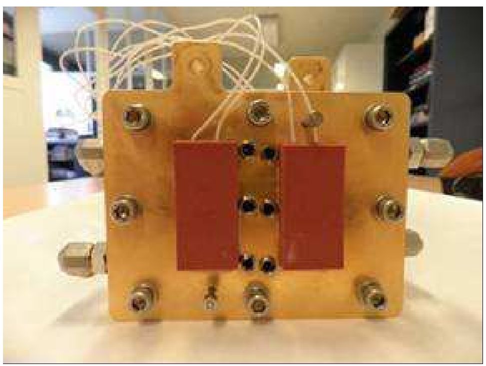
<u>Experimental report SC- 3007</u> : Water management in operating fuel cells by SAXS

The aim of this experiment was first to demonstrate that the water management in fuel cells can be studied *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.

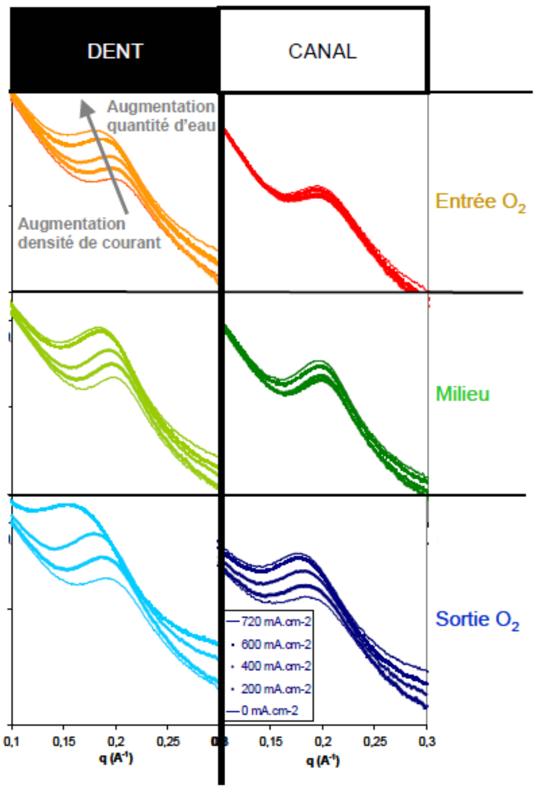
SAXS beamtime on the D2AM beamline was asked because it was possible to get a x-ray beam with a size smaller than 500 µm and preliminary experiments revealed that a spectrum could be acquired in typically 1 s. Therefore, it becomes possible to analyze the behavior in front of the gas distribution channels 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. We have decided to use metallic end plates that will also act a gas distributors and current collectors. In order to avoid, these metallic end-plate have been covered with a thin layer of gold by electrodeposition in order to avoid any corrosion during the experiments (the membranes are highly acidic ion exchange materials). 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 membrane. Thick membranes were used to obtain a very good signal from the polymer. The active area was 25 cm^2 which corresponds to a good compromise for controlling the fuel cell behavior and being close to industrial stacks. The metallic 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 kept the system gas tight for security reasons and for controlling the fuel cell behavior.



<u>Figure 1</u>: Fuel cell designed for in situ and in operando SAXS experiments.

After optimization of the SAXS beam, installation of the complete device (fuel cell test bench, gas inlet, gas outlet, gas humidification, computer control…), security check and so on, we have run three main types of experiments according to the proposal. First, we have completely dried the fuel cell and used the gas humidification system to equilibrate the membrane at different humidity levels in order to get references at equilibrium. Second, we have run the fuel cell under different current load until equilibrium from 0A to 0.8A/cm^2 (The upper limit was defined by our equipment and fuel cell which show that the fuel presented very good electrochemical performance despite the use of a very thick membrane compared to industrial prototypes.



The obtained SAXS data are shown in figure 2 demonstrate the feasibility of the study of a fuel cell by SAXS. The data presented were obtained in front of the channels (right) and in front of the rib (left) in each position of the cell and for different current densities. It appears clearly that the behaviors are different in each of the 6 positions which is a clear evidence of the heterogeneous behavior of the water management in operating fuel cells. For instance close to the gas inlet we do not observe any effect of the current density on the membrane water content (correlated to the peak position and intensity) while a significant effect is observed under the rib.

The third experiment was conducted on a membrane equilibrated in well-defined conditions and the maximum current load (800 mA/cm^2) was asked to the fuel cell and the SAXS data were recorded as a function of time in each of the 6 positions until the cell reach in performance equilibrium and the SAXS data are constant. The results are also very interesting and a complete analysis is still under progress.

As a summary, despite many technical and experimental problems, we can consider that this series of experiments is a full success. The results will be published probably as two different papers (one devoted to the rib/channel different behaviors and one for the analysis of the transient behaviors) as soon as the data treatment will be completed. This data treatment corresponds to the extraction of the water concentration profiles across the membrane and a comparison with the modeling approach conducting at CEA. The water concentration profiles determination is difficult because of the experimental problem encountered in recording the references spectra at equilibrium.