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 concerning the system ageing referring to specific mission profiles. To this purpose, long-term tests are commonly performed. Among different applications, this paper will focus on two micro-cogeneration (μ-CHP) durability tests, based on the same load demand. The first test is realized in 1,000 hours while the second one is reduced to 500 hours resulting in a compressed profile. We observed that the respective global voltage degradation rates are similar. Consequently a reflection is proposed to support accelerated tests protocol development.*

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

1. Introduction

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

In this framework, new approaches based on prognostic and health management (PHM) methods [1-3] are introduced to evaluate the remaining useful life (RUL). An example of RUL prediction is given in [2], where the performance of prediction of an echo state networks is analyzed on the voltage degradation. The main objective of the PHM is to reach the FC lifespan by improving the efficiency of the system control and by supporting maintenance and recovery actions. To develop PHM algorithms, several long duration tests are required resulting into extremely time consuming and very costly procedures. Then to reduce these constraints, the development of new accelerated stress test (AST) protocols to support lifetime prediction is an open question. The main contributions available in the literature are summed up in [4]. Authors reported the main stress factors influencing the PEMFC ageing that are commonly exploited to accelerate the degradation mechanisms. From this analysis, it results that high temperature condition is a common stress factor. Moreover, in the literature, different dynamic stress conditions like relative humidity (RH) cycles, improper start and stop and sudden load variations are also used to accelerate the degradation-failure modes [4]. Nevertheless, to be consistent with the cell components ageing induced by real load conditions the main stressors must be opportunely increased. According to [4], load cycling is the simplest mode to induce ageing acceleration. In this framework, previous studies focused on load cycles effects can be found in Bae et al. [5] and Jeon et al. [6-7] works. These authors confirmed that increasing the load variation frequency can have a deep impact in FC ageing, especially for membrane degradation [6].

In this paper the ageing effects induced by a real load profile are underlined. Their impact on system durability is analyzed by comparing the results obtained by compressing the reference cycle profile. For this purpose a first long duration test was performed during 1,000 hours. Subsequently the same load profile was scaled to 500 hours. Experimental activity is presented in the next section (2), while tests results are presented in section 3. Results analysis and perspectives for future works aimed on stack ageing acceleration are proposed in section 4.

2. Context and Experimental Set-up

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

2.1. Test bench description

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

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**		

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

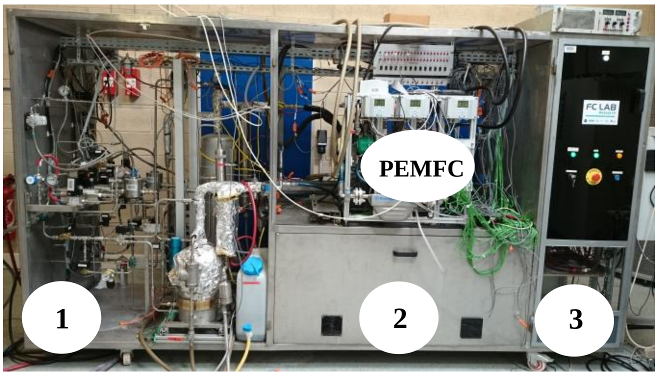

Figure 1: 10 kW test bench

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

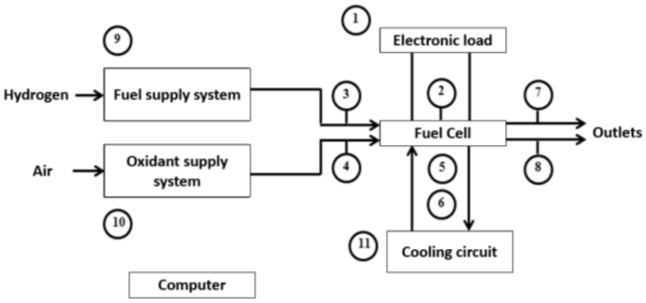

Figure 2: Locations of the different sensors on the test bench

Table 2: Sensor list

Measurements	Sensor
Current (I)	1
Stack voltage (V)	2
Cell voltages (V_n)	2
Stack temperature (Ts_{eau})	6
Hydrogen inlet temperature (TH_2in)	3
Hydrogen outlet temperature (TH_2out)	7
Air inlet temperature ($T_{Air}in$)	4
Air outlet temperature ($T_{Air}out$)	8
Hydrogen inlet relative humidity (HRH_2in)	3
Hydrogen outlet relative humidity (HRH_2out)	7
Air inlet relative humidity ($HR_{Air}in$)	4
Air outlet relative humidity ($HR_{Air}out$)	8
Hydrogen inlet pressure (PH_2in)	3
Hydrogen outlet pressure (PH_2out)	7
Air inlet pressure ($P_{Air}in$)	4
Air outlet pressure ($P_{Air}out$)	8
Pressure drop (ΔP)	3-4
Hydrogen inlet flow rate (DH2in)	3
Air inlet flow rate ($D_{Air}in$)	4
Water flow rate (Deau)	5

2.2. PEMFC Stack Specifications

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

Table 3: 8-cell PEMFC nominal specifications

Specifications	Value
Active area	220 cm²
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/cm²
***Both at anode and cathode sides**	

2.3. Long duration (1,000 hours) test

The experimental profile proposed in this section and illustrated in figure 3 is based on on-field measurements performed within the framework of a demonstration project where, Electricity of France (EDF) was partner [9]. The reported µ-CHP load profile simulates the behavior of a stationary PEMFC application during a complete year. Consequently the resulting load profile is obtained by combining in series 4 sub-period of 250 hours; one for each season. The specific PEMFC utilization (power demand) is linked to each of them as reported below.

- *Winter*: Steady state operating conditions at the 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.
- *Summer:* Duty cycle [11 cycles; 1 cycle per day] of which 12 hours at half power and 12 hours at Open Circuit Voltage (OCV). During OCV, the current value is closed to zero and gases are supplied at low flow rates.
- *Autumn* : similar to spring but reversed
    1. Steady state at half power conditions.
    2. Load cycling [5 cycles; 1 cycle per day].

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

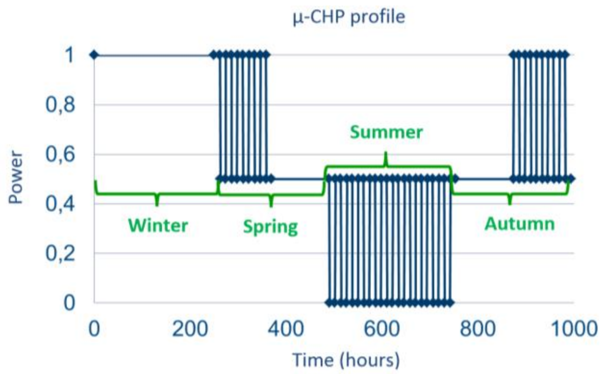

Figure 3: µ-CHP profile given in power versus time

2.4. Reduced (500 hours) test

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

- *Winter:* 125 hours at the maximal power.
- *Spring:* 5 cycles (2 per day) between the maximal power and the half power and 60 hours in steady state conditions at the half power value.
- *Summer*: 11 duty cycles (2 per day) with 6 hours at half power and 6 hours at OCV.
- *Autumn*: 60 hours at half power and 5 cycles between the half power and the maximal power.

During the test a new PEMFC stack of the same technology is used (c.f. table 3). Measurements are scheduled at the end of each sub-periods.

3. Experimental Results

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

3.1. Long-duration test results

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

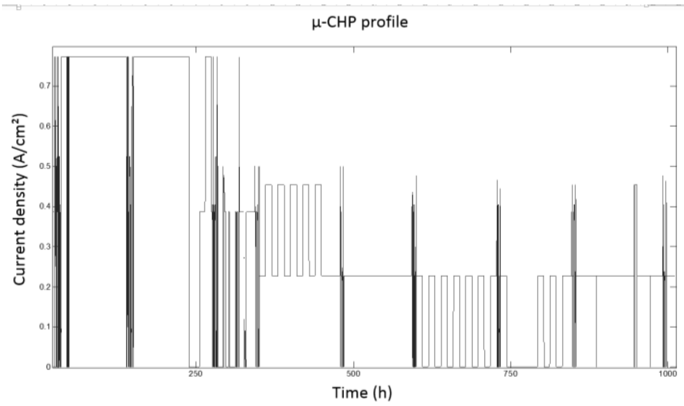

Figure 4: 1,000 hours µ-CHP load profile

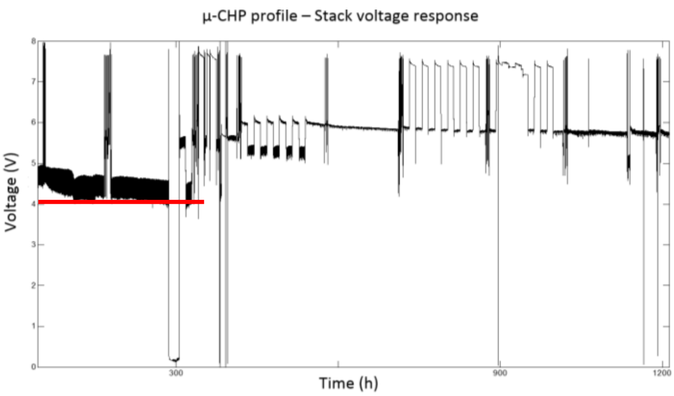

Figure 5: Stack voltage response (1,000h test)

It is possible to observe that the maximal power condition is associated to an initial current density value of 0.77 A/cm², while the half power state corresponds to 0.38 A/cm². As a first result, the maximal power operations severely affected the system performance during the winter sub-period, resulting in a stack critical voltage drop, as shown in figure 5. The stack voltage response decreases under the warning level (red line in figure 5).
Consequently to perform the whole test, the current density values corresponding to the maximal and half load modes were changed. The new ones are fixed to 0.45A/cm² and

0.23 A/cm², 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, resulting: 0.09, 0.23 and 0.38 A/cm².

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.

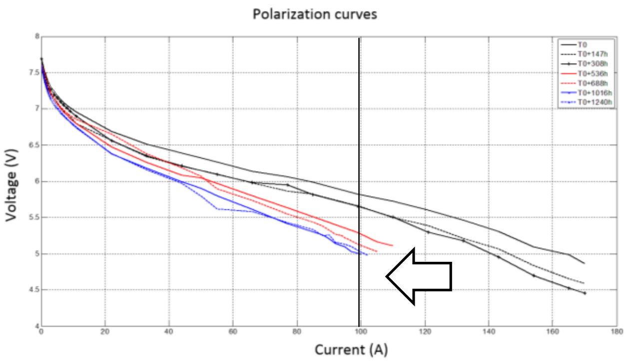

Figure 6: Ageing impact on the polarization curves (1,000 hours test)

Figure 7 represents the EIS curves evolution with ageing for a current density value of 0.38 A/cm². A frequency range comprised between 5 kHz and 100 mHz is considered for measurements. The spectra evolution underline a sensible growth of both activation and diffusion losses, in accordance with the polarization curves evolution presented in figure 6.

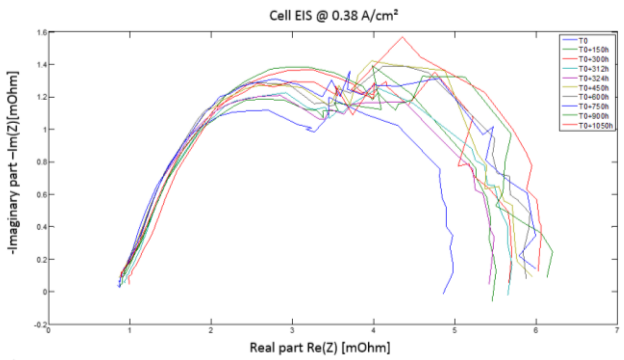

Figure 7: Ageing impact on the EIS measurements (1,000 hours)

For a better evaluation of the voltage degradation, the voltage drop is given depending on the time, in table 4. Values are evaluated at the same current density value of 0.38 A/cm^2. After 1,016h of operation a global voltage degradation rate of 597 µV/h is observed for the whole stack, corresponding to 75 µV/h/cell. The partial information at 147, 308, 536 and 688h are also reported to compare the voltage degradation rate during the different sub-periods.

Table 4: Degradations of the 8-cell stack @ 0.38A/cm²

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

3.2. Reduced test results

The current density profile of the 500 hours experiment is reported in figure 8, while the corresponding stack voltage is given in figure 9. A maximal current density of 0.45 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 underlining that the current densities values considered in both 1,000 hours and 500 hours profiles are the same, except for the winter steady state conditions. Finally, the same metrics proposed in the previous paragraph are considered.

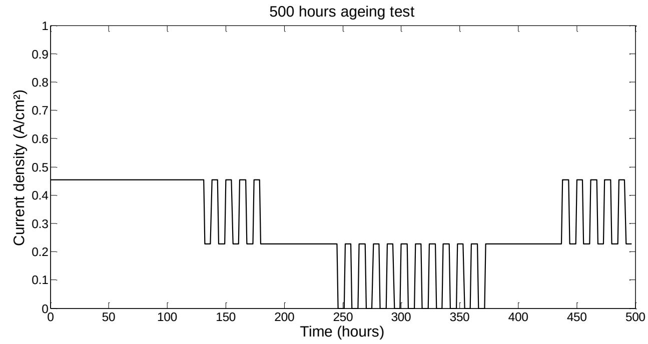

Figure 8: 500 hours µ-CHP load profile

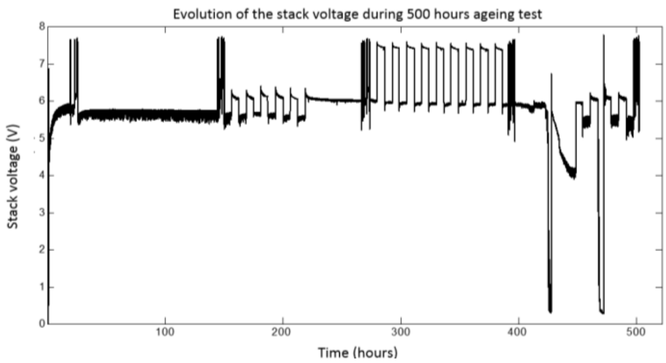

Figure 9: Stack voltage response (500 hours test)

Figure 10 illustrates the evolution of polarization curves during the 500 hours, while EIS evolutions at 0.38 A/cm² are shown in figure 11. Comparing figure 6 and 10, we observe that the polarization curve obtained at 500 hours in the case of the long duration test (red curve in fig.6) is quite similar to the polarization curve measured at the end of the 500 hours test (green curve in fig.10). While considering the impedance spectra acquired at 0.38 A/cm² both in long-duration and reduced test (fig. 7 and 11), we 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.

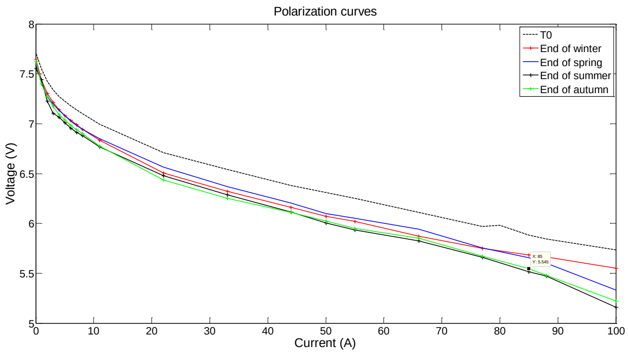

Figure 10: Ageing impact on the polarization curves obtained during the reduced test (500 hours)

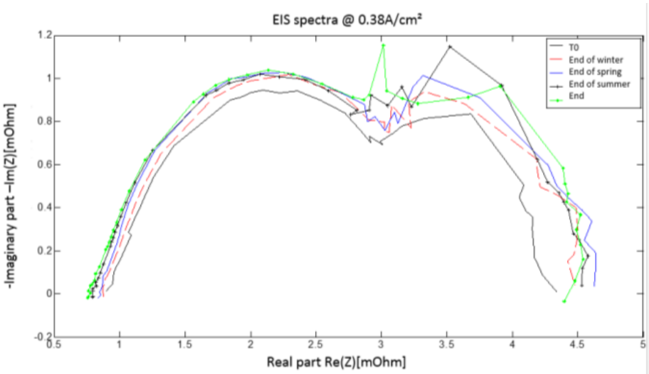

Figure 11: Ageing impact on the EIS measurements

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

Table 5: Degradations of the 8-cell stack @ 0.38A/cm²

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

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.

The results reported in tables 4 and 5 underline that steady state conditions at high current density have an initial deep impact on voltage degradation. Nevertheless this effect seems to be partially recovered during the spring sub-period. It is important to notice that data presented in tables 4 and 5 are obtained for the same current density value of 0.38 A/cm². This can explain the similar voltage rate values obtained in case of high load condition in both the tests. Indeed, if long duration test response showed a critical voltage drop (see fig. 5), this phenomenon was mainly due to the high current density impact on voltage degradation. Nevertheless, this effect was mainly reversible and then, it was recovered by performing the polarization curve. Consequently a similar voltage degradation rate can be observed at 0.38 A/cm^2 in both tests. Moreover by analyzing the voltage rate behavior during the different seasons' sub-periods it is possible to state that summer load variations (between OCV and half load values) also have an important impact, while during spring and autumn sub-periods a partial recovery is observed. To have a better understanding, the unit-less voltage drops evaluated at 0.38 A/cm^2 are presented in figure 12. The voltage drop values are normalized with respect to the reference voltage value measured at j=0.38 A/cm^2 and t=T_0. The red line in figure 12 represents the global voltage degradation rate. We can observe that at the end of both tests, the related voltage drops are closed to this line. The voltage degradation rate increasing and partial recovery are represented by the black (1,000h) and blue (500 hours) full-lines' slope variations. The light blue circled-line indicates the probable (simulated) voltage degradations obtained in case of the 500 hours profile repetition. This line is proposed to underline the voltage rate conservation. Indeed in case of real ageing the effects induced by adding a second 500 hours cycle could not be linear, inducing a global voltage rate variation.

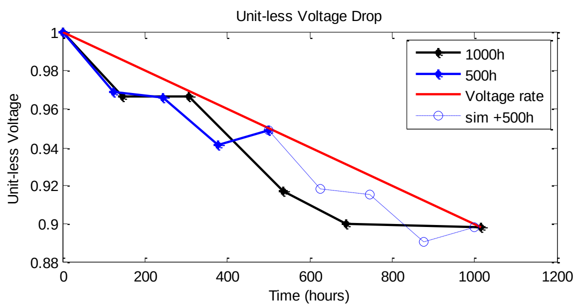

Figure 12: Unit-less voltage drop and degradations' rate comparison for 1,000h and 500 hours tests

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

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

5. Conclusion

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

6. Acknowledgement

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7. References

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