrochemical and physical properties of materials for SOFC [4]. Additionally, in the field of control the majority of work is devoted to increase in SOFC overall availability and durability [5–8].

Various approaches to condition monitoring have been proposed with the aim to detect the onset of fault and hence allow for the design of accommodation actions [3,9]. However, for safety

reasons and maintenance purposes, the information of particular practical value is how long the system will operate prior to its end-of-life (EoL). The importance of anticipating the remaining useful life (RUL) of SOFC stacks is twofold. Firstly, it can help the control system in accommodating the control strategies in order to extend the achievable lifespan. Moreover, it has been shown in [10] how an optimal operating point that respects degradation of the system can significantly reduce the electricity cost. Secondly, the information about RUL is central in predictive maintenance of the deployed devices. Having RUL information at hand, the maintenance staff can efficiently prepare for the system overhaul in due time, thus minimising the downtime of power generation, maximising availability of the system, and hence reducing the cost of power conversion.

However, performing RUL prediction for a particular system is a difficult task. The issue has been a topic of interest over different fields, from mechanical systems [11], to electrochemical ones [12]. So far, relatively little attention has been devoted to the development of algorithms for predicting SOFC RUL. Some work has been done in the area of proton exchange membrane (PEM) fuel cells. Although PEM and SOFC differ in many aspects, it is

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worth screening the main ideas applied to PEM. For example, in [13], the authors discussed and summarised challenges related to PEM fuel cell prognostics and RUL predictions. The authors identify the importance of suitable health indicators for RUL prediction and address the definition of the EoL point of fuel cells.

Works related to RUL prognosis in a PEM aim at modelling temporal evolution of stack voltage, voltage related power output, or efficiency due to degradation [13]. The most common approach is to consider voltage as state of health (SoH) indicator and perform RUL upon evident drift in stack/cell voltage. Such a characterisation of SoH is often justified because voltage is directly associated with power output, and hence the efficiency of power conversion. Relatively few works define SoH differently; for instance, in [14] the authors employ electro-chemical surface area. Various empirical models are then employed in order to predict voltage drop in the future and to estimate RUL. The employed models range from simple ones with linear, polynomial, and exponential structures [15], to complex structures common in machine learning society [16–19].

An alternative approach is via electrochemical impedance spectroscopy (EIS), which is a widely adopted tool for characterisation and diagnostics of fuel cells [20–22]. For that purpose, the fuel cell current or voltage is perturbed by sinusoidal signals. By measuring the voltage response of the fuel cells, the corresponding impedance is then evaluated and employed for diagnostic purposes. In the context of prognostics, EIS measurements are usually employed for parameter estimation of an equivalent circuit model of the fuel cell [23], and simple linear, or polynomial, models describing impedance [24]. Then the identified models are utilised to evaluate SoH and RUL prediction [25]. A more thorough review of the EIS based techniques for diagnostics and prognostics of PEM is available in [13,26].

To the best of the authors' knowledge, only the works in [27–29] address issues related to SOFC prognostics. Here cell/stack voltage is utilised as a single SoH indicator. However, it is widely known that the output voltage is directly affected by operating conditions imposed on the system. Therefore, it is difficult to distinguish whether the voltage drop is caused by degradation of the fuel cells, a change in load condition, or if it is a response of the system due to the controller.

The approaches mentioned are tested under stationary operation only, which is in practice a rather restrictive condition. The issues related to non-stationary operation are omitted. On the other hand, the approaches employing EIS measurements for RUL prediction inherit two main drawbacks of the EIS measurements. One is that the fuel cells need to be perturbed during operation at a particular operating condition. The second is that additional electronics are required, which augments the final cost of the system.

In this paper, we propose a novel approach for RUL prediction of SOFC stacks. The distinctive property of the approach to prognostics is that it is invariant to the operating conditions of the system, thus rendering the need for constant operation unnecessary. The solution below addresses three important aspects of prognostics:

- (i) estimation of SoH of an SOFC stack,
- (ii) prediction of SoH, and
- (iii) definition of EoL of the stack and RUL prediction.

Firstly, an appropriate SoH indicator is identified. The key is the observation that many degradation processes can manifest in severe increase of anodes Ohmic and/or polarisation resistances [30–34]. Therefore, an estimation of internal resistance carries vital information regarding the SoH of the stack.

Internal resistance is a resultant of many concurrent chemical processes across the stack. As direct measurements are precluded,

the only way to gain insight is to reason from available instrumentation mounted around the stack and a mathematical model of system dynamics. Our idea is based on two steps. In the first step we estimate the internal resistance of the SOFC stack from available instrument readings. More specifically, we reconstruct the internal aggregated Ohmic stack ASR by using an Unscented Kalman filter (UKF). Since this parameter describes an internal resistance of the stack, it is directly associated with efficiency of power conversion. Therefore, the maximal loss of efficiency that defines RUL of the stack can be easily correlated with the terminal value of ASR.

To predict future evolution of SoH, an additional drift model is needed. It describes incremental increase in ASR over time. Its corresponding parameters can be evaluated on-line in order to track changes in trends. The identified model is then employed for SoH prediction. Finally, RUL is inferred by comparing the predicted SoH indicator with the EoL criterion, i.e. the maximal allowed ASR value at which power conversion becomes unsatisfactory.

The paper is organised as follows. In the second section a lumped SOFC stack model is briefly described. The third section describes SoH estimation, as well as SoH and RUL prediction. Experimental validation performed on a 10 kW SOFC stack is described in Section 4. Finally, the last section contains concluding remarks.

2. The stack model

Various models describing the dynamic behaviour of SOFC fuel cells are available in the literature [35,36]. Depending on the purpose, these models range from very detailed 3D dynamic models, e.g. [37,38], indispensable during the design stage of the system in question, to simpler ones describing only dominant dynamics. For our application, it is essential for the model to be capable of simulating voltage output of the loaded stack and thermal conditions at the anode outlet.

As discussed in the introduction, the degradation directly affects the internal resistance of the stack. Due to an increase in internal resistance, one can observe voltage drop and increase in stack outlet temperature. Therefore, our model needs to cover only the dominating thermodynamic and electrochemical phenomena.

The stack is modelled as a lumped parameter model with energy balance written as follows [39]:

$$K_s \frac{dT}{dt} = \dot{E}_{in}(T_{in}) - \dot{E}_{out}(T) - IU(T) \quad (1)$$

where T is the outlet gas temperature, $\dot{E}_{in}$ and $\dot{E}_{out}$ denote energy flows in and out of the stack, respectively, K_s denotes lumped heat capacity of the stack, U is stack voltage and I is load current. The inlet/outlet energy flows $\dot{E}$ are defined as [39]:

$$\dot{E}_\xi = \sum_i \dot{n}_i^\xi h_i(T_i^\xi) \quad (2)$$

where $\dot{n}_i^\xi$ denotes molar flow of the i th gas component, $i \in \{\text{H}_2, \text{H}_2\text{O}, \text{CH}_4, \text{CO}, \text{CO}_2, \text{O}_2, \text{N}_2\}$, at corresponding locations where $\xi \in \{\text{in}, \text{out}\}$, i.e. stack inlet/outlet, and with corresponding temperatures T_i^ξ , and h_i is enthalpy of the gas.

The steam methane reforming (SMR), water-gas shift (WGS) and hydrogen oxidation reactions are considered to be in thermodynamic equilibrium. This assumption is easily justified and often employed in the modelling of SOFC systems [39–43]. In [44] the authors studied the issue in deep for a particular real system and concluded that the equilibrium was reached during the operation. This result is of direct importance for us as in this paper we use the same experimental facility as in [44]. The reforming equilibrium is solved analytically assuming total methane reforming [45]. In the natural gas employed in this study the percentage of higher

hydrocarbons is very low, hence they are discarded from the model. In turn, molar flow $\dot{n}$ of the i^{th} gas component at the anode outlet $\dot{n}_i^{\text{out}}$ can be calculated from inlet molar flow $\dot{n}_i^{\text{in}}$ and the extent of SMR x , WGS y , and hydrogen oxidation reactions z :

$$\begin{aligned}\dot{n}_{\text{H}_2\text{O}}^{\text{out}} &= \dot{n}_{\text{H}_2\text{O}}^{\text{in}} - x - y + z \\ \dot{n}_{\text{H}_2}^{\text{out}} &= \dot{n}_{\text{H}_2}^{\text{in}} + 3x + y - z \\ \dot{n}_{\text{CH}_4}^{\text{out}} &= \dot{n}_{\text{CH}_4}^{\text{in}} - x \\ \dot{n}_{\text{CO}}^{\text{out}} &= \dot{n}_{\text{CO}}^{\text{in}} + x - y \\ \dot{n}_{\text{CO}_2}^{\text{out}} &= \dot{n}_{\text{CO}_2}^{\text{in}} + y\end{aligned}\quad \begin{aligned}\dot{n}_{\text{N}_2}^{\text{out}} &= \dot{n}_{\text{N}_2}^{\text{in}} \\ z &= \frac{N_0 I}{n_e F}, \\ K_{\text{WGS}}(T) &= \frac{\dot{n}_{\text{CO}_2}^{\text{out}} \cdot p_{\text{H}_2}^{\text{out}}}{\dot{n}_{\text{CO}}^{\text{out}} \cdot p_{\text{H}_2\text{O}}^{\text{out}}},\end{aligned}\quad (3)$$

where N_0 is number of cells, F is Faraday's constant, and n_e is number of free electrons. The temperature dependent WGS equilibrium constant is K_{WGS} :

$$\log K_{\text{WGS}} = AT^4 + BT^3 + CT^2 + DT + E. \quad (4)$$

The parameters A, B, C, D , and E are documented in Table 1 [46]. Furthermore, in K_{WGS} from (3) all the components are gases. Here we assume that concentration of each gas component is directly proportional to its flow rates. Moreover, it can be easily shown that the methane conversion rate at low pressures and high temperatures is nearly 100% [47], hence we can put $x = \dot{n}_{\text{CH}_4}^{\text{in}}$. Therefore, one can omit the SMR equilibrium from calculations. Finally, y is the only unknown and can be easily calculated from the K_{WGS} equation, see [45] for details. Note that usually there is some nitrogen present also on the fuel side. Ideally nitrogen does not react, hence $\dot{n}_{\text{N}_2}^{\text{out}} = \dot{n}_{\text{N}_2}^{\text{in}}$.

Nernst voltage of a single cell U_0 is evaluated by employing [48, p. 269]

$$U_0 = 1.2586 - 0.000252T + \frac{RT}{2F} \ln \frac{p_{\text{H}_2} p_{\text{O}_2}^{0.5}}{p_{\text{H}_2\text{O}}} \quad (5)$$

where R is the universal gas constant, p_{H_2} , p_{O_2} and $p_{\text{H}_2\text{O}}$ are hydrogen, oxygen and water vapour partial pressures inside the stack. The partial pressures of the gases inside the stack are approximated from the input/output partial pressures that can be easily evaluated from (3):

$$p_j = \frac{1}{2} \left(\frac{\dot{n}_j^{\text{in}}}{\dot{n}_{\text{tot}}^{\text{in}}} + \frac{\dot{n}_j^{\text{out}}}{\dot{n}_{\text{tot}}^{\text{out}}} \right)$$

where $j \in \{\text{H}_2, \text{H}_2\text{O}\}$ and $\dot{n}_{\text{tot}}^{\text{in/out}}$ are total anode inlet/outlet molar flows. Clearly, the oxygen partial pressure p_{O_2} is calculated at the cathode side in the same manner.

The temperature variation of lumped Ohmic ASR of a single cell can be expressed by the second-order Steinhart-Hart equation [48,40]:

$$r = \gamma \exp \left[\frac{30000}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right) \right] \quad (6)$$

where γ is Ohmic ASR at reference temperature T_0 .

The ASR model (6) is fairly simple. As shown in [49], the full ASR model is parameterised by several physical properties of materials, such as porosity and tortuosity. Assuming simple structure (6), the information regarding the different degradation phenomena is lumped into a single parameter, i.e. γ . Furthermore, the ASR is a function of other operating conditions such as gas mixture. However, in [50, p. 272] the approximation of an ASR by simple structure is elaborated. The approximation partially incorporates activation and concentration polarisations and holds for mild fluctuations in fuel utilisation. In this case, the temperature has dominant impact on the value of ASR as given in (6).

Table 1

Coefficients for temperature dependent estimation of equilibrium constants in (4) [46].

Const.	WGS
A	-2.6312×10^{-11}
B	1.2406×10^{-7}
C	-2.2523×10^{-4}
D	1.95027×10^{-1}
E	-66.139488

Finally, the stack voltage U can be expressed as

$$U = N_0 \left(U_0 - r \frac{I}{A} \right) \quad (7)$$

where A is the active area of a single cell.

The lumped dynamic model of the stack reads as follows:

$$\begin{aligned}K_s \frac{dT}{dt} &= \dot{E}_{\text{in}}(T_{\text{in}}) - \dot{E}_{\text{out}}(T) \\ &\quad - IN_0 \left(1.2586 - 0.000252T + \frac{RT}{2F} \ln \frac{p_{\text{H}_2} p_{\text{O}_2}^{0.5}}{p_{\text{H}_2\text{O}}} \right. \\ &\quad \left. - \gamma \exp \left[\frac{30000}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right) \right] \frac{I}{A} \right).\end{aligned}\quad (8)$$

3. State-of-health estimation and remaining useful life prediction

The evaluation of the remaining useful life consists of three steps: (i) SoH estimation, (ii) drift model identification and (iii) RUL prediction. The first step is achieved by employing the SOFC stack model (8) for non-linear state estimation described in the previous section. At the beginning of the operation, when the stack is new, $\gamma = \gamma_0$. With progressing degradation, γ starts to increase. To estimate the current value of γ from operating data, the UKF is applied. Secondly, we consider the reconstructed γ as a SoH indicator, and evaluate its future trend. To do so, one needs the model of drift in γ . Assuming linear structure of the drift model in γ , we apply another Kalman filter (KF) in order to estimate the parameters of the assigned model. Finally, RUL is predicted by employing the identified drift model of γ . The structure of the algorithm is shown in Fig. 1.

Note that the two filters (UKF and KF) could, in principle, be lumped in one single filter. However, the degradation phenomena on one hand, and the thermodynamics and electrochemistry on the other, run on different time scales. The degradation runs on the slowest time scale. Systems incorporating process dynamics of very different time scales are commonly known as *stiff systems*, and require sophisticated numerical solutions [51]. Since the focus of this paper is on SoH estimation and RUL prediction of SOFCs, the filter is split serially into two subsequent filters, whereas the first one operates on the fast scale and the second one on the slow scale. Prior to employing the estimated $\hat{\gamma}_k$ for drift model identification, the signal is appropriately downsampled and, therefore, the problems related to numerical instabilities are avoided.

3.1. Extended stack model for SoH estimation

The model (8) is discretized by employing Euler's method,

$$\frac{dT}{dt} \approx \frac{T(t + \Delta t) - T(t)}{\Delta t},$$

where Δt is a discretization step. Furthermore, the time evolution of γ in (6) is modelled as a time varying discrete stochastic process:

$$\gamma_{k+1} = \gamma_k + v_k \quad (9)$$

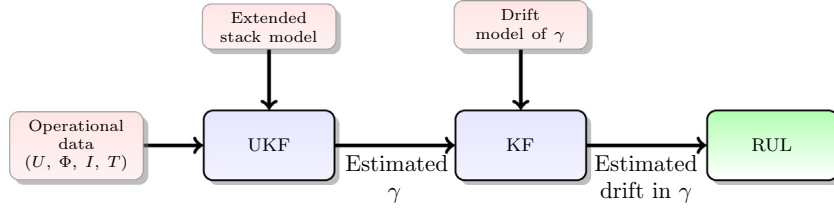


Fig. 1. Algorithm structure for SoH estimation and RUL prediction.

where v_k is zero mean normal random variable, where k denotes time step.

The model described in Section 2 is rewritten in state-space form with state vector $\mathbf{x}_k$,

$$\mathbf{x}_k = \begin{bmatrix} T_k \\ \gamma_k \end{bmatrix}, \quad (10)$$

where T_k is the outlet gas temperature from (8) and γ_k is the temperature invariant parameter of Ohmic ASR from (6). The model equations in state-space can then be written as

$$\begin{bmatrix} T_{k+1} \\ \gamma_{k+1} \end{bmatrix} = \begin{bmatrix} T_k + \frac{\Delta t}{K_s} \left(\dot{E}_{in,k} - \dot{E}_{out,k} - I_k N_0 \left(1.2586 - 0.000252 T_k + \frac{RT_k}{2F} \ln \frac{p_{H_2} p_{O_2}^{0.5}}{p_{H_2O}} - \gamma_k \exp \left[\frac{30,000}{R} \left(\frac{1}{T_k} - \frac{1}{T_0} \right) \right] \times \frac{I_k}{A} \right) \right) \\ \gamma_k \end{bmatrix} + \mathbf{v}_k \quad (11a)$$

and the output vector $\mathbf{y}_k$ is

$$\mathbf{y}_k = \begin{bmatrix} U_k \\ T_k \end{bmatrix} = \begin{bmatrix} N_0 \left(1.2586 - 0.000252 T_k + \frac{RT_k}{2F} \ln \frac{p_{H_2} p_{O_2}^{0.5}}{p_{H_2O}} - \gamma_k \exp \left[\frac{30,000}{R} \left(\frac{1}{T_k} - \frac{1}{T_0} \right) \right] \times \frac{I_k}{A} \right) \\ T_k \end{bmatrix} + \mathbf{w}_k. \quad (11b)$$

In (11), the variable $\mathbf{v}_k$ is used to describe model mismatch that arises due to modelling assumptions and simplifications. It is modelled as zero-mean normal random Gaussian process with, e.g. $\mathbf{v}_k \sim \mathcal{N}(\mathbf{0}, \mathbf{Q})$. Similarly, $\mathbf{w}_k$ models measurement noise as zero-mean normal random variable $\mathbf{w}_k \sim \mathcal{N}(\mathbf{0}, \mathbf{R})$.

The above model, written in compact mode, has the following state-space form:

$$\begin{aligned} \mathbf{x}_{k+1} &= \mathbf{f}(\mathbf{u}_k, \mathbf{x}_k) + \mathbf{v}_k \\ \mathbf{y}_k &= \mathbf{h}(\mathbf{u}_k, \mathbf{x}_k) + \mathbf{w}_k, \end{aligned} \quad (12)$$

where $\mathbf{u}_k$ is input vector, $\mathbf{f}(\cdot)$ is a set of non-linear state transition Eqs. (11a), $\mathbf{h}(\cdot)$ is a set of output Eqs. (11b), and $\mathbf{y}_k$ is output vector.

The UKF for estimating γ employs the model (12) and real measurements $\mathbf{y}_{m,k}$ on the system in order to update the state vector $\hat{\mathbf{x}}_{k|k}$:

$$\hat{\mathbf{x}}_{k|k} = \hat{\mathbf{x}}_{k|k-1} + \mathbf{Z}_k (\mathbf{y}_{m,k} - \hat{\mathbf{y}}_{k|k-1}) \quad (13)$$

where $\hat{\mathbf{x}}_{k|k-1}$ and $\hat{\mathbf{y}}_{k|k-1}$ denote the predicted state and output by the model, and $\hat{\mathbf{x}}_{k|k}$ is the corrected state updated by measurements, and $\mathbf{Z}_k$ is Kalman gain. In such a way, by combining the model and measurements, the UKF aims to correct the states of the model

to best match the measured data. Therefore, by combining the dynamic model of the stack and measurements on the real system, the UKF recursively identifies the value of γ . More details about the KF algorithm can be found in e.g. [52].

3.2. Identification of the drift model

For the purpose of drift model identification, we presume that degradation processes progress over time during system operation. The role of operating conditions have been quite extensively

researched and a number of evidences have been collected that clearly show that improper operational modes rapidly shorten

the life of the SOFC stack [53,54]. Furthermore, in this work we assume that the stack is not subjected to critical or damaging conditions that can lead to abrupt breakdown.

For our purpose, the following simple model of drift in γ is assumed

$$\gamma(t) = a_k t + b_k. \quad (14)$$

Note that the drift model (14) is selected purely on empirical analysis of data and the parsimonious principle, which prefers economic models to accomplish the desired level of prediction. Moreover, such a linear model can be viewed in the aspect of a Taylor series. Taylor expansion is a commonly employed method for linearisation of non-linear functions, and the first two terms are employed for function approximation. Therefore, it is always possible to approximate trajectory of γ by linear function with limited accuracy conditioned by properties of Taylor expansion. Nevertheless, for better understanding of such a model, and perhaps better results, more basic research related to SOFC degradation phenomena is required. More complex models can be found in the literature, e.g. [55].

Since the model (14) is linear, conventional KF can estimate its parameters with sufficient accuracy. In this case, parameter $\hat{\gamma}_k$,

here denoted “measurement” $\hat{\gamma}_{k,m}$, is the output of the UKF described in Section 3.1.

Rewriting the model (14) as a discrete state-space model with two states,

$$\mathbf{s}_k = \begin{bmatrix} \gamma_k \\ a_k \end{bmatrix},$$

results into

$$\mathbf{s}_{k+1} = \begin{bmatrix} 1 & 1 \\ 0 & 1 \end{bmatrix} \mathbf{s}_k + \boldsymbol{\varphi}_k \quad (15)$$

$$\gamma_k = [1 \quad 0] \mathbf{s}_k + r_k,$$

where a_k is an unknown gradient of linear function. Similarly to the previously described UKF, $\boldsymbol{\varphi}_k \sim \mathcal{N}(\mathbf{0}, \boldsymbol{\Phi})$ defines process noise, and $r_k \sim \mathcal{N}(0, R_\gamma)$ measurement noise. Note that the measurement variance R_γ is directly known from the output of the UKF.

Similarly to (13), the state vector $\mathbf{s}_k$ is *predicted* and *corrected* following the standard KF algorithm:

$$\hat{\mathbf{s}}_{k|k} = \hat{\mathbf{s}}_{k|k-1} + \mathbf{K}_k(\hat{\gamma}_{k,m} - \hat{\gamma}_{k|k-1}) \quad (16)$$

where $\hat{\mathbf{s}}_{k|k-1}$ and $\hat{\gamma}_{k|k-1}$ denote the predicted state and output by the model, and $\hat{\mathbf{s}}_{k|k}$ is the *corrected* state updated by “measurement” $\hat{\gamma}_{k,m}$, and $\mathbf{K}_k$ is Kalman gain.

One has to stress an important aspect of the model (15). Note that in the state-space form, the degradation rate, i.e. slope of the linear function a_k , is a modelled unknown time variant random process and is therefore capable of adjusting to time varying trends. For example, if the degradation rate increases, the stack begins to degrade faster, and the KF employing this model will adjust to the new rate and therefore still be able to predict RUL correctly.

3.3. Prediction of the remaining useful life

The concept of the RUL defines the remaining life-span of a component with the aim of minimising catastrophic failure events [56]. However, in the context of SOFC, we understand the notion of RUL as the time until the performance of the SOFC stack becomes unsatisfactory. The time moment when the SOFC is unable to perform adequately denotes its EoL.

Depending on the application and the end user, EoL criteria can differ. For instance, EoL can be defined as a drop in output voltage (e.g. 10%), a drop in power output, or drop in efficiency in power conversion. Given the nominal operating conditions and EoL criterion, γ_{RUL} can be easily calculated from (5)–(7). For instance, let $a \in [0 \ 1]$ define minimal nominal acceptable voltage output U_{RUL} ,

$$U_{RUL} = a \cdot U_{T0},$$

where U_{T0} is the nominal output voltage at $T = T_0$. Then, having the nominal operating conditions defined, γ_{RUL} can be calculated as

$$\gamma_{RUL} = \frac{N_0 U_0 - a U_{T0}}{N_0 \exp \left[\frac{30,000}{R} \left(\frac{1}{T} - \frac{1}{T_0} \right) \right] \frac{1}{A}} \quad (17)$$

where U_0 is Nernst potential (5) and T is nominal stack operating temperature. From here onwards, we understand RUL as time span until the γ reaches terminal value γ_{RUL} .

Having γ_{RUL} defined by EoL criterion, RUL can be calculated employing the drift model (15). At each time moment the KF from Section 3.2 outputs state estimate $\mathbf{s}_k$ with its corresponding covariance $\mathbf{P}_k$. Therefore, the outcome of the RUL prediction is uncertain as well, since it arises from the uncertain state estimation and uncertain future evolution of the ASR modelled by process noise $\boldsymbol{\varphi}_k$.

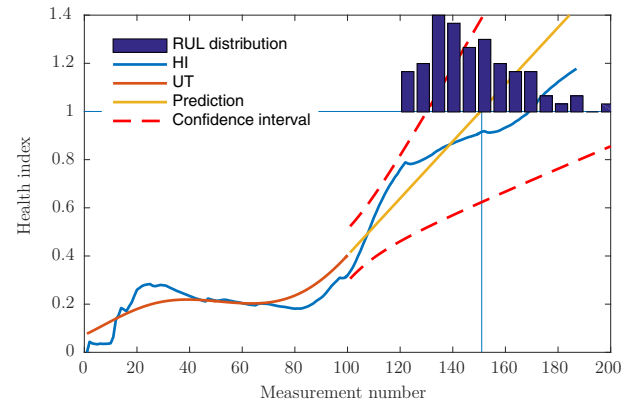


Fig. 2. Basic idea of RUL prediction: drift model in corresponding health index (HI) (—) is identified up to current measurement (e.g. 100). From that moment on, evolution of HI is predicted (—) with corresponding confidence interval (---). Having terminal value of HI (e.g. 1), RUL distribution can be evaluated.

The fundamental problem of finding an RUL distribution employing model (15) falls within the field of first passage time (FPT) probabilities, see e.g. [57]. Finding FPT distribution has been of interest to scientists in a variety of fields from microbiology [58] and ecology [59] to economics [60]. Given the degradation model (15), the FPT probability that γ will surpass the terminal value γ_{RUL} at prediction time step k , i.e. $P(T = k)$, reads

$$P(T = k) = P(\gamma_n \geq \gamma_{RUL}, \gamma_{n-1} < \gamma_{RUL}, \dots, \gamma_1 < \gamma_{RUL}).$$

The above probability can, in principle, be evaluated by calculating the so called orthant probabilities for each time step k [61]. However, by employing the solution described in [61], the dimension of orthant becomes larger and larger with growing k . Consequently, the computational time rises, rendering the approach inapplicable for our needs.

Due to the problems described above, the approximate RUL distribution was estimated via Monte Carlo (MC) simulations. To do so, a finite realisation of state variables $\mathbf{s}_k$ and noise variables $\boldsymbol{\varphi}_k$ was generated at each time moment k . Then, for each sample from the generated set, an open loop simulation of the model (15) was performed until the termination criterion was reached, i.e. γ_k reaches terminal value γ_{RUL} . The RUL is simply the number of recursions it took before the terminal criterion was reached. Fig. 2 demonstrates the basic idea of applying KF for RUL prediction.

4. Experimental evaluation

The experimental evaluation of the proposed approach was performed on an SOFC system shown in Fig. 3a, see [62,63]. The system is comprised of two modules: a stack and a balance of plant (BoP). The 80-cell stack is sealed in an insulating housing and connected to the BoP module. The system can generate up to 10 kW of electric power from a single 80-cell SOFC stack. The feasible operating range for the unit in question is defined by the following variables: an air flow rate in the range of 700–1600 l/min and a natural gas flow rate up to 35 l/min, while the maximum operating temperature is 850 °C at the exhausts and 800 °C at the inlets.

The system is instrumented with a number of sensors that provide measurements ranging from concentrations of gases to pressures, flows, and finally electrical measurements. The study below is performed on an excerpt from operational data gathered during a longer experiment. During this time, the system is operated at, and near, nominal operating conditions with planned step changes in stack current, fuel flow (fuel utilisation) and air flow (air utilisation) as a design of experiment type test which varied the

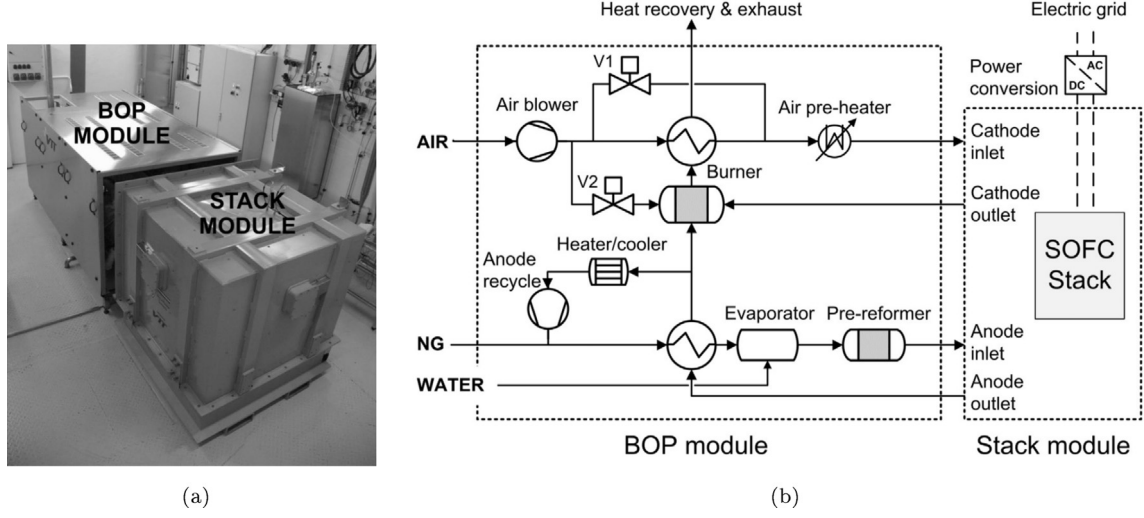


Fig. 3. The SOFC power generating unit installed at VTT (a), and a schematic representation (b) [62].

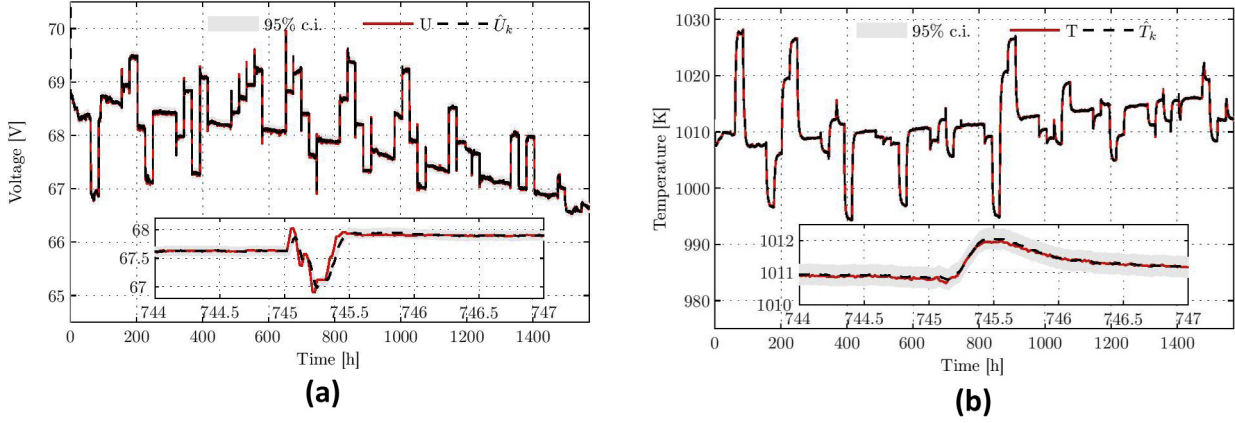


Fig. 4. Stack output measurements (U, T) and their corresponding UKF predicted values ($\hat{U}_k, \hat{T}_k$). One can note that UKF accurately estimates the states of the model, even under degradation phenomena occurring inside the stack. Detailed views on the figures demonstrate proper application of the UKF.

overall power output in the range of 15%. Moreover, during this experiment the stack underwent a considerable degradation, which makes the collected data ideal for our study. More details on the experiments can be found in [62,64]. Signals were sampled at a 1 min sampling rate.

Remark 1. Note that the terminal value of γ , i.e. γ_{RUL} , is simply its value at the end of our experimental session and is not associated with EoL criterion of the stack in any way. Moreover, performance assessment corresponds to a fraction (≈ 1500 h) of the true overall operation horizon of the stack. For validation purposes of the RUL algorithm, we consider the beginning of life at the start, and the end of life at the end of the experiment.

4.1. Estimation of the γ parameter

As explained in Section 3.1, an UKF that estimates γ was built. Process and measurement covariances, $\mathbf{Q}$ and $\mathbf{R}$, respectively, were heuristically estimated from measured data in the pre-processing stage:

$$\mathbf{R} = \begin{bmatrix} 0.1 & 0 \\ 0 & 0.01 \end{bmatrix}, \quad \text{and} \quad \mathbf{Q} = \begin{bmatrix} 0.01 & 0 \\ 0 & 10^{-6} \end{bmatrix}. \quad (18)$$

The process noise covariance $\mathbf{Q}$ defines variance of additive noise acting on state vector (10). Note that noise variance acting on the second state is relatively low with respect to the first state. In a way this could be interpreted as slow variation of γ over time, which is in accordance with slow degradation rate. A more systematic approach for estimation of matrices (18) can be found in, e.g. [11,65].

Fig. 4 shows measured and filtered outputs from (11b). More specifically, Fig. 4a shows the stack measured output voltage U , and the output of the UKF, $\hat{U}_k$ with corresponding 95% confidence interval (CI). Likewise, Fig. 4b shows cathode outlet temperature T , and corresponding filtered value $\hat{T}_k$, again accompanied with 95% CI. More importantly, from the figures it is easily seen that the UKF successfully estimates states of the model and handles the degradation drift occurring inside the stack. Detailed views in the figures demonstrate a very accurate and reliable performance of the UKF, i.e. the estimated mean values, denoted by $\hat{U}_k$ and $\hat{T}_k$, capture the dynamics of the measurement, while the noise is filtered out and enclosed within the corresponding CI.

The most important result of the designed UKF is the estimated state of ASR γ . In principle, γ is invariant to the operating conditions and is affected only by slow degradation phenomena, hence making it an ideal candidate as a SoH indicator. Fig. 5 shows the

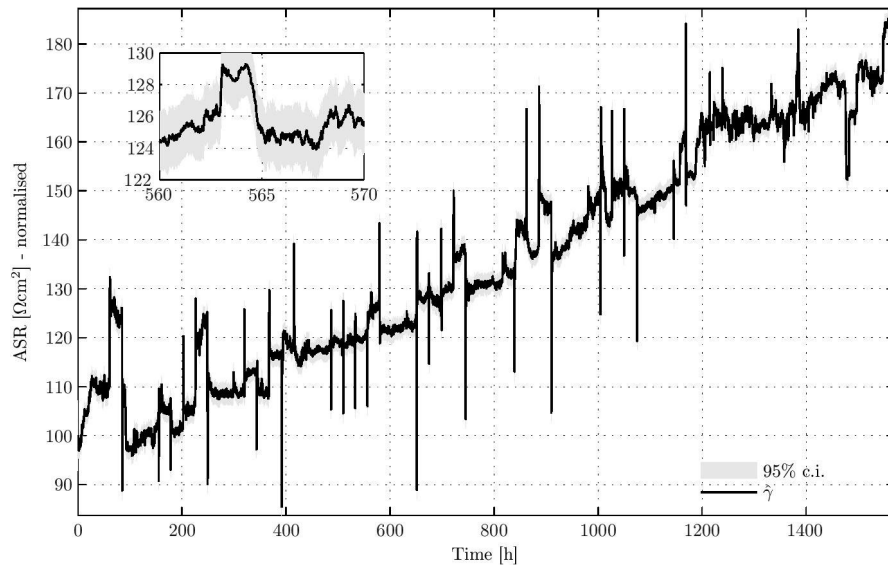


Fig. 5. Estimation of stacks ASR at reference temperature. It is apparent that the estimation $\hat{\gamma}$ encloses drift, which can be easily exploited for RUL predictions.

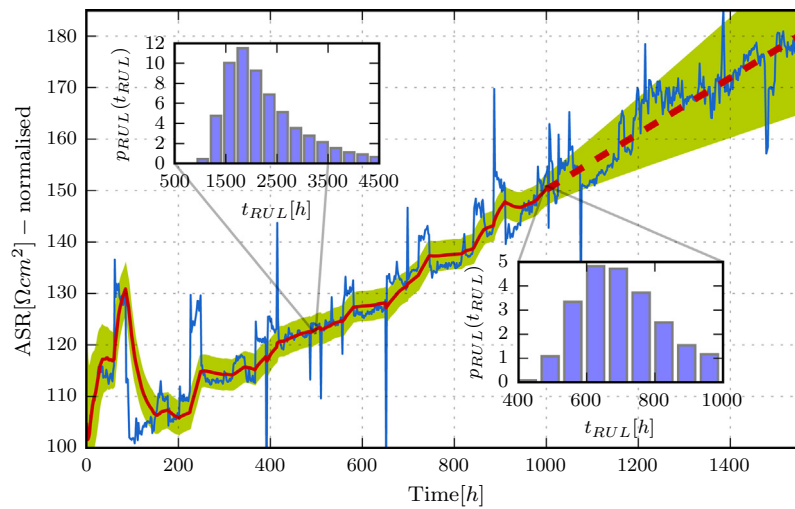


Fig. 6. Prediction of the increase in ASR: employing KF the trend and model parameters are identified. The plot shows output of UKF (—) overlapped by the prediction of the drift model in red (—), together with 95% CI at each time moment k . Dashed bold red curve (—) shows predicted value of γ at $t \approx 1100$ h. The detailed views show the distribution of RUL prediction, i.e. $p_{RUL}(t_{RUL})$. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

estimated value $\hat{\gamma}$ with the associated CI, that is the second state of the designed UKF. Evidently the estimated value encloses temporal drift, from which RUL will be inferred later on. From Fig. 5, one can observe that some dynamics are still present in the estimated value. It can be shown that the spikes and step responses are correlated with some of the input variables, for example current, fuel flow, etc. Clearly, these responses are a result of model and plant mismatch. However, the estimation error due to mismatch is low.

4.2. Identification of the drift model and RUL prediction

The following Figs. 6 and 7 show the results of γ and RUL predictions, respectively. RUL distribution is evaluated based on 10,000 MC simulation runs at each time step. The terminal value of γ_{RUL} corresponds to the value of γ at the end of the experiment.

Drift in γ is modelled by employing the model from Section 3.2 in KF algorithm with process covariance

$$\Phi = 10^{-10} \times \begin{bmatrix} 0 & 0 \\ 0 & 7 \end{bmatrix}.$$

Note that the drift parameter a_k belongs to state vector $\mathbf{s}_k$, so that its correct value is to be identified and is not considered constant. Note also how covariance Φ defines process noise acting only on the state that corresponds to the gradient parameter a_k of the drift model. In such a way, only the drift parameter of the model is corrected with respect to the measured value of γ .

Fig. 6 compares the output of the UKF (—), overlapped with the prediction of the drift model (—), together with 95% CI at each time moment k . The detailed views show the distribution of RUL prediction, i.e. $p_{RUL}(t_{RUL})$, at two selected time instances, i.e. $t \approx 500$ and $t \approx 1100$. Up to $t \approx 1100$ h, the KF from Section 3.2 is employed in order to filter out the trend while simultaneously identifying model parameters. The identified model is then employed to predict the trend of γ until the end of the experiment, as marked by the dashed line. Note that the variance of the prediction increases with increasing prediction horizon. In the case when the current state of the SOFC is far away from its terminal condition, such as the case shown in detail in Fig. 6 at $t \approx 500$ h, our model predicts γ with great uncertainty. Consequently, the RUL

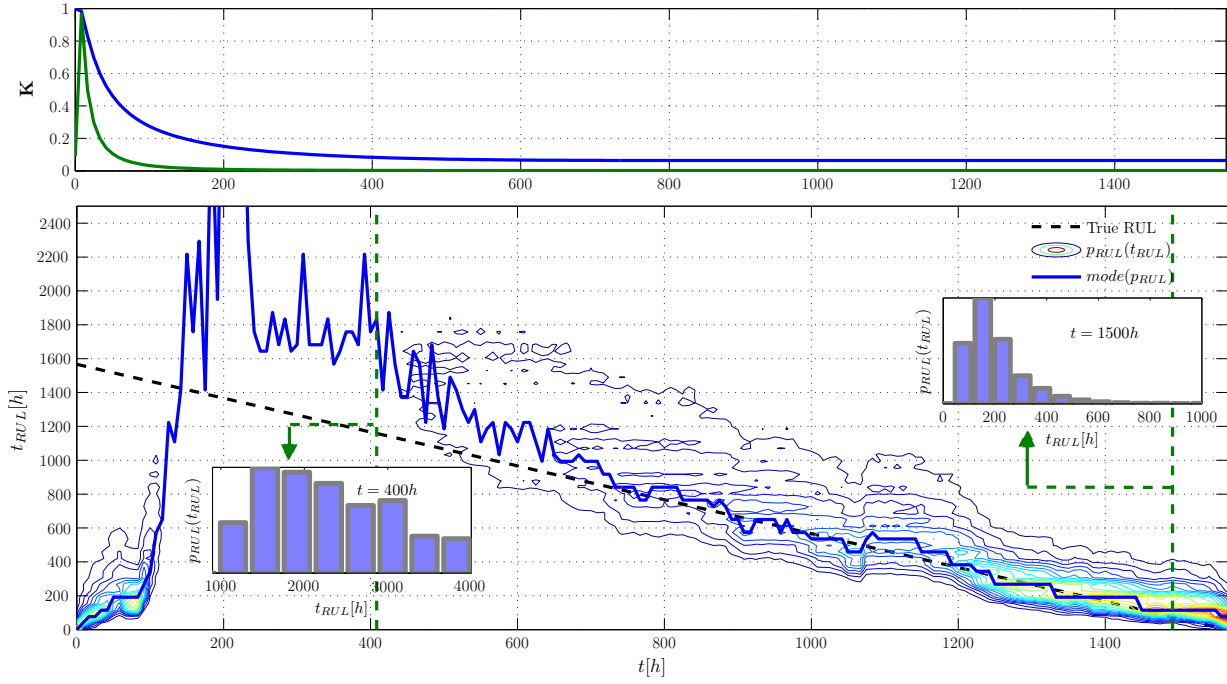


Fig. 7. RUL predictions for the entire duration of the experiment. The contour plot contains three elements: (i) true RUL (black dashed), (ii) RUL distribution at time moment t with support $t_{RUL} \in \{1, 2, \dots\}$, and (iii) mode of the corresponding RUL distribution marking the most probable t_{RUL} . The upper plot on the Fig. shows values of Kalman gains K_k .

distribution $p_{RUL}(t_{RUL})$ has a rather high uncertainty, as predicted RUL values fall within range from 1500 h to 4500 h.

As the time runs, the stack deteriorates further and γ increases, hence the drift model predicts end of life within a shorter prediction interval. Consequently, prediction becomes more and more accurate, as shown in the second detailed view in Fig. 6, close to $t \approx 1100$ h. In this case, the model becomes more confident and predictions are more precise, as shown by RUL distribution $p_{RUL}(t_{RUL})$. In this specific case, the model predicts approximately 600 h of RUL, which corresponds to the end of our experiment.

Fig. 7 shows RUL predictions for the entire duration of the experiment. The contour plot contains three elements: (i) true RUL, (ii) RUL distribution for each time moment k with support t_{RUL} , and (iii) mode of the corresponding RUL distribution.¹ The upper plot on the Fig. 7 shows values of Kalman gains K_k . The detailed views in Fig. 7 show RUL prediction at two particular time instances. At $t \approx 400$ h, the drift model parameters are highly uncertain, and so is the RUL prediction. This can be observed by the lack of contour lines, which is due to the fact that the distribution evaluated at $t = 400$ h is so flat that its values are lower than values of plotted contour lines. Conversely, at $t \approx 1500$ h, the KF has converged, and drift parameters are significantly less uncertain. Consequently, the RUL prediction distribution is narrower, as shown in the detailed view and depicted by coloured contour lines. Note that the True RUL depicted at running time instance t is simply

$$\text{True RUL} = T_{\text{END}} - t,$$

where T_{END} is the time marking the EoL (in our case EoL is the end of experiment, i.e. ~ 1500 h).

Fig. 6, and particularly Fig. 7, deserve some additional comments. At the beginning of the experiment (roughly the first 200 h), the parameter γ exhibits a sort of non-monotonous behaviour. In particular, during the first ≈ 100 h it increases, during the subsequent ≈ 100 h it decreases, and then, after 200 h, starts increasing again. Such a performance can hardly be explained by physical arguments. Instead, one has to seek for explanation in the fact that the process

model (11) is not perfect but contains some modelling errors. These errors are best seen as spikes during abrupt changes in electrical load. Modelling error is reckoned to be the reason for the non-typical time evolution of the estimated γ during the first 200 h. This has a significant impact on the initial estimates of the RUL, which look unrealistic. Namely, the second KF evaluates the trend parameters of the γ model (14). One should recall that the KF requires some time to stabilise the estimates, or more precisely, to bring the reconstruction error in γ to the level defined by noise (ideally to zero). The transient in KF response is affected by the evolution of the parameter γ so that a certain amount of time and new data is needed to smooth out the influence of the non-monotonic initial section in the γ time series on calculated trend. That is why at the beginning the estimated trend is also non-monotonous. The consequence is a completely unrealistic prediction of RUL. After cca. 200 h the trend parameters commence indicating a stable trend and hence a rather accurate RUL distribution. To evaluate whether the KF has converged, one can observe the behaviour of Kalman gain. From the K_k values given in Fig. 7, it is clear that the KF converged within the initial 200 h. In summary, for the reasons above one has to ignore the initial response of the RUL estimator.

5. Conclusion

The paper presents a new approach to the evaluation of degradation in a SOFC stack, which provides accurate and reliable estimates of future degradation trends and the remaining useful life. The important property of the proposed algorithm is that it operates irrespective of time varying operating conditions. This brings important practical implications as no special operating requirements are requested, which is not the case with the existing algorithms, and no intrusion in the process operation is needed. Furthermore, the algorithms are computationally simple and ideal for implementation on a low cost control system. Note that performing 10,000 MC simulations is highly excessive, as they were only employed in order to obtain highly accurate approximations of RUL distribution. Fewer MC simulations are required to obtain the required information for practical use.

¹ Mode of the distribution $p_{RUL}(t_{RUL})$ refers to t_{RUL}^* for which $p_{RUL}(t_{RUL}^*) \geq p_{RUL}(t_{RUL})$.

The major novelties of the paper are threefold:

- (i) a SoH indicator based on estimated ASR of the stack is proposed;
- (ii) end of life of the stack is associated with the terminal value of the ASR (which is in turn related to the efficiency of the energy conversion);
- (iii) RUL predictions are based on an adaptive data-driven degradation model, which is capable of adjusting to new conditions, i.e. faster or slower degradation.

From the experimental evaluation of the proposed approach, four major conclusions can be drawn:

- (i) RUL prediction is heavily influenced by the quality of the SoH indicator. Clearly, the better the process model that is employed, the lesser the impact of non-modelled dynamics on the corresponding estimation.
- (ii) The proposed degradation models are capable of adjusting to a change in degradation rate, i.e. model parameters are corrected to the current trend in SoH. However, this property is twofold: if the quality of the SoH estimation is low, RUL prediction may be misleading, as in the early hours of the experiment.
- (iii) Special care should be devoted to interpretation of RUL predictions. In the beginning, when degradation is not yet evident and the KF is yet to converge, the drift model is unreliable. Therefore, the predictions should be taken with some caution.
- (iv) The approach is invariant to the varying operating conditions as it is validated on real experimental data of a loaded SOFC system under varying operation.

There are a number of questions that remain to be addressed. Firstly, the noise covariance estimation should be refined. Secondly, the degradation model can be enhanced by incorporating more degradation physics. Then, RUL distribution, which is now evaluated via MC, should be revisited and a potential (approximative) analytic or numerical solution should be sought. Finally, the basic model of the SOFC stack could be updated in order to achieve more accurate estimation of the γ .

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Appendix A

The appendix illustrates the general structure of the Kalman filter, in particular how its Kalman gain K_k in (16) is obtained. Note that the notation of the matrices are generic and not to be directly associated to the ones as above. Let the linear state-space model of a system be

$$\mathbf{x}_{k+1} = \mathbf{A}\mathbf{x}_k + \mathbf{B}\mathbf{u}_k + \mathbf{w}_k \quad (\text{A.1})$$

$$\mathbf{y}_k = \mathbf{C}\mathbf{x}_k + \mathbf{D}\mathbf{u}_k + \mathbf{v}_k \quad (\text{A.2})$$

where $\mathbf{x}_k$ is the state vector, $\mathbf{u}_k$ is the input vector and $\mathbf{A}, \mathbf{B}, \mathbf{C}, \mathbf{D}$ are matrices of appropriate sizes. The additive components $\mathbf{w}_k$ and $\mathbf{v}_k$ are zero mean Gaussian noises with covariance matrices $\mathbf{Q}$ and $\mathbf{R}$. In the equations, k denotes time instance.

The Kalman filter is an algorithm which employs both measurements and the system model output to estimate the model states $\mathbf{x}_k$. To do so, one can distinguish between two phases, *prediction* and *update*.

A.1. Prediction

At arbitrary time instance k , the state $\hat{\mathbf{x}}_{k|k-1}$ and prediction covariance $\mathbf{P}_{k|k-1}$ are predicted employing the model and estimations from precious time instance $k-1$ denoted by subscript $\bullet_{k-1|k-1}$:

$$\hat{\mathbf{x}}_{k|k-1} = \mathbf{A}\hat{\mathbf{x}}_{k-1|k-1} + \mathbf{B}\mathbf{u}_k \quad (\text{A.3a})$$

$$\mathbf{P}_{k|k-1} = \mathbf{A}\mathbf{P}_{k-1|k-1}\mathbf{A}^T + \mathbf{Q}. \quad (\text{A.3b})$$

A.2. Update

In the update phase, the innovation $\mathbf{e}_k$ (residual between measurement $\mathbf{y}_{k,m}$ and model prediction $\mathbf{C}\hat{\mathbf{x}}_{k|k-1}$) is employed to correct the state from the *prediction* stage employing the following steps:

$$\mathbf{e}_k = \mathbf{y}_{k,m} - \mathbf{C}\hat{\mathbf{x}}_{k|k-1} \quad (\text{A.4a})$$

$$\mathbf{S}_k = \mathbf{A}\mathbf{P}_{k|k-1}\mathbf{A}^T + \mathbf{R} \quad (\text{A.4b})$$

$$\mathbf{K}_k = \mathbf{P}_{k|k-1}\mathbf{A}^T\mathbf{S}_k^{-1} \quad (\text{A.4c})$$

$$\mathbf{P}_{k|k} = (\mathbf{I} - \mathbf{K}_k\mathbf{A})\mathbf{P}_{k|k-1} \quad (\text{A.4d})$$

$$\hat{\mathbf{x}}_{k|k} = \hat{\mathbf{x}}_{k|k-1} + \mathbf{K}_k\mathbf{e}_k. \quad (\text{A.4e})$$

In the above equations, $\mathbf{S}_k$ is called the innovation covariance, and $\mathbf{K}_k$ is optimal Kalman gain. The optimally estimated states of the system are calculated according to (A.4e). More information about the KF, and its non-linear counterpart, the UKF, can be found in [52].

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Chapter 5

Health Aware Control and Performance Optimisation of SOFC Systems

5.1 Introduction

Assuring optimal operating conditions even in the case of changing operating regimes and health conditions is recognized as an important integrative part of the future SOFC systems.

The problem of basic control of fuel cell systems has been quite well elaborated in the literature. Various low-level control structures for SOFC systems are available. A feedforward-feedback control strategy for load following was proposed in [107]. Simulations indicate that the proposed control strategy performs only limited load following while maintaining the burner temperature and fuel utilisation within a safe operational range. Similarly, Carré, Brandenburger, Friede, *et al.* [108] proposed a feedforward control of a SOFC system with AOGR configuration. The experimental validation shows that secure operating conditions as well as maximization of the electrical efficiency is achieved. However, without proper feedback control, larger variations in stack temperatures are observed. Gaynor, Mueller, Jabbari, *et al.* [109] developed controllers that prevent fuel starvation in SOFC systems. It is shown by means of simulation that fuel starvation can be prevented by using rate limiters and reference governors when the load current changes rapidly. On the other hand, Sorrentino and Pianese [110] studied the problem of proper transition between cold start and warmed-up phases. Sendjaja and Kariwala [111] describe decentralized proportional-integral (PI) control for stack voltage and fuel utilisation. The paper concludes that a systematically designed decentralized controller can match or even outperform the performance of advanced control strategies, such as MPC. A control system for a SOFC based hybrid plant, with a tubular SOFC stack and microturbine, is presented in [112]. A combination of feed-forward and feedback PI controllers is shown to keep the stack gradient as well as anode/cathode pressure difference constrained. Considerable attention has been focused also on the design of MPC controllers. For instance, Jacobsen, Spivey, and Hedengren [113] demonstrate successful load following by employing MPC. Similar applications of the MPC can be found [76], [77], [114], [115], where different types of data-driven models are employed. Although effective, the MPCs suffer from one major drawback. They rely on the accuracy of the model employed in the controller design. However, if the system changes, as is the case for the SOFCs when the fuel cells degrade, the predictions of the model become inaccurate, hence the control becomes ineffective.

Several options for optimising the performance of the SOFC system are explored in the

literature. In these studies, the SOFC systems are normally placed within a wider context such as co-generation or smart grid, e.g. [116]. Marchetti, Gopalakrishnan, Chachuat, *et al.* [78] describe a supervisory controller which combines the real-time optimization of electrical efficiency and MPC. The approach is developed and verified on a dynamic model of the SOFC power system. Simulation results showed that near-optimality can be obtained and constraints can be respected despite model inaccuracies and large variations in load demand. The experimental validation of the proposed approach was also performed [117]. It was shown that the real time optimizer, which applies the static model of the system, was able to converge quickly and safely towards the optimum efficiency. Cheng, Jing, Xu, *et al.* [118] employ a feed-forward (FF)-feedback control system, similarly to [107]. The references for these controllers, however, are chosen by employing the results of the optimisation problem carried out off-line. The proposed controller is able to keep the temperatures within the safe range while guaranteeing high efficiency in steady operation as well as during load switching transients. Conversely, a dynamic model of the 1.5 kW SOFC power system was employed in search for optimal operating points for maximum steady state system efficiency. Jiang, Li, and Li [119] employed model-based analysis to provide the optimal open-loop for the system. Similarly, a model-based optimisation approach was applied on the model of a 5 kW SOFC system to find optimal operating points under fluctuating load. The authors of [120] then used the generated data to design static FF maps to maximise system efficiency under varying load conditions. With a similar control objective in mind, Li, Wu, and Zhu [121] developed a hierarchical load tracking control of a grid-connected SOFC power system. The conditions that satisfy maximal efficiency were sought via a non-linear optimisation program.

5.2 Problem Statement

This chapter proposes an upgrade to the low-level controller with a supervisory controller (SC). Generally, the set-points for the low-level controller are arbitrary values within pre-defined boundaries. They are usually defined by the functional limitations of the system. For instance, the maximal stack temperature is prescribed by the manufacturer to be at ca. 1100 K. Another example is the lowest operating temperature that has to be kept above, e.g., 900 K to facilitate internal reforming of the fuel as well as to keep the cell voltage above a certain limit. Yet, the optimal operating point of the system depends not only on the stack itself, but also on its corresponding BoP module. Therefore, finding the optimal operating point of the system, with respect to some criterion, can considerably improve the overall operation of the stack. The SC has the task of adjusting the reference values of the low level controllers. To do so, a criterion function has to be minimised while taking into account system constraints. The criterion function consists of two terms. One refers to the system efficiency, the other refers to the durability.

The actions of the SC are formulated as an optimisation problem, employing the concept of *extremum seeking* [122], where performance optimisation is performed directly on the system itself, thus rendering the need for a precise process model unnecessary. The controller design also takes into account practical aspects of implementation, meaning that all the developed algorithms should be easily implemented on conventional low-cost programmable logic controllers (PLCs). The ideas are first evaluated by means of simulation. The simulation model of the entire process in the nominal condition is developed and implemented in Matlab. Then, a model of degradation, derived from a set of experiments in the laboratory is included in the system simulator.

The remainder of this chapter includes (i.) a short description of the SOFC system for which the control system was developed, (ii.) a brief outline of the corresponding math-

emational model in the nominal operating mode and in the degradation mode, (iii.) the developed control system, (iv.) simulation case studies, (v.) preliminary experimental validation.

5.3 SOFC System and its Corresponding Model

The control system design and validation is carried out on a model of a SOFC system thoroughly described in [93], [123]. The system consists of BoP and stack modules, as shown in Chapter 4, Figure 4.3.

The system model is realised in a state-space form

$$\begin{aligned}\dot{\mathbf{x}} &= \mathbf{f}(\mathbf{x}, \mathbf{u}) \\ \mathbf{y} &= \mathbf{h}(\mathbf{x}, \mathbf{u})\end{aligned}\tag{5.1}$$

where $\mathbf{x}$, $\mathbf{u}$, and $\mathbf{y}$ are state, input, and output vectors, respectively. The $\mathbf{f}(\bullet)$ and $\mathbf{h}(\bullet)$ represent sets of state-transition and output equations. In total there are seven states and ten inputs. The output vector consists of temperatures and molar flows at each component, and output voltage of the stack.

The following section describes the stack degradation model. The stack model itself is described in Chapter 4. Since the BoP module consists of relatively simple components whose models are widely covered in the literature, their description is omitted. The component models of a pre-reformer, heat exchangers, and blowers, used in our work, can be found in [6], [124].

5.3.1 The degradation model

There are a handful of SOFC degradation models available in the literature. For instance, Cayan, Pakalapati, Celik, *et al.* [125] studied contamination mechanisms of the Ni-YSZ anode operated on coal syngas. Additionally, the work reported in [126], [127] implements degradation phenomena dealing with interconnect corrosion, loss of ionic conductivity of the materials, nickel agglomeration in the anode, chromium contamination and formation of insulating phases in the cathode. These models, even though capable of simulating different types of degradation mechanisms, become too complex to be incorporated in control-orientated models.

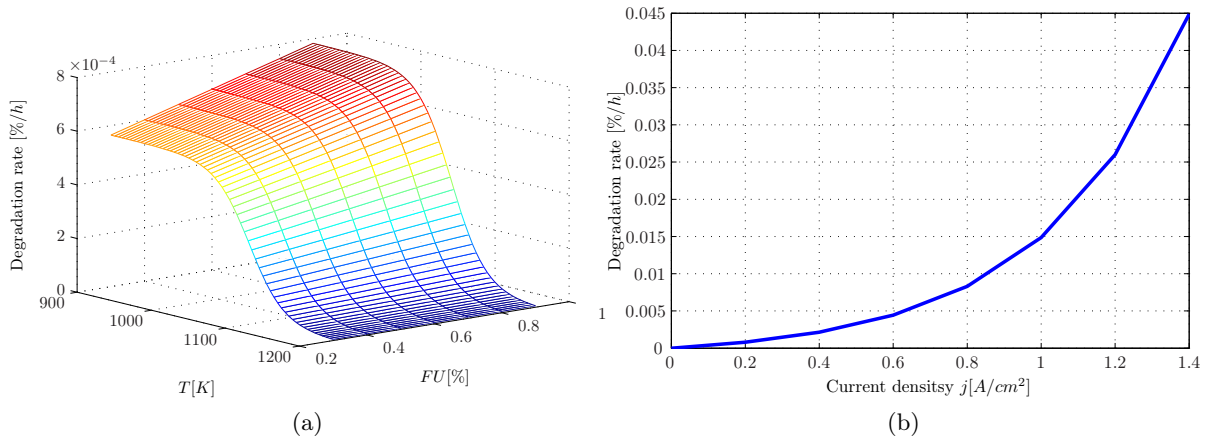


Figure 5.1: Analysis of degradation rate

An example of a degradation model was developed by Zaccaria, Tucker, and Traverso [60], [128], and also incorporated in our lumped stack model:

$$r_d = \frac{dR}{dt} = \frac{0.59FU + 0.74}{1 + e^{\left(\frac{T-1087}{22.92}\right)}} (e^{2.64j} - 1), \quad (5.2)$$

where FU is the stack's fuel utilisation, T is lumped stack temperature, and j is current density. The degradation rate r_d describes the evolution of Ohmic ASR, γ from equation (6) in Chapter 4, as a function of the operating parameters.

The data-driven degradation model (5.2) is not valid for all degradation modes. The model was identified in [129], by employing experimental data thoroughly described and analyzed by [38], [89]. The degradation model includes only a particular set of all possible degradation mechanisms. Nevertheless, it may be used for a more qualitative analysis of the long term operation of the SOFC system as in [130].

Figure 5.1 analyses the degradation model (5.2). In particular, Figure 5.1a shows the degradation rate as a function of stack temperature T and fuel utilisation FU . Close analysis shows that the degradation rate is slower at lower FU . Moreover, the degradation rate drops with increasing temperatures, which is unusual. As discussed above, the degradation model was identified from experimental data, where a specific cathode degradation mechanism was dominant [89]. Furthermore, Figure 5.1b shows the degradation rate as a function of current density j . As expected, the degradation rate increases with higher current densities.

5.4 Control Objectives and Control System Architecture

Running a SOFC system takes rigorous compliance with physical and operational constraints. It should be noted that no particular application of the SOFC system is assumed so that the system is treated as a standalone system. In order to comply with constraints and guarantee safe operation of the system, the following control objectives are established:

- i.) Temperature gradients within the stack should be kept under control. That means the temperature should not be allowed to exceed some upper, yet tolerable bound.
- ii.) Load following should be guaranteed. That means that the control systems should promptly adjust the fuel and air flows in order to allow for possibly non-stationary load conditions imposed directly to the stack.
- iii.) Efficiency should be brought to the maximum.
- iv.) The degradation rate should potentially slow down provided the extended life of the stack is required.

It should be noted that some of the requirements, e.g. maximal efficiency and minimal degradation rate, can be conflicting. Also, actions to accomplish requirements i.) and ii.) are executed on the fast time scale, while the last two require a much longer time period.

A natural way to separate slow from fast processes is to introduce a hierarchy of functions. The proposed control system consists of four interconnected modules as shown in Figure 5.2. The purpose of the low-level controllers is to keep the system running at corresponding operating conditions, defined by the controller set-points. The soft sensors, described in Chapters 2 and 3 provide information on non-measurable process variables to the remaining three modules. Namely, the reconstructed temperatures can be used as controlled variables, while the condition monitoring module, described in Chapter 4, employs the estimations of gas flow components at the anode inlet. The condition monitoring

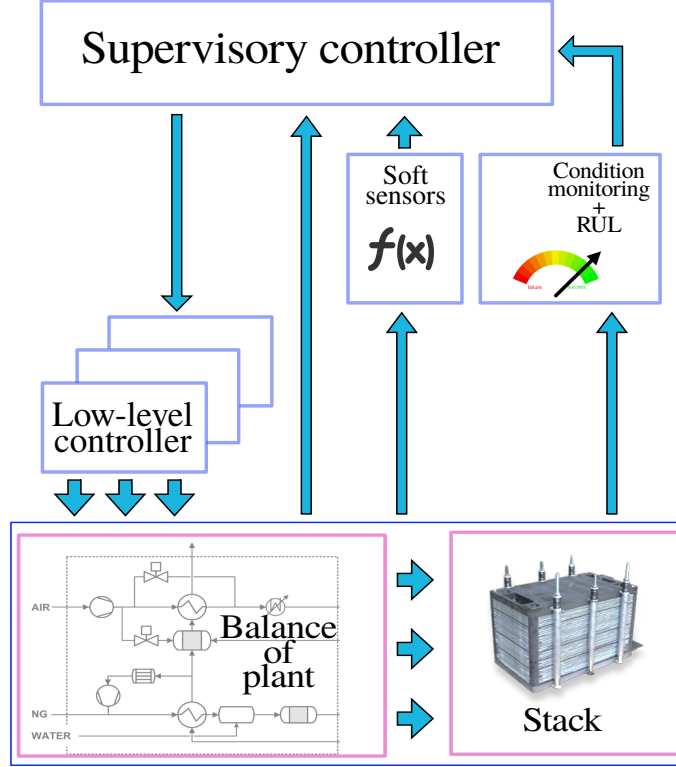


Figure 5.2: The control system architecture aimed to achieve optimal performance indicators.

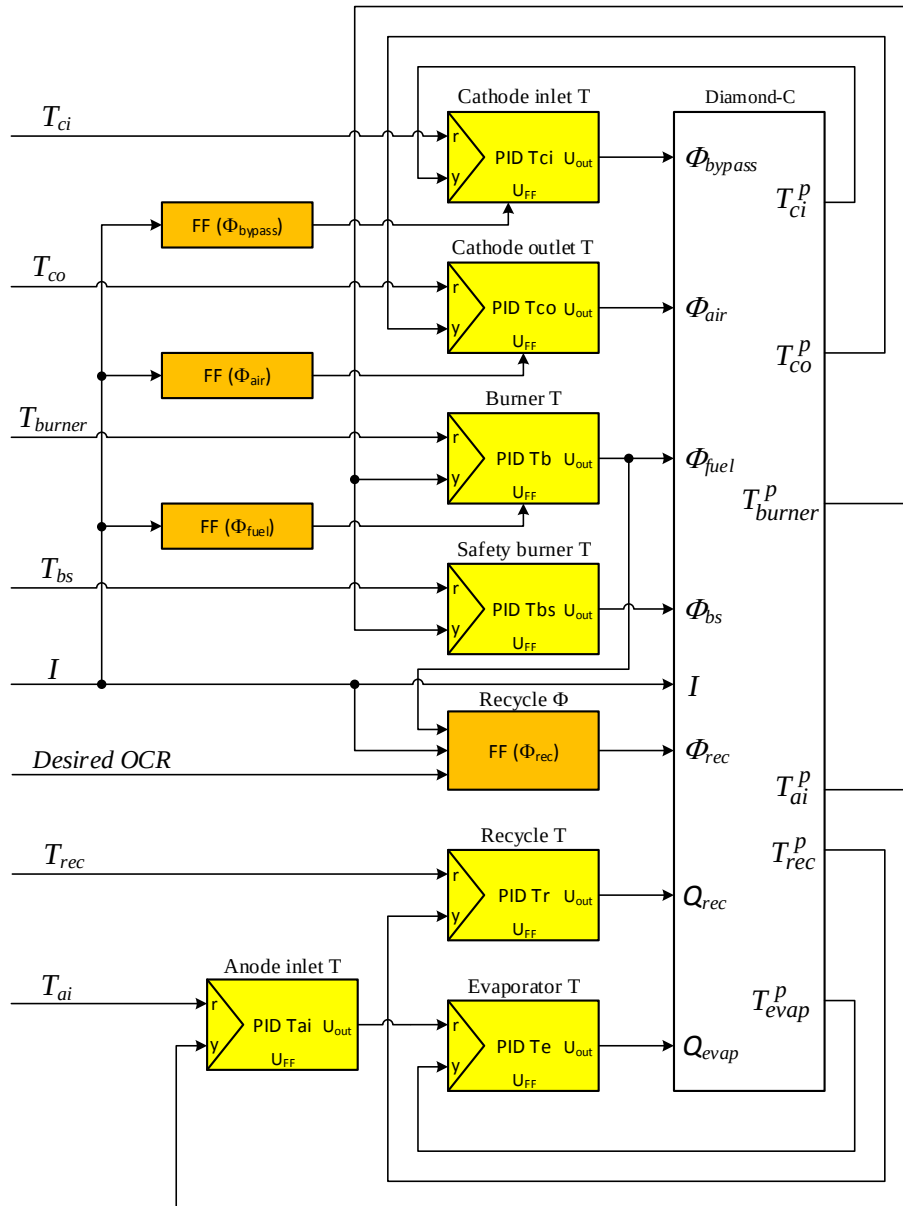
module provides information on the instantaneous degradation rate, i.e. a measure of stack performance deterioration. Finally, this information is fed to the supervisory controller, which calculates the set-points for the low-level controller by employing the information about performance degradation rate and the overall efficiency of the system.

Remark (Note on the system efficiency). The efficiency of the power conversion on the system level depends not only on the efficiency of the stack itself, but also on the power consumption of the BoP components. In order to maximise the electrical efficiency of the system, one needs to minimise the power consumption of the BoP module and maximise the fuel utilisation in the stack. The main electric power consumers are air and recycle blowers, followed by the heaters and coolers. Therefore, the net system efficiency will be achieved when the air and recycle flows are minimal. Another important note on the blower consumption is that the power consumption increases as a quadratic function of the required flow. Finally, the heaters/coolers are preferably turned off and only in use when necessary, so to minimise their consumption. The overall efficiency of the system is then calculated as

$$\varepsilon = \frac{P_s - P_{ab} - P_{rb} - P_{evap} - P_{rec} - P_{ah}}{LHV\Phi_f}, \quad (5.3)$$

where, referring to Figure 4.3, P_s is output power of the stack, P_{ab} and P_{rb} are air and recycle blower power demand respectively, and P_{evap} , P_{rec} , P_{ah} are evaporator, Heater/Cooler, and air pre-heater consumptions, respectively. The total energy fed to the system is calculated from flow of the natural gas Φ_f , where LHV is the lower heating value of the methane.

To effectively control the system variables under a non-stationary stack current, a combination of feedback and FF control was applied [131]. The FF part depends on the stack current and coarsely controls the main system variables, while the feedback control (under prescribed limitations) slightly corrects the FF actions in order to eliminate control error.



The purpose of this section is to briefly describe the low-level control structure shown in Figure 5.3. A more detailed description can be found in [132], while references [133], [134] describe the operating constraints of the system. Altogether, there are six main controlled variables:

- *Cathode outlet air temperature* T_{co} depends on many process variables, e.g. stack current, cathode inlet air temperature and flow, anode inlet temperature and flow, the ratio of reformed fuel in pre-reformer, etc. However, from all of the manipulative variables, *cathode inlet (air) flow* Φ_{air} is the one with the highest gain, and is therefore chosen as the control variable [131]. Here it should be noted that at lower stack currents, the endothermic reactions within the stack might decrease cathode outlet temperature below cathode inlet temperature. Such a situation may occur if the gross consumption of the heat, produced by the hydrogen oxidation, falls below the total energy demand required for steam reforming of the methane. In this case, the controlled process can reverse its gain and become uncontrollable. In order to prevent such a situation, the minimum stack current limit is set to 40A, the desired oxygen to carbon ratio is decreased at low currents, and the anode inlet temperature is kept above 720K by a controller described later on. The cathode outlet reference temperature is 1000K.
- *Cathode inlet air temperature* T_{ci} might be controlled by an air pre-heater or by an air by-pass valve. In order to increase efficiency and omit the use of an electric pre-heater, the *air by-pass valve* Φ_{bypass} has been chosen as the control variable. Furthermore, the use of the by-pass valve provides the most direct way of controlling the air inlet temperature. To do so, the valve opens and closes to add some cold air to the overheated inlet, which is heated up as it passes through the air heat exchanger. The reference temperature for the T_{co} is 70K lower than the cathode outlet air reference temperature T_{ci} , and increases with lower currents.
- The afterburner is used to heat-up the cathode inlet flow via the air heat exchanger. Its main source of energy is exhaust gas from the stack, therefore the control variable is *fuel flow* Φ_f to stack, and the controlled variable is *afterburner temperature* T_{burner} . An increase of afterburner temperature is achieved by increasing the fuel flow and vice versa. Since the afterburner heats-up the cathode inlet air, its reference temperature is chosen 120K higher than reference cathode inlet temperature.

There is another safety controller which controls the afterburner air valve V2, and prevents overheating of the catalytic burner by manipulating Φ_{bs} . The reference for this controller T_{bs} , i.e. maximal allowed burner temperature, is set at 1150K, and remains inactive in normal operating scenarios.

- *Anode recycle flow* Φ_{rec} is controlled by the FF controller, which manages the stoichiometric conditions inside the stack. Its main purpose, given the required load, is to keep the system in a theoretically carbon formation free operating point by ensuring sufficient OCR at the anode inlet. While providing the steam for methane reforming, the recycle flow also carries some additional energy in the form of heat, and thus helps raise the overall efficiency of the system.
- *Anode inlet temperature* T_{ai} should be kept above some minimum value (820K) which usually occurs at lower stack currents (40A). The temperature is controlled by the *evaporator temperature* T_{evap} . This is achieved by using cascade control where the inner control loop is controlling the evaporator temperature and the outer loop controls anode inlet temperature. Note that if the anode temperature becomes too low, the cathode outlet temperature might decrease severely, and the process might reverse its gain. During normal operation, for stack currents above 80A, the anode inlet temperatures are higher than 720K, thus the evaporator heater power is usually 0.
- The *evaporator outlet gas temperature* T_{evap} should be kept within some limits for the

next stage (pre-reformer). Evaporator outlet gas temperature can be increased by the built-in *electric heater* with power Q_{evap} . In order to increase the system efficiency, the heater should be used only when really needed. Therefore, the temperature limits between 800K and 980K have been chosen as an acceptable temperature range. Note that due to the implemented cascade structure, these limitations need to be recalculated into limitations of the outside loop [135].

- *Anode recycle temperature* T_{rec} is controlled only to satisfy the safety conditions of the measuring apparatus and the recycle blower that are installed in the recycle loop. To facilitate proper measurements and anode recirculation, the electrical Heater/Cooler with power Q_{rec} enables conditioning the gas passing through the pipes. The controller is active only if the temperature exceeds the corresponding upper limit.

Of the FF parts of the controllers, all but the recycle flow Φ_{rec} , are calculated with respect to current demand. Their main purpose is to speed-up the response of the control loop. Based on the measurable disturbance, i.e. current demand, the controllers first roughly adjust the corresponding actuator variable, which is then further controlled by the feedback part of the control loop. Furthermore, the controllers have an anti-windup system (due to limitations of the control variables) as well as manual/automatic control.

5.4.2 Supervisory controller

As discussed in the previous section, the objective of the low-level controller is to keep controlled variables at their reference values. However, the set of reference values, which is formally selected by the system operator, may be suboptimal with respect to the control objective. Furthermore, as the system deteriorates, the initial set at the beginning-of-life (BoL) of the system may become suboptimal. Therefore, a systematic way of adjusting the reference set is essential.

Table 5.1: The set-points of the low-level controllers. The super script p marks process output, otherwise the variable is reference or actuator signal.

Controlled variable	Manipulated variable	Set-point
Cathode air outlet temperature T_{co}^p	Air flow Φ_{air}	T_{co}
Cathode air inlet temperature T_{ci}^p	By-pass valve Φ_{bypass}	$T_{co} - \Delta T(I)$
Afterburner outlet temperature T_{burner}^p	Fuel flow Φ_{fuel}	$T_{ci} + 120K$
Anode gas inlet temperature T_{ai}^p	Evaporator temperature T_{evap}	720K (min)
Evaporator temperature T_{evap}^p	Electric heater Q_{evap}	Set by T_{ai}
Desired OCR	Recycle flow $\Phi_{recycle}$	2.2
Anode recycle temperature T_{rec}^p	Electric heater/cooler Q_{rec}	800K (max)

Table 5.1 lists the pairs of manipulated/controlled variables from Figure 5.3, and their corresponding set-points. It can be seen that the only two remaining *free variables* that define the operating point of the system are desired OCR and cathode outlet temperature T_{co} . The remaining set points are tightly connected with the latter. For instance, the T_{ci} is set by T_{co} and current I (through FF), so that the maximal temperature gradient over the stack is limited. Further, the afterburner temperature T_{burner} is defined as $T_{ci} + 120K$, so that the T_{ci} can be controlled by opening/closing the air bypass valve. To keep the system operating at high efficiency, the anode inlet controller is usually inactive, and only heats up the anode when necessary. Similarly, the T_{rec} is only controlled if needed, and is

inactive during normal operation. Finally, the OCR is set to a constant value, as low as possible, so that stack operates in carbon formation free operating point while keeping the power consumption of the recycle blower minimal.

Optimisation problem

The main objective of the supervisory controller is the optimisation of some steady-state operating performance of the system, while at the same time satisfying some constraints. The controller objective can be formulated as a nonlinear mathematical programming problem. In the general form it reads as follows:

$$\min_{\mathbf{x}} c(\mathbf{x}) \quad (5.4)$$

$$\mathbf{g}(\mathbf{x}) \leq 0, \quad (5.5)$$

where $c(\mathbf{x})$ and $\mathbf{g}(\mathbf{x})$ are non-linear functions, and $\mathbf{x}$ is argument of the functions.

Particular case

In the SOFC system, the argument $\mathbf{x}$ is a variable set that consists of different process variables, e.g. FU, stack current, stack temperature T_{co} , OCR, anode/cathode pressure difference, etc.

The analysis of the performance degradation model described in Section 5.3.1 shows that the degradation rate depends mainly on three factors: (i.) current, (ii.) fuel utilisation, and (iii.) stack temperature. However, the impact of these three variables needs to be re-evaluated when the fuel cells operate as a component in a larger system. For instance, without power management strategy, the stack current can not be regarded as manipulative variable, but only as measurable disturbance. Or, due to the structure of the low-level controller, the fuel utilisation FU becomes a dependent variable. Furthermore, since our degradation model neglects the impact of OCR, while also having a negligible effect on system efficiency, the OCR is set constant. Therefore, the optimisation problem (5.7) in our specific case reads:

$$\begin{aligned} \min_{T_{co}} c(T_{co}) \\ T_{min} \leq T_{co} \leq T_{max} \\ OCR = 2.2, \end{aligned} \quad (5.6)$$

where T_{co} is approximate stack temperature, and $c(T_{co})$ is some cost function. The solution of the said problem needs to satisfy constraints, i.e. $T_{co} \in [T_{min} \ T_{max}]$, and $OCR = 2.2$. The cost function $c(T_{co})$ is defined so that either (i.) degradation rate is minimal, (ii.) power conversion efficiency is maximal, or (iii.) the compromise between the two is achieved:

$$c(T_{co}) = \omega_1 r_d(T_{co}) + \omega_2 (1 - \varepsilon(T_{co})), \quad (5.7)$$

where $r_d(T_{co})$ is the degradation rate of the SOFCs at the given stack temperature T_{co} , and $\varepsilon(T_{co})$ is the efficiency, again at the given stack temperature T_{co} . The weights ω_1 , ω_2 are arbitrary. By adjusting their values, one can modify the cost function to put more emphasis either on minimisation of the degradation rate, or maximisation of efficiency, respectively.

While the $\varepsilon(T_{co})$ can be easily calculated from process variables, i.e. flow of the methane to the system, power consumption of blowers and heaters, and power output of the stack, see Eq. (5.3), the degradation rate r_d has to be obtained otherwise. In our work, the $r_d(T_{co})$ is estimated following the approach described in Chapter 4.

Extremum seeking control. The optimization problem (5.6) is solved on the basis of well-known *extremum seeking* control [122]. Motivation behind the selection of this approach is simple. The conventional approaches solve the optimisation problem on a system model and apply the solution to the real system. However, if the model of the system is inaccurate, then the solution yields sub-optimal performance. Moreover, the system is prone to change due to various degradation phenomena. Since these phenomena usually take place in the porous microstructure of the electrodes, the simplified control-oriented models become insufficient for simulating the behavior of the system in such detail. To make the matter even more complicated, more than one phenomenon can take place at the same time, thus further complicating the implementation of model based optimisation. One of the main advantages of the extremum seeking approach is that no process model is required to optimise the system operation.

Algorithm 5.1: Optimisation procedure.

```

Initialization:
Set the sampling time of the controller  $\Delta t$ .
Set the amplitude of the step excitation  $\Delta T$ .
Set the initial step direction  $d$ .
while in operation do
    if  $t = k\Delta t$  then
        if step then
             $T_{co}(k) = T_{co}(k-1) + d\Delta T$ 
            step = 0
        else if  $c(k\Delta t) < c((k-1)\Delta t)$  then
             $T_{co}(k) = T_{co}(k-1) + d\Delta T$ 
            step = 1
        else
             $d = -d$  ;
             $T_{co}(k) = T_{co}(k-1) + 2d\Delta T$ 
            step = 1
        end
        Check the constraints:
        if  $T_{co}(k) > T_{MAX}$  then
             $T_{co}(k) = T_{MAX}$ 
        else if  $T_{co}(k) < T_{MIN}$  then
             $T_{co}(k) = T_{MIN}$ 
        end
    end
end

```

An instance of the algorithm, described in [136], [137], is proposed to perform the model-free optimization. Pseudo-code for the applied perturbation and observe (P&O) algorithm is illustrated in Algorithm 5.1. All the parameters are initialised at the beginning, where the sampling time of the controller Δt should be long enough (i.) for the system to reach the steady-state operation, and (ii.) so that the fuel cells undergo observable degradation that can be quantified by the SoH estimator. Similarly, the amplitude of the step excitation ΔT should be large enough to reflect on the efficiency as well as degradation rate. In the initial time instance, the algorithm calculates the value of the cost function $c(T_{co})$, and performs initial step change in the optimisation variable, i.e. cathode outlet T_{co} in direction d with amplitude ΔT . At the next time instance, the cost function is re-evaluated and

compared to its value from the previous time instance. If its value is decreased, then the algorithm continues with step changes in the same direction. Alternatively, the direction of search is reversed, and the reference is changed with an amplitude of two steps.

5.5 Evaluation of the Proposed Control System

A major part of the evaluation of the proposed control system is carried out within a simulation environment. The simulations are described and the results are discussed in this section.

Note that the simplicity of the employed degradation model (5.2) does not hinder the overall assessment of the proposed control strategy. To be more specific, the SC calculates the operating point of the system solely by employing information provided by the condition monitoring module and efficiency of the system. The latter makes the proposed approach versatile as the controller adapts to current situations without exact prior knowledge. However, if the relevant information about the ongoing degradation mechanisms is available, the supervisory controller could be designed in a different, more efficient way.

We consider the following five different scenarios:

- $\mathcal{S}_1$ – steady-state operation without SC,
- $\mathcal{S}_2$ – steady-state operation where lifespan is optimised,
- $\mathcal{S}_3$ – operation with fluctuating load without SC,
- $\mathcal{S}_4$ – operation with fluctuating load where lifespan is optimised,
- $\mathcal{S}_5$ – steady-state operation with mixed optimisation criterion.

5.5.1 Analysis of the cost function on system level with implemented low-level controller

The analysis of the performance criteria on the system level with implemented low-level controller is necessary in order to understand the behavior of the supervisory controller. To this aim, the individual contributions to the criterion function (5.7) are evaluated for the only remaining manipulative variable that defines the operating point of the system, cathode outlet temperature T_{co} .

Figure 5.4 shows contributions of individual performance criteria from (5.7). The degradation rate $r_d(T_{co})$ and efficiency $\varepsilon(T_{co})$ are calculated for all the T_{co} within the pre-defined range, provided by stack manufacturer. The load current was kept constant, while the stack reference T_{co} was increasing in a linear manner from minimal to maximal allowed values. Note that enough time was left between two consecutive increases in T_{co} so that the transients behavior of the system was over and only steady-state values were obtained. Two facts are apparent from Figure 5.4:

- (i.) The efficiency of the power conversion with increasing reference temperature of the stack first increases and then decreases;
- (ii.) According to the degradation model (5.2), the degradation rate drops with increasing temperature.

The explanation of such behavior of the system is as follows. The stack heats up due to hydrogen oxidation and the reaction rate of this reaction is conditioned with stack current.

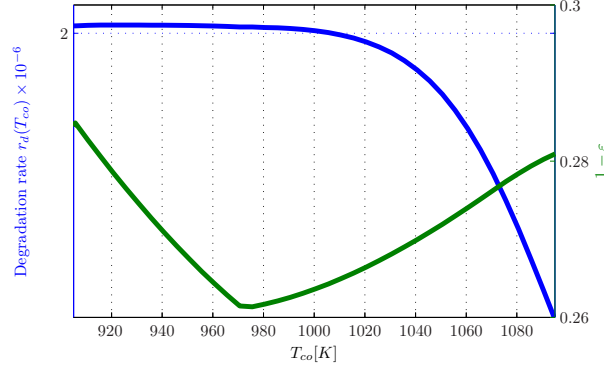


Figure 5.4: Analysis of performance criteria on the system level with implemented low-level controller. The degradation rate is estimated via the SoH algorithm elaborated in Chapter 4.

Hence, at a constant stack current, only a limited amount of heat is available within the stack. Therefore, in order to achieve lower or higher temperatures of the stack, two opposing actions are applicable: (i.) less air is fed to the system to cool it down, air blower consumption is reduced, and efficiency increases; (ii.) if heat production due to hydrogen oxidation is insufficient, more fuel is required to heat up the system, and efficiency drops. The analysis shows that the system efficiency can be improved by 2% by only selecting the proper operating point. The location of the minimum shifts left/right with different loads.

The degradation rate is estimated by employing the UKF described in Chapter 4. Therefore, the influence of different process variables in (5.2) are lumped into *one* parameter. For instance, in the temperature range $T_{co} \in (900, 970)K$, the degradation rate is dominated by FU , whereas at higher temperatures, the temperature dependent term dominates, see Figure 5.1. The effect of FU is mild on the system level and is barely observable.

5.5.2 Results and discussion

Table 5.2 sums up the basic information about the operating point of the system. During all the simulations, the values of these essential process variables were either (i.) constant or fluctuating with negligible amplitude (constant load), or (ii.) fluctuating according to required load, as to satisfy load following conditions. For the former case, the load profile, plotted in Figure 5.5, was based on the standard load profile of the residential houses [138].

Table 5.2: Basic data regarding the operating point of the system.

	Nominal value	Unit
Stack current	100	A
Fuel flow	15	l/min
Air flow	400	l/min
Cathode outlet temperature	1000	K
Anode inlet temperature	720	K

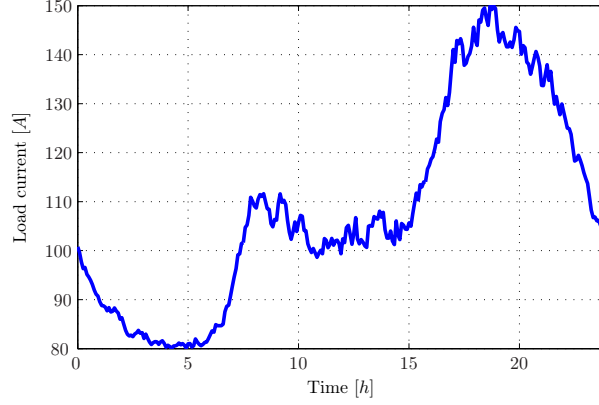


Figure 5.5: Daily profile of load demand.

5.5.2.1 The minimisation of the degradation rate

Constant operation

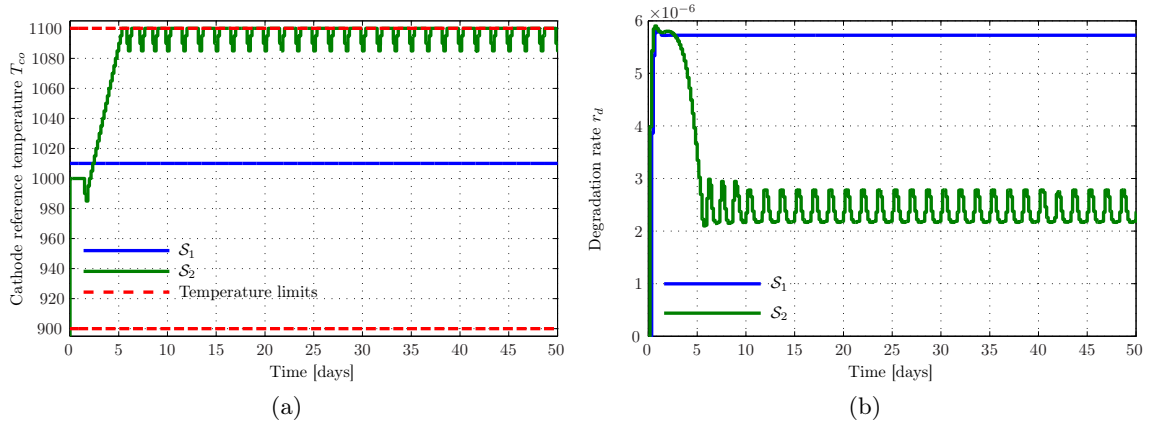


Figure 5.6: Analysis of SC performance with constant load conditions: the SC reduces the degradation rate by increasing the T_{co} . The SC continues perturbing the system when the optimum is reached.

Figure 5.6 compares system performance under two scenarios: with the system operating in constant operating point $\mathcal{S}_1$, and with the operating point defined by the SC $\mathcal{S}_2$. In both cases the stack current was kept at 130 A, while the stack temperature T_{co} was 1010 K for the case $\mathcal{S}_1$. The selected starting temperature for the SC was 1000 K, with a sampling time of $\Delta t = 4$ hours, and a step size of $\Delta T = 5K$. The total duration of the simulation considers 500 days of operation, however only the initial 50 days are shown, so that it is easier to read the figure. Figure 5.6a shows the cathode outlet temperature T_{co} for both cases. Observing the case $\mathcal{S}_2$, it is easily seen that the SC begins to increase the temperature and thus minimising the degradation rate, which is in accordance with Figure 5.4. The temperature settles around the maximal allowed operating temperature of the system, denoted here by red dashes. Furthermore, the Figure 5.6b shows the optimisation criterion for the duration of the simulation. In case $\mathcal{S}_1$, the degradation rate is constant, while it drops significantly in the case of $\mathcal{S}_2$. Another important aspect of

employing *extremum seeking* control is obvious from the figure. The system is constantly perturbed, even when the SC reaches the optimum. This can be observed in both the temperature as well as in the degradation rate, which fluctuate around optimum.

Varying load

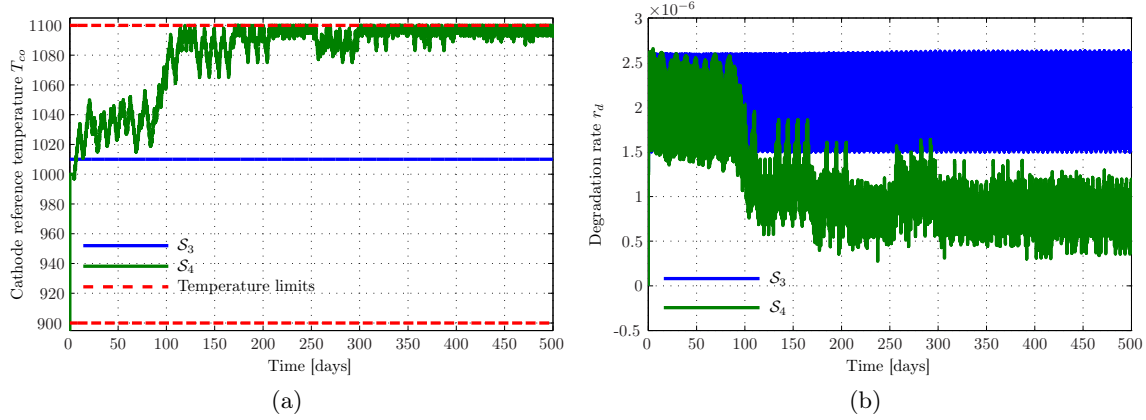


Figure 5.7: Analysis of SC performance with varying load: the SC optimises the performance of the system by minimising the degradation rate; however, it takes a longer time compared to $\mathcal{S}_2$ as it has no information about the load demand.

Figure 5.7 shows the performance of the SC under varying load. The parameters of the SC are the same as for the simulations $\mathcal{S}_1$ and $\mathcal{S}_2$. Similarly to $\mathcal{S}_1$, the temperature in $\mathcal{S}_3$ was kept constant at 1010 K. Figure 5.7a plots the reference temperatures for both scenarios. In the case of $\mathcal{S}_4$, the SC started to increase the system temperatures in order to minimise the degradation rate. However, the convergence rate of the optimiser is much slower, as the ever changing current during the day, and day after day, reflects in the degradation rate, thus becoming a disturbance in the optimisation criterion. Namely, as discussed in Section 5.3, the degradation rate depends not only on the variables that the controller has control over, but also on the disturbances, such as current. Therefore, the simple optimisation may sometimes be incapable of handling such a situation, and neglects this effect. Yet, from Figure 5.7a, it can be observed that the temperature set point reaches optimum, but only after cca. 100 days. The effect of the current disturbance on the degradation rate can be observed in Figure 5.7b. In simulation $\mathcal{S}_3$, the degradation rate follows the dynamics of the load current around a specific mean value, which is defined by mean stack current and constant temperature; while during the simulation $\mathcal{S}_4$, the SC gradually decreases the degradation rate to the minimum. The controller, however, has no control over the current demand, and therefore, the degradation rate continues to oscillate accordingly. Furthermore, it is intuitively clear that degradation rate dictates the speed of convergence. If degradation is fast, the health estimation algorithm requires less time to confidently estimate the impact of change in T_{co} on the degradation rate. Furthermore, the convergence rate could be improved by incorporating measurable disturbance, i.e. current, in the criterion function.

Practical implications

The above Figures 5.6 and 5.7 illustrate the performance of the SC from the control view point. In both cases, the SC reaches the optimal operating point which corresponds

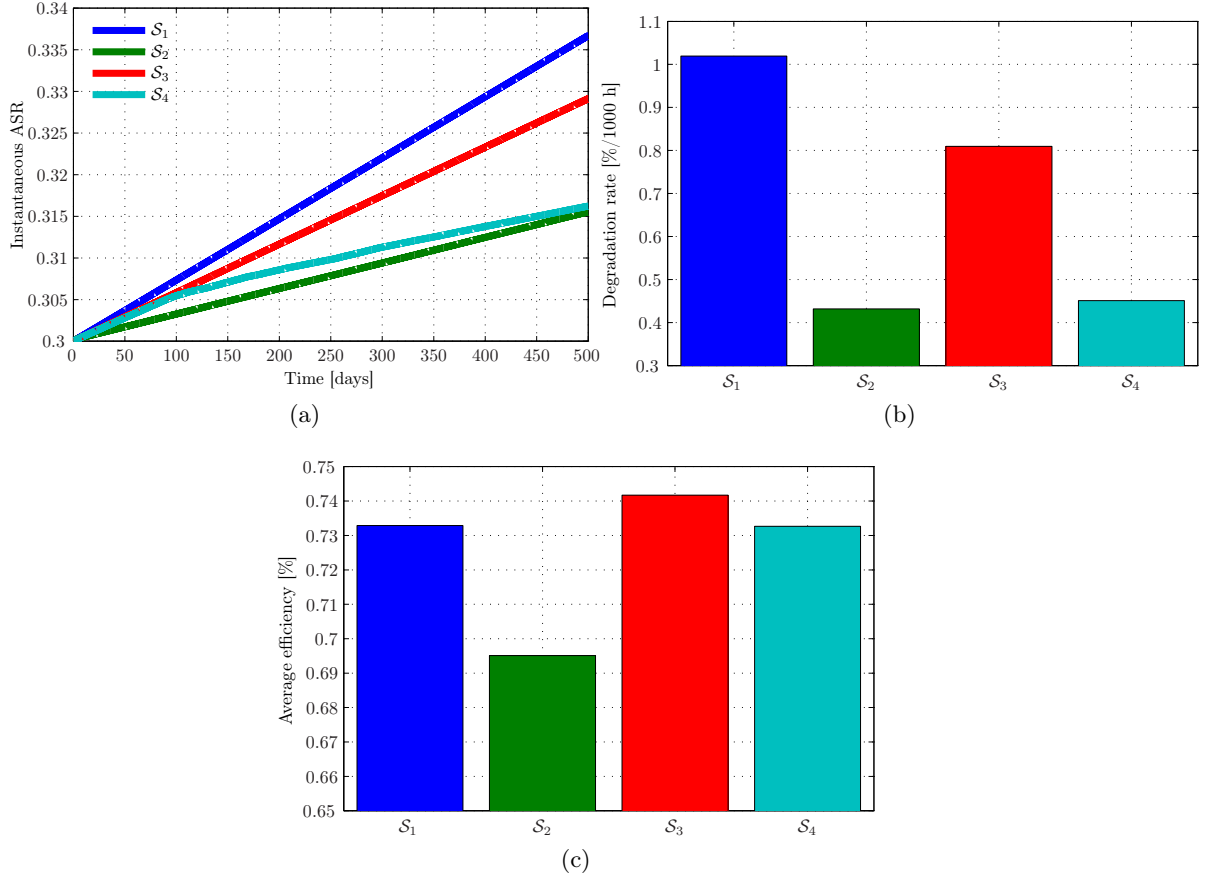


Figure 5.8: Analysis of system performance: (a) the evolution of stack ASR throughout the 500 days long simulations, (b) average degradation rate in %/1000 h, and (c) average system efficiency.

to the implemented degradation rate model. Even though the oscillating load current in scenario S_4 significantly hinders the performance of the SC, the optimum is still reached.

Figure 5.8 examines the implications of the applied control strategy from the end-user point of view. Figure 5.8a shows the evolution of ASR throughout all four simulation scenarios. In all the cases, at $t = 0$ the ASR was $0.3 \Omega cm^2$. It can be easily seen that the degradation was significantly faster in cases S_1 and S_3 , where the operating point of the system was kept constant at an inappropriate temperature. The degradation rate in Figure 5.8b displays the average degradation rate calculated from Figure 5.8a. When changing from S_1 to S_2 , the degradation rate dropped from 1%/1000 h to 0.43%/1000 h. Similarly, the degradation rate dropped from 0.85%/1000 h to 0.45%/1000 h for the case S_3 to S_4 . Finally, Figure 5.8c compares average system efficiency. As expected, average efficiency dropped for the cases where the SC optimised the operating point of the system. This behavior is due to continuous changes in the T_{co} , which require either some additional air to be fed to the system to cool it down, or some additional fuel to be burned in the afterburner to heat up the system. In either case, the figure clearly shows that the optimal operating point of the system with respect to degradation rate contradicts the optimum with respect to system efficiency.

5.5.2.2 Compromise between degradation rate and efficiency

In this scenario, the compromise between the two performance criteria is considered, see (5.7). The weights $\omega_1 = 1$ and $\omega_2 = 30 \cdot 10^{-6}$ were set for the simulation. The resulting optimisation criterion $c(T_{co})$ is shown in Figure 5.9. The selection of the weights is arbitrary, hence ω_1 was selected so that the contribution of the efficiency to the cost function was in the same range as that of the degradation rate $r_d(T_{co})$, i.e. approximately $\omega_1(1 - \varepsilon) \in (0.9, 1.1) \times 10^{-5}$. From the Figure, one can observe one local minimum at $T_{co} = 975$ K, and the global minimum at the maximal viable temperature $T_{co} = 1100$ K.

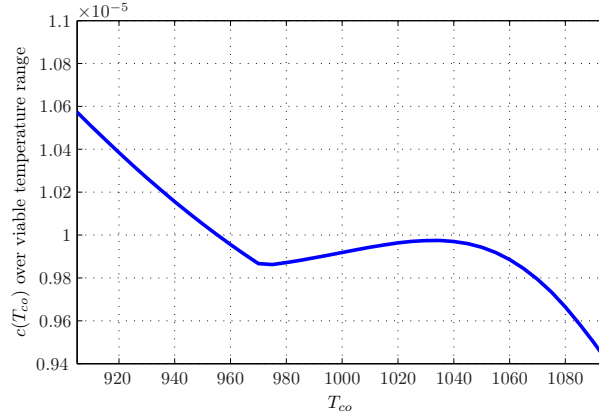


Figure 5.9: Value of the optimisation criterion over the viable temperature range of the T_{co} . The function shows a local minimum around $T_{co} = 975$ K.

Figure 5.10 analyses the performance of the SC. As before, the viable operating range of the stack is selected, $T_{co} \in [900, 1100]$ K, marked in red dashes. The duration of the simulation, however, is limited to 13 days, just enough time for the SC to converge to the optimum. Figure 5.10a shows the T_{co} and Figure 5.10b shows the corresponding value of $c(T_{co})$. From the moment the supervisory controller was turned on, after the first day of simulations, it initially decreased the stack temperature. Consequently, $c(T_{co})$ was increased. However, at $t = 2$ days, the SC reversed the strategy and started to gradually increase T_{co} . This action gradually decreased the value of $c(T_{co})$. After 4 days, the SC reached the minimum of $c(T_{co})$, as can be seen in Figure 5.10b. The final set-point of the stack temperature fluctuates around local minimum at approximately 975 K. This temperature corresponds well with the local minimum in Figure 5.9. From that moment on, the operating point of the system fluctuated around this local minimum. Figure 5.10c and 5.10d analyse both the performance criteria, the efficiency and the degradation rate, respectively. From the Figures it can be seen that efficiency improved by around 2% and the degradation rate decreased slightly.

Note that the final set-point for the controller is, by far, a suboptimal with respect the optimisation criterion show in Figure 5.9. The difficulties arise due to the simplicity of the implemented optimisation procedure, and fact that we are dealing with non-convex optimisation problem [139]. The non-convex optimisation is generally problematic. As opposed to the convex optimisation, the criterion function exhibits local minima. Consequently, the optimisation algorithm, however advanced it may be, can get "stuck" in one of the said local minima. Nevertheless, at least in our specific case, if the performance of the SC becomes unsatisfactory, the optimisation criterion can be adjusted by selecting different values of ω_1 and ω_2 , to either put more or less emphasis on system durability. In such a way, $c(T_{co})$ can be adjusted to resemble the ones in Figure 5.4. Clearly, selecting

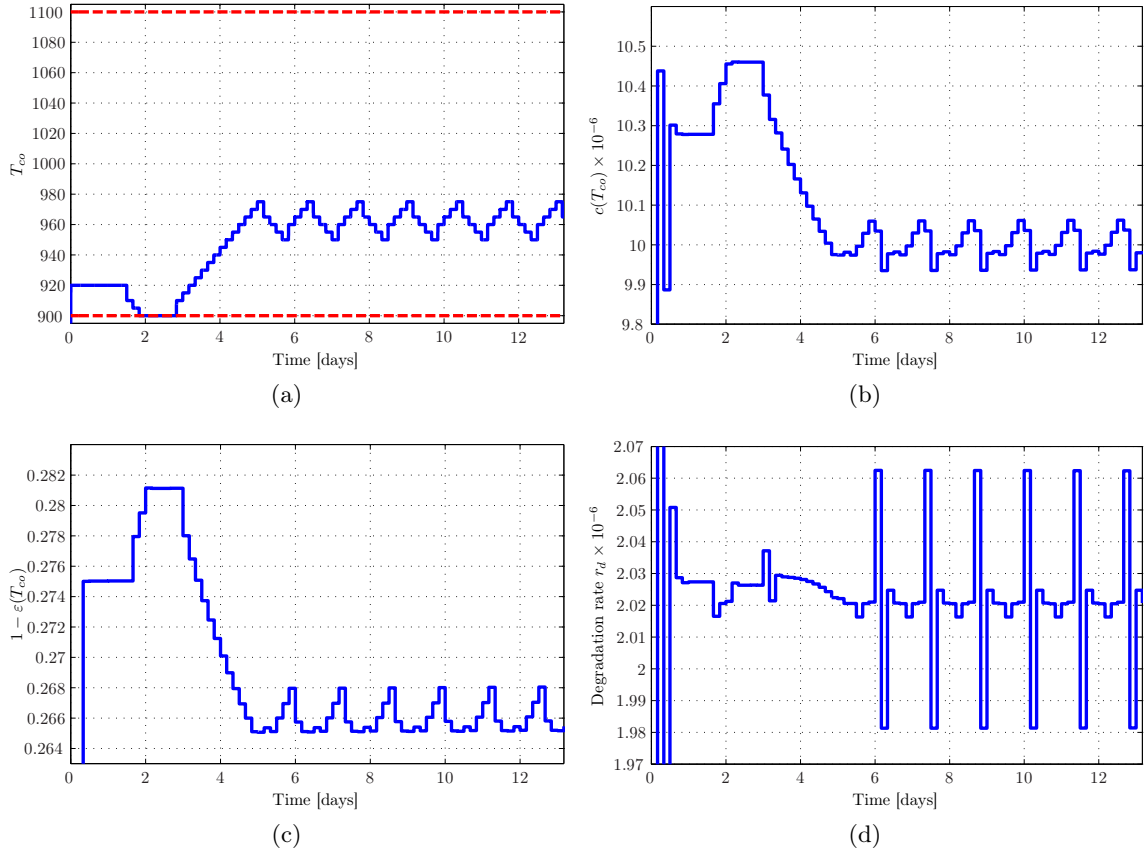


Figure 5.10: Analysis of SC performance: (a) the temperature reference T_{co} , (b) the evolution of the optimisation criterion $c(T_{co})$, (c) the evolution of system efficiency, and (d) the evolution of the degradation rate for the duration of the experiment. The SC gradually decreased the value of $c(T_{co})$, thus improving the overall performance of the system.

$\omega_2 = 0$, higher temperatures and slower degradation can be achieved at the expense of the system's efficiency. This would cause the system to behave as in the scenario $\mathcal{S}_2$.

5.5.3 Preliminary experimental evaluation

The preliminary experimental evaluation of the proposed control strategy was performed on a refurbished SOFC system described in Section 4. While the BoP module remained the same, the stack module was modified significantly. The old stack was replaced with two planar, anode-supported, low-temperature SOFC stack towers with 119 fuel cells each. Together the stacks provide nominal electrical power of approximately 6 kW. The two towers share the same manifolds, one for fuel and one for air feed. For the research purposes, the stacks were installed with two individual active loads, so that each of them could be controlled accordingly.

The above described control system was implemented on a standard PLC. The low-level controllers were properly tuned, the corresponding references, described in Table 5.1, were set, and the SC controller was activated to minimise the degradation rate by adjusting the stack temperature T_{co} .

During the initial testing of the system and tuning of the low-level controller, the stacks underwent severe degradation. Therefore, with rapid performance loss, the total load was decreased to 40% in order to have the system running as long as possible. The

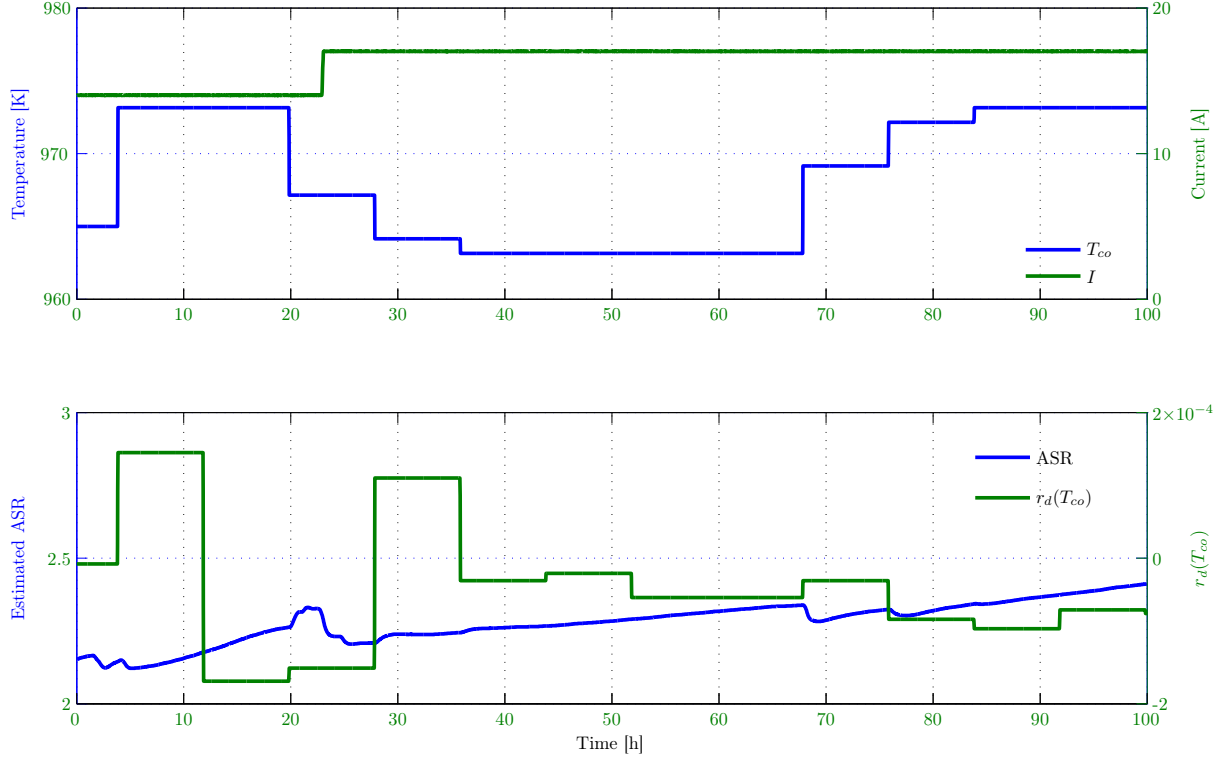


Figure 5.11: The analysis of the SC performance. The SC firstly increased the temperature, and the degradation increased. Then the temperature was decreased to decrease the degradation rate. Finally, the SC increased the temperature once again.

SC parameters were set at $\Delta t = 9h$, and $\Delta T = 3K$. The viable temperature range for the SC was set to $T_{co} \in [963.1 \ 973.1]$. The selected range was severely restrictive so to prevent hazardous situations should the SC fail.

Figure 5.11 shows an excerpt from the experimental data-set. The plot analyses only the variables relevant for this discussion. Namely, the T_{co} set point, the estimated ASR and corresponding degradation rate, i.e. $r_d(T_{co})$, and stack current I . There are three main events during the experiment: (i.) first the SC increased the temperature, (ii.) then the temperature was decreased, and (iii.) the temperature was increased again. Observing the Figure, one can see that the degradation rate firstly increased with increased temperature. In the next stage, the SC decreased the T_{co} all the way down to the lower limit. During this time, the degradation rate dropped. However, during this transition period the stack current was increased for 3 A, at roughly $t = 21$ h. Therefore, it is difficult to determine the reason for the drop in the degradation rate. One could argue that, since the current increased, the degradation rate dropped due to decrease in temperature, and not due to the increase in current (generally, higher currents cause faster degradation). However, this conclusion is ambiguous.

The stack temperature then remained at the lowest boundary, until around $t = 69$ h. Around that time the stack temperature was again increased to the maximal value and the degradation rate increased slightly. The action of the SC was probably caused by some unmeasurable disturbance. As discussed in Chapter 4, the estimated ASR depends on the plant and model mismatch. In turn, the mentioned disturbance coupled with model mismatch, reflects in the estimated degradation rate $r_d(T_{co})$, hence changing the functionality of the SC.

It is important to know that the effect of disturbances on the estimated value of $r_d(T_{co})$ can be alleviated. By adjusting the parameters of the algorithm that calculates the derivative of estimated ASR introduced in Chapter 4, these kind of disturbances can be filtered. During the above experiment, the parameters of the degradation rate estimator were selected so that the $r_d(T_{co})$ was more sensitive. This was due to the fact that the stack was practically at the EoL, and the sampling time of the SC, i.e. Δt , was relatively short. Unfortunately, higher sensitivity of the $r_d(T_{co})$ estimate caused mentioned difficulties.

5.6 Conclusion

This chapter discusses the design of a control system which incorporates information from the software sensors and SoH estimator. The control logic consists of two layers, low-level and supervisory control. The low-level control system is designed to minimise the electric consumption of the BoP components keeping the stack operating in safe regime. The supervisory controller optimises the said operating point. With respect to the preferences of the end user, the optimisation objective can be adjusted to (i.) minimise the degradation rate of the stack and so increase its total lifespan, (ii.) maximise the efficiency of the system, or (iii.) seek for the compromise between the two objectives.

The proposed control system is validated on a model of a 10 kW SOFC system. Five simulated scenarios are presented showing that a lower degradation rate can be achieved by maintaining the system at an appropriate operating point. According to the implemented degradation model, the degradation rate could be reduced from 1%/1000h to 0.43%/1000h with the system operating at a constant load of approx. 9 kW. Accordingly, the system with an SC optimised operating point could experience a 57% increase in total lifespan. A similar conclusion can be drawn for the cases with fluctuating load, with a total lifespan increase of about 50%. The mentioned conclusions, with respect to an increase of lifespan, are rather illustrative and applicable only to the specific experiment under which the degradation model (5.2) was built. A more general model is required for more realistic conclusions.

Nevertheless, a short (yet insufficient) experimental validation of the proposed control system shows promising preliminary results. Although drawing unambiguous conclusions is unreasonable due to limited experimental capabilities, one major conclusion is indisputable: *“The developed control system has a major impact on the degradation rate, and, in turn, to the total life-span of the system.”*

There are many aspects of the SC yet to be investigated:

- (i.) The selection of parameters of the SC:

The sampling time of the SC is crucial for minimisation of the degradation rate. The sampling time needs to be just long enough for the actions of the SC to be reflected in the calculated degradation rate.

- (ii.) The optimisation variable:

The proposed approach builds on the assumption that temperature is the most dominant factor on the degradation rate and is therefore optimised. However, extending the set of the "free knobs" (optimisation variables) may improve the overall performance of the system. For instance, optimising the fuel utilisation of the stack can increase the total efficiency of the system, while at the same time increasing the degradation rate.

- (iii.) Incorporating information from different diagnosis modules:

The proposed approach considers only information provided by the non-intrusive SoH estimator. While this approach provides adequate information about the ongoing degradation rate, the underlying degradation mechanism remains unknown. Employing different diagnosis algorithms, such as EIS, may bring additional information from which the control system can adequately adjust the system operating point.

(iv.) Extensive experimental validation:

The simplicity of the developed modules make them versatile and can be easily adapted for new components. Therefore, the already developed and PLC implemented modules can be easily put to use in similar systems and extensively tested.

Chapter 6

Conclusions

This dissertation addresses condition monitoring, prognostics, and health-aware control of the SOFC system. The notable original contributions include

- *the development of a temperature soft sensor* that enables direct control over stack gradients by providing on-line estimation of minimal and maximal stack temperatures,
- *the estimator of fuel gas composition* that enables close monitoring of the fuel gas composition flows at different locations inside the SOFC system,
- *the SoH estimator and RUL predictor* which employs information from soft and physical sensors, and a mathematical model of the stack to estimate concurrent SoH of the fuel cells,
- *the development of health-aware control for SOFC system* which, by employing the information from the above modules, keeps the system operating in safe region while at the same time minimises the degradation rate, maximises the efficiency, or seeks for a compromise between the two.

The temperature soft sensors are able to reconstruct minimal and maximal stack temperatures, thus enabling a more direct thermal management of the SOFC stack. While the causal relationship among input and output process variables is found by reasoning and understanding of the underlying physical processes, the model structure, as well as its corresponding parameters, are derived directly from experimental data. Sub-models that map input variables to internal temperatures, and input variables to outlet temperatures are identified by employing subspace identification theory. Then, the sub-models are merged into one simple model. It is shown that the derived model, with a rather simple structure, is able to reconstruct internal temperatures with high accuracy. The approach is applicable to the systems assembled on automated line, where the structure, materials, and operating regimes coincide. In this case, the model parameters can be identified on an instrumented reference system and further implemented on the deployed units.

The benefits of a fuel gas composition estimator are twofold. First, the estimation of gas composition can provide information required by the control system in order to keep the stack operating within a safe regime. Having this information, the control system is able to adjust operating parameters such as OCR or fuel utilisation to appropriate values with high accuracy without having to perform actual composition analysis on-line. Second, the information provided by this soft sensor is essential for the SoH module. Namely, the actual quantities of gas flows at the anode inlet is required by on-line estimation of the ASR, and thus SoH of the stack. The main novelty of the approach is the simple structure of

the said estimator which, compared to the classical approach, does not require numerical optimisation. The final form of the proposed estimator is simple matrix equation which can be easily implemented on a standard PLC.

In a way, the SoH estimator can also be considered as a soft sensor. By employing the lumped model in UKF algorithm, the estimator calculates the ASR from easily accessible process variables. The ASR carries vital information about the SoH of the stack, as the majority of degradation phenomena manifest in an increase of stack losses on a macro scale. The developed approach is non-intrusive, which means that the information it provides may be acquired from operational data only, and no external excitation is needed. Furthermore, the estimator is designed to be independent of operating conditions, at least in the nominal operating conditions of the system. Besides the SoH of the stack, the developed algorithm also predicts its RUL. Then, a model describing evolution of the the estimated ASR is identified. The on-line updated model then employs Monte Carlo simulation to predict future evolution of stack SoH, and so to obtain RUL.

The control system integrates all the developed modules together. The two-level control strategy follows two main objectives: (i.) maximisation of the power conversion, and (ii.) maximisation of the lifespan of the stack. While many control systems aim only at maximising the system efficiency, the proposed control system is one of the first to address the maximisation of the SOFC lifespan. First, the low level controller is designed so minimal electrical power is consumed in the BoP module. While the controller is designed so that minimal stoichiometric parameters such as fuel/air utilisations are always satisfied, the noncritical variables are only kept within the predefined range. Second, the SC aims at finding a set of references for the low level controller such that the degradation rate is minimised. Extensive simulations are carried out to demonstrate the performance of the designed controllers. On simulations, the validation of the proposed approach shows promising results.

The control system, and all the supportive modules, have been designed to facilitate their implementation on standard industrial equipment. Despite the vast effort that was put into the experimental validation of the system, full analysis and clear conclusions cannot be drawn due to the issues related with the high degradation rate of the system – the voltage of the fuel cells dropped below the feasible limit before the system was tuned and fully integrated. Nevertheless, a short experimental campaign encourages further tests.

Further work

There are numerous open questions that have yet to be answered. Of them, the issues of highest interest are the following:

- **The reconstruction of temperature gradients without in-situ measurements on an instrumented reference system.**

The developed temperature soft sensors provide information about internal stack temperatures by employing actual in-situ measurements to identify the described model. However, in cases when the in-situ measurements in a reference system are unavailable, the information can be acquired in an alternative manner. For instance, by employing a more complex model of the stack and estimating its states through some filtering technique, e.g. UKF, one may be able to reconstruct not only minimal and maximal temperatures, but also the full temperature gradient across the stack.

- **Expanding the equilibrium calculation for the gas composition estimator.**

The gas composition estimator builds on the assumption of thermodynamic equilibrium of methane reforming and water gas shift reactions. This equilibrium provides fairly accurate estimates of the fuel gas composition. However, as explained

in Chapter 2, this simple equilibrium neglects the presence of possible crossover (anode to cathode or vice versa) leakages. In this case, there are some other reactions (e.g. direct hydrogen and/or methane oxidation) that take place in anode/cathode compartments. Considering these additional reactions within the equilibrium could further improve the accuracy of the estimates.

- **Advanced prognostics.**

While the developed approach enables RUL predictions, one could improve its accuracy by incorporating degradation physics in the stack model. Having a set of parameters that describe the degradation rate as a function of input process variables might facilitate a more accurate estimation of the SOFC stack in question. In turn, this would result in a more precise RUL prediction.

Another open issue is the derivation of the RUL probability density function (PDF). In Chapter 4 the RUL distribution is obtained by employing MC simulations. A more suitable derivation of the RUL PDF is required.

- **Incorporating more information about ongoing degradation mechanisms and identifying mitigation strategies.**

The developed control strategy aims at minimising the degradation rate by manipulating stack temperature. While the temperature is the major factor in many degradation mechanisms, it is by far not the only one. Therefore, identifying the dominating degradation mechanisms, developing a corresponding mitigation strategy, and, finally, incorporating this information in the control system remains an open question. Nevertheless, the developed control system is the first demonstration of maximising the lifespan of the SOFC stack by employing an appropriate control strategy.

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Publications Related to the Thesis

Journal papers

1. Boštjan Dolenc, Pavle Boškoski, Martin Stepančič, Antti Pohjoranta, Đani Juričić, "State of health estimation and remaining useful life prediction of solid oxide fuel cell stack", *Energy convers. manage.*, vol. 148, p. 993-1002, 2017.
2. Boštjan Dolenc, Darko Vrečko, Đani Juričić, Antti Pohjoranta, Cesare Pianese, "On-line gas composition estimation in solid oxide fuel cell systems with anode off-gas recycle configuration", *J. power sources*, vol. 343, p. 246-253, 2017.
3. Boštjan Dolenc, Darko Vrečko, Đani Juričić, Antti Pohjoranta, Cesare Pianese, "On-line estimation of internal stack temperatures in solid oxide fuel cell power generating units", *J. power sources*, vol. 336, p. 251-260, 2016.

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Biography

Boštjan Dolenc was born on 20th July 1989 in Postojna, Slovenia. After completing the grammar school at the School center Postojna in 2008, he enrolled as an undergraduate student at the Faculty of Electrical Engineering, University of Ljubljana. During his studies, he received the scholarship at Jožef Stefan Institute. In 2011/2012 he also participated in Erasmus student exchange at *Universitat Politècnica de València*, Valencia, Spain.

After graduation in 2013, he enrolled in doctoral study programme at Jožef Stefan International postgraduate school. He was also accepted to the Young Researchers Programme of the Slovenian Research Agency under the supervision of prof. dr. Đani Juričić. In 2015 he was awarded Ad Futura scholarship for Ph.D. student exchange and spend a period of seven months at *Università degli Studi di Salerno*, Italy. There, he studied under the supervision of prof. dr. Cesare Pianese.

During his time as a Ph.D. student, his research has been focused on the development of control orientated algorithms of technical systems, including SOFCs. The main results include the development of the so-called software sensors, condition monitoring algorithms, and the development of the control algorithms. He also participates in the integration of the developed algorithms to the comprehensive control system, which focuses on the safe operation, durability, and longevity of the SOFC system. Furthermore, he has also engaged in study and development of the diagnosis algorithms based on electrochemical impedance spectroscopy. This includes extraction of the relevant information from measured data.

In addition, he also contributed to the development of diagnostic algorithm for other technical systems, more particularly rotational machinery. The latter consists of development and application of signal processing and statistical techniques for fault detection and isolation.

