Experimental report 02-01 801 : Gestion de l'eau dans une pile à combustible au cours du fonctionnement

The aim of this experiment was first to demonstrate the feasibility of evidencing the membrane swelling during PEMFC in operando by small-angle X-ray Scattering (SAXS). Similar Neutron experiments have been previously performed at the Laboratoire Léon Brillouin (LLB, Saclay, France) using a specially designed aluminium cell which was completely transparent to neutrons. Despite the high quality of the obtained results, it appeared that the spatial resolution was not sufficient (neutron beam size of typically 1 cm) to obtained very local information. The gases are distributed by 1.2 mm channels in the bipolar plates separated by 0.8 mm ribs used to collect the current. It follows that with a 1 cm neutron beam it was not possible to evidence any swelling differences in front of the channels and the ribs. Moreover, while the neutrons are very efficient to characterize the distribution of water within the membrane under stationary conditions, the count time necessary to obtain a reasonable statistic was around 2 minutes which is not sufficient to get information on the transient behaviors. SAXS beamtime on the D2AM beamline was asked because it was possible to get a x-ray beam with a size smaller than 500 µm and preliminary experiments revealed that a spectrum could be acquired in typically 1 s. Therefore, it becomes possible to analyze the behavior in front of the gas distribution channels and the current collecting ribs separately and to follow the evolution of the scattering profiles when a current load is applied to the fuel cell. In addition to the beam size and the flux, the synchrotron source was also necessary to get high energy x-rays in order not to be completely absorbed by the cell and the quantity of water that can fill the gas distribution channels. The first problem was to build a fuel cell transparent to x-rays (Figure 1). We have decided to use a cell design very closed to the one used in the laboratory to evaluate the performance which is made of graphite plates as gas distributors and current collectors. Holes have been drilled in the graphite plates. The membrane-electrode assemblies have been prepared by CEA using thick Nafion membranes (Nafion 117 with a thickness of 200 µm under swollen state) and state-of-art electrodes deposited onto the gas diffusion layer. Thick membranes were used to obtain a very good signal from the polymer. The active area was 25 cm2 which corresponds to a good compromise for controlling the fuel cell behavior and being close to industrial stacks. The metallic end-plates have been prepared with 6 traversing holes in order to be transparent to x-rays by series of two holes close to the gas inlet, the middle of the cell and close to the gas outlet. Each time a hole is in front of a channel and of a rib. Kapton windows were used to close each hole to keep the system gas tight for security reasons and for controlling the fuel cell behavior.

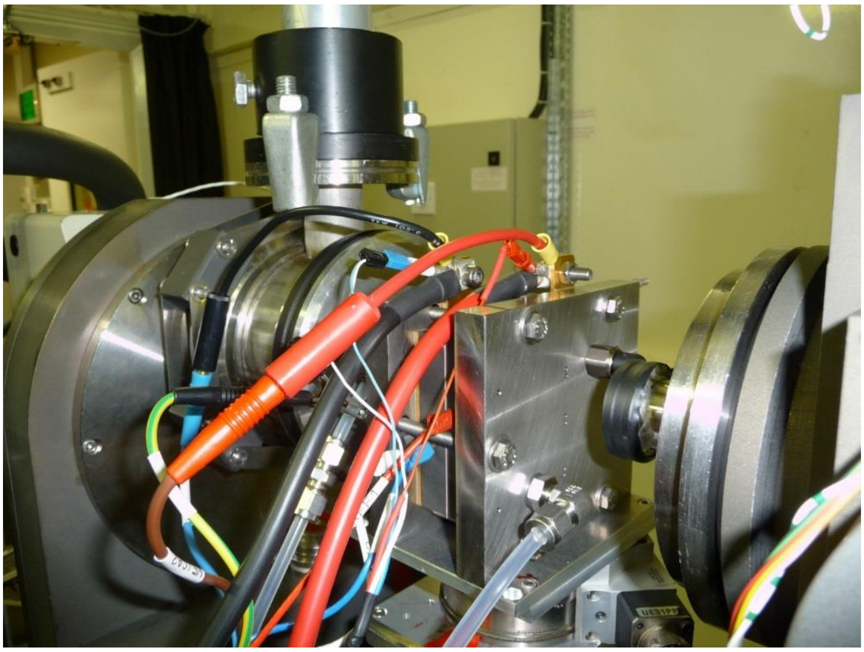

Figure 1. Dedicated single cell for SAXS experiments on D2AM beamline.

The cell was operated at room temperature for sake of simplicity and total back-pressure (at the outlets) was fixed at 1 bar.

Many problems were encountered with the beamline and only three shifts were ok.

We faced some difficulties to avoid flooding of the cell and condensation of the water on the windows which absorbs a huge part of the incident beam.

Finally, we managed to see the evolution of the ionomer peak and demonstrates that it is possible to follow operando the hydration state of the membrane using SAXS (Figure 2).

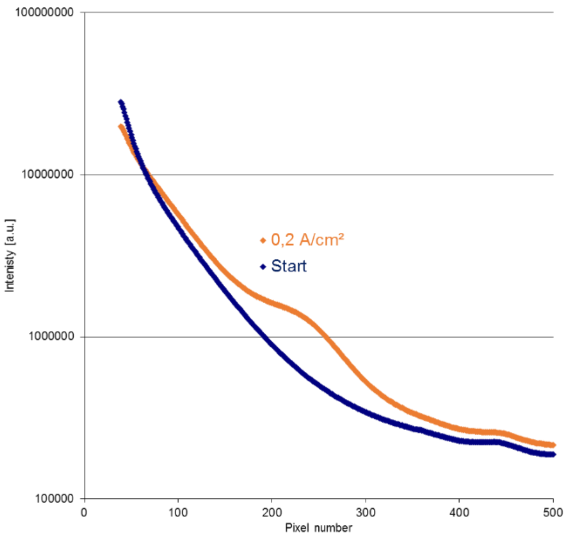

Figure 2. Raw SAXS spectra obtained at room temperature just after starting the cell and after few minutes à 0.2 A/cm². The membrane is dry at the beginning and swells as current is produced, inducing a shift of the ionomer peak towards small angles along with an increase in intensity.