ESRF	**Experiment title:** Locale Structure of the Filler Network in Reinforced Rubber Nanocomposites	**Experiment number**: SC-3238
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Names and affiliations of applicants (* indicates experimentalists):

*Dr. JESTIN Jacques ; Laboratoire Léon Brillouin, CEA de Saclay ; 91191 Gif sur Yvette

*Dr. BOUE François ; Laboratoire Léon Brillouin, CEA de Saclay ; 91191 Gif sur Yvette

*M. BOUTY Adrien ; Laboratoire Léon Brillouin, CEA de Saclay ; 91191 Gif sur Yvette

*Dr. OBERDISSE Julian ; Laboratoire Charles Coulomb, Pl. Eugène Bataillon ; 34095 MONTPELLIER 5

*M. BAEZA Guilhem ; Laboratoire Charles Coulomb, Pl. Eugène Bataillon ; 34095 MONTPELLIER 5

Report:

Thanks to the good eletronic contrast between Styrene Butadiene Rubber and silica, we successfully measured on ID02 the dispersion of fillers in SBR-Silica nanocomposite systems.

We worked on two kind of systems. First were industrial systems : the filler is an agglomerated silica (supplied by Rhodia) and composites are prepared by mixing directly in an internal mixer the rubber and the silica. Second were model systems : the filler is a silica nanosphere in a colloïdal suspension (Ludox type) and composites are prepared by solvent casting after mixing the rubber and the silica in a good solvent.

Our experimental parameters were :

- The silica fraction (6 fractions from 8%v. to 21%v. for industrial systems, 4 fractions from 1 to 16%v. for model systems),
- The type (OCTEO or Si69) of agent tuning the dispersion, and its quantity (1, 2 or 4 nominal quantities),
- The type of matrix (Non Functionnalized or D3-Functionalized matrix).

That represented all together 160 samples.

For each of our samples, we measured the scattering intensity on three configurations :

- E=12,46keV , Sample-Detector distance = 1m to scan the q range $[10^{-2} ; 5*10^{-1}]$ $Å^{-1}$
- E=12,46keV , Sample-Detector distance = 10m to scan the q range $[10^{-3} ; 5*10^{-2}]$ $Å^{-1}$
- The USAXS setup with the Bonse-Hart camera to reach the q_{min} value 10^{-4} $Å^{-1}$. Knowing the large scales of organisation of the filler (up to 1µm), such a q_{min} value was necessary to caracterize accurately the dispersion.

The 2D-data recorded were then processed, using the software SAXS-Utilities avalaible on place. As our systems were isotropic, the 2D scattering pattern were also isotropic. That's why we reduced these 2D data $I(q_x;q_y)$ to 1D data I(q), with $q=sqrt(q_x^2+q_y^2)$: the scattering signal was thus radially averaged around the

center of scattering. Then, for each sample, the 1D-data I(q) for the 3 configurations were linked together. At the end of this process, we obtained the absolute scattering intensity of the sample I(q) on the entire q-range [10^{-4} ; $5*10^{-1}$] $Å^{-1}$. We finally obtained the signal of silica by substracting the one of the pure matrix.

We show a part obtained spectra representative of our main results in Non Functionalized matrices.

On Fig.1a and 1b below, we can follow the evolution of silica dispersion as a function of Phi_{Silica} for a given type and quantity of agent in industrial systems :

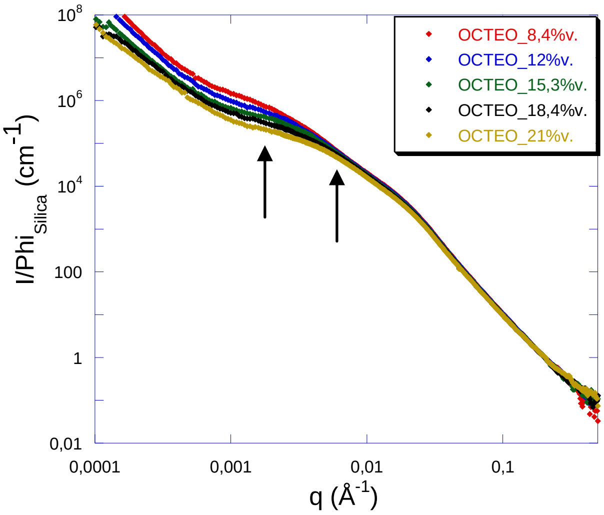

Fig.1a : Silica dispersion tuned by 1 nominal quantity of OCTEO in industrial systems as a function of Phi_{Silica}

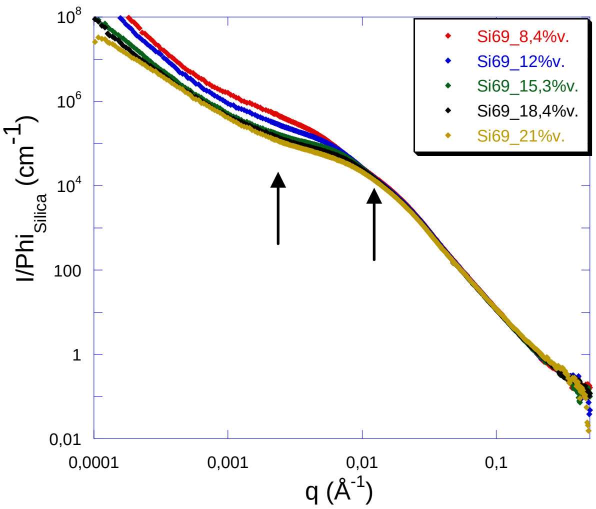

Fig.1b : Silica dispersion tuned by 1 nominal quantity of Si69 in industrial systems as a function of Phi_{Silica}

The separation of the overlayed I/Phi_{Silica} (q) curves happens at higher q with Si69 ($5*10^{-3}<q<1*10^{-2}$ $Å^{-1}$; arrows on Fig.1b) than with OCTEO ($3,5*10^{-3}<q<6,4*10^{-2}$ $Å^{-1}$; arrows on Fig.1a). We interpret this as a better dispersion of silica with Si69 : agglomerated silica is grinded in smaller aggregates with Si69. An accurate analyze is in progress to explain the lowering of the intensity as Phi_{Silica} increases.

We show on Fig.2a and 2b the silica dispersion in some model systems :

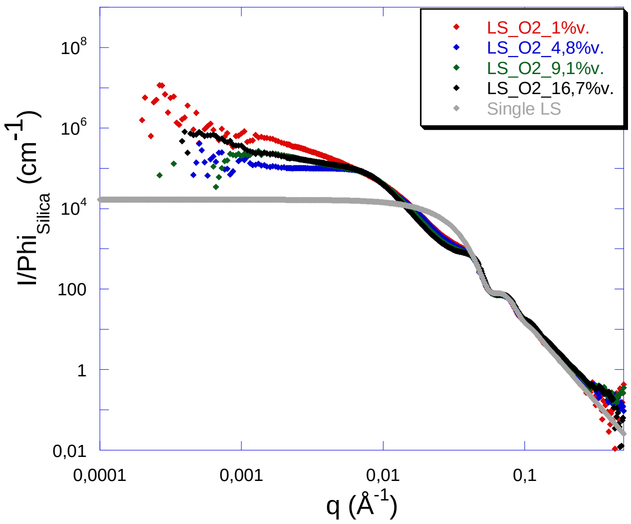

Fig.2a : Silica dispersion tuned by 2 nominal quantities of OCTEO in model systems as a function of Phi_{Silica}

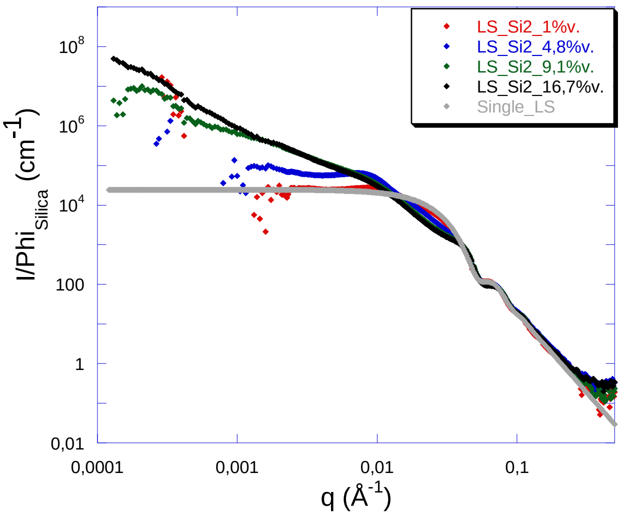

Fig.2b : Silica dispersion tuned by 2 nominal quantities of Si69 in model systems as a function of Phi_{Silica}

In model systems, nanoparticles generally organize in bigger aggregates, as intensities at small q are higher than the one of a single particle (Gray curves : Single LS). For low fractions (1 and 4,8%), this increase at small q is less important with Si69 (Fig.2b, red and blue curves) than with OCTEO (Fig.2a, red and blue curves), showing that the aggregation is more moderated with Si69. The analyze of the structure of the corresponding aggregates is in progress.