ESRF	**Experiment title:** Characterisation of the nanoporous structure of carbon aerogels by SAXS.	**Experiment number**: SC-964
Beamline: BM2	**Date of experiment**: from: June 5, 2002 to: June 9, 2002	**Date of report**: January 2004
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Report:

The aim of this experiment was to characterise the multiscale morphology of supercritically dried aerogels, before and after pyrolysis. The influence of solvent (water or organic solvent) and catalyst (single step basic, or double step base-acid catalysis) on the structure of resorcinol-formaldehyde aerogels was investigated. Information obtained by SAXS was compared to that obtained by usual methods used for measuring surface areas and pore size distributions. For some samples, the structure was also investigated by WAXS experiments performed in the Laboratoire de Spectrométrie Physique, Grenoble. This project was a part of the PhD work of D. Langohr. The goal of the thesis was to understand the link between the structure of the material and its capacity to store hydrogen because adsorption in carbon nanostructures would be the most suitable way to store hydrogen.

SAXS measurements were performed at the D2AM beamline. Ultra-small angle measurements (USAXS) were made using an incident energy of 7.9 keV, the distance between sample and detector being D = 210 cm. In this configuration the beam-stop, a cross-hair of 300 µm platinum wire, yielded measurements at q values between *ca*. 9×10^{-4} and 10^{-2} Å^{-1}. In a second configuration, providing data in the range ~ 4×10^{-3} Å^{-1} = q = 0.7 Å^{-1}, the incident energy was set to 16 keV and the sample to detector distances were 157 and 30 cm. In this case, the beam-stop was a lead disk of 2 mm diameter. In all cases, an indirect illumination CCD detector (Princeton Instruments) with effective pixel size of 42.5 µm, cooled by a Peltier effect device, was used. Intensity curves $I(q)$, obtained by azimuthal averaging, were corrected for grid distortion, dark current, sample transmission and also for background scattering. Thin slices (0.8 mm) of organic or carbon aerogels were placed in the sample holder without mica windows.

The most important results can be summarised as follows:

- SAXS curves show that the aerogel structure strongly depends on the synthesis parameters (solvent and/or catalysis conditions) because they control the gelation mechanism.
- Anisotropic patterns (fig.1) are observed at the mesoscale for aerogels obtained in acetone just above the gelation threshold.
- By combining SAXS and WAXS experiments (fig.2), we were able to show the existence of narrow nanopores (about 9 Å) in addition to larger ones, around 22 Å, revealed by adsorption. To the best of

our knowledge, it is the first time that such nanopores closely related to the ultimate carbon structure is experimentally established by a method other than gas adsorption. Because their extend depends on the sol-gel reaction, it is likely that they may be related to the degree of polymer cross-linking.

- The surface area determined by SAXS (by means of the invariant Q and the limiting Porod domain shown in Fig. 2) was compared to that obtained by nitrogen adsorption (77K). In some cases the two values were significantly different. When the SAXS surface area is larger (*e.g.*, carbon aerogels synthesised in acetone), it suggests that the nanoporosity is not fully accessible to nitrogen molecules at 77K. When the SAXS surface area is smaller than the adsorption one, it is likely that adsorption occurs in the narrow micropores that are not taken into account in the surface limited by the Porod domain.

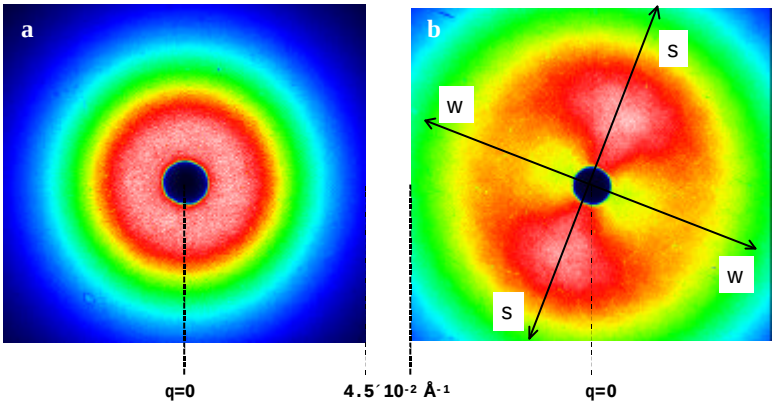

Figure 1. SAXS patterns obtained for carbon aerogels synthesised in water (a) and in acetone (b). The arrows indicate the two directions (w=weak and s=strong) of anisotropy.

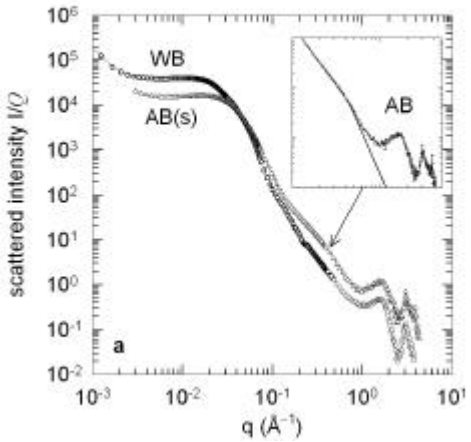

Figure 2. Combined SAXS and WAXS curves. (a) Carbon aerogels synthesised in water (WB) and in acetone (AB); for the sake of clarity the curve AB(w) is omitted; insert shows the Porod plot.

<u>Publications</u>

- 7th International Symposium on Aerogels (Alexandria, 2-5 November, 2003) oral presentation by F. Ehrburger-Dolle

- "Anisotropic high surface area carbon aerogels" S. Berthon-Fabry, D. Langohr, P. Achard, D. Charrier, D. Djurado, F. Ehrburger-Dolle, submitted for publication in J. Non Cryst. Solids (November 2003).