INSIGHT

"ImplementatioN in real SOFC Systems of monItoring and diaGnostic tools using signal analysis to increase tHeir lifeTime"

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Testing protocols and data acquisition specifications for stack testing

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Document Abstract

In the frame of the INSIGHT project, and most particularly of WP2 dedicated to testing, one task, WT2.2, aims to define and select the most relevant testing protocols and specifications to characterize SRU/SS/Stacks. The objective is the evaluation of all techniques considered for system monitoring and detection of faults/failures selected in task 2.1 which will also lead to the lifetime estimation.

In order to save testing resources, hence experiments costs, means of accelerated testing relevant for the application area will be used. The test protocols will consider routine durability tests over 1000 h, and the on-purpose introduction of some faults as identified in Task 2.1. Three main faults were identified: *fuel starvation*, *gas leakage* and *carbon poisoning*.

Therefore, different modules are suggested which can be combined individually. 2 durability modules and different fault modules are suggested. The testing protocols are based on the suggestions from the EU-SOCTESQA project, which aimed at harmonizing SOFC/SOEC testing.

Each test program follows a standardized sequence consisting of: start-up and initial characterization, the actual durability period, and a final characterization. The durability modules contain the most important degradation initiating set of conditions, starting with baseline operation at constant conditions, followed by the introduction of the fault incidents.

Those testing protocols are used to perform SRU, short stacks and stacks characterizations in WP2 but also as input for system testing, both in lab and on field in WP6.

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1. Objective

The aim of this task is to define and select the most relevant testing protocols and specifications to characterize SRU/SS/Stacks. The objective is evaluation of all techniques considered for system monitoring and detection of faults/failures selected in task 2.1 which will also lead to the lifetime estimation. Besides, it will give to WP3 specifications for monitoring (EIS, THD, stack signals).

In order to save testing resources, hence experiments costs, means of accelerated testing relevant for the application area will be used. The test protocols will consider routine durability tests over 1000 h, and the on-purpose introduction of some faults as identified in Task 2.1. Three main faults were identified: *fuel starvation*, *gas leakage* and *carbon poisoning*. It might be impossible to execute all faults within one single test due to degradation.

Therefore, different modules are suggested which can be combined individually. 2 durability modules and different fault modules are suggested. The testing protocols are based on the suggestions from the EU-SOCTESQA project, which aimed at harmonizing SOFC/SOEC testing.

Each test program follows a standardized sequence consisting of: start-up and initial characterization, the actual durability period, and a final characterization. The sequences of the test programs are illustrated in figures Figure 1-Figure 4 for each test program and detailed operating procedures are outlined in section 0. The durability modules contain the most important degradation initiating set of conditions, starting with baseline operation at constant conditions, followed by the introduction of the fault incidents.

Those testing protocols are used to perform SRU, short stacks and stacks characterizations in WP2 but also as input for system testing, both in lab and on field in WP6.

2. TEST program I – fault I 'Fuel starvation'

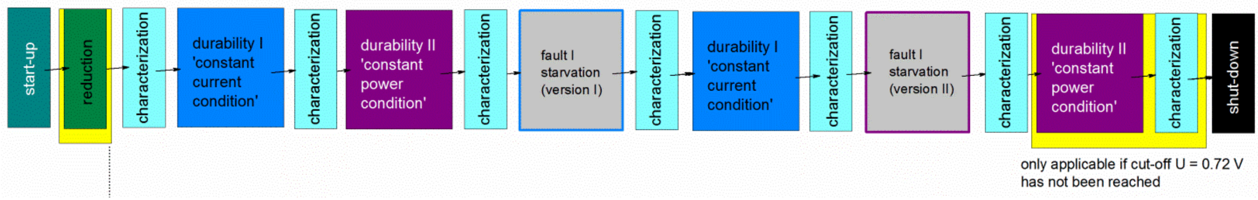

Figure 1 Overview of test program for investigation of fault I 'Fuel starvation'. Yellow highlighted modules are only applied if necessary or possible.

The detailed test program is illustrated in Figure 1. The test program begins with the start-up (1°C min^{-1}) of pre-reduced stacks[1] with a constant total fuel flow rate of 0.36 Nl h^{-1} cm^{-2} (0.216 Nl h^{-1} cm^{-2} H_2 and 0.144 Nl h^{-1} cm^{-2} N_2) and an initial characterization based on the guidelines given from the SOCTESQA-project (iV + EIS at OCV and 0.5 A cm^{-2}). First the module 'durability I' at constant current conditions is performed (nominal operating conditions 0.4 A cm^{-2}, 80% FU, 60/40 H_2/N_2 fuel composition as done at SP IT). As it is purpose of the INSIGHT project to investigate stacks close to real operating conditions a second module 'durability II' at constant power conditions is added. These modules serve as 'long-term' baselines for further characterization and fault investigations.

Afterwards, the first of three relevant faults *'Fuel starvation'* will be introduced. The designed experiment targets to simulate the break-down of the fuel supply (hardware fault) under constant operating conditions of the short-stack. '*Fuel starvation'* version I will be caused by stepwise

[1] Additionally, if other cell tests are performed i.e. SRU tests a standard reduction procedure is outlined in Section 0

throttling of the fuel supply, with regular monitoring of each step (10 h per step are proposed for sufficient investigation time for the individual detection methods: THD + EIS + signals). Each step decreases the fuel supply by 5%, so that a characterization at 80% - 85% - 90% - 95% FU will be investigated. If failure (i.e. cell voltage lower than 0.6 V) is observed, the fuel supply will be increased to nominal conditions (80% FU) and kept for 10 h. Then a characterization of the stack is performed for evaluating the degree of recovery after the fault. Thereafter we suggest performing of an additional durability module (durability I + OPTIONAL durability II) in order to correlate degradation behavior of a stack at 'fault free' conditions and 'faulty' conditions for life time prediction.

In order to investigate *'Fuel starvation'* version II under constant power operating conditions, a different experiment will mimic the effect of a possible software-fault. Constant power mode operations in a large stack are achieved by regulating the current according to the state-of-health voltage of the stack and adjust the fuel flow for constant FU. The second simulation of fault I is causing fuel starvation by increasing the current during operation while maintaining constant fuel flow. Aside from fuel starvation, this allows investigation of the resulting change of power density (which cannot be avoided).

This testing period lasts for ca. 820 h (+ 220 h optional including durability II if applicable after fault).

Detailed specifications for each module are outlined in section 0.

3. TEST program II – 'Carbon deposition'

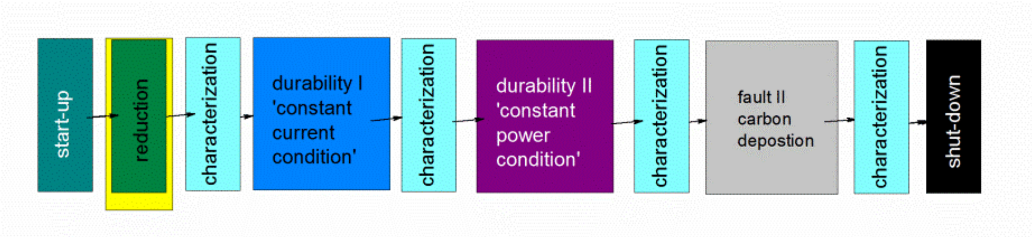

Figure 2 Overview of test program for investigation of fault II 'Carbon deposition'. Yellow highlighted modules are only applied if necessary.

Test program II includes the same start-up procedure and initial testing period as outlined in test profile I (module: characterization + durability I/II). However, in order to monitor and detect '*carbon deposition'*, the test will be performed in a fuel composition based on methane and steam with a S/C-ratio = 2. After 'the baseline' test period, carbon deposition will be caused on purpose for verifying different fault detection methods. The carbon deposition will be introduced by stepwise decrease of the S/C ratio (max. 1/1). 5 steps are considered: 2 – 1.75 – 1.5 – 1.25 – 1.00 and 10 h per step are designed. However it may be necessary to adjust the testing time if failure (i.e. fast voltage decrease/break down of cell voltage) is detected earlier.

As recovery strategy for carbon deposition the stack will be set back to nominal operating conditions for 24 h. Intermediate and regular characterizations after each durability module, as well as after each fault introduction will allow life time assessment and evaluation of counter-acting methods for each particular failure. The shut-down procedure is identical to TEST program I. The complete testing sequence is designed for approx. 570 h.

Alternatively, modules of TEST profile II can be added to TEST profile I if the health-state of the stack allows combining them. A schematic description for the proposed TEST program III can be taken from Figure 3. In that way, testing time of the short stack can be shortened. The combined profile lasts approx. 1200 h.

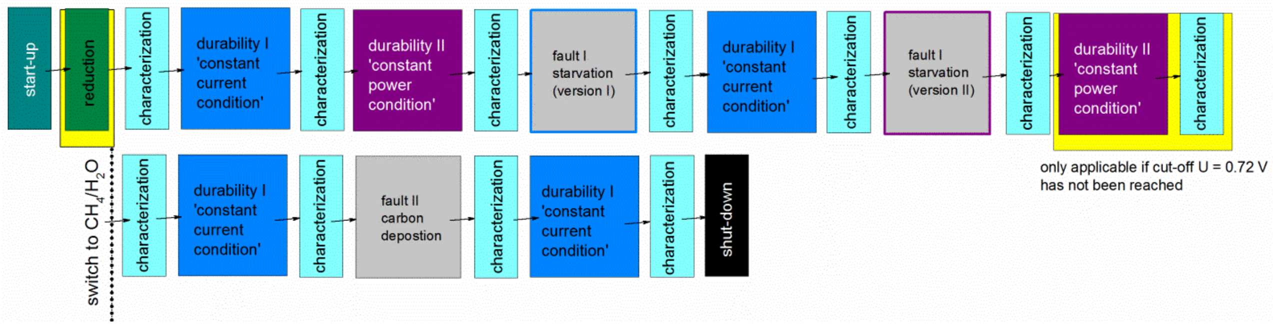

Figure 3 Overview of test program combining fuel starvation and carbon deposition

4. TEST program III – 'Leakage'

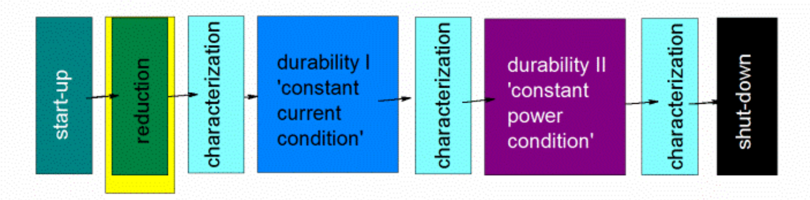

Figure 4 Overview of test under 'leak' conditions.

For investigating the third fault which is supposed to simulate 'leakage' a long-term test with on purpose defective sealing is suggested. The initial long-term sequence which is performed under the same conditions as the former test programs allows evaluating the differences of degradation and responses of the monitoring signals.

5. Specification Test modules

Table 1 specification test modules

module	details	Specifications	monitoring	time	Remark
start-up	start-up	(j=0 A cm^{-2}, e-load and power supply OFF) F_{compr}: needs to be specified (at RT) $f_{N2,neg,in}$: 0 to 0.144 Nl h^{-1} cm^{-2} (N_2) [**ONLY for pre-reduced cells**: $f_{H2,neg,in}$: 0 to 0.216 Nl h^{-1} cm^{-2} (H_2)] $f_{Air,pos,in}$: 0 to 2.124 Nl h^{-1} cm^{-2} (Air) T_{TP}: RT to 750 °C (1 K min^{-1}) (through controlling T_{oven}) $T_{PH,neg}$: RT to 750 °C (1 K min^{-1}) (when applicable) $T_{PH,pos}$: RT to 750 °C (1 K min^{-1}) (when applicable) temperature reference = oxygen electrode outlet	$T_{neg,in}$ $T_{pos,in}$ $T_{neg,out}$ $T_{pos,out}$ T_{PH}	14	for pre-reduced stacks 60% H_2 balanced with N_2
	sintering	holt t=3 h		3	stabilization

Reduction	Reduction procedure	$f_{H2,neg,in}$: 0 to 0.0072 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0 to 0.3528 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 0 to 4 Nl h^{-1} cm^{-2} (Air) Hold t=1 h	$V_{RU,i}$ (T_{TP}/T_{BP}/ $T_{neg/pos,in/out}$ can be monitored to identify possible gas burning behavior due to leakage)	1	2%/H_2 in N_2
		$f_{H2,neg,in}$: 0.0108 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.3492 Nl h^{-1} cm^{-2} Hold t=0.7 h		0.7	3%/H_2 in N_2
		$f_{H2,neg,in}$: 0.0144 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.3456 Nl h^{-1} cm^{-2} Hold t=0.5 h		0.5	4%/H_2 in N_2
		$f_{H2,neg,in}$: 0.0288 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.3312 Nl h^{-1} cm^{-2} Hold t=0.5 h		0.5	8%/H_2 in N_2
		$f_{H2,neg,in}$: 0.0576 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0. 3024Nl h^{-1} cm^{-2} Hold t=0.2 h		0.2	16%/H_2 in N_2
		$f_{H2,neg,in}$: 0.1152 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.2448 Nl h^{-1} cm^{-2} Hold t=0.2 h		0.1	32%/H_2 in N_2

		$f_{H2,neg,in}$: 0.18 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.18 Nl h^{-1} cm^{-2} Hold t=1 h		1	50%/H_2 in N_2

constant current characterization	IV	Recommended parameters flows $f_{H2,neg,in}$: 0 to 0.216 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0 to 0.144 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 0 to 4 Nl h^{-1} cm^{-2} (Air) current control j: 0 to j_{max} j is varied stepwise 0.025 A cm^{-2} per step 0.5 min hold time per step j_{max} is reached when $V_{RU,min}$ is not any more above 0.65 V, i.e., $V_{RU,min}$ @ j_{max} ≤ 0.65 V and $V_{RU,min}$ @ (j_{max} -0.025) > 0.65 V hold time at j_{max} as short as possible	$V_{RU,i}$ V_{stack} T_{TP} $T_{neg,out}$ $T_{pos,out}$	0.5	initial iV-curve
	EIS	Recommended parameters: DC current: I= **0 A cm^{-2}** / **0.5 A cm^{-2}** (included due to power density evaluation) @ 0.5 A cm^{-2} : (77.5) 80% FU $f_{H2,neg,in}$: 0.27 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.09 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2} Hold t=10-30 min (temperature stabilization before EIS) Frequency range: **100 kHz – 20 mHz** (start from 100 kHz) Number of frequency points per decade: 7 (Note: 4 points are also acceptable at low frequency in order to shorten the measurement time) EIS should be performed in galvanostatic mode.	Z' Z'' |Z| ϕ T_{TP} $T_{neg,out}$ $T_{pos,out}$ $V_{RU,i}$	7.5	test specifications have been chosen based on the 'SOCTESQA' -project

		Amplitude of AC current: $\bar{\boldsymbol{I}}$ = 1 A (Amplitude of AC voltage: $\bar{V}$ = 10-30 mV per RU) (NOTE: parameters can be adapted when appropriate in order to obtain for example impedance data of good quality) Set DC current stepwise to 0 after EIS			
	THD	Recommended parameters: DC current i= 0.5 A cm^{-2} Frequency range: **1 Hz – 20 mHz** (start from 1 Hz) Frequency amplitude: **5% and 10 %** (0.025 and 0.05 A cm^{-2}) Number of frequency points: 10	%THD T_{TP} $T_{neg,out}$ $T_{pos,out}$ $V_{RU,i}$	5	initial THD

durability I	constant current operating conditions	DC current = 0.4 A cm^{-2} (77.5) 80% FU, 10% OU $f_{H2,neg,in}$: 0.216 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.144 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2} Recommended impedance parameters at Frequency range: **100 kHz – 20 mHz** (start from 100 kHz) Number of frequency points per decade: 7 (NOTE: parameters can be adapted when appropriate in order to obtain for example impedance data of good quality) Recommended parameters: Frequency range: **1 Hz – 20 mHz** (start from 1 Hz) Frequency amplitude: **5% and 10 %** (0.025 and 0.05 A cm^{-2}) Number of frequency points: 10	$V_{RU,i}$ V_{stack} T_{TP} $T_{neg,out}$ $T_{pos,out}$ ($f_{neg,out}$) ($f_{pos,out}$) optional: EIS monitoring each 48 h (4 IS for reference) alternate: THD monitoring each 48 h (4 measurement s for reference)	200	baseline for ‘normal operating conditions of SS’ before fault detection

durability II	constant power operating conditions	Power density = 0.3 W cm^{-2} and 0.8 V (80% FU) $f_{H2,neg,in}$: regulate flow for (77.5) 80% FU $f_{N2,neg,in}$: balance flow to a net flow of 0.36 $f_{Air,pos,in}$: keep air flow at 10% OU During operation regulate current and flow to keep 80% FU (and balance with with N_2 to have equal net-flow) Recommended impedance parameters at Frequency range: **100 kHz – 20 mHz** (start from 100 kHz) Number of frequency points per decade: 7 (NOTE: parameters can be adapted when appropriate in order to obtain for example impedance data of good quality) Recommended parameters: Frequency range: **1 Hz – 20 mHz** (start from 1 Hz) Frequency amplitude: **5% and 10 %** (0.025 and 0.05 A cm^{-2}) Number of frequency points: 10	$V_{RU,i}$ V_{stack} T_{TP} $T_{neg,out}$ $T_{pos,out}$ ($f_{neg,out}$) ($f_{pos,out}$) optional: EIS monitoring each 48 h (4 IS for reference) alternate: THD monitoring each 48 h (4 measurements for reference)	200	baseline for 'normal operating conditions of SS' before fault detection

fault I – fuel starvation simulation of ' break down of fuel flow	80%	DC current = 0.4 A cm^{-2} (77.5% FU) $f_{H2,neg,in}$: 0.216 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.144 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	step 1: 80%
	85%	$f_{H2,neg,in}$: 0.203 Nl h^{-1} cm^{-2} (82.4% FU) $f_{N2,neg,in}$: 0.135 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	step 2: 85%
	90%	$f_{H2,neg,in}$: 0.191 Nl h^{-1} cm^{-2} (87.6% FU) $f_{N2,neg,in}$: 0.127 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	step 3: 90%

	95%	$f_{H2,neg,in}$: 0.181 Nl h^{-1} cm^{-2} (92.5% FU) $f_{N2,neg,in}$: 0.121 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	step 4: 95%
	recovery	increase flow to DC current = 0.4 A cm^{-2} (77.5% FU) $f_{H2,neg,in}$: 0.216 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.144 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2} hold = 10 h		10	recovery

fault I – fuel starvation simulation of software fault – increase in current but no increase in flow	80%	Power density = 0.3 W cm^{-2} $f_{H2,neg,in}$: regulate flow for 80% FU $f_{N2,neg,in}$: balance flow to a net flow of 0.36 $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	step 1: 80%
	85%	keep set-flow for initial 80% FU increase current for 85% → power density will be affected	EIS, THD	10	step 2: 85%
	90%	keep set-flow for initial 80% FU increase current for 90% → power density will be affected	EIS, THD	10	step 3: 90%
	95%	keep set-flow for initial 80% FU increase current for 95% → power density will be affected	EIS, THD	10	step 4: 95%
	recovery	go to step 1 conditions		10	recovery

fault II - leakage	Leakage fault	operation of faulty stack under same conditions as proposed in module 'durability I' and 'durability II'	$V_{RU,i}$ V_{stack} T_{TP} $T_{neg,out}$ $T_{pos,out}$ ($f_{neg,out}$) ($f_{pos,out}$) optional: EIS monitoring each 48 h alternate: THD monitoring each 48 h		

fault III – carbon deposition	S/C = 2	DC current = 0.4 A cm^{-2} (77.6% FU) $f_{CH4,neg,in}$: 0.0539 Nl h^{-1} cm^{-2} $f_{H2O,neg,in}$: 0.1078 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.1983 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	
	S/C = 1.75	$f_{CH4,neg,in}$: 0.05907 Nl h^{-1} cm^{-2} (75.5) $f_{H2O,neg,in}$: 0.10338 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.19755 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	
	S/C = 1.5	$f_{CH4,neg,in}$: 0.06498 Nl h^{-1} cm^{-2} (73.6) $f_{H2O,neg,in}$: 0.09756 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.19746 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	
	S/C = 1.25	$f_{CH4,neg,in}$: 0.0722 Nl h^{-1} cm^{-2} (71.3) $f_{H2O,neg,in}$: 0.09025 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.19755 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	
	S/C = 1.00	$f_{CH4,neg,in}$: 0.08122 Nl h^{-1} cm^{-2} (68.7) $f_{H2O,neg,in}$: 0.08122 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.19756 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2}	EIS, THD	10	
	recovery	increase flow to $f_{CH4,neg,in}$: 0.0539 Nl h^{-1} cm^{-2} $f_{H2O,neg,in}$: 0.1078 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.1983 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2} hold t = 24 h		24	recovery

shut-down		Change fuel gas to reducing flushing gas (always maintaining reducing atmosphere at negative electrode (e.g. N_2+5%H_2): $f_{H2,neg,in}$: 0.018 Nl h^{-1} cm^{-2} $f_{N2,neg,in}$: 0.342 Nl h^{-1} cm^{-2} $f_{Air,pos,in}$: 4 Nl h^{-1} cm^{-2} (j=0 A cm^{-2}, e-load and power supply OFF) T_{TP}: 750 °C to RT (-2 K min^{-1}) $T_{PH,neg}$: 750 °C to RT (-2 K min^{-1}) (when applicable) $T_{PH,pos}$: 750 °C to RT(-2 K min^{-1}) (when applicable) when reaching 300 ^{o}C: increase of air flow $f_{Air,pos,in}$: 4 to 5 Nl h^{-1} cm^{-2} at RT: turn off flows	$V_{RU,i}$ V_{stack} T_{TP} $T_{neg,out}$ $T_{pos,out}$ $T_{neg,in}$ $T_{pos,in}$	20	