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IMPROVED LIFETIME STACKS FOR HEAVY DUTY TRUCKS THROUGH ULTRA-DURABLE COMPONENTS

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DELIVERABLE REPORT

D2.1: INITIAL PROTOCOL DEFINITION FOR HEAVY-DUTY ACCELERATED STRESS TESTS AND LOAD PROFILE TESTS	
Due Date	30 April 2021
Author (s)	Thomas Mittermeier, Thilo Lehre, Claudia Schwarz, Severin Vierrath, Adam Hodgkinson, Jonathan Sharman, Silvain Buche, Deborah Jones
Workpackage	WP 2, Heavy-duty stack degradation assessment and lifetime prediction
Workpackage Leader	Bosch
Lead Beneficiary	IMTEK
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Date released by Coordinator	12 May 2021

DISSEMINATION LEVEL		
PU	*Public*	***X***
PP	*Restricted to other programme participants (including the Commission Services)*	
RE	*Restricted to a group specified by the consortium (including the Commission Services)*	
CO	*Confidential, only for members of the consortium (including the Commission Services)*	

NATURE OF THE DELIVERABLE		
R	*Report*	***X***
P	*Prototype*	
D	*Demonstrator*	
O	*Other*	

SUMMARY	
Keywords	*heavy-duty accelerated stress test, heavy-duty load profile test*
Abstract	*Work package 2 of the IMMORTAL project aims to define and perform a set of stack and laboratory cell ageing tests, accelerated and load profile tests, which reflect real heavy-duty truck operation. In this deliverable a first definition of accelerated stress tests (ASTs) and load profile tests (LPTs) is presented. The single cell ASTs are based on light-duty vehicle ASTs as proposed by the U.S. Department of Energy. For the short-stack AST a novel procedure focusing on platinum dissolution is proposed based on voltage cycling via the electrical load on a test bench in hydrogen (anode) and limited air supply (cathode). The definition of the LPT includes load cycling, short stop, cold soak, characterization and short stop to high load.*
Public abstract for confidential deliverables	*As above*

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INITIAL PROTOCOL DEFINITION FOR HEAVY-DUTY ACCELERATED STRESS TESTS AND LOAD PROFILE TESTS

CONTENTS

1 INTRODUCTION

IMMORTAL aims to develop exceptionally durable and high power density MEAs well beyond the current state of the art up to TRL4 by building on understanding of fuel cell degradation pathways specific to heavy-duty truck operation and developing lifetime prediction models from extensive real-life stack operation, accelerated stress test and load profile cycles on short stacks. IMMORTAL encompasses OEMs, tier 1 suppliers, and leading industrial and academic/research organisation partners with long expertise in fuel cell science and technology. Work package 2 of the project aims to define and perform a set of stack and laboratory cell ageing tests, accelerated and load profile tests, which reflect real heavy-duty truck operation.

In this deliverable a first definition of accelerated stress tests (ASTs) and load profile tests (LPTs) is presented. For that purpose protocols have been harmonized among the project partners.

2 ACCELERATED STRESS TESTING (AST)

The selected accelerated stress tests are based on light duty ASTs proposed by the U. S. Department of Energy (DOE)[1] with slight adaptations. The following tables gather the protocols used in each group. Any differences between them were conserved to see the impact of that difference on the AST outcome.

Table 1. Catalyst: platinum dissolution AST

Metric		DOE	JMFC	Bosch	IMTEK	CNRS
Cycle	*Type*	Square wave	Square wave	Square wave	~Square wave	Square wave
	Potential	0.6 – 0.95 V	0.6 – 0.95 V	0.6 – 1.0 V	0.6 – 0.95 V	0.6 – 0.95 V
	Time	6 s	6 s	4 s	8 s	6 s
	Number	30,000	30,000	10,000	30,000	30,000
Anode	*Feed*	H_2 200 sccm	H_2 200 sccm	H_2 200 sccm	H_2 200 sccm	H_2 200 sccm
	T	80°C	80°C	80°C	80°C	80°C
	p	amb	amb	amb	amb	amb
	RH	100%	100%	100%	100%	100%
Cathode	*Feed*	N_2 75 sccm	N_2 75 sccm	N_2 800 sccm	N_2 75 sccm	N_2 75 sccm
	T	80°C	80°C	80°C	80°C	80°C
	p	amb	amb	amb	amb	amb
	RH	100%	100%	100%	100%	100%

Table 2. Catalyst support: carbon corrosion AST

Metric		DOE	JMFC	Bosch	IMTEK	CNRS
Cycle	*Type*	Triangle sweep	Triangle sweep	Triangle sweep	Triangle sweep	Triangle sweep
	Potential	1.0 – 1.5 V	1.0 – 1.5 V	1.0 – 1.5 V	1.0 – 1.5 V	1.0 – 1.5 V
	Time	2 s	2 s	2 s	2 s	2 s
	Number	5,000	5,000	500	5,000	5,000
Anode	*Feed*	H_2	H_2	H_2 200 sccm	H_2	H_2
	T	80°C	80°C	80°C	80°C	80°C
	p	amb	amb	amb	amb	amb
	RH	100%	100%	100%	100%	100%
Cathode	*Feed*	N_2	N_2	N_2 800 sccm	N_2	N_2
	T	80°C	80°C	80°C	80°C	80°C
	p	amb	amb	amb	amb	amb
	RH	100%	100%	100%	100%	100%

Table 3. Membrane: chemical degradation AST

Metric		DOE	JMFC	IMTEK	CNRS
Cycle	*Type*	OCV-hold	OCV-hold	OCV-hold	OCV-hold
	Potential	OCV	OCV	OCV	OCV
	Time	500 h	500 h	100 h	500 h
	Number	1x	1x	1x	1x
Anode	*Feed*	H_2 λ=10 @ 0.2 A/cm²	H_2 λ=10 @ 0.2 A/cm²	H_2 60 sccm/cm²	H_2 λ=10 @ 0.2 A/cm²
	T	61°C	61°C	61°C	61°C
	p	150 kPa abs	150 kPa abs	150 kPa abs	150 kPa abs
	RH	30%	30%	30%	30%

Cathode	*Feed*	air λ=10 @ 0.2 A/cm²	air λ=10 @ 0.2 A/cm²	Air 60 sccm/cm²	air λ=10 @ 0.2 A/cm²
	T	61°C	61°C	61°C	61°C
	p	150 kPa abs	150 kPa abs	150 kPa abs	150 kPa abs
	RH	30%	30%	30%	30%
Cell temp		T_{cell} = 90°C	T_{cell} = 90°C	T_{cell} = 90°C	T_{cell} = 90°C

Table 4. Membrane: combined chemical and mechanical degradation AST

Metric		DOE	JMFC	IMTEK	CNRS
Cycle	*Type*	Square wave	Square wave	Square wave	Square wave
	Potential	OCV @ 0% RH – OCV @ 100% RH	OCV @ 0% RH – OCV @ 100% RH	OCV @ 0% RH – OCV @ 100% RH	OCV @ 0% RH – OCV @ 100% RH
	Time	30 s – 45 s	30 s – 45 s	30 s – 45 s	30 s – 45 s
	Number	x-over > 15mA/cm² 20,000	x-over > 15mA/cm² 20,000		
Anode	*Feed*	H_2 40 sccm/cm²	H_2 40 sccm/cm²	H_2 40 sccm/cm²	H_2 40 sccm/cm²
	T	Dry – 90°C	Dry – 90°C	Dry – 90°C	Dry – 90°C
	p	amb	amb	amb	amb
	RH	0% - 100%	0% - 100%	0% - 100%	0% - 100%
Cathode	*Feed*	Air 40 sccm/cm²	Air 40 sccm/cm²	Air 40 sccm/cm²	Air 40 sccm/cm²
	T	Dry – 90°C	Dry – 90°C	Dry – 90°C	Dry – 90°C
	p	amb	amb	amb	amb
	RH	0% - 100%	0% - 100%	0% - 100%	0% - 100%
Cell temp		T_{cell} = 90°C	T_{cell} = 90°C	T_{cell} = 90°C	T_{cell} = 90°C

2.1 Short stack AST

In the first set of protocol definitions for IMMORTAL, a novel procedure for a short stack (SSt) AST focusing on platinum dissolution is proposed. This SSt AST is based on voltage cycling via the electrical load on a test bench in hydrogen (anode) and limited air supply (cathode). Originally, a similar procedure was proposed for diagnostic means by Toyota Motor Company (TMC).[2] The original idea from TMC is to perform voltage scans (similar to cyclic voltammetry) on a stack with limited air supply. The resulting stack current and voltage as well as cell voltage signals are recorded. The voltage scan rate must be chosen to be high enough to cause substantial capacitive currents in the cells, while the limited air supply needs to ensure that those capacitive currents remain offset by the faradaic oxygen reduction currents, so that the absolute currents always remain in the electronic load operating regime (typically, overall reductive currents on cathodes). This procedure results in comparably "easy" requirements (see below) for the respective test bench, rendering it feasible for inter-laboratory testing.
Test bench prerequisites:

- Electrical load voltage ramp
- Sampling rate ≥10 Hz (current, stack voltage, cell voltage)
- Current accuracy (approx.): at 0.05 V/(cell·s): ≤50 mA

Qualitatively, in this procedure, the operation of fuel cells is reflected more realistically than with AST procedures involving hydrogen and nitrogen atmosphere in anode and cathode, respectively, due to the presence of hydrogen and air in anode and cathode. Absolute currents in this AST are expected to be lower than in real operation. Experimental work on this procedure was not part of the present project period, therefore experimental proof of concept is yet to be achieved.

While the procedure will be further developed in the next project period, Table 5 lists guidelines for the first implementation of the method on a testbench.

Table 5. Guidelines applicable conditions for the here proposed Short stack AST. Note that maximum and minimum voltage are variables in the specific test protocol.

		Anode			Cathode		
Voltage scan rate	**Stack temperature**	**Feed**	**Pressure**	**RH**	**Feed**	**Pressure**	**RH**
$V\ s^{-1}\ cell^{-1}$	°C	$sccm_{H2}\ cell^{-1}\ cm^{-2}$		%	$sccm_{air}\ cell^{-1}\ cm^{-2}$		%
0.02 - 0.2	80 (or max. allowed if lower than 80 °C)	≥3	ambient	90	0.2 - 4.0	ambient	90

3 Load Profile Testing (LPT)

In IMMORTAL, long-term Load Profile Tests (LPTs) are supposed to serve as a reference against which the developed ASTs can be compared and to which the resulting acceleration factors can be calibrated. Therefore, it is necessary to develop and utilize load profile tests relevant to heavy duty vehicle (HDV) applications. One possible approach for the first LPT iteration in IMMORTAL (as also anticipated in the project proposal) is to derive the initial LPT from input originating in other public funded projects, especially from FCH-JU funded H2HAUL.[3] However at the due date of this report, no relevant LPT is available yet from H2HAUL, rendering the search for potential further input necessary (see Table 6).

Table 6. Potential references for initial IMMORTAL LPT development.

Reference	**comment**	**Applicable for IMMORTAL D2.1**
H2HAUL*	No information available yet	×
GIANTLEAP*	Modal drive cycle, based on inter-/intra-city bus (range extender)	×
INSPIRE* / GAIA* /ID-FAST*	Modal drive cycle, based on automotive use case	×
VOLUMETRIQ*	Transient drive cycle, based on LDV use case	×
AVL / Bosch / FPT internal	Limited availability at beginning of project	limited

*FCH-JU funded projects

Table 6 shows that at the initial stage of IMMORTAL a practical and HDV-relevant adaption of LPTs developed in earlier public-funded projects is not feasible (while it may become relevant in the further course of IMMORTAL's WP2, e. g., for D2.4). Therefore, for D2.1, it was necessary to rely on the limited available input from the industry partners and to derive IMMORTAL's own initial LPT. For this purpose, the operation states and stressors that should be reflected by the here-defined LPT were discussed and decided in the project consortium. Specifically, these are:

- Load cycling
- Idling, stand-by
- Bleed down

In IMMORTAL, an LPT runtime of 1,000 – 2,000 h is envisaged. This is between 3 – 7 % of the IMMORTAL lifetime target of 30,000 h. At 3 – 7 % of predicted lifetime, it is required to utilize an accelerated drive cycle in order to produce reliably quantifiable aging rates. This renders the application of a transient drive cycle (which is in principle a real driving profile) impossible, thus creating the necessity to utilize a modal drive cycle (which is a combination of conditions and transitions representative to operation) instead. Based on Bosch's internal degradation budgets and available reference data and including available input from the project partners, an initial IMMORTAL LPT is proposed and described in the following.
Throughout IMMORTAL WP2, it is vital that the chosen ASTs and LPTs can be correlated via acceleration factors specific to the most relevant degradation mechanisms. In order to allow for comparison with degradation rates observed in ASTs, the here-proposed LPT was divided into three individual test blocks (Table 7). The first and second test blocks feature load cycling between two levels each (L ↔ H and LL ↔ H), while the third block involves a combination of these load cycles.

Table 7. Definition of three individual test blocks of the LPT, including dwell and transition times.

segment	**<1,080 h**	**1,080 ≤ LPT time <1,200 h**	**1,200 ≤ LPT time <1,800 h**
load levels	2	2	3
repeat unit	L ↔ H: 0.1 A/cm^2 ↔ 1.5 A/cm^2	LL ↔ H: 0.05 A/cm^2 ↔ 1.5 A/cm^2	5x L ↔ H 1x LL ↔ H
dwell time low	15 s*		
dwell time high	3 s*+ 106 s once every hour		
ramps	Ramp (current) up: 30 s, ramp (current) down: 16 s		

*may have to be adapted to test set-up-specific requirements

According to the partners' additional input for expected HDV drive requirements, further elements to be considered in the LPT described here are:

- Open circuit voltage (OCV) and air bleed down (=short stop in Figure 1) every 8 h
- Cold soak and characterization every week

A comparison of this LPT to the procedure proposed in GIANTLEAP yields approximately 10-times more transients between idle and nominal load in IMMORTAL.[4] This reflects the difference in application between the two projects well (range extender in GIANTLEAP, propulsion in IMMORTAL).
Figure 1 depicts a flow chart of the LPT, including the individual test modules, which are further described below. Note that the times given for the individual blocks of the LPT (<1,080 h, 1,080 ≤ LPT time <1,200 h and 1,200 ≤ LPT time <1,800 h, shown in Table 6 and Figure 1) originate from estimates of the expected degradation rates and may be adapted during the course of the LPTs run during IMMORTAL.

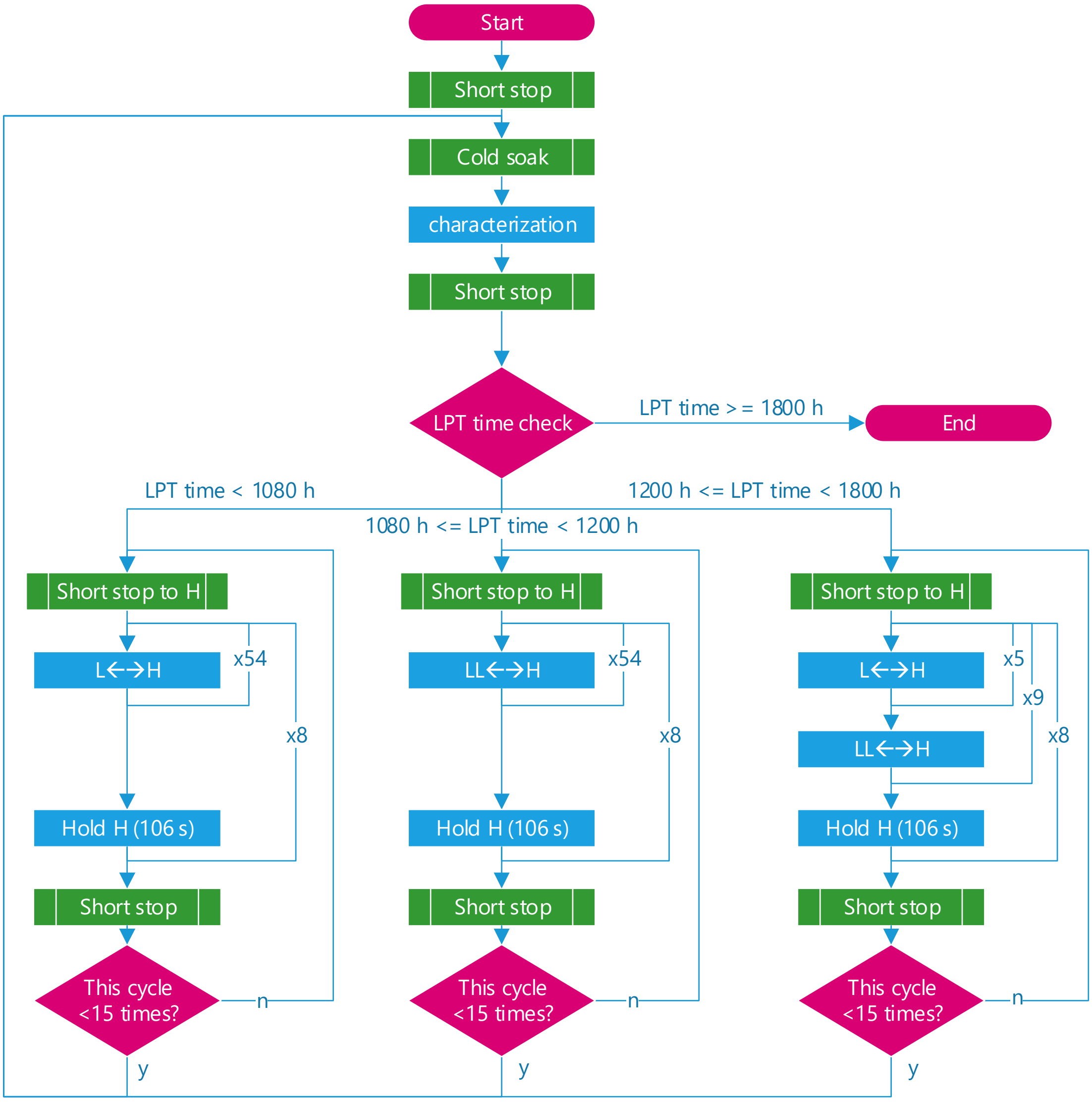

Figure 1. Flow chart of LPT procedure. Each test module is described in the text below.

3.1 Load cycling (L ↔ H, LL↔ H)

Table 8 and Table 9 define the operating conditions for the load levels L, LL and H shown in Table 7 for short stack and subscale single cell experiments, respectively.

Table 8. Definition of operating conditions during LPT load cycles to be applied in short stack experiments.

	current density	coolant stack inlet temperature	anode stack outlet pressure	anode stack inlet gas temperature	anode stack inlet dew point	anode stoichiometry	anode hydrogen composition (dry gas base; rest: N2)	cathode stack outlet pressure	cathode stack inlet gas temperature	cathode stack inlet dew point	cathode stoichiometry
	A/cm²	°C	bar$_a$	°C	°C	-	%	bar$_a$	°C	°C	-
H	1.50	65.0	1.80	70	58.0	1.50	70	1.70	70	58.2	1.80
L	0.10	65.0	1.50	70	58.0	9.00	70	1.30	70	58.2	10.6
LL	0.05	65.0	1.50	70	58.0	18.0	70	1.30	70	58.2	21.2
OCV	0.00	65.0	1.50	70	58.0	Flow as above	70	1.30	70	58.2	Flow as above

Table 9. Definition of operating conditions during LPT load cycles to be applied in subscale single cell experiments.

	current density	cell temperature	anode cell outlet pressure	anode cell inlet gas temperature	anode cell inlet dew point	anode stoichiometry	anode hydrogen composition (dry gas base; rest: N_2)	cathode cell outlet pressure	cathode cell inlet gas temperature	cathode cell inlet dew point	cathode stoichiometry
	A/cm²	°C	bar$_a$	°C	°C	-	%	bar$_a$	°C	°C	-
H	1.50	65	1.50	70	58.0	1.50	70.0	1.20	70	58.2	1.80
L	0.10	65	1.50	70	58.0	Flow as above	70.0	1.20	70	58.2	Flow as above
LL	0.05	65	1.50	70	58.0		70.0	1.20	70	58.2	
OCV	0.00	65	1.50	70	58.0		70.0	1.20	70	58.2	

Comparing Table 8 and Table 9 reveals that only in case of short stack testing are the different load levels associated with different media supply parameter sets. After discussion in the IMMORTAL-consortium, for more robust operation during LPTs, it was decided to keep media supply parameters constant in the case of single cell testing (at the value of the highest load point). Therefore, the parameter sets for short stack and single cell testing were adjusted such to be sufficiently representative of HDV operation and result in projected comparable water management conditions (Bosch internal estimate).

In Figure 2, the transitions between load levels (modules L ↔ H, LL↔ H in Figure 1) are shown schematically. Transitions will be performed as suggested in Stack-Test TM D-02.[5] In short, transitions between different load levels will be performed utilizing ramp times defined in Table 7 for both electrical load and media supply parameters and using defined ramp times. When decreasing current, a delay time will be applied after the start of current ramp until beginning of media supply ramp (see first transition in

Figure 2), while in the case of increasing current, the ramp for media supply is started first, and the current ramp is only applied after another defined delay time (see second transition in Figure 2).

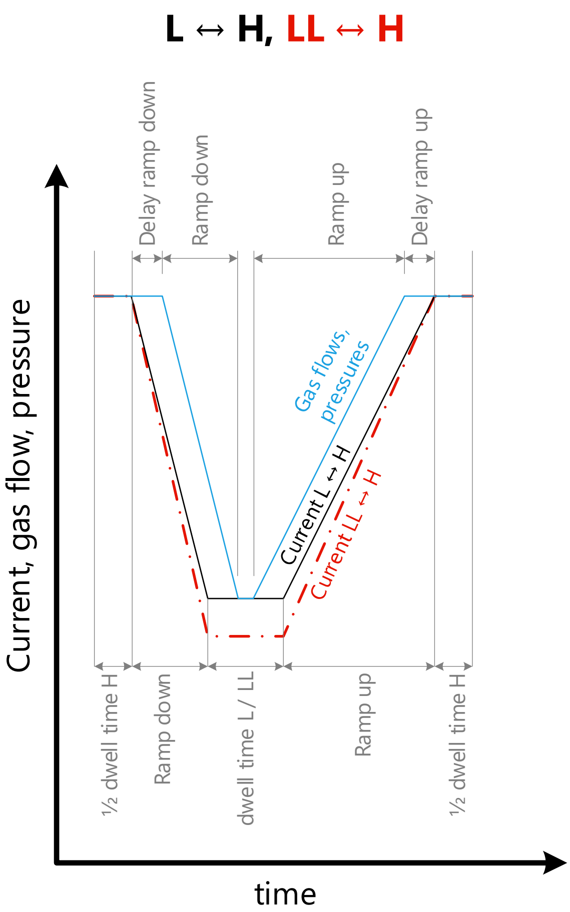

Figure 2. Illustration of transition between load different load levels to be applied during LPT. Note that the blue line qualitatively indicates all media supply parameters listed in Table 8.

3.2 Short stop

The short stop procedure shown in Table 10 and Figure 1 is based on the first part of the performance recovery procedure described in the PEMFC EU harmonisation protocols.[6]

Table 10. Definition of short stops (see Figure 1) during LPTs to be applied in subscale single cell and short stack experiments.

short stop	(assumption: coming from LPT H)	**delay**	**ramp**
1.	any checks to assure defined initial conditions	-	
2.	set electrical load to 0 A (then main contactors off)	-	s. Figure 2
3.	Set media for LPT state L (simultaneous ramps)	s. Figure 2	s. Figure 2
4.	reduce pressures (simultaneous ramps; cath. open, an. 1.2 bara)	-	10 s*
5.	stop air flow to cathode; min flow dry nitrogen (check/adapt voltage alarms)	-	-
6.	wait until U_cell_max < 0.2 V	-	-
7.	wait	600 s	-

*may be adapted to test set-up-specific requirements

3.3 Cold soak

The cold soak procedure shown in Table 11 and Figure 1 is based on the performance recovery procedure described in the PEMFC EU harmonisation protocols and on the U. S. Department of Energy performance recovery protocol.[1,6]

Table 11. Definition of cold soaks (see Figure 1) during LPTs to be applied in subscale single cell and short stack experiments.

cold soak	(assumption: coming from short stop)	**delay**	**ramp**
1.	any checks to assure defined initial conditions	-	-
2.	reduce anode pressure (open)	-	10 s*
3.	stop hydrogen flow on anode; purge with nitrogen	Test bench specific	-
4.	reset (humid) air flow on cathode to initial value. Stop cathode nitrogen flow.	Test bench specific (ensure replacement of H_2 with N_2 in anode first)	-
5.	wait until U_cell_max < 0.2 V	-	-
6.	stop cathode humidification	-	-
7.	set coolant temperature to room temperature	-	-
8.	wait until coolant temperature stack outlet <30 °C	-	-
9.	stop gas flow on anode and cathode	-	-
10.	wait	Adjust to assure frequency of 1 characterization block per week	

*may be adapted to test set-up-specific requirements

3.4 Characterization

Table 7 and Figure 1 define a characterization module for every week of LPT. Here, we propose only one polarization curve protocol for compatibility between IMMORTAL project partners as well as with external data. Specifically, Table 12 shows the conditions for a polarization curve to be performed in between LTP testing. The polarization curve conditions are based on the AutoStack Core project,[7,8] while the procedure described in the Stack-Test protocols shall be applied.[9] Further characterization tests (e. g., leak checks, other polarization curves, cyclic or linear scan voltammetry, electrochemical impedance spectroscopy, etc.) may be performed as available.

Table 12. Definition of operating conditions to be applied during polarization curves used in between LPT cycles (as indicated in Figure 1). Conditions are based on AutoStack Core conditions,[7,8] while the procedure described in Stack-Test shall be applied.[9] While the minimum current density for the application of the stoichiometries listed below is 0.2 A/cm² (and at currents below keeping the same flows as for 0.2 A/cm²), it may be necessary to adapt this value to higher current densities for robust testing (to be examined in pre-tests).

coolant stack inlet temperature	anode stack inlet pressure	anode stack inlet gas temperature	anode stack inlet dew point	anode stoichiometry	anode hydrogen composition (dry gas base; rest: N2)	cathode stack inlet pressure	cathode stack inlet gas temperature	cathode stack inlet dew point	cathode stoichiometry
°C	bar a	°C	°C	-	%	bar a	°C	°C	-
68.0	2.20	73	48.2	1.40	100	2.00	73	52.7	1.60

3.5 Short stop to H

The procedure described in Table 13 is the reverse of the short stop procedure described in Table 10.

Table 13. Definition of transition from short stop to high load (see Figure 1) during LPTs to be applied in subscale single cell and short stack experiments.

short stop to H	(assumption: coming from short stop)	**delay**	**ramp**
1.	any checks to assure defined initial conditions	-	-
2.	set air flow for LPT state L	-	-
3.	stop nitrogen flow on cathode	10 s*	-
4.	reset voltage alarms	-	-
5.	Set media for LPT state H (simultaneous ramps)	-	see Figure 2
6.	connect electrical load and ramp up to LPT state H	s. Figure 2	see Figure 2

*may be adapted to test set-up-specific requirements

4 CONCLUSIONS AND FUTURE WORK

Throughout the project, AST and LPT protocols will be adapted to improve the suitability for heavy duty fuel cell application, which will be presented in another public deliverable report.

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