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Electrochemical Quality Assurance of Solid Oxide Electrolyser (SOEC) Stacks

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High temperature solid oxide cells (SOC) are highly efficient and environmentally friendly electrochemical systems for the H₂/H₂O and/or CO/CO₂ redox reactions. The cells can be operated reversely either in electrolysis (SOEC) or fuel cell (SOFC) mode which facilitates this technology for power-to-gas-to-power application in renewable energy storage systems. However, the successful market introduction and public acceptance of the SOEC technology require high quality, reliability and reproducibility of the corresponding cells and stacks. Therefore, in the European funded project “Solid oxide cell and stack testing, safety and quality assurance” (SOCTESQA) pre-normative test modules and programs for high temperature solid oxide cells and stacks have been developed. Different EU project partners have tested identical SOC stacks in several testing campaigns with the same test programs. The paper presents and compares the results of the stacks in SOEC operation which have been obtained by application of the three most important test modules, e.g. current-voltage characteristics, electrochemical impedance spectroscopy and operation at constant current. The results are analyzed and discussed in context to the test input parameters, e.g. gas temperatures and steam supply stability. Quality aspects like repeatability and reproducibility among the different partners and among different test methods are statistically evaluated and discussed.

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The solid oxide electrolyser cell (SOEC) technology originates from the well-known solid oxide fuel cell (SOFC) technology, which has been developed during the last four decades. The cells can be operated reversely either in electrolysis or fuel cell mode which makes them very attractive for energy storage of excess renewable energies (e.g. wind or solar) by H₂ or H₂+CO syngas (so-called “power-to-gas”).

SOECs consist of a porous ceramic-metal (cermet) negative fuel electrode, a dense oxygen ionic conductive zirconia based electrolyte and a porous ceramic oxide positive air electrode (Figure 1). The cells are usually operated at temperatures of about 700–900°C. Both reactants H₂O and/or CO₂ can be electrolyzed, however the present paper concentrates only on the electrolysis of steam. At the fuel electrode steam is reduced to H₂ following Reaction 1. The resulting O²⁻ ions are conducted through the solid ceramic electrolyte. At the air electrode the O²⁻ ions are oxidized to oxygen molecules as mentioned in Reaction 2.

The overall endothermic reaction for electrolysis of steam into H₂ and O₂ is:

As an example Figure 1 shows the microstructure of an SOEC single cell¹ with the corresponding electrochemical reactions. The cell was originally developed for SOFC application and is mechanically supported by a 300 μm thick porous fuel gas electrode substrate.

At thermodynamic equilibrium the electrochemical reaction of splitting of pure water (partial pressure of water is 1) is accomplished with the standard Gibbs free reaction enthalpy of reaction Δ^oG^o(T),

defined as the difference between the standard enthalpy of reaction Δ^oH^o(T) and the required standard heat demand T · Δ^oS^o(T) (see Eq. 4):

$$\Delta^o G^o(T) = \Delta^o H^o(T) - T \cdot \Delta^o S^o(T) \quad [4]$$

All values are dependent on temperature. However, the required standard enthalpy of reaction Δ^oH^o(T) remains nearly constant above 100°C whereas the standard Gibbs free enthalpy Δ^oG^o(T) decreases with increasing temperature.² The required standard entropic heat demand T · Δ^oS^o(T) for the endothermic decomposition reaction is lower in the vapor phase of water compared to the liquid phase and increases with increasing temperature. This heat demand can be supplied by the internal resistivity losses of the cells under operation, which is described below (Eq. 15) in more detail. Another option is the usage of heat from external sources, e.g. renewable solar heat or waste industrial heat. In this case, the high temperature electrolysis (HTE) requires less electrical electrolysis power compared to low temperature electrolysis (LTE).³

Figure 1. Microstructure of an SOEC with corresponding electrochemical reactions for water electrolysis.

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For practical HTE the steam partial pressure usually is lower than 1. Hence, the molar Gibbs free enthalpy of reaction $\Delta^r G(T, p)$ has to be described as a function of temperature T and partial pressures p of the reactant gases:

$$\Delta^r G(T, p) = \Delta^r G^0(T) + RT \cdot \ln \frac{p_{O_2}^{1/2} \cdot p_{H_2}}{p_{H_2O}} \quad [5]$$

with R : Gas constant, F : Faraday constant and $\Delta^r G^0(T)$: Standard molar Gibbs free enthalpy of reaction with pure H_2O .

Thus, the theoretical thermodynamic reversible cell voltage $V_{rev}(T)$ can be calculated according to the Nernst equation (z : number of electrons: 2):

$$V_{rev}(T, p) = \frac{\Delta^r G(T, p)}{zF} = V_{rev}^0(T) + \frac{RT}{2F} \cdot \ln \frac{p_{O_2}^{1/2} \cdot p_{H_2}}{p_{H_2O}} \quad [6]$$

The measurable open circuit voltage (OCV) differs from the reversible cell voltage by irreversible losses η_0 . The open circuit voltage of a single electrolysis cell at $750^\circ C$ with a steam partial pressure of 0.8 is about 0.9 V.

$$OCV = V_{rev} - \eta_0 \quad [7]$$

In order to achieve higher voltages several cells are assembled in series and form a stack by using interconnect plates between them. The corresponding cell-interconnect units of a stack are called repeat units (RUs):

$$V_{Stack} = \sum_n V_{RU}^n \quad [8]$$

For electrolysis operation electrons from a voltage source (potentiostat) which is connected in the current circuit of the SOEC are pumped from the air electrode to the fuel electrode according to Eqs. 1 and 2. Due to the resistances of the cell or the stack repeat unit the electrolysis voltage $V(I)$ under electrical current I is the sum of the OCV and the resulting overvoltages ΔV_{total} :

$$V(I) = OCV + \Delta V_{total}(I) = OCV + \int I R_{total}(I) dI \quad [9]$$

with

$$R_{total}(I) = R_{Polarisation}(I) + R_{Ohmic}(I) + R_{Diffusion}(I) \quad [10]$$

The resistances of a cell or stack repeat unit can be distinguished between polarization, ohmic and diffusion resistance. The corresponding equations are:

$$R_{Polarisation} = \frac{RT}{zFI}, \quad [11]$$

$$R_{Ohm} = \sum_k \frac{1}{\sigma_k^{kor}} \cdot d_k \cdot \frac{1}{A_k} \quad [12]$$

with

$$\sigma_k^{kor} = \sigma_k \cdot \frac{1 - \frac{p_k}{2}}{(1 - p_k)^2}, \quad [13]$$

and

$$R_{Diffusion} = \frac{RT}{zF} \ln \left(1 - \frac{j}{j_L} \right) \quad [14]$$

with I : electrical current, j : current density, σ_k^{kor} : specific conductivity corrected by porosity, σ_k , d_k , A_k , p_k : specific conductivity, thickness, area and porosity of cell /stack component k of repeat unit, j_L : limiting current density.

Due to the resistance overvoltage losses dissipative heat is generated within the cell or stack repeat unit which increases with increasing current density. If this heat loss equals the required thermodynamic entropic heat demand for water splitting (Eq. 4) thermal equilibrium or the so-called "thermo-neutral" operating mode is reached. In this case the electrical energy input equals the molar enthalpy of reaction

$\Delta^r H(T)$ and the resulting thermo-neutral voltage V_m is defined as:

$$V_m(T, p) = \frac{\Delta^r H(T, p)}{2F} \quad [15]$$

At a temperature of $750^\circ C$ the standard molar enthalpy of reaction $\Delta^r H^0(750^\circ C)$ for steam electrolysis (steam partial pressure of 1) is about 248 kJ/mol⁴ which results in a thermo-neutral voltage of 1.28 V. Operating the cell or RU at voltages below the thermo-neutral voltage requires heat input from an external source in order to maintain constant temperature ("endothermal mode"). On the other hand, operation at voltages higher than V_{th} results in resistivity heat losses which are higher than the required standard molar entropic heat demand ("exothermal mode").

The corresponding electrolysis power P or electrolysis power density p of a SOEC stack at a given current I or current density j is formulated by:

$$P(I) = V(I) \cdot I \quad [16]$$

and

$$p(j) = V(j) \cdot j \quad [17]$$

The area specific resistance (ASR), which is dependent on the current or the current density can be determined from the difference in voltage $\Delta V(j)$ divided by the corresponding difference in current density Δj :

$$ASR(j) = \Delta V(j) / \Delta j \quad [18]$$

The reactant gas utilization U_{gas} for water electrolysis is very often called steam conversion (SC) rate. In general U_{gas} is defined as the ratio of current I to the theoretical maximum current I_{theory} . For an SOEC stack with N repeat units I_{theory} can be calculated from the corresponding gas flow rates $f_{i,in}$ (slpm) and molar volume $V_{i,m}$ of the reactant component i in the stack.

$$U_{gas} = \frac{I}{I_{theory}} \times 100\% \quad [19]$$

$$I_{theory} = \frac{F}{N} \cdot \sum_{i=1}^n z_i \times \frac{f_{i,in}}{V_{i,m} \times 60s} \quad [20]$$

The electrical efficiency $\eta_{el,LHV}$ of an SOEC stack is defined as the ratio of the enthalpy or lower heating value (LHV) flow of produced fuel gases $f_{i,out}$ to the electrical power P_{el} consumed by the stack for the electrochemical reaction:

$$\eta_{el,LHV} = \frac{\sum_{i=1}^n LHV_i \times \frac{f_{i,out}}{V_{i,m} \times 60s}}{P_{el}} \quad [21]$$

For laboratory operations usually the electrical power consumed by the steam generator, the gas preheaters, the furnace and the control unit of the test station is not considered. For the calculation of the efficiency of an SOEC system the electrical power consumption of these components has to be taken into account.

The absolute or relative degradation (ΔX , ΔX_{rel}) of a quantity X (e.g. p , V , ASR ...) within the time from t_1 to t_2 is calculated as the difference between the final value $X(t_2)$ and the initial value $X(t_1)$:

$$\Delta X = X(t_2) - X(t_1) \quad [22]$$

and

$$\Delta X_{rel} = \frac{X(t_2) - X(t_1)}{X(t_1)} \times 100\% \quad [23]$$

The absolute or relative degradation rate of quantity X during the time interval $(t_2 - t_1)$ is then calculated by:

$$\frac{\Delta X}{\Delta t} = \frac{\Delta X}{t_2 - t_1} \quad [24]$$

and

$$\frac{\Delta X_{rel}}{\Delta t} = \frac{\Delta X_{rel}}{t_2 - t_1} \quad [25]$$

Absolute degradation rates are typically expressed per hour and relative degradation rates per 1000 hours. For shorter test durations extrapolation of the degradation to 1000 h can be quite inaccurate. In this case it is recommended to express the relative degradation rate according to the performed test duration, e.g. to 500 h, as in the present paper. An entire survey and description of the formulary can be found in the SOCTESQA document: “Test Module 00: General SOC Testing Guidelines”.⁵

Quality Assurance of SOC Cells and Stacks

Only few documents can be found in the literature, which address the description and specification of test procedures for SOFCs and SOECs. The most relevant ones were developed for SOFC in the previous “FCTESQA” project, and of the Technical Committee 105 of the International Electrotechnical Committee (IEC). The latter has published a technical specification⁶ and a terminology document⁷ on SOFC testing. In contrast to SOFC nearly no standardization document for SOEC exists. Some activities have been undertaken in the European funded “RELHY”⁸ and “ADEL”⁹ projects. An entire survey of the existing pre-normative test documents for solid oxide cells is given in Ref. 10.

However, none of these documents includes detailed specifications of SOC cell/stack assembly units, e.g. for dynamic thermal cycling operation or for the combined fuel cell/electrolysis mode. Moreover established advanced characterization techniques, e.g. the electrochemical impedance spectroscopy (EIS) have not yet been integrated thoroughly in the test protocols. Additionally, several issues related to statistical data analysis and data presentation could not be resolved. Only few results were experimentally validated by round robin tests.

Therefore the EU-funded project “SOCTESQA” (Solid oxide cell and stack testing, safety and quality assurance) which started in May 2014 and ended in April 2017 focused on the development of standardized and industry wide test modules and programs for SOC cells/stacks.^{11–15} New application areas which are based on the operation of the SOC cell/stack assembly in the fuel cell (SOFC), in the electrolysis (SOEC) and in the combined SOFC/SOEC modes have been addressed. This covers the wide range of the SOC energy conversion systems, e.g. stationary SOFC μ -CHP, mobile SOFC auxiliary power unit (APU) and SOFC/SOEC power-to-gas systems. Moreover, the project addressed safety aspects, liaised with standardization organizations and established contact with relevant industrial SOFC/SOEC stakeholders. As such, this collaborative project essentially helps to accelerate the development and the market penetration of hydrogen and fuel cell energy systems in Europe.

Altogether 18 test modules (Table I) were identified¹⁶ in order to address the above mentioned energy conversion systems. A “General testing guideline”⁵ and the most important test modules (bold in Table I) were developed in the SOCTESQA project. These are: Start-up,¹⁷ current-voltage characteristics,¹⁸ electrochemical impedance spectroscopy,¹⁹ reactant utilization,²⁰ reactant gas composition,²¹ temperature sensitivity,²² operation under constant current,²³ operation under varying current,²⁴ thermal cycling²⁵ and shut-down.²⁶

The developed test modules are generic and contain all important information which is necessary for the reliable and reproducible testing of cells and stacks. In order to form test programs several of these test modules are connected in series according to the above mentioned energy conversion applications. These application specific test programs were applied by the different project partners (DLR, CEA, DTU, ENEA, JRC, EIFER, NTU) on identical SOC stacks. In this project, short stacks with 5 repeat units (RUs) and fuel gas electrode supported cells (ASC) were used. The short stacks were provided by an established stack supplier (ElringKlinger AG, Germany).

For the development of the test modules several iterative testing campaigns have been performed. In the first step all terms, nomenclatures and specifications of the SOC stacks and test stations were defined. In the next step the test modules were drafted followed by four successive testing campaigns in order to validate and optimize the test modules. These validation processes covered the most important quality assurance aspects, e.g. the evaluation of the results in terms of repeatability of the different testing campaigns and the reproducibility of the results among the partners. Another important issue is the reproducibility of the different test methods, e.g. jV-characteristics, EIS-spectra and high current interruption (HCI). The corresponding standard deviation s of N measured values x_i with a mean value $\bar{x}$ is calculated according to:

$$s = \sqrt{\frac{1}{N} \cdot \sum_{i=1}^N (x_i - \bar{x})^2} \quad [26]$$

The present paper compares the results of the three most important test modules obtained by the different project partners in SOEC mode. These are current-voltage characteristics (TM 03), electrochemical impedance spectroscopy (TM 04) and operation under constant current (TM 12). Electrochemical properties e.g. open circuit voltage (OCV), area specific resistance (ASR), power density, reactant utilization and impedance values of both stack and repeat units (RUs) are presented. Moreover, the results are analyzed and discussed in context to the test input parameters, enabling a so-called sensitivity analysis. The reproducibility of the different methods, e.g. jV-characteristics, EIS and operation under constant current is analyzed. Finally, important aspects for the determination of reproducible degradation rates of SOC stacks will be presented and discussed.

Experimental

For the development of the test procedures it is necessary to use reproducible SOC stacks which fit as best as possible to the test stations of the different test laboratories. Therefore, detailed specification lists of SOC stacks of different stack suppliers²⁷ and of the test stations of the project partners^{28–30} were established and compiled. For the validation of the test modules it was decided to use SOFC/SOEC short stacks in the light weight design with 5 repeat units (RUs). The stack design is based on stamped metal sheet bipolar plates made of ferritic steel “Crofer22APU” from ThyssenKrupp AG.³¹ The stacks were provided by ElringKlinger AG (Germany) and were developed both for stationary and mobile applications, e.g. single-family household

Table I. List of identified and developed (bold) test modules.¹⁶

Test Module 00: General testing guideline

TM 01	Leakage test	TM 10	Pressure sensitivity
TM 02	Start-up	TM 11	Mechanical load sensitivity
TM 03	Current-voltage characteristics	TM 12	Operation under constant current
TM 04	Electrochemical impedance spectroscopy	TM 13	Operation under varying current
TM 05	Current interruption	TM 14	Thermal cycling
TM 06	Cyclic voltammetry	TM 15	Redox cycling
TM 07	Reactant utilization	TM 16	Shut-down
TM 08	Reactant gas composition	TM 17	Vibration test
TM 09	Temperature sensitivity	TM 18	Emergency stop

Figure 2. Experimental setup for electrochemical characterization of SOEC stacks.

systems (μ -CHP) or auxiliary power units (APU). Fuel gas electrode supported cells (Figure 1) with an active area of about 84 cm^2 with Ni-YSZ and LSCF electrodes¹ are integrated in the stacks by high temperature metal brazing. These cells were supplied by CeramTec GmbH (Germany) and have been originally developed for SOFC operation (so-called “anode supported cells”, ASC). For each campaign, all short stacks were fabricated by ElringKlinger AG in one batch, which ensures a high reproducibility with performance differences between the stacks below 5%.²⁷

Figure 2 shows the experimental setup for the electrochemical characterization of the SOC stacks. In the first step, the interfaces between the SOC stack and the test station were specified, ideally as close as possible to the stack hardware boundaries, e.g. stack bottom and top plates. In the next step, the interfaces were classified in media, electric, thermal and mechanic types which are defined by different physical input (yellow) or output (green) parameters. It is very important to control the test input parameters and measure the test output parameters at the interfaces between the stack and the test station as accurately as possible.³² The gas inlet parameters are flow rate, composition, temperature and pressure. The gas inlet and outlet temperatures were measured with thermocouples which are located just below the stack. The stack temperatures were measured with thermocouples attached to the top plate and the bottom plate of the stack. This enables to measure possible temperature gradients over the height of the stack. The temperature of the stack top plate was selected as reference for the stack temperature. The electrical current probes are connected to the top and bottom plates and the voltage probes are attached to the individual repeat units of the stacks.

The stacks were operated in electrolysis mode with furnace temperature of 750°C , a fuel gas composition of 80 vol.% H_2O + 20 vol.% H_2 and air. The flow rates were 0.125 slpm/RU H_2 + 0.5 slpm/RU H_2O for the fuel electrode and 1.0 slpm/RU air. All gases were controlled very accurately by mass flow controllers (MFCs). However, different steam generators (e.g. direct water injection, controlled evaporation mixtures and bubblers) and different water qualities (e.g. multi stage purified and reverse osmosis) were used at the different test laboratories. Moreover, variations in the fuel gas quality ranging from 4.6 to 6.0 grade and in the air quality ranging from grade [1:1:1] to [1:5:1] according to ISO8573-1:2010³³ among the partners laboratories existed. Finally, due to different gas preheating systems the gas inlet temperatures among the different test laboratories varied in the range of 690°C to 750°C (see Figure 4). These differences in stack input parameters of the different laboratories will be discussed below in the results section.

The mechanical load on the stacks was 500 N. For the development of the current-voltage characteristics test module (TM 03) the electrical current was increased and decreased slowly with a rate of 70 mA/s. For SOEC operation a negative current convention is recommended in order to separate from the SOFC operation mode (see Figure 3). Different data sampling rates between 1 s and 30 s were chosen by the different laboratories in order to evaluate the influence of possible

voltage fluctuations on the electrochemical results, e.g. ASR values. The open circuit voltages (OCV), power densities (p), area specific resistances (ASR) and steam conversion rates of the repeat units of the stacks were determined from the current-voltage curves. For the ASR determination the j -V curves were fitted by different methods e.g. the interval method, the global ASR determination, the linear regression and the polynomial regression.³²

The EIS spectra for the development of test module TM 04 (“Electrochemical impedance spectroscopy”) were measured with an impedance analyzer (“Zahner, Type IM6” for instance) in combination with an electronic load. For SOEC a power supply was integrated in the current circuit. In order to minimize high frequency artefacts the electrical current and voltage probes were twisted separately to each other.³⁴ An AC amplitude of 1 A (12 mA/cm^2) with a frequency range of 10 mHz to 500 kHz was applied to the stacks. EIS spectra were recorded near OCV conditions (-12 mA/cm^2) and at moderate DC current density of -240 mA/cm^2 . In order to determine the resistances of the stack components the electrochemical impedance spectra of the stack repeat units were fitted with an equivalent circuit model. The low frequency resistances of the spectra are compared to the ASR values of the j -V-curves. Additionally, the high frequency resistances of the EIS are compared to theoretical ohmic resistance calculations. These calculations are based on adding the ohmic resistances of the components of a repeat unit according to Eq. 12 and Eq. 13.

For the validation of test module “Operation under constant current” the stacks were operated at 750°C with a fuel gas mixture of 80 vol.% H_2O + 20 vol.% H_2 and air as oxidant gas. The long term tests with 500 h duration were performed at -0.5 A/cm^2 . The degradation rates are calculated according to Eq. 22 to Eq. 25.

Results

Current-voltage characteristics.—This section focuses on the quality issues of the j -V-characteristics of the stacks in SOEC mode. Figure 3 shows the j -V-curves and the measured top plate temperatures with increasing current density for the 5-cell short stacks operated at 750°C with a fuel gas composition of 80 vol.% H_2O + 20 vol.% H_2 and air in the different laboratories.

All stacks show the same open circuit voltage (OCV) of about 4.5 V (which corresponds to an average OCV of the RUs of 900 mV). This value is in good agreement with the theoretical reversible voltage of 909 mV calculated with the Nernst equation (Eq. 6) based on a fuel gas composition of 80 vol.% H_2O + 20 vol.% H_2 . Hence, it can be concluded that the irreversible losses η_0 according to Eq. 7 are very low. The maximum variation of the OCV of the different stacks is $\pm 20 \text{ mV}$ or $\pm 0.4\%$. This low value proves a high reproducibility of the steam flow supplied by the steam generators of the

Figure 3. Current-voltage curves of different SOEC stacks at 750°C with 80 vol.% H_2O + 20 vol.% H_2 and air.

different laboratories. Due to the high steam content in the fuel gas the *OCV* is much lower compared to stacks operated in SOFC mode with $H_2 + N_2$ mixture.^{14,35,36} Moreover, the *OCV* in SOEC is much less sensitive toward changes in the fuel gas composition compared to SOFC. Therefore, the stacks have also been operated in SOFC mode, which leads to reproducible stack *OCVs* of 6.0 V or 1.2 V per RU. Hence, a high gas tightness of the different stacks has been proven.¹⁴

The stack voltages increase almost linearly with increasing current density. This can be explained by low activation polarization losses due to the high steam content in the fuel gas. Especially at higher electrical current densities voltage instabilities occur, which is explained in detail below. The best performance was achieved by stack #1 with an electrolysis voltage of about 6.6 V at a current density of -770 mA/cm^2 . At this operating point the stack electrolysis power is -427 W which corresponds to a power density of -880 mW/cm^2 of the RUs and a high steam conversion rate of 90%. The electrical stack efficiency was 70.6%. Especially at higher electrical current densities differences among the stacks can be observed. Stacks #2, #4, and #5 have almost the same behavior whereas the performance of stack #3 is slightly lower. The maximum difference in stack electrolysis voltage at a current density of -500 mA/cm^2 relative to the average value of 5.80 V is 103 mV or 1.8%. The standard deviation among the stacks is only 72 mV which proves a high reproducibility. This high reproducibility was also confirmed in SOFC operation of the stacks, where power densities of the RUs with 50 vol% $H_2 + 50 \text{ vol}\% N_2$ at 500 mA/cm^2 between 400 mW/cm^2 and 415 mW/cm^2 were measured.¹⁴ The differences in SOEC and SOFC are far below the performance quality threshold value of 5% given by the stack supplier.²⁷

It is very likely that the variation in electrolysis performance can be explained by differences of stack temperatures. Figure 3 shows that the top plate temperature of stack #1 is higher compared to the other stacks, which explains its better performance. However, among the other stacks no clear correlation between performance and stack temperature can be observed. Therefore, in the next step the performance data were correlated to the measured gas inlet temperatures. Figure 4 shows the stack electrolysis voltage at -500 mA/cm^2 as a function of the air inlet temperature (color filled dots) and the fuel gas inlet temperature (white filled dots).

Similar to SOFC operation¹⁴ Figure 4 shows that the gas inlet temperatures have a significant influence on electrolysis performance of the stacks. High gas inlet temperatures result in low electrolysis voltages and vice versa. This correlates very well with the decrease of the resistances of the stack repeat units with increasing temperature (Eqs. 9 and 10).³⁶ No difference between the influence of the air inlet temperature and the fuel gas inlet temperature on the electrolysis voltage can be identified. For a reliable and reproducible jV-characterization

Figure 4. Stack electrolysis voltage at -500 mA/cm^2 as a function of the air inlet (filled dots) and fuel gas inlet (open dots) temperatures.

of SOEC stacks it is obligatory to accurately monitor the gas inlet temperatures.

Moreover, the temperatures of all stacks decrease with increasing current density. This behavior is caused by the endothermic reaction of the water electrolysis according to Eq. 4, whereas in SOFC a continuous increase in temperature is observed.³⁵ In Figure 3 all temperature curves show local minima at current densities around -550 mA/cm^2 . Beyond this current density the stack temperature increases again, which can be explained by the higher dissipative heat generation within the stacks. According to the theory the stack temperature at thermo-neutral voltage (Eq. 15) should be the same as at *OCV*. Figure 4 shows that none of the stacks reaches this temperature. At the highest current densities the electrolysis stack voltages are about 6.5 V, which corresponds to 1.3 V per repeat unit. However, this value is almost the theoretical thermo-neutral voltage of 1.28 V. This discrepancy can be explained by two reasons. Firstly, the temperature measured at the stack top plate does not reflect the real temperature inside the stack. In SOFC for example, measured temperatures inside the stacks of other research groups^{37,38} are much higher compared to the temperatures of the stack end plates. Secondly, the thermal inertia of the stack due to its specific heat capacity has to be taken into account. This thermal inertia can cause differences of the electrochemical stack or cell behavior with increasing and decreasing electrical current. Figure 5 shows the resulting hysteresis between increasing and decreasing jV-curves of stack #2. The corresponding current change rate was 70 mA/s resulting in an overall recording time for the jV-curve of 30 min.

Figure 5 shows that the hysteresis in stack voltage is negligible whereas the stack top plate temperatures with increasing and decreasing current densities are quite different. At current densities below -400 mA/cm^2 the temperatures with increasing current density are higher compared to the temperatures with decreasing current density. At high current densities this behavior is vice versa. This rather complex temperature behavior is caused by the endothermic H_2O splitting reaction, the dissipative heat generated by the resistances of the stack repeat units and the thermal inertia of the stack itself. In order to minimize the hysteresis behavior it is recommended to measure jV-curves with low electrical current increase/decrease change rates, thus reaching quasi-thermal equilibrium at each current step.

Figure 6 shows the jV-curves for repeat unit 3 of the short stacks with increasing current density operated at 750°C with a fuel gas composition of 80 vol.% $H_2O + 20 \text{ vol}\% H_2$ and air in the different laboratories. These RUs in the middle of the short stacks were selected in order to evaluate the reproducibility of the results of the jV-characterization and of the corresponding electrochemical impedance spectra (see below).

Figure 5. Increasing and decreasing current-voltage curves of SOEC stack #2 at 750°C with 80 vol.% $H_2O + 20 \text{ vol}\% H_2$ and air.

Figure 6. Current-voltage curves of RU 3 of different SOEC stacks at 750°C with 80 vol.% H₂O + 20 vol.% H₂ and air.

Area specific resistance (ASR).—Almost all current voltage curves in Figure 3 show voltage fluctuations which are caused by instable vaporization of water in the steam generator. This low signal-to-noise ratio (SNR) is a well-known issue in SOEC operation. The voltage fluctuations increase with increasing current density. Differences between the stacks can be explained by different humidification systems, e.g. controlled evaporation of water-gas stream mixtures or water bubbles, and different data logging rates. For example, the data logging rate of stack #2 with high voltage instabilities was 10 times faster compared to stack #3. The voltage fluctuations of the repeat units of the measured SOEC stacks can reach maximum values of nearly 40 mV. It is recommended to minimize the voltage instabilities by optimization of the humidifier operating parameters, e.g. the vaporization temperature, the water quality and the carrier gas composition for the steam.

However, the voltage fluctuations have a strong impact on the ASR determination according to Eq. 18. The four mostly used ASR calculation procedures are the interval method, the global ASR determination, the linear regression and the polynomial regression. Figure 7 shows the comparison of the ASR values of stack #2 which were determined with the different methods from the jV-curve of Figure 5.

The most common method for the determination of the ASR values is the interval method. Two specific points are selected in the current voltage curve in order to calculate the slope of the curve at a specific current density. In the case of this stack, both a small interval of 25 mA/cm² and a large interval of 100 mA/cm² were selected at a current density of -240 mA/cm². The corresponding ASR values are 2.76 Ω·cm² and 4.80 Ω·cm². In this case, the above mentioned fluctuations of the stack voltage cause significant different ASR values. Therefore, this method is not suitable for the reliable determination of the ASR.

An alternative method is the enlargement of the interval. For the so-called global ASR the interval is spanned over the entire j-V curve so that the error of the interval method is reduced as best as possible. This method leads to an ASR of 2.88 Ω·cm². Another method which is very similar to the global ASR is the linear regression which is mathematically fitted to the j-V-curve. The corresponding algorithms are already integrated in most of the established software programs, e.g. “Microsoft Excel” or “OriginLab”. The ASR can be calculated with the corresponding linear equation, which has a result in this case of 3.0 Ω·cm². However, both methods do not take care of the non-linearity of the j-V curve and are susceptible to errors the more the entire characteristic curve is non-linear.

The last method is the ASR determination by a regression using a polynomial. In contrast to the linear regression the j-V curve is fitted through a higher degree equation with the polynomial regression, usually a so-called “Taylor” polynomial. This enables also to describe

Figure 7. Comparison of ASR values of the stack #2 from current-voltage curve of Figure 5 at a current density of -240 mA/cm² determined with different methods.

non-linear j-V curves. The corresponding equation for describing the j-V curve of stack #2 of Figure 5 is:

$$V(\text{Stack}) = 5.1298 \cdot 10^{-17} \cdot j^6 + 8.9313 \cdot 10^{-14} \cdot j^5 + 6.1229 \cdot 10^{-11} \cdot j^4 + 2.2512 \cdot 10^{-8} \cdot j^3 + 4.8456 \cdot 10^{-6} \cdot j^2 - 2.5013 \cdot 10^{-3} \cdot j^1 + 4.4875 \quad [27]$$

The ASR can be determined by calculation of two points of the j-V curve with the polynomial equation and calculating the slope within the corresponding interval. Figure 7 shows the corresponding ASR values at -240 mA/cm² with a small and a large interval of 25 mA/cm² and 100 mA/cm², respectively. The ASR values are 3.17 Ω·cm² and 3.15 Ω·cm², which reflects a high accuracy of this method. Therefore, it is recommended to use the polynomial regression method for the reliable and reproducible determination of ASRs of SOEC cells and stacks.

Electrochemical impedance spectroscopy.—Figure 8 shows the electrochemical impedance spectra of repeat unit 3 of stacks #1, #2 and #3 at low DC current density of -12 mA/cm², 750°C and process gases of 80 vol.% H₂O + 20 vol.% H₂ and air. The results of the impedance spectra of RU 3 show a good consistency among the different laboratories. The spectra of RU 3 of stacks #1 and #2 are quite uniform whereas stack #3 differs from them. This is in good agreement with the above mentioned lower electrolysis performance of stack #3 observed in the jV-curves in Figure 6.

In the Nyquist diagram the high frequency impedances R_{HF} at about 10 kHz represent the ohmic resistances of the RUs. Usually the inter-

Figure 8. Impedance spectra of RU 3 of stacks measured in different laboratories at 750°C, -12 mA/cm² with 80 vol.% H₂O + 20 vol.% H₂ and air.

cept of the x-axis is used for determining R_{HF} . The ohmic resistances of all three RUs are approximately 190–200 $\text{m}\Omega\cdot\text{cm}^2$ and therefore quite reproducible. Hence, the contact resistances of the cells in the stacks are very low and no detrimental secondary phases were formed during cell / stack fabrication or during operation. This is in good agreement with results obtained in SOFC mode at *OCV* and at the same temperature.^{35,36} Especially in the high frequency range signal disturbances are often observed and reported in literature.³⁴ These artefacts were eliminated in the measured spectra by minimization of the inductive interferences by twisting both the current probes of the stack and the voltage probes of the RUs to each other (Figure 2).

Moreover, the ohmic resistances were validated with theoretical calculations with the ohmic layer model according to Eq. 12 and Eq. 13. With this model the resistances of the components of a repeat unit are added. Figure 9 shows the scheme of a stack repeat unit with the individual components, namely Crofer interconnect sheet, Cr-Mn-spinel oxide layer, Cr-evaporation protection coating, air electrode contact layer, LSCF air electrode, doped CeO_2 diffusion barrier layer, YSZ electrolyte, Ni-YSZ fuel electrode, Ni-YSZ substrate support and fuel electrode contact Ni-mesh. The thickness and porosity of the stack repeat unit components are added in brackets. The theoretical calculation of the ohmic resistance along the current path in Figure 9 results in a value of 180 $\text{m}\Omega\cdot\text{cm}^2$. This value is slightly smaller compared to the measured ones in Figure 8 which can be explained by the assumption of ideal interfaces between the different layers of the components of the repeat unit. However, post-test analysis of stacks operated in SOFC at DLR³⁹ and results of other research groups⁴⁰ have shown that especially the electrical contact of the air electrode may contain additional ohmic contributions. In this context the material and structure of the air electrode contact paste can play an important role.^{41–43}

Altogether three frequency dependent processes can be identified in the impedance spectra in Figure 8. These three impedances can hardly be seen as depressed half-circles and are similar to those observed in SOFC.^{35,36,44,45} Variations of the fuel and air electrode gas compositions have shown, that the steam reduction to H_2 at the fuel electrode occurs at high frequencies (500 Hz–2 kHz) and that the oxidation of O^{2-} -ions to O_2 at the air electrode can be attributed to the middle frequency range (50 Hz–500 Hz). This is consistent with the results of other research groups.^{46–49} However, due to the strong overlapping it is difficult to separate the impedances of the fuel and air electrode processes from each other. In Fig. 8 the electrodes impedance of stack #3 seems higher compared to stack #1 and stack #2. It is therefore very likely that the lower performance of RU 3 of stack #3 in Figure 6 is caused by higher electrodes resistances due to lower electrocatalytic activity.

Figure 9. Scheme and electrical current path across a stack repeat unit with the individual components for the ohmic layer model.

The impedance arc at low frequencies (20 mHz–500 Hz) can be attributed to diffusive and convective gas phase transport processes in the fuel electrode support substrate and/or in the fuel electrode itself.^{32,45–47} Similar to SOFC, this impedance strongly depends on the fuel gas composition and the fuel gas velocity^{50,51} and has a minimum at 50% steam in the fuel gas.⁵² The gas concentration impedances of the spectra in Fig. 8 are nearly the same. The observed small differences can be explained by variations in the fuel gas composition of the different test stations supplied to the stacks. In contrast to the fuel electrode, the gas concentration/transport impedance at the air electrode is usually negligible and can only be observed in the case of very low air flow rates.

The overall resistances of the repeat units 3 at low frequencies near the x-axis vary between 460 $\text{m}\Omega\cdot\text{cm}^2$ and 500 $\text{m}\Omega\cdot\text{cm}^2$. The higher value of the RU 3 of stack #3 can be attributed to the above described higher electrodes polarization impedance. In contrast to SOFC operation nearly all spectra in Fig. 8 show instabilities especially in the low frequency range. As mentioned above in the jV-curves these instabilities are caused by fluctuations of the steam supply of the fuel gas humidification systems. These steam fluctuations have a high impact on the low frequency instabilities of the spectra and can reach levels which make the spectra unusable for evaluation with an equivalent circuit. It is therefore necessary to minimize voltage fluctuations by stabilizing the steam supply of the steam generator as best as possible.

In Figure 10 the EIS spectra of the RUs 3 of the three stacks at a higher current density of $-240 \text{ mA}/\text{cm}^2$ are shown. Figure 10 shows that the ohmic resistances under electrical current load are about 10 $\text{m}\Omega\cdot\text{cm}^2$ higher compared to the spectra near *OCV*. Since the ohmic resistance only depends on temperature this increase can easily be explained with the above mentioned electrical current induced temperature decrease of the repeat units (Figure 3) due to the endothermic reaction. For all three RUs 3 an increase of electrodes impedances (50 Hz–2 kHz) with increasing current density can be observed. This is in good agreement with the results of other research groups.⁴⁶ The RU 3 of stack #3 shows the strongest electrodes polarization impedance increase. In contrast to that are the gas concentration impedances of the fuel electrodes decreased under an electrical current load of $-240 \text{ mA}/\text{cm}^2$. This is in good agreement with the above described strong reduction of this impedance element with decreasing H_2O -concentration till 50% of steam in the fuel gas.⁵² The calculated steam conversion rate at $-240 \text{ mA}/\text{cm}^2$ is 28% which results in an overall fuel composition of about 43% H_2 and 57% H_2O . Hence, this is nearly the operating point where the minimum in the gas concentration impedance is reached.

The overall resistances of the repeat units of stack #1 and stack #2 at $-240 \text{ mA}/\text{cm}^2$ are almost the same as at *OCV*, which is in good agreement with the linear jV-behavior in Figure 3. Due to the higher electrodes polarization resistance the overall impedance of the RU 3 of stack #3 is about 60 $\text{m}\Omega\cdot\text{cm}^2$ higher compared to the spectrum at *OCV*. This is consistent with the highest ASR value of stack #3 in Figure 3.

Figure 10. Impedance spectra of RU 3 of different stacks at 750°C, $-240 \text{ mA}/\text{cm}^2$ with 80 vol.% H_2O + 20 vol.% H_2 and air.

Reproducibility of different resistance determination methods.—

In order to prove the reproducibility of the different characterization methods, the results of the EIS spectra are compared to the j-V curves. Figure 11 shows the correlation between the low frequency impedances R_{LF} of the EIS and the corresponding ASR values of some stack repeat units from the j-V curves. The ASR values were determined with the above mentioned polynomial regression. For the validation of the reproducibility of both methods results at very low current density of -12 mA/cm^2 (filled dots) and higher current density of -240 mA/cm^2 (open dots) were used.

In Figure 11 most R_{LF} and ASR values are in the range $425 \text{ m}\Omega\cdot\text{cm}^2$ to $575 \text{ m}\Omega\cdot\text{cm}^2$. The values at -240 mA/cm^2 tend to be higher than the values at -12 mA/cm^2 , which is consistent with the above described lower temperature at higher current densities. For a high reproducibility of both methods the R_{LF} and ASR values should be the same, reflected in Figure 11 with the theoretical dashed line. Figure 11 shows that most values are located very near to this theoretical ideal line. The overall standard deviation from the ideal value is $12.3 \text{ m}\Omega\cdot\text{cm}^2$, which corresponds to 2.5%. This proves a high reproducibility of the two EIS and j-V methods in terms of determination of the overall resistance. In this context, especially the optimization of the EIS setup (Figure 2) and the determination of the ASRs by polynomial regression (Eq. 27) of the j-V curves have been very efficient. Moreover, the ΔV and Δj intervals for calculation of the ASR values from the j-V curves have to be in the same range as the alternating ΔV or Δj excitation amplitude of the EIS. Hence, a high reproducibility between jV-characterization and EIS is obtained by consequent application of the above mentioned pre-normative test procedures “Test Module 03: Current-voltage characteristics”¹⁸ and “Test Module 04: Electrochemical impedance spectroscopy”¹⁹ of the European project SOCTESQA.

Operation under constant current.—Figure 12 presents the electrolysis voltages of three stacks (#10, #11, #12) tested in different laboratories recorded during 500 h of operation. The stacks were operated in SOEC mode at 750°C with a fuel gas mixture of 80 vol.% H_2O + 20 vol.% H_2 and air at a current density of -0.5 A/cm^2 . The data acquisition period was 300 s.

Figure 12 shows that the applied electrical current value of stack #11 and stack #12 is very reproducible and stable at -0.5 A/cm^2 during the complete SOEC operation. In contrast, lower electrical current stability with a maximum deviation of $\pm 0.9 \text{ mA/cm}^2$ ($\pm 0.18\%$) is observed for stack #10. The electrolysis voltage evolution of all three stacks with time is similar. Stack #12 shows slightly lower degradation compared to the other two stacks. Moreover, strong differences in the voltage stabilities can be observed, which are caused by the above mentioned fluctuations of the steam supply of the different test stations. The highest voltage instabilities are observed for stack #10 whereas stack #12 shows the highest signal-to-noise ratio.

Figure 11. Correlation between low frequency impedances of EIS and ASR values of j-V curves of RUs of three different stacks at -12 mA/cm^2 and -240 mA/cm^2 .

Figure 12. Long term behavior of three stacks of different laboratories in SOEC mode operated for 500 h at 750°C , 80 vol.% H_2O + 20 vol.% H_2 and -0.5 A/cm^2 .

For the calculation of the degradation rates according to Eq. 24 and Eq. 25 it is necessary to select representative voltage values $V(t_2)$ and $V(t_1)$. Similar to the jV-curves and the EIS spectra voltage fluctuations due to instable steam supply can cause great errors in the degradation values. Therefore it is obligatory to eliminate the voltage instabilities either by manually averaging several voltage data or by mathematical linear or polynomial regression of the long term data. The corresponding formula and degradation rates are listed in Table II.

Table II shows that the degradation rates of stack #10 and stack #11 are quite similar with values between $290 \mu\text{V/h}$ and $300 \mu\text{V/h}$. This corresponds to a relative degradation value of about 2.5%/500 h. For these two stacks all three degradation calculation methods result in reproducible degradation values, which can be explained by the almost linear increase of the electrolysis voltage during the 500 h of operation. In contrast, stack #12 shows a lower degradation of about $80 \mu\text{V/h}$ compared to the other two stacks. Additionally, a non-linear degradation behavior can be observed in Figure 12. Therefore, the degradation value with the linear regression calculation method of $91 \mu\text{V/h}$ is not accurate and not applicable for this stack. With the averaging and the polynomial regression methods almost reproducible degradation values between 0.67%/500 h and 0.7%/500 h are obtained. Hence, similar to the above described ASR determination from the jV-curves, it is recommended to use the polynomial regression method for degradation calculation of long term operation tests which contain voltage instabilities.

The differences in the degradation rates between the three stacks can be explained by different qualities or purities of the process gases and of the used water for the steam fed to the stacks. For stack #12 multi stage purified water with low contaminants, H_2 with purity grade 5.0 and compressed air with purity class [1:5:1] according to ISO8573-1:2010³³ were used. In contrast, for stack #10 and stack #11 steam from reverse osmosis purified water, H_2 with purity grade 6.0 (stack #10) and 5.0 (stack #11) and air with quality class of [1:1:1] (stack #10) and [1:4:1] (stack #11) were supplied to the stacks. It is very likely that the higher level of contaminants in the steam of stack #10 and stack #11 has a significant influence on the higher degradation rates observed in Figure 12. This assumption is confirmed by other research groups, which report an increase in degradation rates of SOECs with higher impurities in the fuel gas.⁵³ However, no correlation between air quality and degradation rate can be observed in the durability test in Figure 12, which is in contrast to other literature results.⁵⁴ This discrepancy can be explained by the limited test durability of only 500 h. Therefore, ongoing activities concentrate on testing of SOEC stacks with much longer durability of more than 5000 h.⁵⁵

Reproducibility of different degradation determination methods.—The following section addresses the reproducibility of different degradation determination methods. For this purpose the

Table II. Degradation of SOEC stacks calculated with different mathematical methods.

Stack	Calculation method	Equation	Degradation ($\mu\text{V/h}$)	Degradation (%/500h)
Stack #10	averaging	$\bar{V}(t) = \frac{1}{n} \cdot \sum_{i=0}^n V(ti)$	291.4	2.50%
	linear	$V(t) = 2.8784E-4 \cdot t + 5.8747$	287.8	2.45%
	polynomial	$V(t) = -6.0072E-17 \cdot t^6 + 8.5546E-14 \cdot t^5 - 4.4255E-11 \cdot t^4 + 9.9648E-9 \cdot t^3 - 1.0612E-6 \cdot t^2 + 3.9029E-4 \cdot t + 5.8649$	288.4	2.46%
Stack #11	averaging	$\bar{V}(t) = \frac{1}{n} \cdot \sum_{i=0}^n V(ti)$	294.0	2.46%
	linear	$V(t) = 2.9918E-4 \cdot t + 5.9401$	299.2	2.50%
	polynomial	$V(t) = 1.3611E-16 \cdot t^6 - 2.1602E-13 \cdot t^5 + 1.3108E-10 \cdot t^4 - 3.7432E-8 \cdot t^3 + 4.8369E-6 \cdot t^2 + 1.0452E-4 \cdot t + 5.9383$	301.5	2.52%
Stack #12	averaging	$\bar{V}(t) = \frac{1}{n} \cdot \sum_{i=0}^n V(ti)$	80.1	0.67%
	linear	$V(t) = 9.1004E-5 \cdot t + 5.9305$	91.0	0.76%
	polynomial	$V(t) = -2.8490E-16 \cdot t^6 + 4.4262E-13 \cdot t^5 - 2.6108E-10 \cdot t^4 + 7.0983E-8 \cdot t^3 - 7.9711E-6 \cdot t^2 + 1.7716E-4 \cdot t + 5.9591$	84.1	0.70%

above reported degradation values from the long term operation under constant current are compared with the degradation rate determined by jV-characterization. Figure 13 shows the comparison of the voltage degradation values of the three stacks of Figure 12 determined by steady state operation and jV-characterization. The values of both methods are almost the same, showing a high reproducibility of both methods. The observed differences can be explained by different stack temperature evolution of both methods. In steady state operation the stack achieves thermal equilibrium whereas in jV-characterization this is not the case (see Figure 5). This issue can be minimized by recording jV-curves with very low electrical current increase/decrease change rates.

Summary

The present paper presents the most important aspects and recommendations for quality assurance of SOEC stacks by application of electrochemical test modules. The results of the most important test modules, particularly current-voltage characteristics, electrochemical impedance spectroscopy and operation under constant current, obtained at different test laboratories are compared and discussed. In the first step it is essential to properly define and control all interfaces between SOEC stack and test station. The most important input parameters in terms of performance are the stack temperature and the fuel gas and air inlet temperatures. High gas inlet temperatures result in low electrolysis voltage and vice versa. For jV-characterization a rather complex temperature behavior with endothermic, thermoneutral and

exothermic region is observed in electrolysis mode. It is recommended to measure jV-curves with low electrical current increase/decrease change rate in order to minimize the hysteresis behavior by reaching quasi-thermal equilibrium at each current step. Another important test input parameter in SOEC operation is the stability of the steam supply of the steam generator. Steam supply instabilities lead to voltage fluctuations which have a strong impact on the quality of the jV-curves, EIS spectra and long term operation. Therefore, it is recommended to stabilize the steam supply as best as possible, e.g. by accurate control of the temperature of the steam subsystem and/or to add N_2 to the $\text{H}_2\text{O}+\text{H}_2$ fuel gas mixture. Especially the ASR determination from jV-curves is very sensitive on voltage instabilities and can be overcome most successfully by polynomial regression method.

For electrochemical impedance spectra possible high frequency artefacts, which are caused by inductance interference of the connected current and voltage wires can be minimized by twisting the corresponding wires to each other. The results of the EIS spectra show a good consistency among the different test laboratories. Moreover, the ohmic and overall resistances of the spectra correspond very well to theoretical calculations and to the ASR values of the jV-curves, respectively. Hence, by consequent application of the pre-normative test modules “TM03: Current-voltage characteristics” and “TM04: Electrochemical impedance spectroscopy” a high reproducibility between both test methods as well as between different laboratories is obtained.

For the reliable determination of degradation rates a very high stability and monitoring accuracy of all input parameters during long term test is obligatory. Moreover, similar to the above described ASR determination from the jV-curves, it is recommended to use the polynomial regression method for degradation calculation of long term operation tests which contain voltage instabilities. The degradation values calculated from different test methods, e.g. jV-curves and steady state long term operation, are in good agreement, which proves the reproducibility of the applied test modules. The observed differences can be explained by different stack temperature evolution of both methods.

The results have already been transferred to standards developing organizations, e.g. the International Electrotechnical Commission (IEC). The usage of the corresponding standard documents⁵⁶ will be very beneficial for the improvement of the quality assurance of the SOFC/SOEC technology, thus promoting its successful introduction and acceptance into the global energy market.

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Figure 13. Degradation rate of three stacks of different laboratories determined by steady state operation and jV-characterization operated for 500 h in SOEC mode at 750°C, 80 vol.% H_2O + 20 vol.% H_2 and -0.5 A/cm^2 .

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