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# Long-term tests duration reduction for PEMFC µ-CHP application

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Abstract. Proton exchange membrane fuel cells (PEMFC) are extremely promising devices. Nevertheless some technological constraints concerning system durability and reliability costs, still limit their large-scale production. In this framework lifetime prediction and durability enhancement studies are main concerned. To solve this issue, methods based on Prognostic and Health Management (PHM) are developed. It is worth noting that these methods usually require to establish a consistent database 17 concerning the system ageing referring to specific mission profiles. To this purpose, long-term tests are commonly performed. 18 Among different applications, this paper will focus on two micro-cogeneration ( $\mu$ -CHP) durability tests, based on the same load 19 demand. The first test is realized in 1,000 hours while the second one is reduced to 500 hours resulting in a compressed profile. 20 We observed that the respective global voltage degradation rates are similar. Consequently a reflection is proposed to support accelerated tests protocol development.

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23 Keywords: PEM Fuel Cell ; µ-CHP ; Durability ; Long-term tests ; Accelerated tests ; Prognostic and Health Management.

# 1. Introduction

26 Proton exchange membrane fuel cell (PEMFC) appears nowadays to be a promising energy device to face energy 27 transition challenges. Nevertheless, high cost and low 28 29 durability are still the main bottlenecks for their 30 deployment. So, research activities mainly focus on ageing 31 characterization to enhance the system lifetime.

32 In this framework, new approaches based on prognostic and health management (PHM) methods [1-3] are 33 introduced to evaluate the remaining useful life (RUL). An 34 35 example of RUL prediction is given in [2], where the 36 performance of prediction of an echo state networks is analyzed on the voltage degradation. The main objective of 37 the PHM is to reach the FC lifespan by improving the 38 efficiency of the system control and by supporting 39 maintenance and recovery actions. To develop PHM 40 41 algorithms, several long duration tests are required 42 resulting into extremely time consuming and very costly procedures. Then to reduce these constraints, the 43 development of new accelerated stress test (AST) protocols 44 45 to support lifetime prediction is an open question. The main 46 contributions available in the literature are summed up in 47 [4]. Authors reported the main stress factors influencing the 48 PEMFC ageing that are commonly exploited to accelerate 49 the degradation mechanisms. From this analysis, it results that high temperature condition is a common stress factor. 50 51 Moreover, in the literature, different dynamic stress 52 conditions like relative humidity (RH) cycles, improper 53 start and stop and sudden load variations are also used to accelerate the degradation-failure modes [4]. Nevertheless, 54

by real load conditions the main stressors must be 56 57 opportunely increased. According to [4], load cycling is the 58 simplest mode to induce ageing acceleration. In this 59 framework, previous studies focused on load cycles effects 60 can be found in Bae et al. [5] and Jeon et al. [6-7] works. 61 These authors confirmed that increasing the load variation 62 frequency can have a deep impact in FC ageing, especially 63 for membrane degradation [6]. 64 In this paper the ageing effects induced by a real load 65 profile are underlined. Their impact on system durability is analyzed by comparing the results obtained by compressing 66 the reference cycle profile. For this purpose a first long 67 duration test was performed during 1,000 hours. 68 Subsequently the same load profile was scaled to 500 69 70 hours. Experimental activity is presented in the next section 71 (2), while tests results are presented in section 3. Results 72 analysis and perspectives for future works aimed on stack 73 ageing acceleration are proposed in section 4.

to be consistent with the cell components ageing induced

#### 74 2. Context and Experimental Set-up

The proposed experimental activity has been done in the 75 76 framework of the French ANR project PROPICE "Prognostics and Health Management of PEM Fuel Cell 77 78 Systems" [8]. The project aims at establishing a robust 79 prognostic approach for PEMFC in order to determine the 80 Remaining Useful Lifetime (RUL). During this project, 81 both micro-cogeneration (µ-CHP) and transport profiles are 82 tested. Tests durations are fixed to 1,000 hours. This paper 83 focus on the  $\mu$ -CHP profile tests.

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### 2.1. Test bench description

2 A high power test bench (10 kWe), which main3 characteristics are given in table 1, has been used for4 experimental activity.

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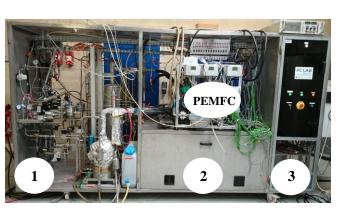
Table 1: Specifications of the 10 kW test bench

Technical Specifications		Range
Cooling circuit		[20-80]°C
Inlet gas temperature *		[20-80]°C
Inlet gas humidification (RH)*		[0-100]%
Water flow rate		[0-20]Nl/min
Gas flow rate	Air	[0-500]Nl/min
	Hydrogen	[0-100]Nl/min
Gas pressure*		[0-2.5]bar
Current		[0-1000]A
*Both at anode and cathode sides		

### 7

8 A picture representing the considered 10 kWe test bench is
9 proposed in figure 1. Both the inlet and outlet gas
10 properties are controlled, such as their flow rate, pressure,
11 temperature and humidification. All the operating variables
12 concerning fluids (1), electrical (2) and thermal flows (3)
13 are recorded at a 1 Hz acquisition frequency all along the
14 test by National Instruments<sup>™</sup> platform (3).

15



# 16 17

18 19 Figure 1: 10 kW test bench

20 The gas flows, the cooling system and the electrical
21 connections are schematized in figure 2, together with the
22 different sensors locations. For a better understanding the
23 sensors' list is reported in table 2.

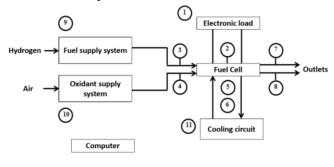


Figure 2: Locations of the different sensors on the test bench

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Table 2: Sensor list

Measurements	Sensor
Current (I)	1
Stack voltage (V)	2
Cell voltages (V <sub>n</sub> )	2
Stack temperature (Ts <sub>eau</sub> )	6
Hydrogen inlet temperature (TH <sub>2</sub> in)	
Hydrogen outlet temperature (TH <sub>2</sub> out)	7
Air inlet temperature (T <sub>Air</sub> in)	4
Air outlet temperature (T <sub>Air</sub> out)	8
Hydrogen inlet relative humidity (HRH <sub>2</sub> in)	3
Hydrogen outlet relative humidity (HRH <sub>2</sub> out)	7
Air inlet relative humidity (HR <sub>Air</sub> in)	4
Air outlet relative humidity (HR <sub>Air</sub> out)	8
Hydrogen inlet pressure (PH <sub>2</sub> in)	3
Hydrogen outlet pressure (PH <sub>2</sub> out)	7
Air inlet pressure (P <sub>Air</sub> in)	4
Air outlet pressure (P <sub>Air</sub> out)	8
Pressure drop ( $\Delta P$ )	3-4
Hydrogen inlet flow rate (DH2in)	3
Air inlet flow rate (D <sub>Air</sub> in)	
Water flow rate (Deau)	5

### 28 **2.2.PEMFC Stack Specifications**

29 Two 8-cells' PEMFC stacks of 1 kWe power are used for
30 experimental activity; one for each test. The nominal
31 specifications of the considered stacks are given in the
table 3.
33

Table 3: 8-cell PEMFC nominal specifications

Specifications	Value	
Active area	220 cm <sup>2</sup>	
Dimensions	220 x 160x 186	
Cooling (water) flow rate	2 l/min	
Anode/cathode stoichiometry	1.5/2	
Anode/cathode inlet pressure	150/150 kPa	
Pressure drop *	30 kPa	
Temperature	80°C	
Relative humidity anode/cathode	50/50 %	
Current density	0.5 A/cm <sup>2</sup>	
*Both at anode and cathode sides		

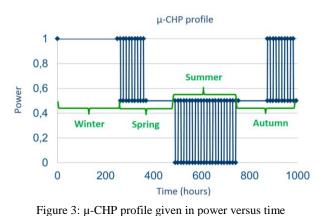
### 35 **2.3.Long duration (1,000 hours) test**

36 The experimental profile proposed in this section and 37 illustrated in figure 3 is based on on-field measurements 38 performed within the framework of a demonstration project 39 where, Electricity of France (EDF) was partner [9]. The 40 reported µ-CHP load profile simulates the behavior of a 41 stationary PEMFC application during a complete year. 42 Consequently the resulting load profile is obtained by combining in series 4 sub-period of 250 hours; one for each 43

season. The specific PEMFC utilization (power demand) is 1 2 linked to each of them as reported below.

- 3 Winter: Steady state operating conditions at the 4 maximal power demand for a period of 250 hours.
  - *Spring* : composed by 2 steps
    - 1. Load cycling [5 cycles; 1 cycle per day] by alternating maximal power and half power operating conditions.
    - 2. Steady state operation at the half power demand for a period of 120 hours.
- 11 Summer: Duty cycle [11 cycles; 1 cycle per day] of 12 which 12 hours at half power and 12 hours at Open 13 Circuit Voltage (OCV). During OCV, the current 14 value is closed to zero and gases are supplied at low 15 flow rates.
  - Autumn : similar to spring but reversed
    - Steady state at half power conditions. 1.
    - Load cycling [5 cycles; 1 cycle per day]. 2.

19 During the test, both the polarization curves and the 20 electrochemical impedance spectra (EIS) at low, mean and 21 high current values are measured once a week. The same 22 measurements are performed at the beginning and at the 23 end of the test. 24



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#### 28 2.4. Reduced (500 hours) test

29 The 500 hours ageing test is obtained by scaling in time 30 domain the µ-CHP load profile applied for long duration test. In the load profile compression procedure, the load 31 32 variation conditions and, in particular the ramps assumed to 33 switch the different load values are kept the same as in the 34 original profile, accordingly to stack dynamic limitations. On the contrary the sub-periods related to the steady state 35 36 conditions are reduced as reported below. The new 37 compressed profile is still composed by four sub-periods 38 (seasons).

- 39 Winter: 125 hours at the maximal power. \_
- 40 Spring: 5 cycles (2 per day) between the maximal \_ 41 power and the half power and 60 hours in steady state 42 conditions at the half power value.
- 43 Summer: 11 duty cycles (2 per day) with 6 hours at half
- 44 power and 6 hours at OCV.

Autumn: 60 hours at half power and 5 cycles between 45 \_ 46 the half power and the maximal power.

47 During the test a new PEMFC stack of the same technology

is used (c.f. table 3). Measurements are scheduled at the 48 49 end of each sub-periods.

#### **3. Experimental Results** 50

51 To perform the experiments, the power demand profile 52 shown in figure 3 is translated into a current density profile. 53

#### 54 **3.1.Long-duration test results**

55 The resulting load profile obtained for the 1,000h ageing 56 test is shown in figure 4, while the corresponding stack 57 voltage response is given in figure 5.

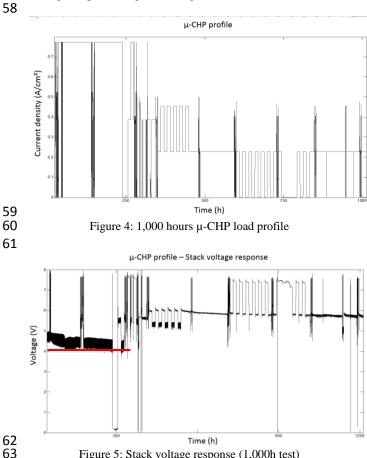


Figure 5: Stack voltage response (1,000h test)

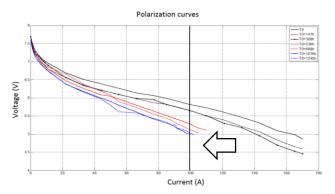
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65 It is possible to observe that the maximal power condition is associated to an initial current density value of 0.77 66 67 A/cm<sup>2</sup>, while the half power state corresponds to 0.38 68 A/cm<sup>2</sup>. As a first result, the maximal power operations 69 severely affected the system performance during the winter 70 sub-period, resulting in a stack critical voltage drop, as shown in figure 5. The stack voltage response decreases 71 72 under the warning level (red line in figure 5).

- 73 Consequently to perform the whole test, the current density
- 74 values corresponding to the maximal and half load modes
- 75 were changed. The new ones are fixed to 0.45A/cm<sup>2</sup> and

0.23 A/cm<sup>2</sup>, respectively. As a consequence, the limits for the polarization curve measurements are changed as shown in figure 6. Afterwards, the 3 reference values considered to perform the EIS measurements are also changed, 5 resulting: 0.09, 0.23 and 0.38 A/cm<sup>2</sup>. 6

The impact of ageing on the polarization curves is presented in figure 6, where their evolutions are reproduced at different steps of the reference load profile.



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11 Figure 6: Ageing impact on the polarization curves (1,000 hours 12 test)

14 Figure 7 represents the EIS curves evolution with ageing for a current density value of 0.38 A/cm<sup>2</sup>. A frequency 15 range comprised between 5 kHz and 100 mHz is 16 17 considered for measurements. The spectra evolution 18 underline a sensible growth of both activation and diffusion 19 losses, in accordance with the polarization curves evolution 20 presented in figure 6. 21

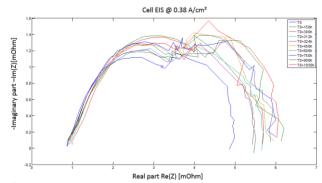




Figure 7: Ageing impact on the EIS measurements (1,000 hours)

25 For a better evaluation of the voltage degradation, the 26 voltage drop is given depending on the time, in table 4. 27 Values are evaluated at the same current density value of 28 0.38 A/cm<sup>2</sup>. After 1,016h of operation a global voltage degradation rate of 597  $\mu V/h$  is observed for the whole 29 30 stack, corresponding to 75 µV/h/cell. The partial 31 information at 147, 308, 536 and 688h are also reported to 32 compare the voltage degradation rate during the different 33 sub-periods.

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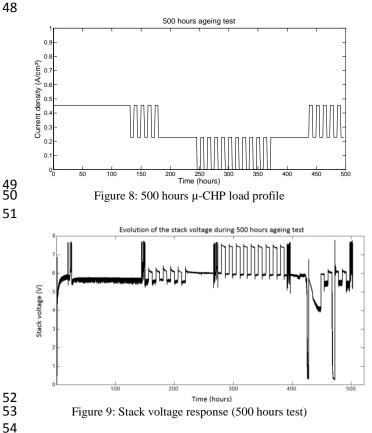
Table 4: Degradations of the 8-cell stack @ 0.38A/cm<sup>2</sup>

Time	Degradation (µV/cell/h)
[T0; T0+147h]	170
[T0; T0+308h]	81
[T0; T0+536h]	116
[T0; T0+688h]	109
[T0 ; Tend]	75

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#### 37 **3.2. Reduced test results**

38 The current density profile of the 500 hours experiment is 39 reported in figure 8, while the corresponding stack voltage 40 is given in figure 9. A maximal current density of 0.45 41  $A/cm^2$  is considered and a value of 0.23  $A/cm^2$  is considered for the half load conditions. So it is worth 42 43 underlining that the current densities values considered in 44 both 1,000 hours and 500 hours profiles are the same, except for the winter steady state conditions. Finally, the 45 46 same metrics proposed in the previous paragraph are 47 considered.

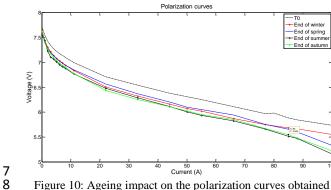


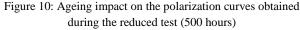
55 Figure 10 illustrates the evolution of polarization curves 56 during the 500 hours, while EIS evolutions at 0.38 A/cm<sup>2</sup> 57 are shown in figure 11. Comparing figure 6 and 10, we 58 observe that the polarization curve obtained at 500 hours in 59 the case of the long duration test (red curve in fig.6) is 60 quite similar to the polarization curve measured at the end 61 of the 500 hours test (green curve in fig.10). While 62 considering the impedance spectra acquired at 0.38 A/cm<sup>2</sup> 63 both in long-duration and reduced test (fig. 7 and 11), we

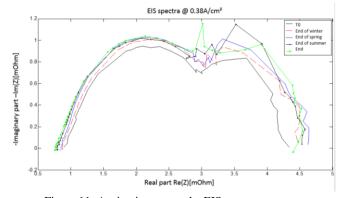
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observe that the same performance losses (activation and
 diffusion) are influenced. Losses intensity are lower in case
 of the reduced duration test, but this comportment can be
 due to the fact that comparing the first spectra at T0 the
 second stack shows the best behavior.







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Figure 11: Ageing impact on the EIS measurements

14 As for long duration test, the voltage drop versus time is reported in table 5. After 500 hours of operations, a voltage 15 degradation rate of 611 µV/h is observed for the whole 16 17 stack, corresponding to 76 µV/h/cell. The information at 18 the end of each season are also reported to compare the 19 voltage rate variation during the different sub-periods. The 20 global voltage degradation rate in both the tests is similar. This point is consistent with the polarization curve 21 22 similitude at 500 hours. For more details, a deeper analysis 23 is presented in the next section.

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Table 5: Degradations of the 8-cell stack @ 0.38A/cm<sup>2</sup>

Time	Degradation (µV/cell/h)
[T0; T0+125h]	184
[T0; T0+245h]	103
[T0; T0+377h]	116
[T0 ; Tend]	76

# 4. Results Analysis and Suggestions

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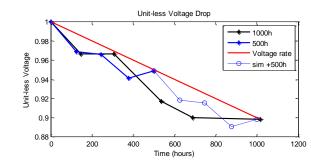
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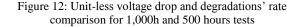
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Results reported in paragraphs 3.1 and 3.2 exhibit the
impact of load cycle profile in system ageing. Tests
performed both at 1,000 and 500 hours mainly show that
even if the test duration is reduced, the global voltage
degradation rate is the same.

34 The results reported in tables 4 and 5 underline that steady 35 state conditions at high current density have an initial deep 36 impact on voltage degradation. Nevertheless this effect 37 seems to be partially recovered during the spring sub-38 period. It is important to notice that data presented in tables 39 4 and 5 are obtained for the same current density value of 40 0.38 A/cm<sup>2</sup>. This can explain the similar voltage rate values 41 obtained in case of high load condition in both the tests. 42 Indeed, if long duration test response showed a critical 43 voltage drop (see fig. 5), this phenomenon was mainly due 44 to the high current density impact on voltage degradation. 45 Nevertheless, this effect was mainly reversible and then, it 46 was recovered by performing the polarization curve. 47 Consequently a similar voltage degradation rate can be observed at 0.38 A/cm<sup>2</sup> in both tests. Moreover by 48 49 analyzing the voltage rate behavior during the different 50 seasons' sub-periods it is possible to state that summer load 51 variations (between OCV and half load values) also have 52 an important impact, while during spring and autumn sub-53 periods a partial recovery is observed. To have a better 54 understanding, the unit-less voltage drops evaluated at 0.38 55  $A/cm^2$  are presented in figure 12. The voltage drop values 56 are normalized with respect to the reference voltage value 57 measured at j=0.38 A/cm<sup>2</sup> and t=T<sub>0</sub>. The red line in figure 58 12 represents the global voltage degradation rate. We can observe that at the end of both tests, the related voltage 59 drops are closed to this line. The voltage degradation rate 60 61 increasing and partial recovery are represented by the black 62 (1,000h) and blue (500 hours) full-lines' slope variations. 63 The light blue circled-line indicates the probable 64 (simulated) voltage degradations obtained in case of the 65 500 hours profile repetition. This line is proposed to underline the voltage rate conservation. Indeed in case of 66 67 real ageing the effects induced by adding a second 500 68 hours cycle could not be linear, inducing a global voltage 69 rate variation.





For a long duration test of 1,000 hours, the stack operates 1 2 at quite long stationary conditions. These operations induce 3 both irreversible and recoverable voltage losses. In case of Δ 500 hours test, the steady state conditions durations are 5 reduced, while the load variation are kept the same. This 6 procedure seems to conserve the global voltage degradation 7 rate, suggesting that load variations and OCV conditions 8 are the main stressing factors for irreversible degradations. 9 Nevertheless, accelerating the cell ageing means increasing 10 the voltage degradation rate. Consequently, the results 11 presented in this work do not represent an accelerated test but characterize an important support in AST procedure 12 13 development.

14 For a better understanding, future studies will be oriented to check the real impact of stationary conditions duration in 15 16 voltage degradation with respect to the load dynamics. 17 Only quantifying this contribution will be possible to 18 evaluate if in case of load variations, the steady state 19 duration can be neglected in voltage degradation rate 20 evaluation. This information will be fundamental in AST 21 procedure development, indicating the limit conditions to 22 respect in steady state duration reduction during the test. Subsequently the ageing acceleration and then the voltage 23 24 degradation rate growth will be induced stressing the load 25 variation conditions. For this purpose, a first suggestion is to increase the number of load variation repetitions 26 27 proportionally to the reference cycle dynamics. This 28 procedure can be also implemented by increasing the 29 reference load cycle frequency, which means accelerating the real load dynamics. These considerations are in 30 31 accordance with AST works presented in [4-7]. These 32 proposals will allow the development of a suitable AST 33 procedure able to link accelerated and real ageing 34 consistently with real load profile dynamics. Therefore it 35 will be finally possible to reduce the actual test duration and costs for durability analysis and lifetime prediction 36 37 application.

### 5. Conclusion

39 This paper presents French ANR project PROPICE experimental results. Two ageing tests are proposed. The 40 first one is a long duration test of 1,000 hours, developed 41 42 for simulating a µ-CHP load demand profile for a complete 43 year. Effects of long duration stationary conditions, OCV 44 operations and sudden load variations are analyzed. The 45 second test of 500 hours reproduces the same load profile 46 scaled in time domain. The effects of the long-term test 47 duration reduction on voltage decay are then analyzed. A 48 similar voltage degradation rate is observed at the test end 49 in both cases. Moreover, the partial voltage drops analysis 50 suggests that it is possible to induce similar voltage decays by reducing the steady-state operations' duration and 51 52 considering the same load variations. These results are then 53 assumed to be a key point to propose new methods to 54 reduce long duration tests by increasing the real load cycle 55 frequency.

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- 58 6. Acknowledgement
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- 62 *experimental campaigns*.

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