

A1314

BioCORE - Thermodynamic evaluation of a biogas powered reversible SOC system

Stephan Herrmann (1), Michael Geis (1), Maximilian Hauck (1), Felix Fischer (1), Sebastian Fendt (1), Matthias Gaderer (2), Hartmut Spliethoff (1)

(1) Lehrstuhl für Energiesysteme, Technische Universität München
Boltzmannstr. 15, DE-85748 Garching/Germany

(2) Professur für Regenerative Energiesysteme, Technische Universität München
Schulgasse 16, DE-94315 Straubing/Germany

Tel.: +49-89-289-16279

Fax: +49-89-289-16271

stephan.herrmann@tum.de

Abstract

In this work the new BioCORE process is evaluated. BioCORE converts a continuously produced biogas (50% CH₄, 50% CO₂) stream in a Reversible Solid Oxide Cell (RSOC) system to produce either electricity or biomethane (=Synthetic Natural Gas; SNG), depending on the shortage or excess availability of electricity, for example from photovoltaic and wind power. The analysis is performed based on a RSOC model, which has been validated against experimental stack data, and further component models created in Aspen Plus. Different process designs are analyzed, which provide different amounts of purified excess CO₂ for industrial purposes. The analysis also covers the heat and electricity demand of the biogas production, as well as gas cleaning. Furthermore, the effect of raising the operating pressures up to 15 bar is studied. Due to rigorous exergetic system optimization and cascaded heat integration, in combination with consumption of excess heat in a steam cycle, the process shows very high conversion efficiencies. At a SOC operating temperature of 765 °C and current density of 0.5 A/cm² in the “generation” mode (electricity production) the exergy efficiency reaches up to 80.6%. During “storage” mode operation (biogas upgrading by electrolysis and methanation) the current density is increased to 1 A/cm² for operating at thermo-neutral voltage, and an exergy efficiency of 85.9% is obtained.

The authors did not wish to publish their full contribution in these proceedings and possibly have published it in a journal. Please contact the authors directly for further information.

1. Introduction

Biogas is a valuable resource that has gained significant attention throughout the last decades, especially in Germany. However, to date the efficiency of electricity generation from biogas, mainly in internal combustion engines, is still well below 50%. Furthermore, biogas plants are often operated in baseload instead of providing operational flexibility to the electricity grid. In an attempt to solve this issue, this work provides a case study to evaluate the thermodynamic potential and limitations of different reversible Solid Oxide Cell (SOC) system configurations operated on biogas. The main aim of this approach is to provide a large quantity of balance power from the existing biogas plants. For this purpose first of all an overview over the current state of the art biogas based SOC systems, as well as reversible SOC systems, is given. Then a new system concept is introduced and evaluated, which shows the potential to increase the efficiency of electricity generation from biogas in SOC.

2. Literature review on biogas based Gas-to-Power and Power-to-Gas processes

State of the art of biogas fed SOFC systems

Operation of Solid Oxide Fuel Cells (SOFC) fed with biogas has been widely studied in the scientific literature. For example, Chiodo et al. [1] determined a maximum DC efficiency of 61.76%_{LHV} (~60% AC) for SOFC operated on steam-reformed biogas at a comparably high temperature of 800°C and a current density of 0.25 A/cm². All other reforming options lead to far lower efficiencies between 37-56%_{LHV}. Curletti et al. [2] found typical system efficiencies in the range of 50-62%_{LHV} for biogas-SOFC systems with carbon dioxide separation, and determined a maximum efficiency of 70%_{LHV} for a pressurized gas turbine-SOFC hybrid configuration with 90% fuel utilization in the SOFC. EU projects like SOFCOM have shown the possible utilization of biogas from waste water treatment plants in SOFC [3]. Additionally, some SOFC producers already offer commercial units capable of generating electricity from biogas with electrical efficiency between 50-60%_{LHV} [4-6]. In 2017, in the frame of the EU funded project DEMOSOFC, a Convion SOFC unit has successfully been installed at a waste water treatment plant near Torino, Italy.

State of the art in biogas upgrading SOEC systems

In the scientific literature a consensus can be found that the thermal integration of synthesis processes, especially methanation, and Solid Oxide Electrolysis Cells (SOEC) by steam production represents an almost ideal combination. For a CO₂-H₂O co-electrolysis system [7] found an electricity-to-SNG (Synthetic Natural Gas) plant efficiency of up to 74.8%_{LHV}. Similarly, the EU project Helmeth aimed at an electrolysis-CO₂-methanation efficiency of >85% based on Higher Heating Value (HHV) [8]. Pozzo et al. [9] investigated an integrated biomass gasification-SOEC-DME (Dimethylether) synthesis plant configuration reaching a biomass+electricity to DME efficiency of 69.5%_{LHV}. Lorenzi et al. [10,11] have studied biogas upgrading by direct co-electrolysis of biogas with steam. They found a maximum biogas+electricity (5-6 MW each) to SNG exergy efficiency of 83% (unpressurized) and 87% (pressurized), respectively. In two similar process configurations Hansen et al. [12] found exergy efficiencies of around 80%.

Recent developments in reversible SOC systems

Recently also reversible operation of SOC has gained increasing attention. Different system configurations have been studied in literature. A summary is shown in Table 1. As

can be deducted from the results a quite fundamental limit of around 70% can be identified for the Round Trip Efficiency (RTE), which is only overcome to a certain extent by operation at low power densities. This is due to the fact that the losses are attributed firstly to fundamental physical effects, such as overpotentials (15-20%), and secondly to system design constraints like Balance of Plant (BoP) related consumption (10-15%). Overpotentials can essentially only be reduced by advancements in materials or larger SOC sizes (leading to higher cost).

Table 1: Properties of RSOC systems in the literature.

Source ¹	Key properties ²	max.RTE
H₂-H₂O based RSOC systems		
[13] (Sunfire GmbH)	Fuel cell operation 25 kW, 50% _{LHV} (CH ₄), electrolysis 142.9 kW, 84% _{LHV} (H ₂)	42%
[14] (DLR)	Heat storage in HT-PCM, RTE: 52% (1 bar), 53% (25 bar)	53%
[15] (DLR)	RTE 60% (30 bar/800°C), 55% (1 bar/850 °C), ESC, PCM	60%
[16] (Univ. of Pisa)	Pressurized (74%) and unpressurized (64%) with internal heat storage in SOC stack	74%
[7] (Politecnico di Torino)	Pressurized and unpressurized with air and pure oxygen operation	72%
Hydrocarbon based RSOC systems		
[17] (CSM)	Reversible SOC system with up to 30 bar, O ₂ operation, mixed species tank (evaporation not considered in balance)	-
[18] (DTU, CSM)	Pressurized operation at 20 bar, 250 MW, cavern storage	72%
[19,20] (CSM)	RSOC with up to 20 bar, air operation, evaporation considered, variation of H-C-ratio	72%
[21] (CSM)	Similar to [19,20] with pipeline gases	70%

Therefore, the focus should be set on system design related improvements. Furthermore, to date all systems studied in the literature have been designed as pure storages. Integration of RSOC with biogas plants has not been considered yet.

3. Simulations

Reversible system concept

The reversible system concept is based on the idea of utilizing a usually continuous supply of biogas (alternatively also gasification product gas) at maximum efficiency in a system, which allows an alternation between electricity production and electricity consumption. This behavior is beneficial with regard to compensation of intermittent solar and wind based renewable electricity. For electricity production, the so called “generation mode”, the biogas is converted in the SOC operating as SOFC. The CO₂ generated during the conversion is stored. In the electricity consuming “storage mode”, the SOC is used as a SOEC to produce biomethane (=SNG) from CO₂, biogas, and electricity. A schematic illustration of the system concept is shown in Figure 1.

¹ DLR = Deutsches Zentrum für Luft- und Raumfahrt; CSM = Colorado School of Mines; DTU = Technical University of Denmark

² PCM = Phase Change Material; ESC = Electrolyte Supported Cell

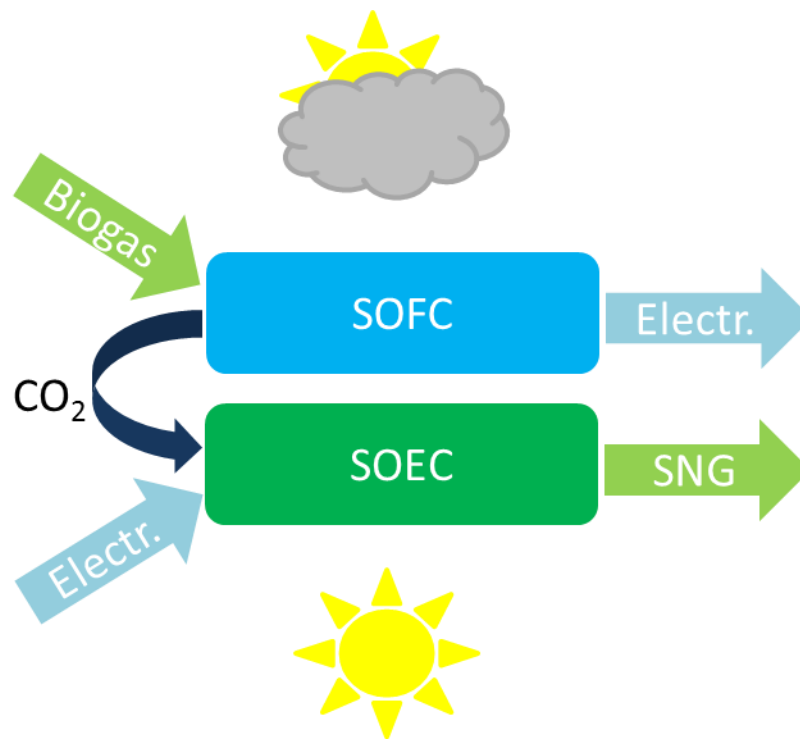


Figure 1: Illustration of the reversible system concept.

SOFC Model

The fuel cell model used in this work has first been created as a pure SOFC model in collaboration within the EU project SOFCOM. It is a 0-dimensional thermodynamic model set up in Aspen Plus. A detailed description of this model has been published in [22]. Furthermore, the original model has been adapted and improved to also accommodate electrolysis operation. The results of this work have been published in [23]. A validation against SOFC stack data can be found in [24]. For further details regarding the model the reader is referred to these publications.

Other component models

The other system components are modelled essentially based on standard Aspen Plus components, such as HeatX blocks (heat exchangers), Gibbs reactors (e.g. reforming, combustion), pressure changers (compressors, pumps, turbines), etc.

Modelling parameters and main assumptions

The main parameters and assumptions are collected in Table 2. All assumptions are based on typical values found in literature, or provided by institutions or companies where available. Many assumptions are chosen conservative, and in no case proven current technical limits are exceeded. In contrast to exact values from the literature some values are rounded for reasons of simplicity, where appropriate. This includes for example the reference pressure for exergy calculations, which is fixed to 1 bara. Absolute temperatures provided in Kelvin are rounded down to full digits.

Table 2: Collection of general simulation parameters.

Parameter	Value	Source
Electric drive/generator efficiency	0.95	Assumption
Isentropic efficiency	0.6 - 0.75	Assumption
DC/AC converter efficiency	0.98	[25]
AC/DC converter efficiency	0.98	[26]
System pressures	1 - 15 bara	Assumption
Heat losses in components	0.0-2.0% of heat transferred	Assumption
SOC operating temperature (standard – maximum)	973 – 1073 K (700 – 800 °C)	[27]
SOC current density (standard – maximum)	0.50 – 0.76 A/cm ² (SOFC) 0.77 – 1.38 A/cm ² (SOEC)	[27]
SOC stack fuel utilization (min – max)	0.4 – 0.85	[27]
FE exhaust recirculation ratio	0.0 - 0.7	
Pressure drop SOC	0.015 (SOFC) - 0.065 bar (SOEC)	[27]
SOC module heat losses	0.3-2.0% of fuel energy	[28]
T ₀ (exergy calculations)	388 K (15 °C)	
P ₀	1 bara	
Methanation catalyst maximum possible (adiabatic) temperature	1000 K (727 °C)	Assumption
Methanation equilibrium final state	558–573 K (285–300 °C)	Assumption
Steam turbine (standard)	45 bara, 450 °C, 0.6, 1 or 2 stage, 0.1 bara	[29]
Steam turbine (advanced)	85 bara, 500 °C, 0.75, 2 stage, 0.1 bara	Assumption
Biogas input (LHV)	802.7 kW	Aspen Plus
Biogas composition	0.5 CH ₄ , 0.5 CO ₂	
Biogas input (HHV)	891.6 kW	Aspen Plus
Biogas exergy	844.7 kW	Calculation
Biogas plant temperature (mesophilic range)	310.15 K (37 °C)	[30]
Biogas plant heat consumption	133 kW _{th}	Derived from [30]
Biogas plant electricity consumption	20 kW _{el}	Derived from [30]

4. Results

The main results of the simulations for the system with state-of-the-art operating parameters (700 °C, 1 bara) in both operating modes are given in Table 3. Only the generation mode operation can be directly compared to the operation of a conventional SOFC system. Regarding the auxiliary consumption especially the air blower has a lower demand compared to conventional SOFC systems, while the Fuel Electrode (FE) recycle blower needs more power because of an overall higher fuel flow. In sum, the net efficiency in generation mode is about 10%-points higher than in typical reference SOFC systems. Including liquefaction of the 218.7 kg/h residual CO₂ the efficiency of power generation

drops from 68.6%_{LHV} to 66.7%_{LHV} due to the power consumption of the required additional compressors (not shown in the table).

Table 3: RSOC system simulation results.

Parameter	SOFC	SOEC
Biogas input		
Biogas feed to SOC system	802.7 kW _{LHV}	802.7 kW _{LHV}
Cell/stack parameters		
Operating voltage	0.814 V	1.308 V
SOC current density	0.501 A/cm ²	0.885 A/cm ²
DC output / input	611.8 kW	-1737.3 kW
Auxiliary consumption		
Inverter loss	-12.2 kW	-35.5 kW
Digester electricity	-20.0 kW	-20.0 kW
Feedwater pump	-0.7 kW	-1.0 kW
Steam turbine	37.8 kW	9.3 kW
FE recycle blower	-12.5 kW	-0.8 kW
Air blower	-12.6 kW	-0.0 kW
Biogas blower	-1.6 kW	-1.6 kW
CO ₂ separation compressor	-39.1 kW	-42.4 kW
Total auxiliaries	-60.9 kW	-92.0 kW
Outputs		
AC net output / consumption	550.9 kW	-1829.3 kW
Digester heat consumption (37 °C)	133.0 kW	133.0 kW
District heating output (80/35 °C)	96.6 kW	57.9 kW
SNG output (LHV)	-	2201.0 kW
SNG output (HHV)	-	2449.5 kW
Wobbe Index (HHV) (10 °C, 1 bara)	-	47.6 MJ/m ³
Electrical/biomethane efficiencies		
LHV efficiency	0.686	0.836
HHV efficiency	0.618	0.931
Exergy efficiency	0.652	0.858
Total efficiency (including digester heating + district heating)		
LHV efficiency	0.972	0.909
HHV efficiency	0.875	0.970
Exergy efficiency	0.676	0.864

Table 4 displays the simulation results for the pressurized system at (765 °C, 15 bara). Compared to the previous case a significantly higher efficiency can be claimed for both generation and storage modes. However, this is tied to more challenging technical parameters and therefore potentially lower system lifetime.

Table 4: Pressurized RSOC system simulation results.

Parameter	SOFC	SOEC
Biogas input		
Biogas feed to SOC system	802.7 kW _{LHV}	802.7 kW _{LHV}
Cell/stack parameters		
Operating voltage	0.929 V	1.280 V
SOC current density	0.509 A/cm ²	1.012 A/cm ²
DC output / input	710.3 kW	-1943.4 kW
Auxiliary consumption		
Inverter loss	-14.2 kW	-39.7 kW
Digester electricity	-20.0 kW	-20.0 kW
Feedwater pump	-0.5 kW	-2.1 kW
Steam turbine	44.7 kW	47.8 kW
FE recycle blower	-0.2 kW	-0.5 kW
Oxygen compressors	9.2 kW	-23.0 kW
Biogas compressor	-25.0 kW	-25.0 kW
CO ₂ separation / Biomethane compressor	-6.2 kW	-0.0 kW
CO ₂ compressor	-18.3 kW	-0.0 kW
Total auxiliaries	-30.5 kW	-62.5 kW
Outputs		
AC net output / consumption	679.8 kW	-2005.9 kW
Digester heat consumption (37 °C)	133.0 kW	133.0 kW
District heating output (120/35 °C)	25.8 kW	23.5 kW
Biomethane output (LHV)	0.0 kW	2394.8 kW
Biomethane output (HHV)		2662.4 kW
Wobbe Index (HHV) (10 °C, 1 bara)		50.2 MJ/m ³
Electrical / Biomethane efficiencies		
LHV efficiency	0.847	0.853
HHV efficiency	0.762	0.948
Exergy efficiency	0.806	0.859
Total efficiency (including digester heating)		
LHV efficiency	1.045	0.908
HHV efficiency	0.941	0.973
Exergy efficiency	0.821	0.864

5. Summary

In this work the new BioCORE process is evaluated. BioCORE converts a continuously produced biogas (50% CH₄, 50% CO₂) stream in a RSOC system. Either electricity or biomethane are produced, depending on the shortage or excess availability of electricity, for example from photovoltaic and wind power. The analysis is performed based on a RSOC model, which has been validated against experimental stack data, and further component models created in Aspen Plus. Different process designs are analyzed, which either convert all CO₂ in the biogas to biomethane or provide purified excess CO₂ for industrial purposes. The analysis also covers the heat and electricity demand of the biogas production, as well as gas cleaning. Furthermore, the effect of raising the operating pressures up to 15 bar is studied. Due to rigorous exergetic system optimization and cascaded heat integration, in combination with consumption of excess heat in a steam cycle, the process shows very high conversion efficiencies. In the pressurized case at a SOC operating temperature of 765 °C and current density of 0.5 A/cm² in the generation

mode the exergy efficiency reaches up to 80.6%. During storage mode operation the current density is increased to 1 A/cm² for operating at thermo-neutral voltage, and an exergy efficiency of 85.9% is obtained.

Acknowledgements

Parts of the models used in this work have been developed within the FCH-JU project SOFCOM GA. Nr. 278798, which is gratefully acknowledged. The current SOFC research at the Chair for Energy Systems is funded within the project SynSOFC by the Deutsche Forschungsgemeinschaft (DFG), which is gratefully acknowledged. Furthermore the authors thank the State of Bavaria for supplementary funding.

References

- [1] CHIODO, V., GALVAGNO, A., LANZINI, A., PAPURELLO, D., URBANI, F., SANTARELLI, M., AND FRENI, S. 2015. Biogas reforming process investigation for SOFC application. *Energy Conversion and Management* 98, 252–258.
- [2] CURLETTI, F., GANDIGLIO, M., LANZINI, A., SANTARELLI, M., and MARÉCHAL, F. 2015. Large size biogas-fed Solid Oxide Fuel Cell power plants with carbon dioxide management. Technical and economic optimization. *Journal of Power Sources* 294, 669–690.
- [3] SANTARELLI, M., BRIESEMEISTER, L., GANDIGLIO, M., HERRMANN, S., KUCZYNSKI, P., KUPECKI, J., LANZINI, A., LLOVELL, F., PAPURELLO, D., SPLIETHOFF, H., SWIATKOWSKI, B., TORRES-SANGLAS, J., and VEGA, L.F. 2017. Carbon recovery and re-utilization (CRR) from the exhaust of a solid oxide fuel cell (SOFC). Analysis through a proof-of-concept. *Journal of CO2 Utilization* 18, 206–221.
- [4] BLOOM ENERGY. 2017. ES-5710 Fuel Cell Data Sheet. <http://www.bloomenergy.com/fuel-cell/es-5710-data-sheet/>. Accessed 14 December 2017.
- [5] CONVION. 2017. Products - Fuel Cell Systems. <http://convion.fi/products/>. Accessed 14 December 2017.
- [6] SUNFIRE. 2017. Power Core. <http://www.sunfire.de/de/produkt-technologie/power-core>. Accessed 14 December 2017
- [7] FERRERO, D. 2016. Design, development and testing of SOEC-based Power-to-Gas systems for conversion and storage of RES into synthetic methane. Dissertation, Politecnico di Torino.
- [8] HELMETH. 2017. European Project. <http://www.helmeth.eu/index.php/project>. Accessed 11 December 2017.
- [9] POZZO, M., LANZINI, A., and SANTARELLI, M. 2015. Enhanced biomass-to-liquid (BTL) conversion process through high temperature co-electrolysis in a solid oxide electrolysis cell (SOEC). *Fuel* 145, 39–49.
- [10] LORENZI, G., LANZINI, A., and SANTARELLI, M. 2015. Digester Gas Upgrading to Synthetic Natural Gas in Solid Oxide Electrolysis Cells. *Energy Fuels* 29, 3, 1641–1652.
- [11] LORENZI, G., LANZINI, A., SANTARELLI, M., and MARTIN, A. 2017. Exergo-economic analysis of a direct biogas upgrading process to synthetic natural gas via integrated high-temperature electrolysis and methanation. *Energy* 141, 1524–1537.
- [12] HANSEN, J.B., FOCK, F., and LINDBOE, H.H. 2013. Biogas Upgrading. By Steam Electrolysis or Co-Electrolysis of Biogas and Steam. *ECS Trans.* 57, 1, 3089–3097.

- [13] SCHWARZE, K., POSDZIECH, O., KROOP, S., LAPEÑA-REY, N., and MERMELSTEIN, J. 2017. Green Industrial Hydrogen via Reversible High-Temperature Electrolysis. *ECS Trans.* 78, 1, 2943–2952.
- [14] SANTHANAM, S., HEDDRICH, M., RIEDEL, M., and FRIEDRICH, K.A. 2017a. Process Design Study of Reversible Solid Oxide Cell (r-SOC) System for Coupling Energy Storage and Hydrogen Economy Supply Chain. *ECS Trans.* 78, 1, 2925–2932.
- [15] SANTHANAM, S., HEDDRICH, M.P., RIEDEL, M., and FRIEDRICH, K.A. 2017b. Theoretical and experimental study of Reversible Solid Oxide Cell (r-SOC) systems for energy storage. *Energy* 141, 202–214.
- [16] DI GIORGIO, P., and DESIDERI, U. 2016. Potential of Reversible Solid Oxide Cells as Electricity Storage System. *Energies* 9, 8, 662.
- [17] KAZEMPOOR, P., and BRAUN, R.J. 2014. Model validation and performance analysis of regenerative solid oxide cells for energy storage applications. Reversible operation. *International Journal of Hydrogen Energy* 39, 11, 5955–5971.
- [18] JENSEN, S.H., GRAVES, C., MOGENSEN, M., WENDEL, C., BRAUN, R., HUGHES, G., GAO, Z., and BARNETT, S.A. 2015. Large-scale electricity storage utilizing reversible solid oxide cells combined with underground storage of CO₂ and CH₄. *Energy Environ. Sci.* 8, 8, 2471–2479.
- [19] WENDEL, C.H., KAZEMPOOR, P., and BRAUN, R.J. 2015. Novel electrical energy storage system based on reversible solid oxide cells. System design and operating conditions. *Journal of Power Sources* 276, 133–144.
- [20] WENDEL, C.H., KAZEMPOOR, P., and BRAUN, R.J. 2016. A thermodynamic approach for selecting operating conditions in the design of reversible solid oxide cell energy systems. *Journal of Power Sources* 301, 93–104.
- [21] REZNICEK, E., and BRAUN, R.J. 2017. Renewable Energy-Driven Reversible Solid Oxide Cell Systems for Grid-Energy Storage and Power-to-Gas Applications. *ECS Trans.* 78, 1, 2913–2923.
- [22] TJADEN, B., GANDIGLIO, M., LANZINI, A., SANTARELLI, M., and JÄRVINEN, M. 2014. Small-Scale Biogas-SOFC Plant. Technical Analysis and Assessment of Different Fuel Reforming Options. *Energy Fuels* 28, 6, 4216–4232.
- [23] HAUCK, M., HERRMANN, S., and SPLIETHOFF, H. 2017. Simulation of a reversible SOFC with Aspen Plus. *International Journal of Hydrogen Energy* 42, 15, 10329–10340.
- [24] HERRMANN, S., HAUCK, M., GEIS, M., FENDT, S., GADERER, M., and SPLIETHOFF, H. 2017. Influence of Operating Parameters and System Design on Efficiency of Biomass and Biogas Based SOFC Systems. *ECS Transactions* 78, 1, 219–227.
- [25] TST. 2017. Wechselrichter Sungrow SG 60KTL - TST Photovoltaik Online Shop. <https://www.photovoltaik-shop.com/wechselrichter-sungrow-sg-60ktl.html>. Accessed 17 October 2017
- [26] SMA, S.T.A. 2017. SUNNY CENTRAL 2200 / 2745 / 2500-EV / 2750-EV. <http://files.sma.de/dl/29394/SCS2200-2475-2500-EV-2750-EV-DDE1743-V12web.pdf>. Accessed 1 December 2017.
- [27] BLUM, L., FANG, Q., DE HAART, L.G.J., MALZBENDER, J., MARGARITIS, N., MENZLER, N.H., AND PETERS, R. 2017. SOC Development at Forschungszentrum Jülich. In *Solid oxide fuel cells 15 (SOFC-XV)*, S. C. SINGHAL, T. KAWADA AND L. STREU, Eds. ECS - The Electrochemical Society, Pennington, NJ, USA, 1791–1804.
- [28] VAN HERLE, J., MARÉCHAL, F., LEUENBERGER, S., AND FAVRAT, D. 2003. Energy balance model of a SOFC cogenerator operated with biogas. *Journal of Power Sources* 118, 1-2, 375–383.



- [29] G-TEAM A.S. 2017a. Steam Turbine - TR HI 150. <https://www.steamturbo.com/steam-turbines/tr-hi-150-5.html>. Accessed 9 November 2017.
- [30] FNR. 2017. Biogas: Faustzahlen. <https://biogas.fnr.de/daten-und-fakten/faustzahlen/>. Accessed 6 November 2017