| ESRF | **Experiment title: Grain Growth in Colloidal Crystals** | **Experiment number**: 26-02-865 |
|---|---|---|
| **Beamline**: BM-26B | **Date of experiment**: from: 09/07/18 to: 13/07/18 | **Date of report**: 24/08/18 |
| **Shifts:** 12 | **Local contact(s)**: Michela Brunelli<br>Daniel Hermida Merino | *Received at ESRF:* |

**Names and affiliations of applicants** (* indicates experimentalists):

Dr. Andrei Petoukhov*, Utrecht University

Nicholas Orr*, Oxford University

Dr. Taiki Yanagishima*, Oxford University

Professor Roel Dullens, Oxford University

Daniël ten Napel*, Utrecht University

## Report:

### Prepared samples of silica and TPM

Previously prepared samples of a variety of sizes of 3-(Trimethoxysilyl) Propyl Methacrylate (TPM) and silica spheres dispersed in organic solvents and water were placed in the beam. We determined that the combinations of solvents and particles had a good enough contrast to be the subject of further investigation.

### Crystal development of high concentration TPM dispersions

Initially, samples containing roughly 30% to 40% volume fraction TPM spheres dispersed in both tetralin and a tetralin/ trichloroethylene mixture were prepared. Flat capillaries of pathlength 0.1 to 0.2 mm and width 2 to 4 mm were used as sample chambers. The sizes of TPM spheres were in the range of 1μm to 1.5μm.

Within seconds to minutes of filling the capillary, iridescence was observed. The X-ray diffraction patterns consisted of hexagonal peaks in a single orientation that were persistent throughout the capillary, both in z and x translations. This indicates the presence of a single crystal and is illustrated in figure 1 Our current thinking is this could be due to flow alignment and or promoted by the confinement generated by the capillary walls. These samples were rotated around an axis perpendicular to the X-ray beam to gain information on the structure. More detailed and quantitative analysis is in progress, but initial results suggest that the structure is dominated by FCC.

The evolution of the single crystal was studied over time and a range of z heights. Upon first inspection, the resulting diffraction patterns show that the sample is becoming more polycrystalline throughout; however, the rate of change appeared to be greatest at the bottom of the capillary. We propose this crystal collapse is due to the gradient in osmotic pressure causing a gradient in the lattice spacing leading to a disruption in long range order.

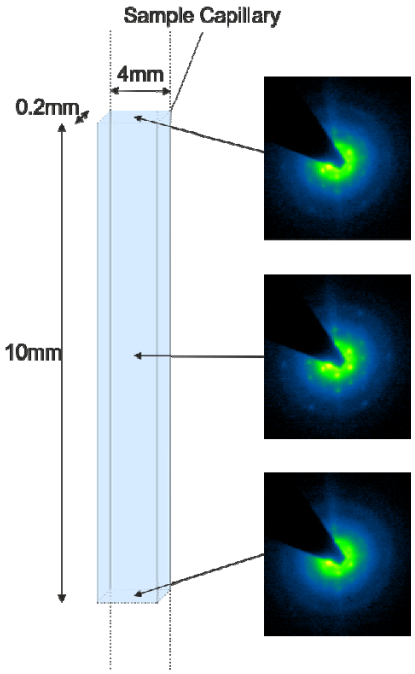


FIG. 1
Initial single crystal formation of 1.1µm TPM spheres. The diffraction peaks are in the same orientation throughout this illustrated region.

**Sedimentation, crystal growth and development of lower concentration TPM dispersions**

We produced some samples of TPM at lower concentrations to the initial samples. Similar scans over time at different z heights were performed. The lowest concentrations did not crystallise over the several hours we observed them. We only saw scattering rings caused by fringes in the particle form factor. An intermediate concentration TPM in tetralin solution was observed to sediment before crystallisation began after roughly 2h. The evolution of the sample within the sediment and higher in the sample was studied over a number of hours, as illustrated in figure 2. Significant crystalline structure was observed.

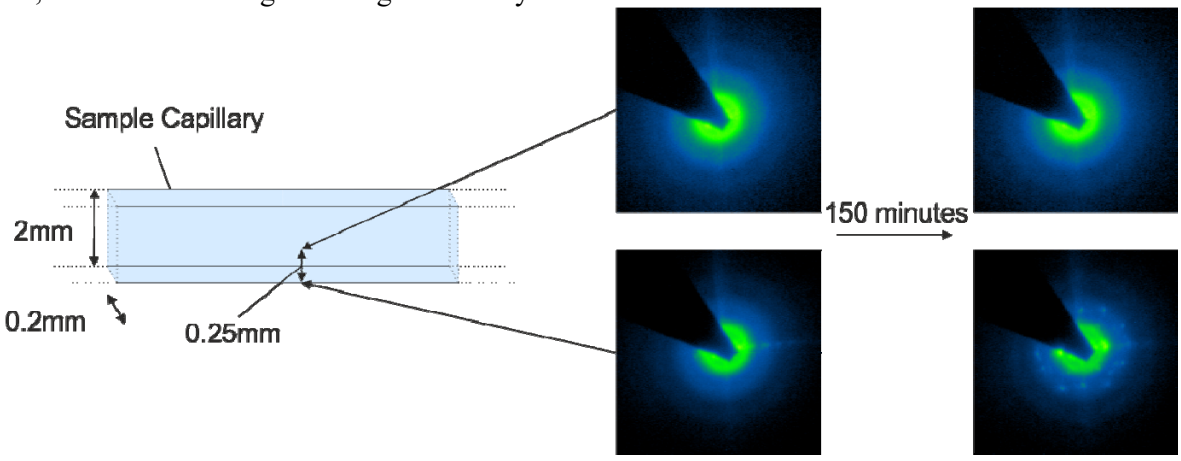


FIG. 2
1.1 µm TPM spheres were placed inside a horizontal capillary. Crystallisation at the bottom of the capillary over 150 minutes is illustrated, shown by the emergence of diffraction peaks. Diffraction peaks were not observed 0.25mm higher in the sample.

**Silica devitrification**

Capillaries containing silica dispersions in water were spun down by a centrifuge to produce a sediment. X-ray diffraction patterns suggest that the sediment is initially amorphous. Over time, the sediment became iridescent at the top, a signature of crystallisation. We jumped at the opportunity to study devitrification dynamics; similarly to before, z scans over time were performed around the crystallising region. Initial inspection indicated substantial crystal growth at the top.

We would like to thank the DUBBLE team and in particular Michela Brunelli and Daniel Hermida Merino for the support throughout our experiment.