Solid Oxide Cell and Stack Testing, Safety and Quality Assurance

Collaborative Project - *FCH JU GRANT AGREEMENT N° 621245*
THEME [SP1-JTI-FCH.2013.5.4]
***Start date**: 01.05.2014 – **Duration**: 36 months*
***Project Coordinator**: M. Lang – DLR*

Test Module 13: Operation under Varying Current

Responsible Author	Karine Couturier (CEA)
Co-Author(s)	Xiufu Sun (DTU) Carlos Boigues-Munos (ENEA) Michael Lang (DLR)

Version: 3.5
Last revision: 02.06.2017

Content

Abbreviations

APU	Auxiliary power unit
ASR	Area specific resistance
CHP	Combined heat and power
nlpm	Normal litr per minute
OCV	Open circuit voltage
RU	Repeating unit
SOC	Solid oxide cell
SOFC	Solid oxide fuel cell
SOEC	Solid oxide electrolysis cell
slpm	Standard litre per minute
TIP	Test input parameter
TM	Test module
TOP	Test output parameter

TM 13 – Operation under Varying Current

1 Objective and Scope

Test Module 13 (TM13) deals with solid oxide cell (SOC) operation under varying current when in galvanostatic conditions. The present document is valid for both fuel cell (SOFC) and electrolyser (SOEC) modes of the cell/stack. SOC assembly units are expected to operate under various loads depending on the electrical, thermal and/or gas demands. Besides the direct coupling with power generators relying on renewable energy sources might generate intermittent load patterns. The aim of this test module is to establish a widely accepted method for SOC dynamic operation under relevant load profiles. A calculation method of the SOC degradation rate is also recommended from the continuous SOC voltages recording as a function of time. The particular interest of this method consists in achieving the voltage evolution/dependency over time and so to know if SOC performance evolves steadily or not. Other TMs, such as TM03 “Current-voltage characteristics” and TM04 “Electrochemical impedance spectroscopy”, also allow to calculate SOC degradation rates on resistances in particular. It is necessary to provide different representations of degradation in order to give a comprehensive representation of the cell/stack assembly unit durability.

This test module addresses SOC cell/stack assembly units, testing systems, instruments and measuring and test methods. Moreover, it is also applicable to the four SOC applications selected in the SOCTESQA project, which will strongly influence the load patterns to be tested:

- Stationary and distributed power generation (SOFC-µCHP),
- Mobile (SOFC-APU),
- H_2 production in power-to-gas (SOEC),
- Electricity storage in power-to-gas-to-power (SOEC/SOFC).

Definition of relevant load patterns related to these applications is supported by recommendations of the SOCTESQA Industrial Advisory Board members. This test module follows the recommendations of the IEC international standard [1] when applicable. All the quantities used in TM13 are defined with their symbols and units in the section 7 of TM00 “General SOC testing guidelines”. The test object for which this TM applies is also described in section 5 of the master document TM00.

2 Test Equipment and Set-up

This part is fully detailed in section 6 of the master document TM00. A complete test system is described with all its different subsystems around as well as the interfaces between the test object and the test system. Some figures are given showing the consequent test input and output parameters locations on the test object as well as their measurement method and accuracy. Finally some advice is supplied in regard to the mounting of the test object in the test system and to the quality of the test environment.

3 Test Input Parameters (TIPs)

There are two types of test input parameters: variable and static. The first type may vary during the duration of the TM while the second type does not vary during the overall duration of the TM. The

SOC assembly units are usually operated in galvanostatic mode (at a constant current for a given operating point).

Table 1 and *Table 2* below list all the relevant test input parameters (operating conditions) useful in TM13. T_{oven} (as well as $T_{PH,neg}$ *and* $T_{PH,pos}$ when available) is controlled and kept constant all along the TM according to the targeted initial operating T_{cell} or T_{stack} (either at OCV before increasing current or at the desired operating current at a chosen current plateau). $T_{neg,in}$ and $T_{pos,in}$ can vary as well as T_{cell} or T_{stack} during the TM. $f_{neg,in}$, $f_{pos,in}$, $f_{i,neg,in}$ and $f_{i,pos,in}$ gas flow rates are also controlled to fix the mole fractions of gas components $x_{i,neg,in}$ and $x_{i,pos,in}$ in each chosen operating mode and in association with the current I, to achieve the targeted gas utilizations at both electrodes ($U_{gas,neg}$ and $U_{gas,pos}$) and the initial voltage as well (initial values of V_{cell}, V_{stack} and $V_{RU,av}$). Then gas flow rates and mole fractions are variable TIPs depending on the tested operating mode (SOFC, SOEC or combined SOFC/SOEC). The absolute current $|I|$ is progressively increased to its first target value with a rate $\Delta I/\Delta t$ to be defined for each application, maintained to this value for a certain operating time $t_{op,1}$ and varied with the same rate to a second target value for another operating time $t_{op,2}$. A load cycle can comprise several plateaus at different current values and is then repeated m times.

Table 1: Static test input parameters during TM13

Description of quantity	**Symbol**	**Unit often used**	**SI unit**
Temperature of the oven	T_{oven}	°C	K
Temperature of the pre-heaters for preheating the negative electrode gas stream	$T_{PH,neg}$	°C	K
Temperature of the pre-heaters for preheating the positive electrode gas stream	$T_{PH,pos}$	°C	K
Rate of current change	$\Delta I/\Delta t$	A s^{-1}	A s^{-1}
Operating time of the plateau *d*	$t_{op,d}$	s, min, h	s
Number of plateaus per cycle	d	-	-
Number of cycles	m	-	-

Table 2: Variable test input parameters during TM13

Description of quantity	**Symbol**	**Unit often used**	**SI unit**
Flow rate of the negative electrode gas stream at cell/stack inlet	$f_{neg,in}$	nlpm, slpm l_n min^{-1}, l_s min^{-1}	m^3 s^{-1}
Flow rate of the positive electrode gas stream at cell/stack inlet	$f_{pos,in}$	nlpm, slpm l_n min^{-1}, l_s min^{-1}	m^3 s^{-1}
Flow rate of component *i* in the negative electrode gas stream at cell/stack inlet	$f_{i,neg,in}$	nlpm, slpm l_n min^{-1}, l_s min^{-1}	m^3 s^{-1}
Flow rate of component *i* in the positive electrode	$f_{i,pos,in}$	nlpm, slpm	m^3 s^{-1}

gas stream at cell/stack inlet		l_n min^{-1}, l_s min^{-1}	
Mole fraction of component *i* in the negative electrode gas stream at cell/stack inlet	$x_{i,neg,in}$	-	-
Mole fraction of component *i* in the positive electrode gas stream at cell/stack inlet	$x_{i,pos,in}$	-	-
Electrical current through the cell/stack	I	A	A

4 Test Output Parameters (TOPs)

Table 3 below shows the list of the test output parameters that are recorded during the overall duration of TM13, mainly the cell/stack assembly unit response e.g. V_{cell}, V_{stack} and $V_{RU,i}$ SOC voltages. In association with the current I and for long-term endurance tests, those TOPs allow the calculation of the SOC cell/stack performance degradation rates on voltage for each current plateau and as described in the master document TM00 (section 10).

T_{cell}, T_{stack}, T_{TP}, T_{BP}, $T_{neg,in}$, $T_{pos,in}$, $T_{neg,out}$ and $T_{pos,out}$ are significant TOPs to be measured when analyzing the stack behavior under stationary and transient thermal conditions. Moreover, these TOPs allow to check the good course of the test (mainly gas tightness keeping in addition to OCV value). $f_{neg,out}$, $f_{pos,out}$, $f_{i,neg,out}$ and $f_{i,pos,out}$ as well as $x_{i,neg,out}$, $x_{i,pos,out}$, $p_{i,neg,out}$ and $p_{i,pos,out}$ are also relevant TOPs to be followed in order to check the performance and healthiness of the stack, especially when analyzing hydrogen production in SOEC operation and gas tightness.

Table 3: Test output parameters during TM13

Description of quantity	**Symbol**	**Unit often used**	**SI unit**
Voltage of the cell	V_{cell}	V	V
Voltage of the stack	V_{stack}	V	V
Voltage of the repeating unit (RU) *i* in the stack	$V_{RU,i}$	V	V
Temperature of the cell	T_{cell}	°C	K
Stack temperature	T_{stack}	°C	K
Temperature of the top plate of the stack	T_{TP}	°C	K
Temperature of the bottom plate of the stack	T_{BP}	°C	K
Temperature of the negative electrode gas stream at cell/stack inlet	$T_{neg,in}$	°C	K
Temperature of the positive electrode gas stream at cell/stack inlet	$T_{pos,in}$	°C	K
Temperature of the negative electrode gas stream at cell/stack outlet	$T_{neg,out}$	°C	K
Temperature of the positive electrode gas stream at cell/stack outlet	$T_{pos,out}$	°C	K

Flow rate of the negative electrode gas stream at cell/stack outlet	$f_{neg,out}$	nlpm, slpm l_n min^{-1}, l_s min^{-1}	m^3 s^{-1}
Flow rate of the positive electrode gas stream at cell/stack outlet	$f_{pos,out}$	nlpm, slpm l_n min^{-1}, l_s min^{-1}	m^3 s^{-1}
Flow rate of component *i* in the negative electrode gas stream at cell/stack outlet	$f_{i,neg,out}$	nlpm, slpm l_n min^{-1}, l_s min^{-1}	m^3 s^{-1}
Flow rate of component *i* in the positive electrode gas stream at cell/stack outlet	$f_{i,pos,out}$	nlpm, slpm l_n min^{-1}, l_s min^{-1}	m^3 s^{-1}
Mole fraction of component *i* in the negative electrode gas stream at cell/stack outlet	$x_{i,neg,out}$	-	-
Mole fraction of component *i* in the positive electrode gas stream at cell/stack outlet	$x_{i,pos,out}$	-	-
Partial pressure of component *i* of the negative electrode gas stream at cell/stack outlet	$p_{i,neg,out}$	mbar, kPa	N m^{-2} (Pa)
Partial pressure of component *i* of the positive electrode gas stream at cell/stack outlet	$p_{i,pos,out}$	mbar, kPa	N m^{-2} (Pa)

5 Derived quantities

The following *Table 4* gives the derived quantities useful for this TM. For each current plateau, they are all calculated from TIPs and TOPs with the equations presented in TM00 - section 10.

Table 4: Calculated derived quantities during TM13

Description of quantity	**Symbol**	**Unit often used**	**SI unit**
Electrical current density through the cell/stack	j	A cm^{-2}	A m^{-2}
Gas utilization at the negative electrode	$U_{gas,neg}$	%	-
Gas utilization at the positive electrode	$U_{gas,pos}$	%	-
Average RU voltage of all RUs in the stack	$V_{RU,av}$	V	V
Average temperature of the stack	T_{av}	°C	K
Degradation rate of cell voltage	$\Delta V_{cell}/\Delta t$	µV h^{-1}	V s^{-1}
Degradation rate of stack voltage	$\Delta V_{stack}/\Delta t$	µV h^{-1}	V s^{-1}
Degradation rate of repeating unit (RU) *i* voltage	$\Delta V_{RU,i}/\Delta t$	µV h^{-1}	V s^{-1}

For long-term endurance test, the degradation rate is reported as the change in voltage (ΔV_{cell}, ΔV_{stack} and $\Delta V_{RU,i}$) over a given time which has to be clearly mentioned (see TM00 – section 10). Indeed, for instance, the first hundred hours are generally disregarded because the cell/stack assembly unit follows a kind of transient operation and the degradation rate can be very high in this period. The degradation rate is expressed in mV per kh (or µV per h), as defined in section 10 of the master document TM00 and reference [2]. It can additionally be expressed in % of the initial voltage value. Moreover, it is either related to the operating time or the number of cycles.

The absolute degradation ΔX of a quantity X within the time from t_0 to t_1 is calculated as the difference between the final value $X(t_1)$ and the initial value $X(t_0)$:

$$\Delta X = X(t_1) - X(t_0)$$

The relative degradation ΔX_{rel} is calculated by dividing ΔX by the initial value $X(t_0)$:

$$\Delta X_{rel} = \frac{X(t_1) - X(t_0)}{X(t_0)} \cdot 100\%$$

The degradation rate (rate of change) of quantity X during the time interval *(t_1-t_0)* is then calculated by:

$\frac{\Delta X}{\Delta t} = \frac{\Delta X}{t_1 - t_0}$ with the unit [unit of *X*/time unit]

$\frac{\Delta X_{rel}}{\Delta t} = \frac{\Delta X_{rel}}{t_1 - t_0}$ with the unit [%/time unit]

Degradation rates are typically expressed by the absolute or relative change per 1000 hours. It is thus advisable to normalize the results to 1000 h time interval. This can simply done by converting the unit of time interval to kh.

For load cycling experiments, degradation rate values can also be related to the number of cycles *m*:

$\Delta X_m = \frac{X(t_1) - X(t_0)}{m}$ and $\Delta X_{m,rel} = \frac{X(t_1) - X(t_0)}{X(t_0) \cdot m} \cdot 100\%$

6 Test Procedure

This specific TM is dedicated to the study of the capability of cell/stack assembly unit to modulate its power according to application-specific load profiles.

6.1 *Critical Parameters and Parameter Controls*

In this specific TM for operation under varying current and in addition to the stability of the operating conditions on each current plateau (temperature, pressure, gas flow rates and current density), a particular attention has to be paid to the accuracy of the current and its rate of change in order to follow the load patterns as close as possible. Also voltage measurement as a function of time has to be sufficiently accurate and voltage signal sufficiently clean to allow degradation rate determination.

Furthermore, it is important to avoid pollution from inlet gases and the test bench itself, since it has a strong influence on the degradation.

6.2 *Operation under varying current*

Realistic testing load profiles have to be defined for each application selected (stationary and distributed power generation (SOFC-µCHP), mobile SOFC-APU application, SOEC for H_2 production (power-to-gas) and combined SOFC/SOEC for electricity storage (power-to-gas-to-power) for instance). In the following subsections, typical load profiles from literature references for these applications are shown. These profiles can be used as examples in order to develop a simplified and practical test profile (as shown in *Figure 3*).

At a relevant temperature, inlet gas compositions and flow rates for each application, realistic loads I, plateau durations $t_{op,d}$ and speed rates for switching from one current to the other $\Delta I/\Delta t$ have to be defined based on literature and manufacturers' recommendations. Also the number of plateaus d and cycles m to be tested has to be fixed. Number of 200 cycles [2] and duration of 5 days [4] which can lead to about 500 cycles depending on the load pattern are mentioned in literature. Fuel/air utilization and steam conversion $U_{gas,neg}/U_{gas,pos}$ are mentioned for each tested load.

1) Check that the stability of the TIPs (T_{oven}, $T_{PH,neg}$, $T_{PH,pos}$, $p_{neg,out}$, $p_{pos,out}$, $f_{neg,in}$, $f_{pos,in}$, $f_{i,neg,in}$ *and* $f_{i,pos,in}$) is acceptable according to the stability criteria given in section 6.5 of the master document TM00;

2) Increase $|I|$ from zero to the first targeted value progressively in accordance with the chosen rate of current application $\Delta I/\Delta t$;

3) Let operation last for $t_{op,1}$ then vary $|I|$ from the first to the second target current value with the chosen rate of current change $\Delta I/\Delta t$ for $t_{op,2}$ operation and so on. When the first cycle is finished, repeat it m times. When cycles last sufficiently long, EIS measurements can usefully be performed and are recommended between each full cycle. It is also recommended to measure and record properly all TIPs and TOPs mentioned in sections 3 and 4. In particular, the evolution of the V_{cell}, V_{stack} and $V_{RU,i}$ voltages and of the T_{cell} or T_{stack} is carefully recorded as a function of time during the overall duration of the TM13. If a leak is suspected at any moment during the test (unexpected temperature evolution, signal instabilities), stack operation can be interrupted by decreasing the current absolute value to zero and OCV value checked. Then, in any operational mode (SOFC, SOEC), an automatic cut-off voltage threshold may also be set, bringing immediately to step 4) even if the foreseen number of cycles m isn't achieved yet. Alternatively, a value of voltage degradation rate (decrease in SOFC mode and increase in SOEC mode) can be defined to stop the test module automatically even if the foreseen number of cycles m isn't achieved yet. An automatic cut-off criteria may also be chosen on T_{cell} or T_{stack};

4) If different from zero at the last plateau, decrease $|I|$ to zero progressively in accordance with the chosen rate of current application $\Delta I/\Delta t$.

Figure 1 below presents the general TIPs' evolution all along this TM for one SOEC or SOFC operating mode (negative or positive current) and combined SOEC/SOFC operation.

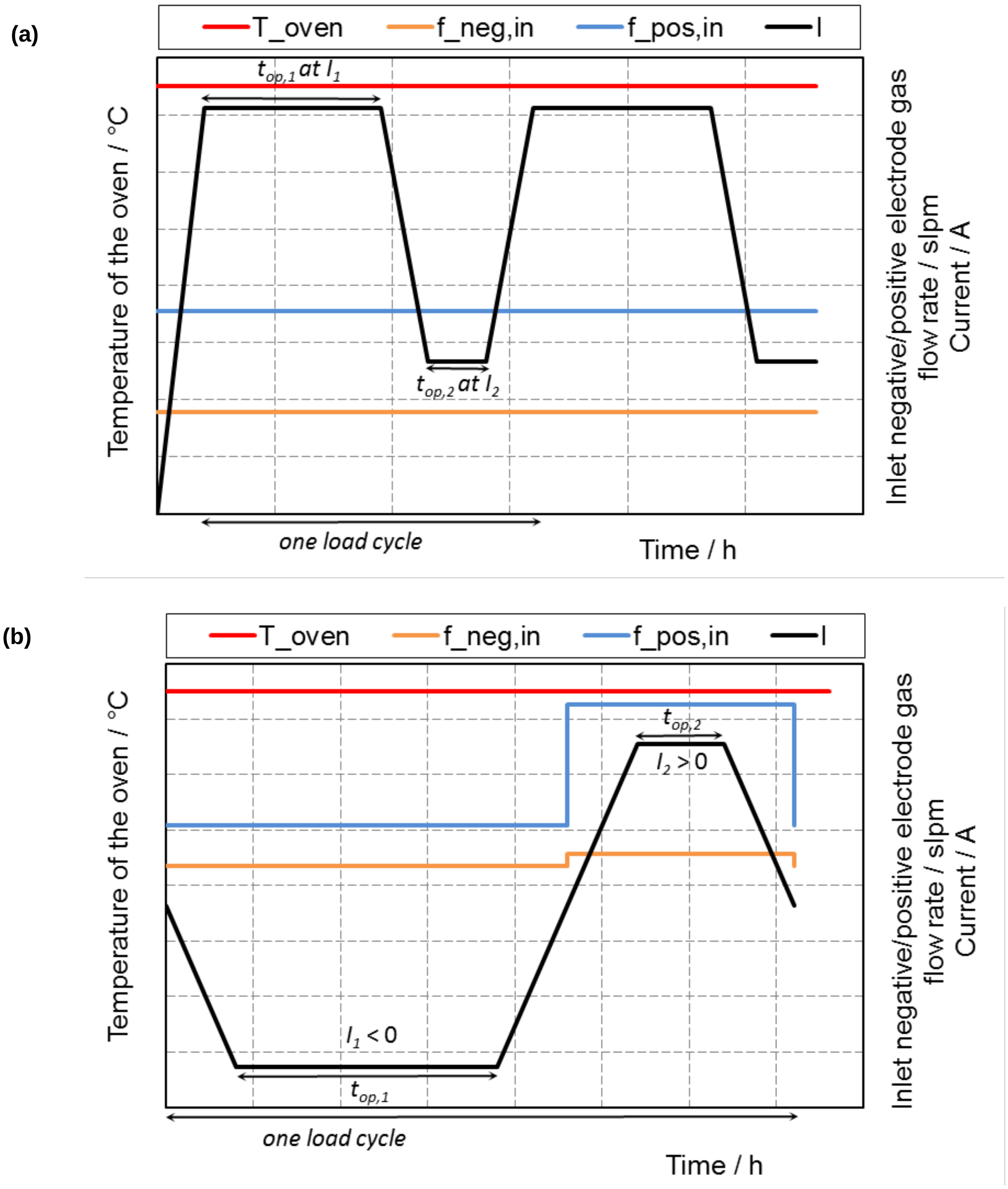


Figure 1: General evolution of TIPs during TM13 for (a) one SOEC or SOFC operating mode and (b) combined SOEC/SOFC operation

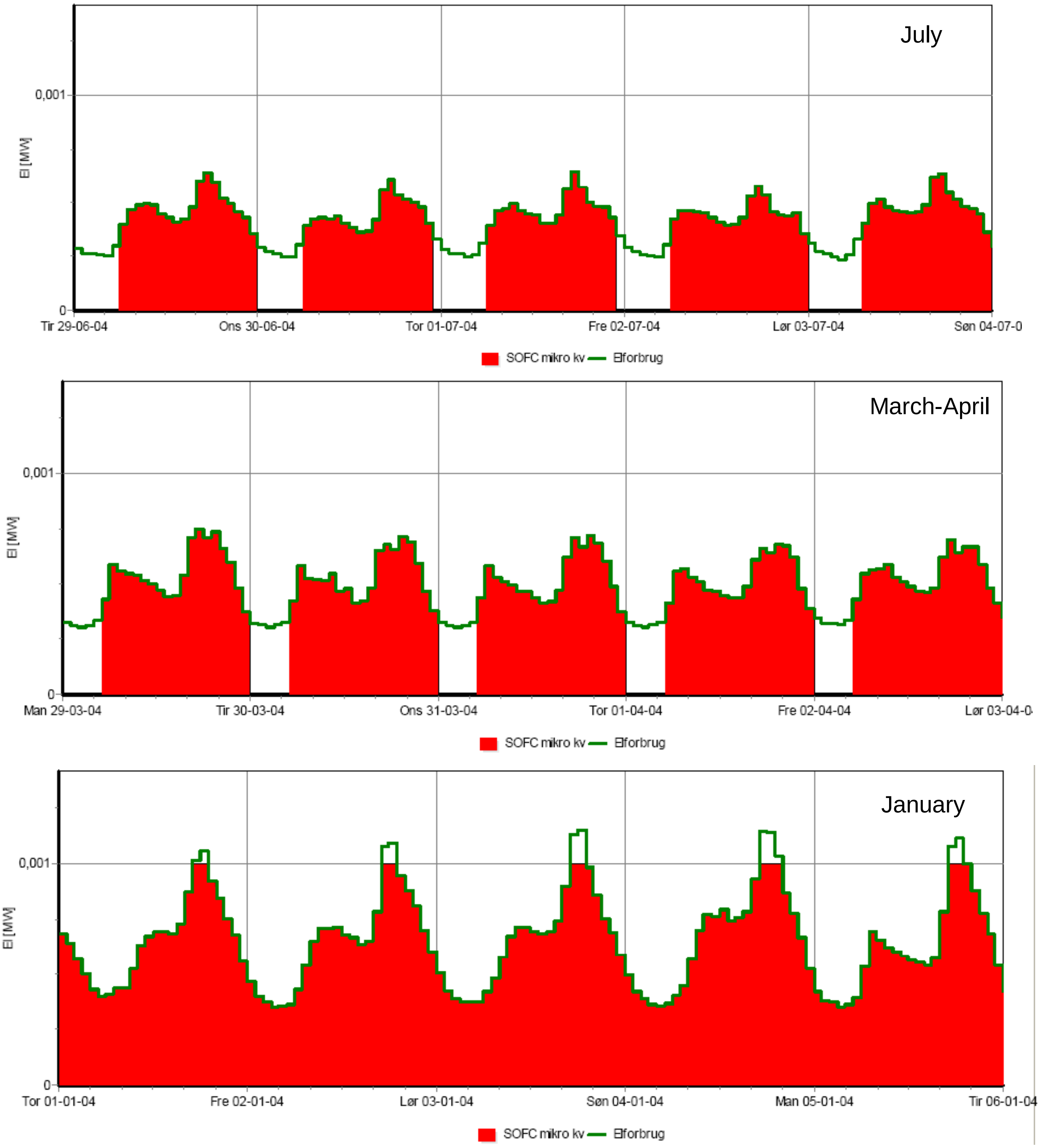
On each current plateau, initial voltages and gas utilizations at the negative and positive electrodes (fuel utilization (SOFC) or steam conversion rate (SOEC) at the negative electrode and air / oxygen utilization (SOFC) at the positive electrode) are defined for each application.

6.2.1 For stationary and distributed power generation (SOFC-µCHP)

Traditionally the control strategy of µCHP units has focused on following either the electricity or the heat demand of the end-user. The irruption of smart and interconnected embedded control systems

(IoT) allows operating the power units in an optimum (economic) point autonomously and this does not need to follow necessarily any of the loads.

Typical load and heat profiles are simulated in reference [2]. Simulation of SOFC operation for an electricity demand controlled unit with no export of electricity is shown in *Figure 2*. The green curves show the electricity demand and the red area is the electricity production in the SOFC unit.



July

March-April

January

Figure 2: Example of load and heat profiles of a SOFC-µCHP (electricity demand controlled unit) [2]

In reference [2], the following simplified load pattern is proposed for SOFC tests simulating a micro-cogenerator in operation (see *Figure 3*).

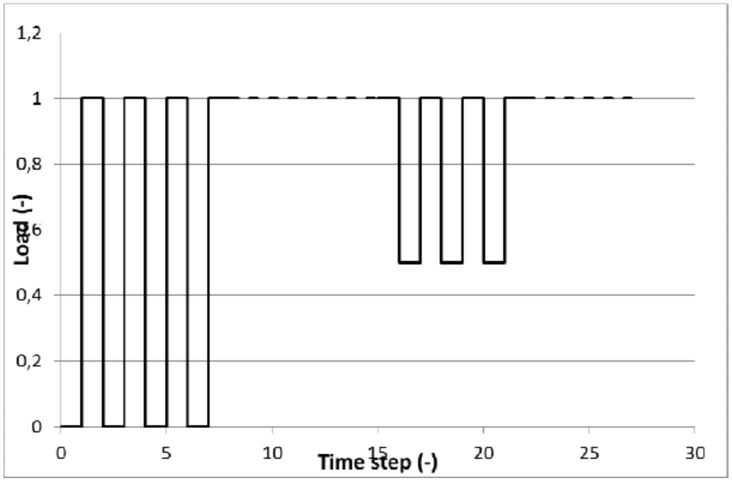
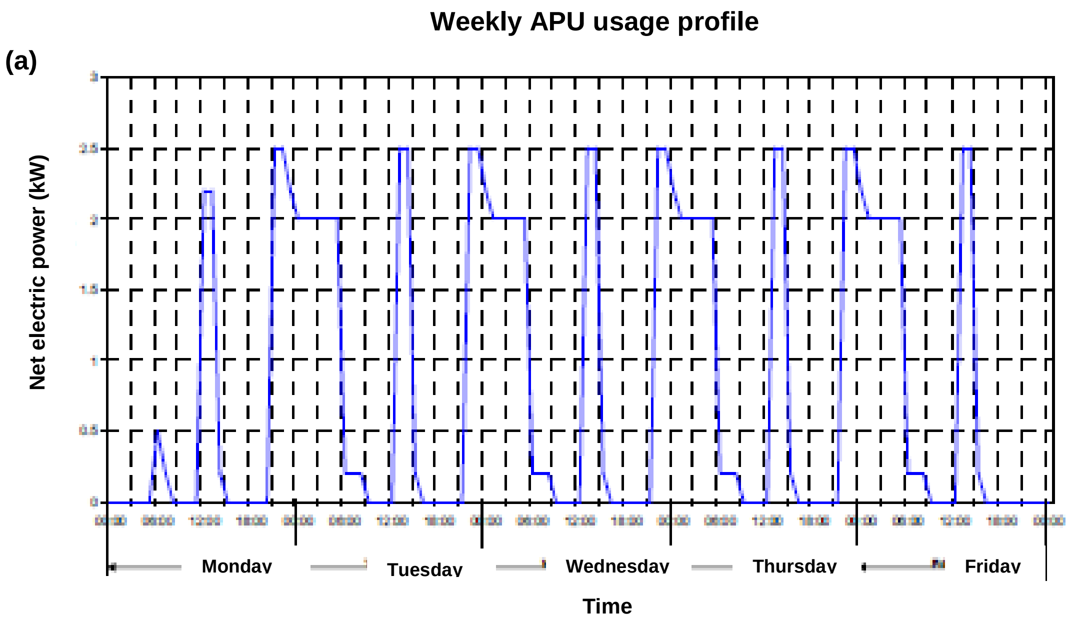


Figure 3: Simplified load profile simulating the operation of a SOFC-µCHP [2]

6.2.2 For mobile SOFC-APU application

A typical usage profile of a truck APU system is supplied in *Figure 4*(a) below with a focus on 48 hours in *Figure 4*(b) and can be used to define relevant testing load patterns.



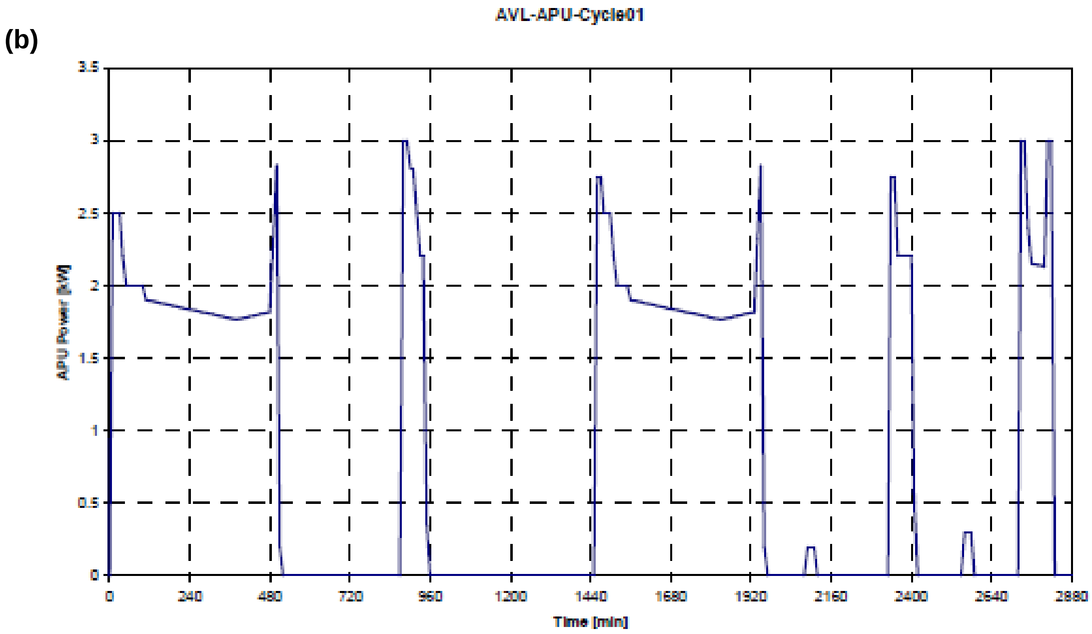


Figure 4: Load profile (a) and focus on 48 hours (b) of a SOFC truck APU [3]

6.2.3 Concerning SOEC for H_2 production (power-to-gas) and combined SOFC/SOEC for electricity storage (power-to-gas-to-power)

When coupling the SOC assembly unit with power generators relying on renewable energy sources, a realistic load profile can be defined based on reference [4] in SOEC mode. The following load profiles have been tested at single cell, SRU and short stack levels (see below) taking into account the renewable energy conversion technologies (converting solar or wind energy to electricity for instance). In order to study the influence of electric and/or thermal effects on degradation, testing of the cell/stack assembly unit is performed with different current densities corresponding to thermoneutral or exothermal operation (galvanostatic mode in *Figure 5* to *Figure 8*). Different rates of current change corresponding to ON/OFF operation of the electrolyser (highest value in *Figure 5* and *Figure 6*) or following load curves of renewable electricity production (lower value in *Figure 7* and *Figure 8*) are also interesting to be tested.

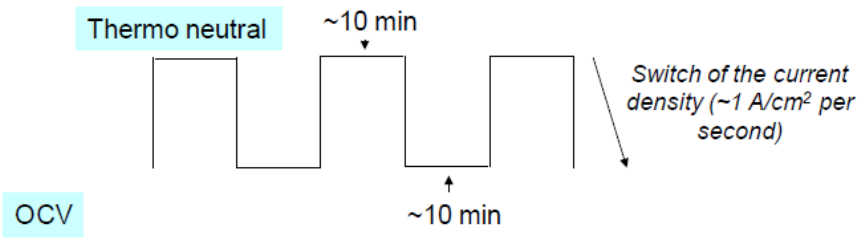


Figure 5: Load profile of a SOEC system with fast switch on/off at thermo-neutral conditions [4]

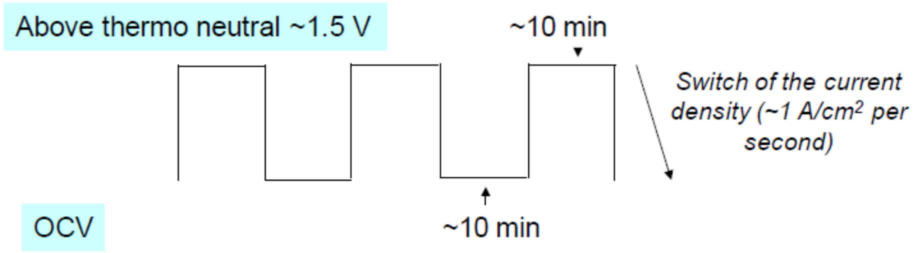


Figure 6: Load profile of a SOEC system with fast switch on/off at exothermal conditions [4]

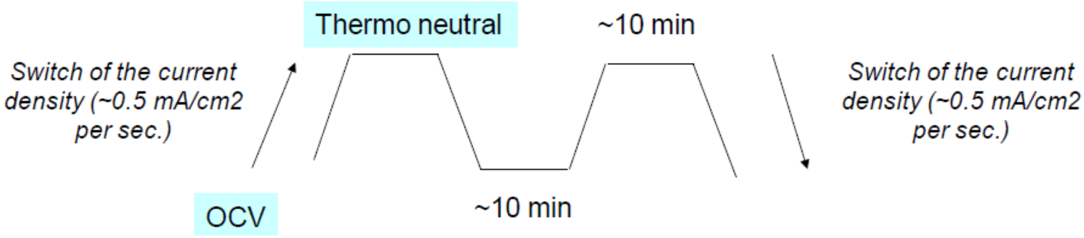


Figure 7: Load profile of a SOEC system with renewable energy source following and thermoneutral conditions [4]

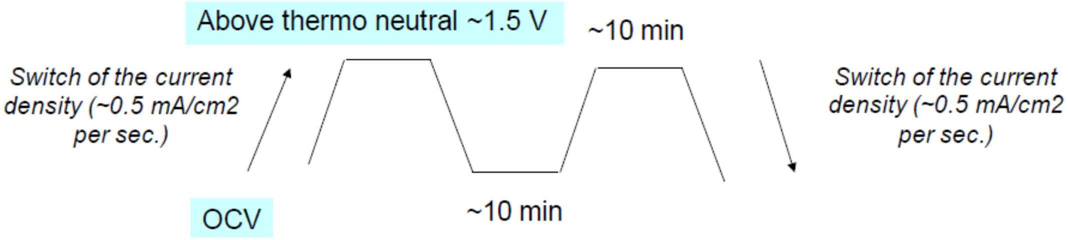


Figure 8: Load profile of a SOEC system with renewable energy source following and exothermal conditions [4]

7 Data Post Processing and representation

Information on reporting of test results is mentioned in section 9 of the master document TM00. In particular, for this TM, a graph presenting the evolution of V_{cell}, V_{stack} and $V_{RU,i}$ voltages as a function of time is highly recommended as well as a graph with the time-evolution of T_{cell} or T_{stack} (see *Figure 9* and *Figure 10* in SOEC and combined SOFC/SOEC modes for instance).

Also the calculated voltage degradation rates can be compared in a graph as shown in *Figure 11*.

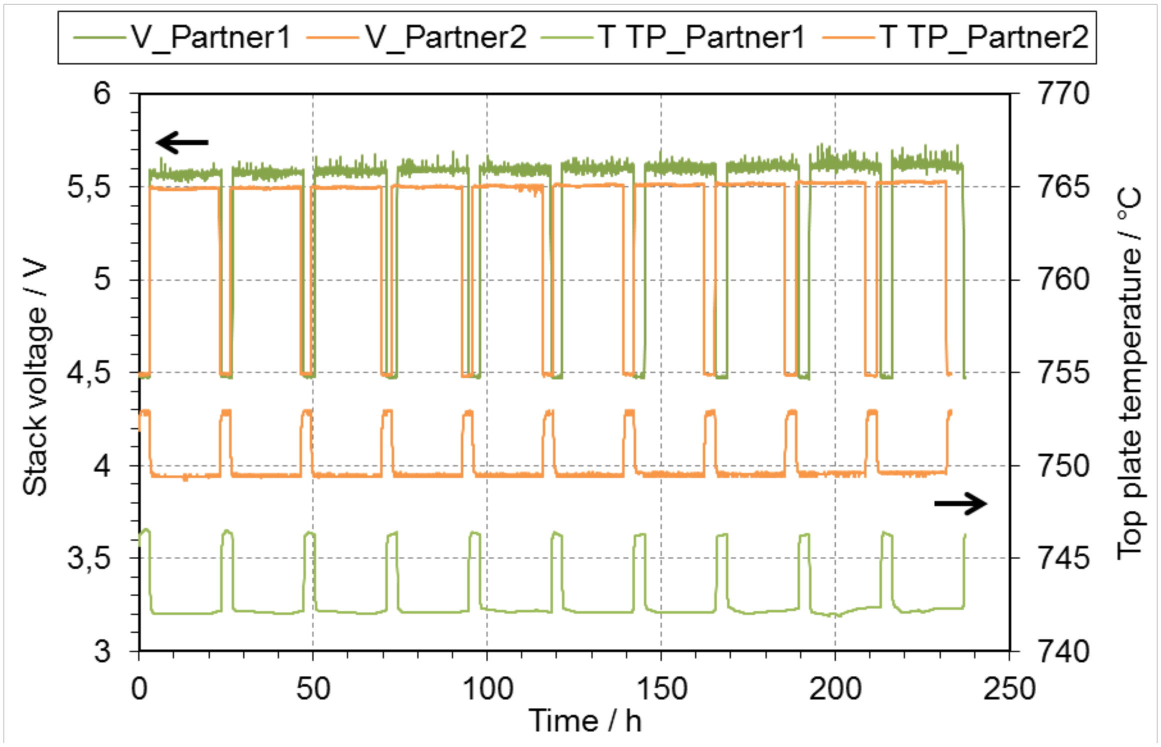


Figure 9: Examples of data representation in SOEC mode during TM13. Stack voltage and top plate temperature as a function of time

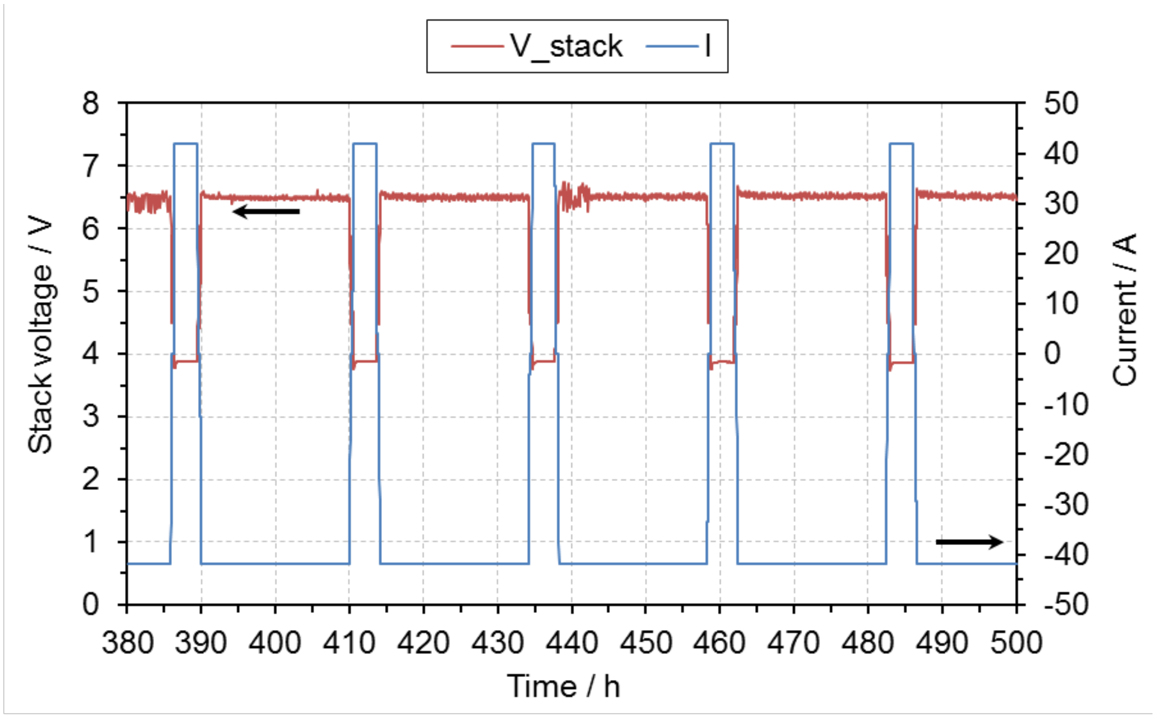


Figure 10: Example of data representation in combined SOFC/SOEC mode during TM13: Stack voltage and electrical current as a function of time

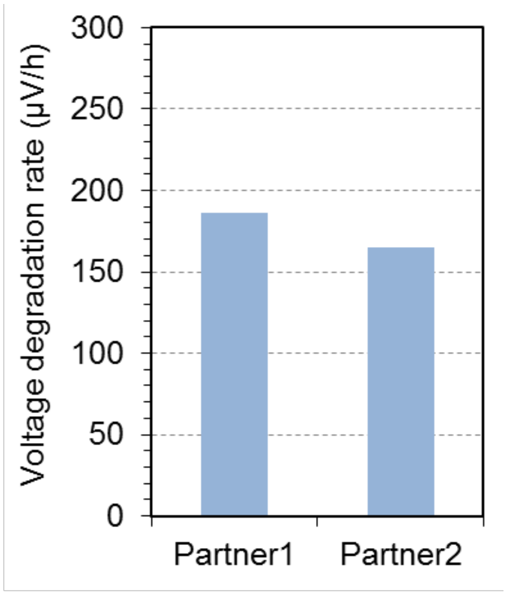


Figure 11: Example of data representation in SOEC mode during TM13: voltage degradation rates between two partners

8 Differences to Existing Procedures

This TM objective is quite new. Few test programs in transient operating conditions can be found in literature [2-4] but existing procedures as reference [1] don't take into account dynamic operation in both modes. Based on those references, the present TM13 fully dedicated to operation under varying current presents in details the relevant TIPs, TOPs and derived quantities with their associated formularies, their evolutions, the test procedure to perform long-term operation in SOFC, SOEC and even combined SOFC/SOEC modes and the different ways to express degradation rates in order to achieve a more comprehensive representation of the cell/stack assembly unit durability.

9 Bibliography

[1] International Electrotechnical Commission (IEC) – Fuel Cell Technologies – Standard 62282 – Part 7-2: Single cell and stack test methods – Single cell and stack performance tests for solid oxide fuel cells (SOFC)

[2] M. Näslund, H. Iskov, Delrapport DGC1: Accelerated lifetime testing and standardization of SOFC systems, Danish Gas Technology Centre, Horsholm 2012

[3] EU - Project METSOFC - Grant agreement number: 211940 - Project final report - 2012, http://www.metsofc.eu/Publications/~/media/Metsofc/downloads/metsofc_final_report.ashx, latest access 12/12/2016

[4] ADEL deliverable D1.5 – Definition of ADEL testing protocol and revised test matrix, Q. Fu, A. Brisse, EIFER, 2013