Post-doctoral position (12 months + 12 months)

Current density and water distribution measurements in operating Proton Exchange Membrane Fuel Cell (PEMFC)

A post-doctoral fellow is offered in the framework of the CEA Flagship Program "Operando" dedicated to transverse activities between the Technological Research Division (DRT) and the fundamental research division (DSM). The CEA has a renowned activity on fuel cell research, in particular operando scattering experiments dedicated to investigate the behavior of fuel cell components in real operating conditions.

The goal of the project is to optimize PEMFC design and operation. The post-doc activity will be focused on the measurement of the current density and of the water distribution in an operating fuel cell :

- to provide a better understanding of PEMFC operation as a function of the operating parameters (Temperature, Gas hydration, Pressure, Gas composition). (first year)
- to investigate the effects of ageing and membrane degradation on the fuel cell performances and water management.

The measurement of the distribution of the current density will be performed using a reliable commercial setup on a full size cell. The CEA developed an approach based on non-intrusive operando Small Angle Neutron Scattering (SANS) experiments used to quantify the water distribution during fuel cell operation both through plane (across the polymer membrane) and in-plane (liquid water at the cell surface). The SANS measurements will be performed at cutting-edge Large Scale Facilities as the high flux European neutron reactor (Institut Laue Langevin, ILL). Complementary high and low resolution neutron imaging experiments could be envisaged to draw a comprehensive 3D picture of water repartition inside the cell.

The candidate should be highly-motivated and self-directed and have a background in physics and/or knowledge on fuel cell tests and their electrochemical characterizations. Some experience with the scattering techniques would be also advantageous.

This post-doctoral fellow will participate to international conferences and will have the opportunity of co-authoring high impact research publications. Knowledge of French and/or English language will be essential.

To apply for this position, please send a CV, names and contacts of two referees and a cover letter to arnaud.morin@cea.fr and sandrine.lyonnard@cea.fr.

The subject is fully described in the following. For further details and questions, please contact: arnaud.morin@cea.fr or sandrine.lyonnard@cea.fr and see http://www-liten.cea.fr/uk/publications/docs/highlights.pdf.

Coupling between the distribution of water and the current density in operating Proton Exchange Membrane Fuel Cell (PEMFC)

GENERAL CONTEXT

Principle of Proton Exchange Membrane Fuel Cell

The proton exchange membrane fuel cell (PEMFC) is an electrochemical converter which produces electricity and heat from hydrogen and oxygen, with water as the only by product during operation (Figure 1). With its fast starting at room temperature, its versatility, its high power density and efficiency, it is considered as a promising energy source as an alternative to fossil fuels, for use in portable, stationary and transport applications [1].

Figure 1 : Schematic representation of a proton exchange membrane fuel cell

Real fuel cells are made of a series of single cells (Fig. 1) assembled into so-called stacks, as displayed in Figure 2a. The stack design, namely cell size and number of unit cells, is adapted according to the targeted operating conditions and application.

Figure 2. (a) Schematic drawing of a PEMFC stack and single cell. (b) Picture of an Hybrid Fuel Cell car integrating a PEMFC stack developed at CEA (inserted picture)

PEMFC is now a commercial product

As a result of intense research and development activities, the PEMFC is a large scale commercial product since few years. Several thousands of small power stationary plants units are installed in

Japan since 4 years. Hyundai and Mercedes have prepared the introduction of fuel cell cars in the market within 2014-2015.

The CEA has developed fuel cell stacks and systems for several years, and successfully implemented them in prototypes of cars, boats or stationary power plants (Figure 2b). The next generation of CEA stack provides a power density as high as 2.5 kW/l and 2 kW/kg thanks to the use of optimized embossed stainless steel bipolar plates integrating the cooling circuit and the gas distribution of both anode and cathode in less than one millimeter in thickness. Durability tests are conducted during several thousands of hours. These specifications are at the best state-of-the-art level. However, these advertisements and good results must not hide the reality. PEMFC is a costly technology and cost reduction often goes at the expense of cost of performance and/or durability. The need of improvement is still crucial and still requires a deep understanding of the system components and behavior.

Water management issue

Current research shows that one of the main issue affecting power output, stability, and lifetime is the amount and distribution of water in the system, which are both strongly affected by the sorption and transport properties of the polymer electrolyte.The water management, i.e., the ability to maintain the dynamic balance of water in the membrane-electrode assembly (MEA) during operation in order to achieve proper membrane hydration without causing electrode flooding [2], is a critical issue affecting both performances and durability of low temperature FCs [3-8]. The great difficulty in developing an effective control of the water management is due to the complexity of the PEMFC operation, resulting from various, correlated, multiphysical, and multiscale phenomena. The overall amount of water within the MEA depends on a number of operating parameters as current density, hydration and flow of the inlet gases, temperature, pressure, etc. But, under given working conditions, the local water content is also related to the spatial heterogeneity of the cell, i.e., the design of the gas distribution channels and the layered structure of the different porous media. Reactants are progressively converted to products from the inlet to the outlet, so that water activity is expected to increase along the in-plane direction. But, at the same time, water is redistributed between anode and cathode through the membrane submitted to chemical and electrical gradients (Figure 3).

The consequence of the coupling of all phenomena is a fully 3D water repartition, inducing heterogeneity in FC performance (current density distribution) [9-13] and degradation [14,15].

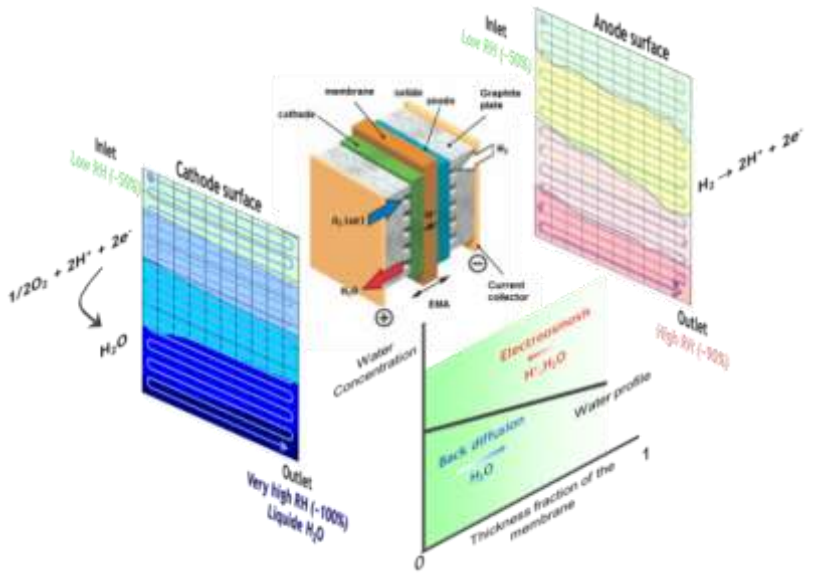

Figure 3. Schematic representation of the 3D water repartition in a PEMFC single cell.

Thus, the need for a fundamental understanding of water transport in the PEMFC membrane-electrode assembly has motivated, over the past few years, the development of new operando diagnostic tools sensitive to the local water content in the components of the cell, namely in the channels of the gas distributors and in the gas diffusion layers, in the anode, the cathode and the membrane constituting the Membrane Electrode Assembly (MEA).

A variety of techniques (magnetic resonance imaging, confocal spectro-microscopy, small-angle neutron scattering,...) are available, with specific limitations: they are more or less intrusive and can be used in more or less representative conditions (cell geometry, membrane thickness, temperature, current density) [16].

UTILITY OF NEUTRONS AND BACKGROUND OF THE PROPOSERS

Neutrons are a very relevant and non-intrusive probe to quantify the water content in the various components of a fuel cell, especially during operation:

i) The metallic materials constituting the fuel cell are transparent to neutrons

ii) The neutrons are extremely sensitive to protons due to the exceptional incoherent cross section of hydrogen atoms with respect to any other atomic species. Therefore, the neutrons probe quite exclusively the properties of protons contained in the ionic phase of the membrane (i.e. charge carriers and hydration protons) and/or in the liquid water located in the components of the MEA and in the gas distribution channels.

Our group has developed since several years a method based on Small angle neutron scattering (SANS). It is the only method that allows to quantify simultaneously the local water content in the membrane (via the direct relation between microstructure and water content of perfluorosulfonated ionomers) and the amount of liquid water outside this component (producing an incoherent proton scattering signal) [16]. Accordingly, the operando technique has provided, up to now, invaluable insights into the water distribution in the working FC [17-26]. A PEMFC transparent to neutrons can be built using standard MEA materials and without significant modification of the state-of-art design (Figure 4a). SANS measurements have thus been carried out under representative conditions of gas stoichiometry, temperature (from -10 to 80 °C), current

density (up to 800 mA.cm^{-2}), pressure (up to 3 bars) and with relative humidity (RH) of fed gases varying from 0% to 100% [20,22,23,25] (Figure 4b).

The local water content in an operating fuel cell has been probed every 30 seconds in front of the rib and the channel of the bipolar plate. These results are currently under publication [26].

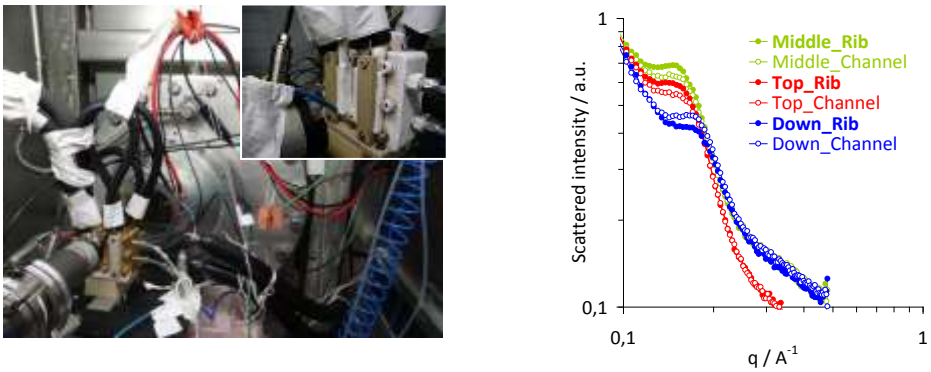

Figure 4. (a) 25 cm² single cell developed at CEA for SANS experiments on D22 beamline at ILL (Picture of the single cell at -10°C inserted). (b) SANS spectra recorded in different areas of an operating PEMFC (30 seconds, sub-millimeter resolution) and used to determine water repartition at 80°C_1.6 bar H2/Air 60/40%RH_St. 1.2/2 counter-flow.

NOVELTY AND GOALS OF THE SUBJECT

Difference in cell design

The previous experiments using SANS were conducted in 25 cm² single cells which are conventionally used worldwide to characterize the performance and the durability of the components of the MEA at the laboratory scale. They allow to test the fuel behavior cell at low cost and on reasonable timescales, in conditions as representative as possible of the real setup, yet slightly different, especially regarding the cell geometry. Basically, the size of the cell, and consequently of the MEA, in a real stack is always larger than those characterized previously by SANS.

The integration of MEA into a real size fuel cell leads to additional effects compared to conventional small single cells :

- It is not possible to reproduce exactly the design of the flow field channel of a stack in a small size single cell, although it is well-known that if significantly affects the water repartition.
- Compared to a single cell, the mechanical stress applied onto the MEA and the local temperature are inherently more heterogeneous in a real stack, which affect both the operation and degradation processes.

Therefore, in the current state of the developments, unveiling the influence of the operating parameters on the water repartition in *full-scale experiments* is required to improve the design of the stacks.

The repartition of liquid water in the channel of a PEMFC stack has been probed by other groups using neutron imaging. Most of these experiments have been conducting at Paul Scherrer Institute and at NIST on the beamline BT2 neutron imaging facility (NIF) which has been financed by General Motors (GM) [16,27,28]. Nevertheless, only the presence of liquid water can be probed with neutron imaging. In other words, it is not possible to quantify the water content in the

membrane, although it is clearly a crucial parameter regarding the performance and durability. Moreover, the time resolution is around 15 minutes, thus much longer compared to SANS experiments. This can lead to some misinterpretation of the water distribution. It is indeed not possible to differentiate water which is accumulating in the probed area and droplets of water which are flowing in front of the beam.

We demonstrated in the last SANS experiments in small single cell that our method allows to reach the time and spatial resolutions relevant to study the behavior of real size PEMFC. So, from now on, efforts can be implemented to perform these studies.

In this post-doc, we plan to perform SANS experiments on a large size single cell with the design and constraints of a prototype stack presenting the best state-of-the-art performance and durability. For the first time, it will be possible to quantify the water content in the membrane and outside the membrane in a real full-scale PEMFC.

Current density distribution

The distribution of the current density will be measured onto the full scale single cell (same design than for SANS experiments) using a commercial device from S^{++}. This kind of measurements is commonly carried out at CEA. The knowledge of the distribution of both current density and water on a common real size cell design is of prime importance for a deep understanding of fuel cell operation and is one of the most original results expected at the end of the post-doc.

Complete 3D characterization of water distribution

We also propose to design a specific single cell in order to perform, for the first time, on the same cell SANS experiments, high resolution and low resolution neutron imaging in order to have a complete view of the three dimensional repartition of water, that-is-to-say water amount within the membrane (through-plane) and within the Anode and Cathode Gas Diffusion Layers (in-plane).

Summary of the goals

This post-doc has two main goals: i/ quantify the effect of *operating parameters and ageing* on the current density and water distribution in a full-scale single PEMFC fully representative of those integrated in cars, boats or stationary power plants have three main goals (Figure 5). ii/ Design a specific cell to conduct SANS, low and high resolution neutron imaging and perform characterizations in representative operating conditions.

The project will provide, for the first time, crucial information on the water management of real-size PEMFC which are required for the improvement of stack prototypes for transportation and stationary applications.

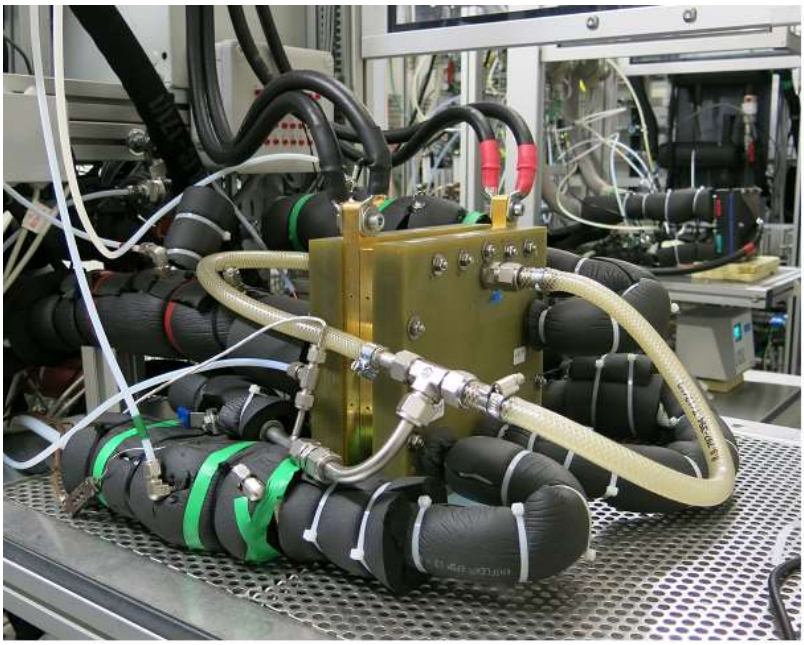

Figure 5. Picture of full-scale single PEMFC

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