Solid Oxide Cell and Stack Testing, Safety and Quality Assurance

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Test Module 12: Operation under Constant Current

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Abbreviations

APU	Auxiliary power unit
ASR	Area specific resistance
CHP	Combined heat and power
nlpm	Normal litre 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 12 – Operation under Constant Current

1 Objective and Scope

This document presents the test module 12 (TM12) which deals with solid oxide cell (SOC) operation under constant current (i.e. galvanostatic conditions) either as a fuel cell (SOFC) or as an electrolyser (SOEC). The aim of this test module is to establish a widely accepted method for SOC steady-state operation when a long-term endurance test is performed. 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 of cell/stack resistances (ohmic and polarization). 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, measuring methods and test methods. Moreover, it is also applicable to the four different SOC applications selected in the SOCTESQA project:

- 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 steady-state operating conditions 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 TM12 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 the 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. TM12 doesn't include any variable TIPs. The SOC assembly units are usually operated in galvanostatic mode (at a constant current for a given operating point).

TM12 starts by setting the following parameters as constants: oven temperature, total pressure (atmospheric or higher) as well as gas flow rates at the negative and positive electrodes and finally the magnitude of the current after a period of stabilization of the first parameters. Those parameters are chosen according to full operating conditions i.e. in relation to relevant 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) for each application.

Table 1 below lists all the relevant test input parameters (operating conditions) useful in TM12. They all belong to the static category. 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} (at OCV before increasing current). Optionally, it is also possible to fix T_{oven} under polarization. $T_{neg,in}$ and $T_{pos,in}$ can vary as well as T_{cell} or T_{stack} during the TM. The following pressure values $p_{neg,out}$, and $p_{pos,out}$ must also be controlled so as to obtain the targeted value at the inlet in relation to the measured pressure drop over the cell/stack assembly unit (Δp_{neg}, Δp_{pos}). $f_{neg,in}$, $f_{pos,in}$, $f_{i,neg,in}$ and $f_{i,pos,in}$ gas flow rates are also controlled to fix the mole fractions of gas components $x_{i,neg,in}$ and $x_{i,pos,in}$ 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_{stack}, V_{cell} and $V_{RU,av}$). The absolute current $|I|$ is maintained to the targeted value for a certain operating time t_{op} [2-5].

Table 1: Static test input parameters during TM12

Description of quantity	**Symbol**	**Unit often used**	**SI Unit**
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 gas stream at cell/stack inlet	$f_{i,pos,in}$	nlpm, slpm l_n min^{-1}, l_s min^{-1}	m^3 s^{-1}
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}
Electrical current through the cell/stack	I	A	A
Pressure of the negative electrode gas stream at cell/stack outlet	$p_{neg,out}$	mbar, kPa	N m^{-2} (Pa)
Pressure of the positive electrode gas stream at cell/stack outlet	$p_{pos,out}$	mbar, kPa	N m^{-2} (Pa)
Temperature of the pre-heater for preheating the negative electrode gas stream	$T_{PH,neg}$	°C	K

Temperature of the pre-heater for preheating the positive electrode gas stream	$T_{PH,pos}$	°C	K
Temperature of the oven	T_{oven}	°C	K
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	$x_{i,pos,in}$	-	-

4 Test Output Parameters (TOPs)

Table 2 below shows the list of the test output parameters that can be recorded during the overall duration of TM12, mainly the cell/stack assembly unit response e.g. V_{stack}, V_{cell} and $V_{RU,i}$ SOC voltages [2,3,4]. 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 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). $f_{neg,out}$, $f_{pos,out}$, $f_{i,neg,out}$ 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 2: Test output parameters during TM12

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 3* gives the derived quantities useful for this TM [1,2]. They are all calculated from TIPs and TOPs with the equations presented in TM00 - section 10.

Table 3: Derived quantities possibly calculated during TM12

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 $(t_1\text{-}t_0)$ which has to be clearly mentioned (see TM00 – section 10). Indeed, for instance, the first hundred hours are generally disregarded ($(t_1\text{-}t_0) < t_{op}$) because the cell/stack assembly unit follows a kind of transient operation and the degradation rate can be very high in this period. This degradation rate is expressed in mV per kh (or µV per h), as defined in the section 10 of the master document TM00 and reference [2]. It can additionally be expressed in % of the initial value.

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 $(t_1\text{-}t_0)$ 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.

6 Test Procedure

This specific TM12 is dedicated to SOC steady-state operation for long-term endurance tests. The aim of this TM consists in recording performance evolution (i.e. cell/stack voltage in galvanostatic mode) over a period of time at constant nominal operating conditions (temperature, pressure, gas flow rates and current) dependent on each application.

6.1 Critical parameters and parameter controls

In this specific TM, particular attention has to be paid to the stability of the operating conditions (temperature, pressure, gas flow rates and current) when constant. Also voltage measurement as a function of time has to be sufficiently clean to allow degradation rate determination during long-term operation.

Furthermore, the importance of avoiding pollution from inlet gases and the test bench itself can be pointed out here, since it has a strong influence on the degradation.

6.2 Operation under constant current

Realistic operating points have to be defined for each selected application (stationary SOFC-µCHP and distributed power generation, mobile SOFC-APU application, SOEC for H_2 production (power-to-gas) and combined SOFC/SOEC for energy storage (power-to-gas-to-power) for instance) in terms of targeted initial V_{cell}, V_{stack} and $V_{RU,av}$ voltages as well as $U_{gas,neg}$ and $U_{gas,pos}$ gas utilizations. At relevant temperatures, inlet gases compositions and flow rates for each application, a realistic current I and operation duration t_{op} have to be defined based on literature and manufacturers recommendations.

Figure 1 below presents the general TIPs' evolution all along this TM.

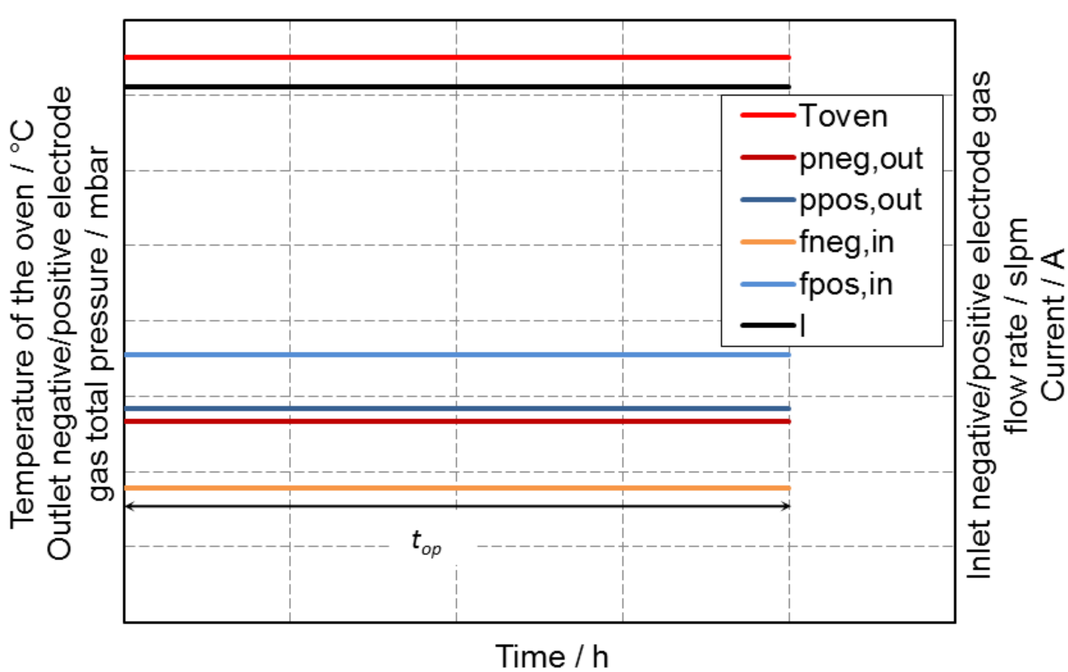

Figure 1: General evolution of TIPs during TM12

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

2) Let operation last for t_{op}. It is 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 TM12. 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. In any operational mode (SOFC, SOEC), an automatic cut-off voltage threshold may also be set, bringing immediately to stop the current density even if the foreseen duration t_{op} is not 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 duration t_{op} is not achieved yet. An automatic cut-off criteria may also be chosen on T_{cell} or T_{stack}.

7 Data Post Processing and Representation

Information on reporting of test results is mentioned in the 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 2* for stack voltage and temperatures evolution in SOEC mode for instance).

Also the calculated degradation rate of the SOC voltage can be presented in a graph as shown in *Figure 3*. If a strong temperature evolution is observed during the operation under constant current, the determination of the degradation rate will be largely influenced. In this case, a temperature correction can be applied:

- either by adapting the oven temperature at the end of the TM12 step in order to bring the stack temperature back to the initial value and take the obtained voltage as the final one,

- or by performing the TM12 step at a constant T_{stack} through constant adjustment of the oven temperature which can be automatically realized through the temperature regulation unit.

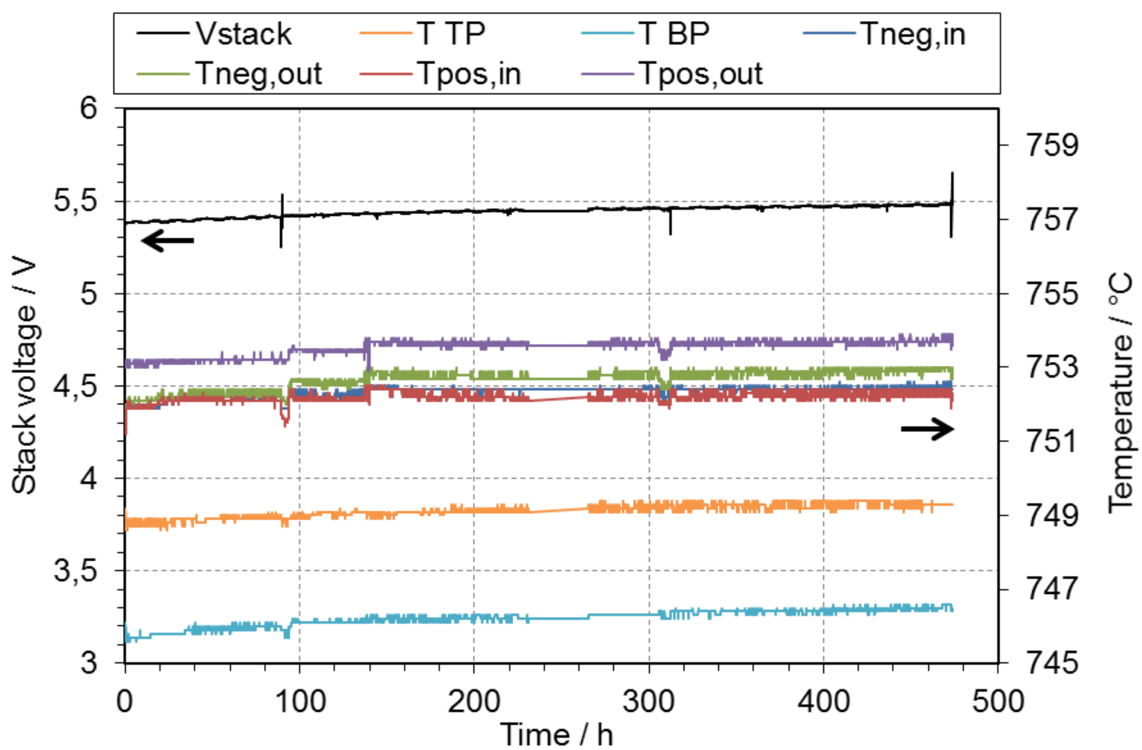

Figure 2: Example of data representation in SOEC mode during TM12: stack voltage and temperatures as a function of time for a constant current of -0.3 A/cm². Temperatures variation of about 1°C in this example

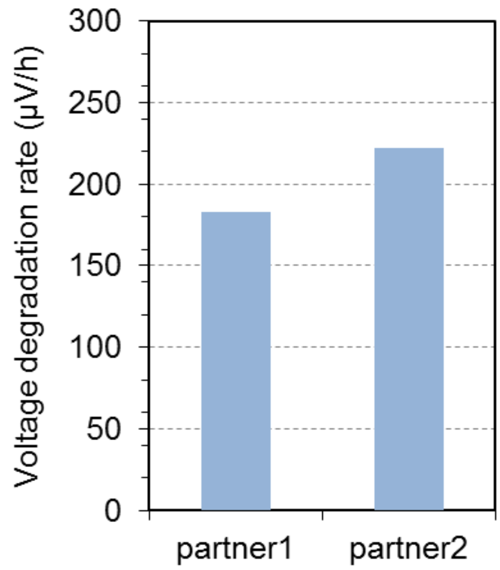

Figure 3: Example of data representation in SOEC mode during TM12: calculated voltage degradation rates between two partners

8 Differences to Existing Procedures

This TM topic is quite common nowadays as shown by references found in literature [2-5]. Nevertheless, existing procedures, as reference [1], remain quite generic. Based on those references, the present TM12 fully dedicated to operation under constant 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 both SOFC and 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] SOFC single cell performance and endurance test modules, Test Modules TM SOFC 01-04LD / 05-08HD, FCTESQA project, 2010.

[4] SOFC stacks performance and endurance test modules, Test Modules TM SOFC 25-28HD, FCTESQA project, 2010.

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