



Prognostic, Diagnostic & Health Management of Fuel Cells – A state of the art

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Motivations

- Part 1 Fuel Cell technology and PEMFC Systems
- Part 2 Behavior and losses of PEMFC
- **Part 3 Prognostics & Health Management**
- Part 4 Ongoing works : diagnostic and health management of FCS
- Part 5 Ongoing works: prognostics of FCS
- **Concluding remarks**







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Motivations





Towards FC systems

- Switching to fuel cell ? Transportation applications
 - The age of utilizing exclusively fossil fuels comes to an end
 - Resource reduction
 - Pollution issues

First alternative: rechargeable batteries

- Significant progress has been made BUT
 - Long duration recharging operation
 - Limited autonomy of the electrical vehicle
 - ⇒ Mostly "hybrid" vehicles : reduce rather than eliminate the dependency on fossil fuels...

Second alternative: fuel cell systems

- When combined with oxygen, hydrogen produces electricity
- Residues: water and heat
- (Theoretical & in-situ) pollutant emissions is zero
- ⇒ Attractive alternative
- ⇒ High energy density (but linked to H2 storage)









Towards FC systems

- Switching to fuel cell ? Stationary applications
 - Increasing interest for the storage of electricity
 - Wide introduction of renewables
 - Intermittency of renewables

First alternative: "classical" solutions

- Electrochemical batteries, flywheels
 - High cost, limited durability, limited energy density
 - → moreover, limited ability to store electricity for long time
- Pumped storage
 - Large scale only at specific places
- Second alternative: hydrogen
 - Exploit the duality between electricity & hydrogen
 - Ability for long duration storage
 - Can be considered at a microgrid level and at a grid level
 - Attractive alternative











Where are development headings?

- Towards enhanced durability
 - Scientific and technological bolts
 - Fuel cell system efficiency
 - Increase it from about 30-40% to about 40-50%
 - Public acceptance
 - Socio-economic aspect: hydrogen-based energy is unknown
 - Strong link with public policies
 - Cost (whole life cycle)

esearch

- Linked to industrial deployment
- Fuel cell system durability (need to increase the lifespan)
 - Ex. for PEMFC systems
 - Common life duration of around 1500 3000 hours
 - Where 5000 hours are required for transportation applications
 - And up to 100000 hours for stationary applications & railways









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Part 1 – Fuel Cell technology and PEMFC Systems





Fuel Cell technology

Is Science Fiction becoming Reality ?

Jules Verne, 1875: "The Mysterious Island"

« ... but after the European mines, [...], the American and Australian mines will for a long time yet provide for the consumption in trade. For how long a time? [...] For at least two hundred and fifty or three hundred years. That is reassuring for us, but a bad look-out for our great-grandchildren! [...] And what will they burn instead of coal? [...] water decomposed into its primitive elements... "

Basic principle discovered and demonstrated in 1839

British physicist William Grove

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Research

For more than a century, the priority given to the development of thermal machines and electrical batteries overshadowed this invention.













Research

Fuel Cell technology

- Principle of a fuel cell
 - What is a Fuel Cell?



- US Fuel Cell Council definition, modified by FC Testing and STandardisation NETwork
 - An electrochemical device that continuously converts the chemical energy of a fuel and an oxidant to electrical energy (DC power), heat and other reaction products. The fuel and oxidant are typically stored outside of the cell and transferred into the cell as the reactants are consumed.
- Main difference with "traditional" battery
 - Fuel is supplied continuously & stored outside







- Taxonomy of Fuel Cell

Oper.

Power

AFC – Apollo (NASA)



PEMFC – Car Appl. (CEA)



SOFC -

Stat. Appl. (MSRI)



	Temp. (°C)	range (W)		
DMFC	20 – 90	1 – 100	Low-power portable applications (mobile phones, computers)	
PEMFC	30 – 100	1 – 100k	Automobile / Transport Low-power stationary appl. (residential sector)	1.1
AFC	50 – 200	500 – 10k	Spaceships	100
PAFC	~220	10k – 1M	Domestic heat & electricity co-generation (CHP)	
MCFC	~650	100k – 10M+	High-power units for CHP, maritime applications	
SOFC	500 – 1000	1k – 10M+	Same as MCFC + Transport	-

Main application area





– PEMFC – operating principle

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Fuel / Fuel Oxidizer: H₂ (pure or reformed) / Air







– Structure

Structure of a single cell

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PEMFC = Polymer Exchange Membrane Fuel Cell





- Structure
 - Structure of a stack

■ Assembly of several cells in series ⇒ to increase the operation voltage







– Whole PEMFC System

The stack within a whole system

- Stack "only" converts energy...
- Prior to the electrochemical reaction
 - How to supply "produce", store, and supply the hydrogen and oxygen?
- Posterior to the electrochemical reaction
 - How to manage the electricity generated?
 - How to manage the heat generated?
 - How to manage the water generated?
- During the electrochemical reaction
 - How to control the process?
 - How to ensure safety of the whole system?
- ⇒ FC System = Stack + Ancillaries





Research









- Some ideas about numerical values... (1)

Efficiency

- Maximal (elec.) efficiency of a FC stack ≈ 55%
- In fuel cell power generators, up to 40% of the produced energy is consumed by all their ancillaries

Volume and prize

- □ Fuel cell stack volume ≈ 30% of the fuel cell system volume (70% is linked to ancillaries)
- □ Fuel cell stack price ≈ ancillaries' price
- □ Platinum price (within catalyst) ≈ only about 5% of the price of a whole PEMFC power generator





- Some ideas about numerical values... (2)

Current

- Current density (A/cm2)
 - Directly linked to performances of FC stack materials (membrane, electrode quality, gas diffusion...)
 - Typically between 0.5 et 1 A/cm² (PEMFC)
- Active area
 - For a given cell type, increasing current implies increasing electrode area

Voltage

- Per cell
 - Thermodynamic limitation: 1,18V at atmospheric pressure and at 80°C
 - Open circuit voltage per cell (I=0A): typically 0.9V
 - Nominal voltage per cell: 700mV
 - Minimum voltage per cell: typically about 400mV
- Stack: linked to the number of cells associated in serial arrangement









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Part 2 – Behavior and losses of PEMFC



PEMFC as a complex system



- Building behaviors models would be of prime importance for design, control, diagnostics, optimization... BUT
- FC = highly multiphysics and multiscale systems
 - Multiphysics = electrical, mechanical, thermal engineering, electrochemistry...
 - Multiscale = from the µm to the m
 - Multiscale = different time constants are involved
 - Electrochemistry ≈ instantaneous
 - Electrical power converter $\approx 10^{-4}$ s
 - Membrane water hydration content $\approx 10^{\circ}$ s
 - Temperature $\approx 10^2 s$
 - − Durability $\approx 10^{5}$ s

High difficulty to access internal parameters

- Specific know-how of the manufacturers
- No sensor available



PEMFC behavior is hard to catch. Even if research increases in this area, a "complete" FC system model is still not available. Some developments at the "stack level".

- Characterization of a stack
 - Two useful "tools"
 - Polarization curve
 - Enables to estimate losses
 - Enables to estimate efficiency
 - Electrochemical Impedance Spectroscopy
 - Enables to build impedance spectra (Nyquist plots)
 - Nyquist plot
 - Enables to estimate internal resistances / impedances of a fuel cell
 - Enables to depict and analyze failure / ageing mechanisms (~ feature for PHM community)







Towards modeling

Electrical behavior - however insufficient / all normal-faulty possible modes...





Other modeling approaches

Electrical equivalence



M.Becherif et al., Journal of Power Sources, 2010.



A.Hernandez et al., Fuel Cells, 2006.



Neural networks



S.Jemeï et al., IEEE Trans. on Ind. Elec., 2008.



D.Hissel et al., Int. Rev. of Elec. Eng., 2008.



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Part 3 – Prognostics & Health Management







State-of-the-art

Research



State-of-the-art

Research







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Part 4 – Ongoing works : diagnostic and health management of FC







Failure mechanisms

– Data to be gathered?

Real data are required to assess the health state of the system

Use of a minimum number of actual sensors (linked to feasibility, cost, reliability, dynamic...)

Measurements technically or economically possible	Measurements technically or economically not really possible	Measurements technically or economically obviously not possible
- Stack current	– Single-cell voltages	– Air flow
 Stack voltage 	– Air / H2 pressures (inlet / outlet)	– H2 flow
 Cooling water temperature 	– Stack internal temperatures	– Channels (air, H2, water) flows
– Air / H2 temperatures (inlet / outlet)		 Current density
– Air compressor speed		– Air/ H2 hygrometry
		 Electrolyte membrane water content
		 Stack impedance using a specific impedancemeter
		 Inlet gases composition
		 Outgoing effluents composition

Failure mechanisms

Degradations of the stack

Taxonomy / nature of the degradation

- Mechanical degradation
 - Mainly due to improper manufacturing processes (crack...)
 - ⇒ Often the cause of early failure
- Thermal degradation
 - Use of the cell outside the its optimal operating range ($T^{\circ}...$)
 - ⇒ Involves changes at micro/nano levels changes in physical properties
- Chemical and electrochemical degradation
 - Presence of contaminants like fuel impurities, air pollutants
 - ⇒ Affects electrode kinetics, conductivity and mass transfer affects FC performance

Taxonomy / localization of the degradation

- Membrane
- Catalyst layers (electrodes)
- Gas diffusion layers
- Bipolar plates

Failure mechanisms

– Degradation modeling?

Parameters reducing the FC lifetime

- Fuel impurities (sulfur, CO for PEMFC, ...)
- Oxidant impurities (oil from the compressor, salt from environment, ...)
- Fuel and oxidant stack starvation (linked to the dynamic and the control of the system)
- Temperature supervision (linked to the system control)
- Hydration supervision for PEMFC (linked to the system control)
- Pressure variations (linked to the system control)
- Peak power demands and current ripples (linked to the control and to the power electronics)
- Open circuit voltage operation for PEMFC (linked to the control)...
- ⇒ Gives an overview of potential causes...

Whatever the degradation is...

- It results in a voltage drop
- ⇒ Gives an idea of potential effect...

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- Impact of compressor failure oxidant circuit
 - Compressor failures
 - □ Oxygen starvation ⇒ consequences on performance and durability

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100 90 80 70 60 50 40 30 20 10 0 Oxygen percentage (%)

- This collapse is linked to current density
- Air hygrometry can increase or decrease the phenomena...

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• 3 current concentrations ; 2 sets of operating conditions

• 5 cm² single cell area ; High oxidant flow (to perform the water evacuation)

- Experiment conditions (Gérard et al. 2010a)
- Impact of compressor failure oxidant circuit

Examples of interactions system / stack

Cell potential (mV) T = 80°C T = 50°C 400 Hr = 50% Hr = 80% 300 l = 100 mA/cm² 200 l = 500 mA/cm² $I = 900 \text{ mA/cm}^2$ 100

Impact of compressor failure – oxidant circuit

- Experiments at constant gas flow and low stoichiometry (Kulikovsky et al. 2004, Liu et al. 2006, Gérard et al. 2010)
 - Potential oscillations
 - Potential can reach near zero (or even negative) values

Examples of interactions system / stack

- Impact of compressor failure oxidant circuit
 - Experiments at constant gas flow and low stoichiometry (2)

Nominal conditions I=0.5 A.cm⁻²

- Impact of compressor failure oxidant circuit
 - Consequences on ageing (3) Post Mortem analysis
 - Visual analysis
 - TEM-FEG analysis

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TECHNOLOGIES

- Air Inlet
- High current density in starvation operation
 - Metallic bipolar plate corrosion
 - Marks of seals caused by hot temperature
 - Platinum particles in the membrane at sample "1" (air inlet, high current density in starvation operation)
 - Active layer structure difference between sample "1" and sample "2"

Air Outlet

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No current density in starvation operation

Impact of the converter – current ripple

DC/DC converter

The output fuel cell current is submitted to the high frequency switching leading to a current ripple
 Impact on durability

Impact of the converter – current ripple

Ageing tests

- º 2 durability tests, same new stacks
 - with ripple (5kHz)
 - without ripple
- 5 cell stack, 220 cm²
- Characterizations every week
 - 4 polarization curves
 - 3 EIS (at 3 different current)

Ageing test nominal conditions				
Cooling temperature	348 K			
Relative humidity	50%			
Gas pressure	1.5 bars			
Hydrogen stoichiometry	1.5			
Oxygen stoichiometry	2			
Nominal current density	110 A (0.5 A.cm ⁻²)			
Ripple current frequency	5 kHz			
Ripple current amplitude	20%			

- Impact of the converter current ripple
 - Stack potential comparison
 - zone 1: 264 µV.h⁻¹
 - zone 2: 387 µV.h⁻¹
 - zone 3: 382 µV.h⁻¹
 - zone 4: 507 µV.h⁻¹

- Impact of the converter current ripple (Gérard et al. 2010)
 - Stack potential comparison

Time (h)

- Impact of the converter current ripple
 - Reversible degradation?
 - EIS before and after the characterization process

Health monitoring

- Residual generation for health estimation (Steiner et al. 2010)
 - Aim: diagnose flooding in PEMFC
 - Procedure: analysis of a residual between
 - experimental pressure drop
 - estimated pressure drop (thanks to an Elman neural network)
 - Step 1: output to be estimated
 - Thanks to a fault tree analysis
 - pressure drop ΔP (Darcy's law)
 - Step 2: feature selection
 - Thanks to a fault tree analysis
 - current /
 - dew point temperature T_{dwpt} (inlet relative humidity)
 - stack temperature T
 - air inlet flow rate Q

- Health monitoring
 - Residual generation for health estimation (2)
 - Step 3: estimation of the pressure drop
 - Elman neural network
 - Step 4: residual generation

Health monitoring

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TECHNOLOGIES

Residual generation for health estimation (3)

Input data

FC Voltage

Pressure drop (actual / estimated)

Residual analysis

- Ageing estimation condition monitoring
 - Estimation of the age of a stack (Hissel et al. 2007)
 - Aim: answering the following questions by carrying out low-cost experimental characterization
 - What is the age of the stack?
 - On which conditions it was operated?

- Ageing estimation condition monitoring
 - Estimation of the age of a stack (2)
 - Hyper-parameters
 - Load conditions
 - Type 1: constant current 50 A
 - Type 2: dynamic current profile (from real car solicitation)

Ageing estimation – condition monitoring

- Estimation of the age of a stack (3)
 - Nyquist plot from both experiments

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Stack 1 : constant current 50 A

- Ageing estimation condition monitoring
 - Estimation of the age of a stack (4)
 - Classification results
 - Typical degradation under static current solicitations
 - Typical degradation under dynamical current solicitations
 - Some "transient" operating points (non-assigned points)

- Non intrusive diagnostic through wavelet packet analysis (1/3)
 - Methodology (Yousfi-Steiner & al., 2010)
 - Use directly the voltage signal to perform health assessment of the FC stack (in real time...)
 - Use time-scale signal treatment methods
 - Here Wavelet Transformation is considered
 - Principle : the time-varying signal is projected on a wavelet basis

- Non intrusive diagnostic through wavelet packet analysis (2/3)
 - Example :
 - Experimental results on a PEMFC stack during cathode flooding
 - Here 7 levels are considered, i.e. 254 packets / measured signal

- Non intrusive diagnostic through wavelet packet analysis (3/3)
 - Example :
 - For each wavelet packet, select specific parameters able to characterize a specific fault class
 - Optimization of the representation space (i.e. selecting the most suitable parameters among the total list of parameters)
 - Classification : here experimental results for flooding / non flooding operating mode in a PEMFC stack

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- Ageing estimation past operating time estimation
 - Estimation of fuel cell operating time (Onanena et al. 2010)
 - Aim
 - Estimate fuel cell operating time
 - Thanks to EIS measurements
 - Procedure
 - Latent regression model
 - Automatically split the spectrum into segments
 - Segments are approximated by polynomials

- Ageing estimation past operating time estimation
 - Estimation of fuel cell operating time (2)
 - Feature selection: 6 hyper-parameters
 - Polarization resistance value
 - Minimal value of the imaginary part in the impedance spectrum
 - Its corresponding real part values
 - Its occurring frequency
 - Internal resistance value
 - Its corresponding frequency of occurrence

- Ageing estimation past operating time estimation
 - Estimation of fuel cell operating time (3)
 - Example of results

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Part 5 – Ongoing works: prognostics of Fuel Cell Systems

Data-driven prognostics of PEMFC

- Echo State Networks for prognostics of FEMFC

Background

- Part of Reservoir Computing (H.Jaeger, 2001)
- Better human brain paradigm than traditional ANN

$$\begin{split} \tilde{x}(n) &= f \big(W_{inp}. u(n) + W_{res}. x(n-1) \big) \\ x(n) &= (1-\alpha). x(n-1) + \alpha. \tilde{x}(n) \\ y(n) &= W_{out}. x(n) + W_{feed}. y(n-1)) \\ y(n) &= f (W_{out}. x(n)) \end{split}$$

Outline

- □ Avoid algorithmic complexity ⇒ structural complexity
- Learning phase in a single step: linear optimization (minimize MSE)

Data-driven prognostics of PEMFC

- Echo State Networks for prognostics of FEMFC
 - Application: prediction of a PEMFC degradation
 - Horizon of prediction: 500,1000 and 2500 tu
 - Structure used: Direct and Parallel approach

Direct approach

Research

Parallel approach

Hybrid prognostics of PEMFC

- Matching empirical degradation models
 - Hypotheses
 - FC aging
 - Irreversible with a long time constant
 - Not measurable directly ⇒ deductible from another variable
 - ⇒ Aging observed through voltage drop
 - / Operating conditions
 - Constant current solicitation
 - Study framework
 - Opening applicative limits: model
 - Non-exact (unknown coefficients), Non-stationary (time varying), Non-linear a priori
 - Non Gaussian noise
 - ⇒ Bayesian Tracking Particle filtering framework

Hybrid prognostics of PEMFC

- Matching empirical degradation models

Formulation

Hidden state model (degradation state)

$$x_k = f\left(x_{k-1}, \theta_k, \nu_k\right)$$

• Observation model \Rightarrow Available measurements $z_k = h(x_k, \mu_k)$

Optimal Bayesian solution

- Initial state distribution is known $p(x_0 | z_0) \equiv p(x_0)$
- Obtaining of $p(x_k | z_{1:k})$ in 2 steps (prediction / update)

$$p(x_{k} / z_{1:k-1}) = \int p(x_{k} / x_{k-1}) \cdot p(x_{k-1} / z_{1:k-1}) \cdot dx_{k-1}$$

$$p(x_{k} / z_{1:k-1}) = \frac{p(z_{k} / x_{k}) \cdot p(x_{k} / z_{1:k-1})}{p(x_{k} / z_{1:k-1})}$$

$$p(x_k / z_{1:k}) = \frac{p(z_k / z_{1:k-1})}{p(z_k / z_{1:k-1})}$$

Solving: particle filter

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Estimations at t = 400 h

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TECHNOLOGIES

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Hybrid prognostics of PEMFC

Example of results —

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TECHNOLOGIES

Accuracy of ± 90h on life time duration of 1000h

Predicted RUL comparison FC 2

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Concluding remarks

The interest of H2 technology

FC are promising energy converters

- High efficiency & low noise level
- Possible heat recovery (especially for high temperature FC SOFC)
- Possibly no dependency to fossil fuels
- Energy density is directly linked to the size & weight of the fuel tanks
- Still issues on system-level
 - Interactions between the FC stack & their ancillaries
 - Reliability & durability, Diagnosis & Prognostic
 - Dedicated ancillaries on a tiny market

• H2

- Best candidate for next generation fuel?
- May play a key role in the future energy economy electricity storage for renewable energies
- Still issues on H2 production, public acceptability, on-board storage, distribution facilities

– PHM of PEMFC – a challenging but exciting task!

Issue: durability

- Increase limited lifespan of FCS
- Ex. of PEMFC systems
 - Common life duration of around 2000 3000 hours
 - Where at least 5000 hours are required for transportation applications...

HOW?

- Observe ageing
- Model the behavior
- Assess current health state
- Predict future health states
- Test, optimize and validate the approaches
- Prepare industrial transfer

Open challenges

- FCLAB ? ... Develop this research field
 - FCLAB has been granted with different funding (ongoing...)
 - Regional Government (Franche-Comte Region) PHM-PAC project (2012)
 - ANR :
 - PROPICE (<u>www.propice.ens2m.fr</u>) (2012). Prognostics and Health Management of PEM Fuel Cell Systems.
 - DIAPASON2 (2010) (https://diapason2.eifer.uni-karlsruhe.de/). Diagnostic of PEM fuel cell systems.
 - European Project
 - FCH-JU SAPPHIRE (<u>https://sapphire-project.eifer.kit.edu/</u>) (2012). System Automation of PEMFCs with Prognostics and Health management for Improved Reliability and Economy.
 - FCH-JU D-CODE (<u>http://www.d-code.unisa.it/</u>) (2009). DC/DC Converter-based Diagnostics for PEM systems.

2014: IEEE PHM 2014 Data Challenge

- http://eng.fclab.fr/ieee-phm-2014-data-challenge/
- Competition opens in December...
- June 22-25, 2014: 2014 IEEE Int. Conf. on PHM (Spokane, WA, USA)

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