ESRF	**Experiment title:** Static and dynamic X-ray scattering of carbon black - elastomer composites	**Experiment number**: 02 01 94
Beamline: BM02	**Date of experiment**: from: 26 September 2001 to: 1 October 2001	**Date of report**: September 2002
Shifts: 15	**Local contact(s)**: Erik Geissler	*Received at ESRF:*

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Report:

Reinforcement of elastomers with colloidal particles is a well-known fact that has been widely investigated but its mechanism still remains not fully understood. Therefore, we intended to investigate a series of well-defined composites. The parameters involved are the type of elastomer (EPR and SBR), whether it is cross-linked (dicumyl peroxide) or not and the type of filler (carbon black, hydroxylated or hydrophobic pyrogenic silicas). SAXS and uniaxial stretching measurements are performed on samples originating from the same batch.The aim of the project was to determine the long range structure and the relaxation times of this series of samples unstrained and strained by uniaxial stretching at several elongations by means of dynamic X-ray scattering (or X-ray photon correlation spectroscopy, XPCS).

Measurements were made at the BM2 beamline. X-ray energy was 7.9 keV and the sample to detector distance was 2.10 m. In order to perform SAXS measurements at ultra small angles (USAXS), background arising from the guard slits has to be reduced. After polishing the tantalum cylinder of the guard slits, the background is concentrated in two perpendicular thin streaks. Therefore a crosshair beamstop consisting of 300 µm platinum wire was used instead of the 500µm diameter disc. To obtain a coherent X-ray beam, the primary slits were closed to 200×200 µm^2 and a circular pinhole (diameter = 10 µm) was used. This experimental set-up and the direct illumination CCD detector (22µm resolution) permitted to reach low q values ($\cong 6 \times 10^{-4}$ Å^{-1}) and to obtain a coherence factor β close to 0.17 [1]. The beam intensity on the sample was close to 2×10^6 photons/s for a 200 mA ring current. For each sample, exposure time was 1 second and 1000 (or 2000) frames were measured.

The results obtained from the analysis of the intensity curves were presented at E-MRS (Strasbourg, 18-21 June 2002, invited lecture) and submitted for publication [2]. A paper dealing with XPCS results is in preparation. Results obtained for carbon black dispersed in EPR is given below as an example.

Publications

[1] **Ultra Small Angle X-Ray Scattering: conditions, limits and results in speckle experiments** F. Livet, F. Bley, F. Ehrburger-Dolle, E. Geissler, D. Lebolloc'h, T. Schulli, SAS2002 XII International conference on Small-Angle Scattering, 25-29 August 2002, Venice (Italy); *submitted to J. Appl. Cryst.*

[2] **Filler networks in elastomers**, F. Ehrburger-Dolle, F. Bley, E. Geissler, F. Livet, I. Morfin, C. Rochas Polymer Networks 2002, 2-6 September 2002, Autrans (France); *submitted to Macromolecular Symposia*

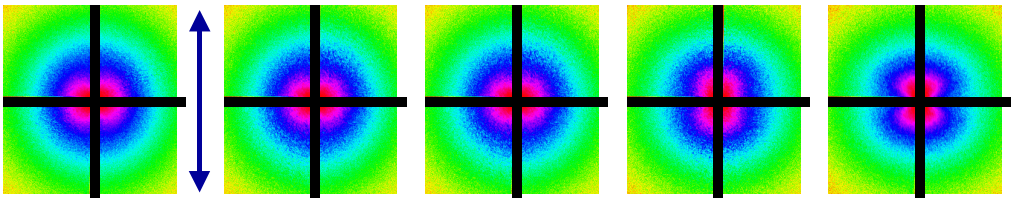

Figure 1. USAXS speckle patterns from carbon black N330 dispersed in uncross-linked EPR. From left to right: before stretching, stretched in the direction of the arrow at ε=0.02, ε=0.07, ε=0.27, ε=0.53. The distance between the centre and any side of the square is equal to 50 pixels (corresponding to 4×10^{-2} nm^{-1}).

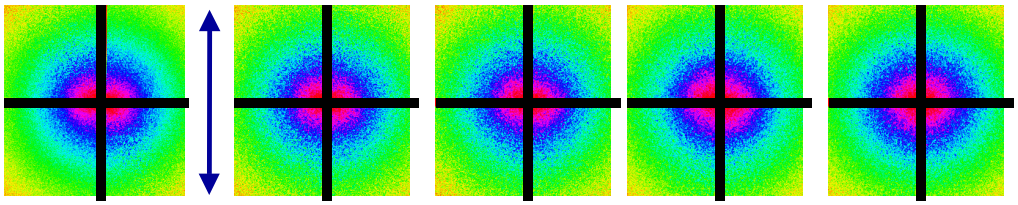

Figure 2. USAXS speckle patterns from carbon black N330 dispersed in cross-linked EPR. From left to right: before stretching, stretched at ε=0.03, ε=0.07, ε=0.13, ε=0.27.

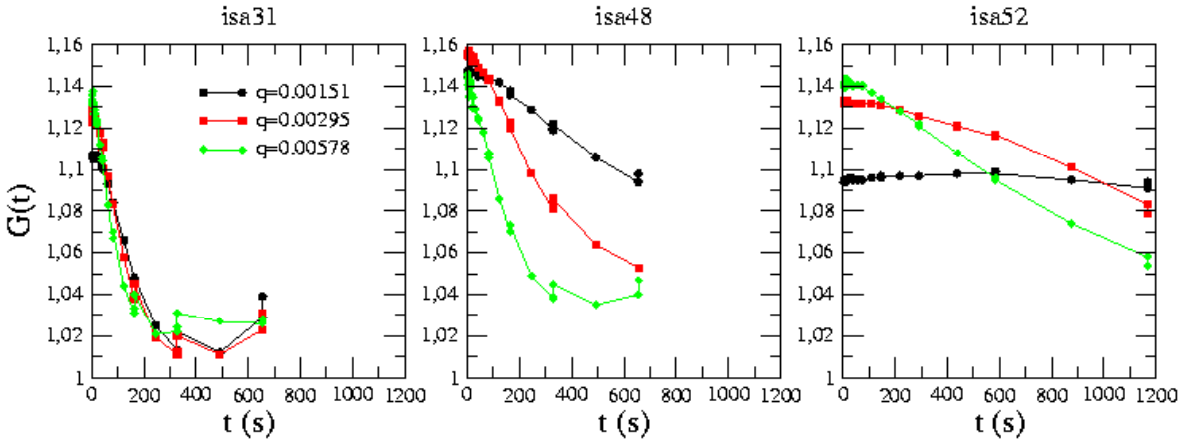

Figure 3. Time correlation function obtained for: uncross-linked N330-EPR stretched at ε=0.07 (isa31, 1000 frames, 1s); cross-linked N330-EPR unstretched (isa48, 1000 frames, 1s) and stretched at ε=0.27 (isa52, 2000 frames, 1s).

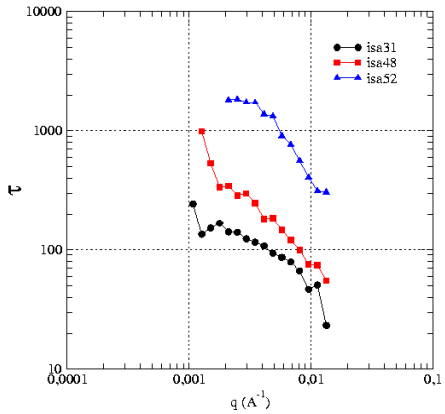

Figure 4. Variation of the correlation time τ as a function of q.

From the analysis of the curves shown in Figure 3 and 4, the following information can be deduced:

- the correlation time τ is much shorter for the uncross-linked composite than for the cross-linked one
- τ increases significantly in the stretched cross-linked sample whereas it remains almost the same for the uncross-linked composite (not shown here)
- τ does not vary as q^{-2}, the absolute value of the exponent is smaller than 2