DUBBLE	**Experiment Title:** Self-Organization/Phase Behaviour of Hollow Silica Cubes	**Experiment number:** 26-02 692
Beamline: BM26B	**Date(s) of experiment:** From: 04.04.2014 Till: 08.04.2014	**Date of report:** May 2014
Shifts: 12	**Local contact(s);** Dr. Guiseppe Portale	
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Report: (max. 2 pages)

In the performed experiment we have studied the crystalline sediments formed in suspensions of hollow silica colloids [1,2] with a cubic shape with microradian x-ray diffraction (μrad XRD) [3]. We prepared samples of differently sized cubic colloids at different solvent conditions (water, ethanol, salt) to induce different inter-particle interactions and osmotic pressures.

The build-up of the microradian scattering setup was performed during an earlier experiment of our collaborators, which allowed for a much more effective use of the beamtime. Still, an exhaustive re-alignment was needed to be able to meet the challenge of this experiment. Fine tuning of the CRLs and other optics has finally resulted in a very high quality focusing of the beam at the detector.
We were pleased with the granted 12 shifts, which allowed us to do most of the planned measurements. We were very pleased with the rotation and translation stage (sample pillar) that was very user-friendly and allowed perfect sample alignment and easy sample rotations.

Detailed studies of the cube sediments revealed that at different solvent conditions, different structures were formed in the sediments. Height scans of the cube sediments were performed and showed that the structures are effected by an increased osmotic pressure in the sediments. Figure 1 shows the diffraction patterns for cubes of 1246 nm edge length (L) at three different salt conditions in water and in ethanol, where the order decreases with increasing depth in the sediment (the top of the sediment is located at 0 mm). The two water samples show that an increase in salt concentration of 35 μM NaCl results in decreased ordering in the sediment as the pattern becomes less defined. For a different salt, tetramethyl ammonium hydroxide (TMAH), that is known to increase the surface charge of the silica cubes, the extend of order in the sediments increases as seen by the increase in sharpness of the peaks and the increase of higher order peaks in the pattern. The cubes dispersed in ethanol display much less ordering and seem to have a hexagonal type order in contrast to the rhombic order of the water samples.

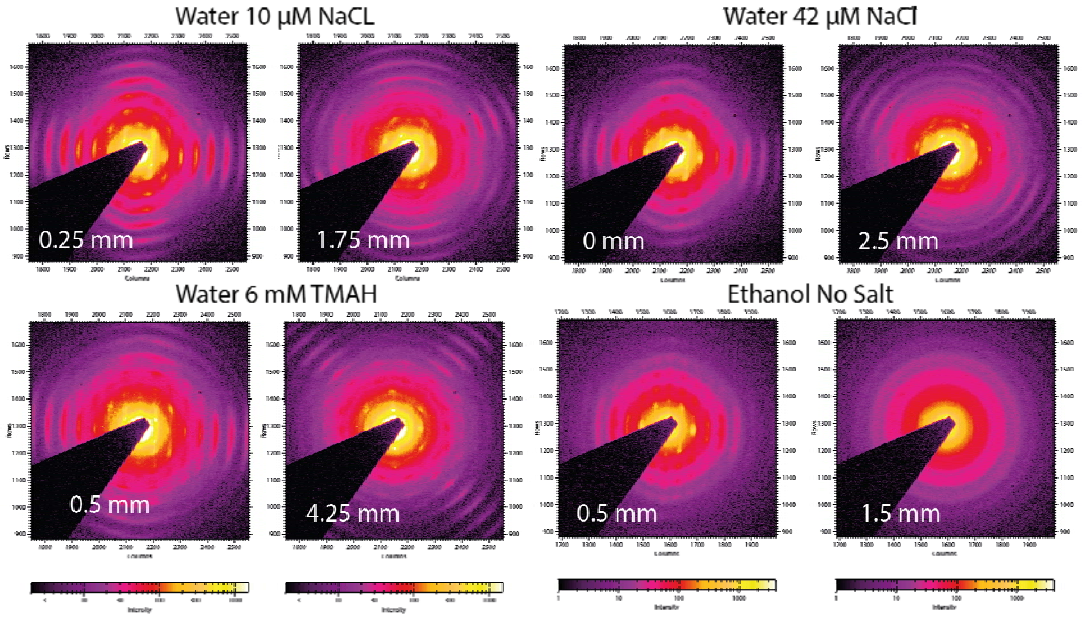

Figure 1. 2D SAXS patterns of cubes with L = 1246 nm at four different solvent conditions at different heights from the sediment top.

The extend of order was investigated in detail by doing rotation scans over 140° (maximum of rotation accessible with the used sample holders) for all samples that showed the presence of Bragg peaks. Figure 2 shows the rotation scans of the 6 mM TMAH in water sample at the position shown above, i.e. 4.25 mm below the top of the sediment. At 35° and 70° different peaks appear in the pattern indicating the presence of a single crystal structure in the sediment. Further analysis will have to be performed to be able to determine the exact crystal structure. The same holds for all other samples where peaks could be observed.

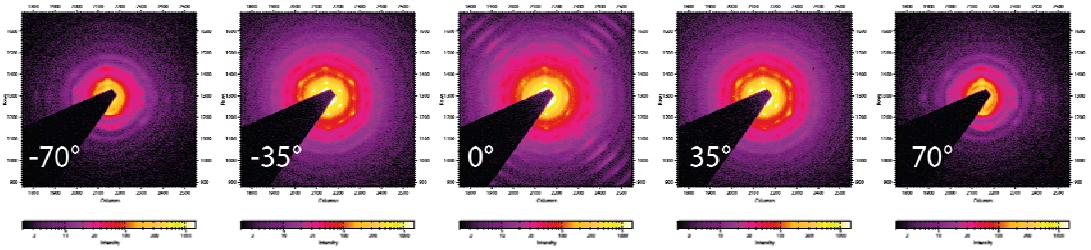

Figure 2. 2D SAXS patterns of the 6 mM TMAH sample at 4.25 mm from the top of the sediment.

To summarize, we highlight that the µrad XRD experiment revealed the presence of ordering in the cube sediments with different structures depending on the exact conditions. Full rotation scans give insight into the exact structure of the sediment.

Finally, we would like to thank G. Portale and D. Hermida Merino for their excellent support.

[1] L. Rossi *et al.*, Soft Matter, 2011, 7, 4139-4142
[2] J.M. Meijer *et al.*, Langmuir, 2012, 28, 7631-7638
[3] J.H.J. Thijssen, et al., Advanced Materials 18, 1662 (2006)