ESRF	**Experiment title:** DYNAMIC STRUCTURAL CHARACTERISATION OF FAT SLURRIES	**Experiment number**: 26-02-789
Beamline: BM26B	**Date of experiment**: from: 13/05/2016 to: 16/05/2016	**Date of report**: 20.06.2016
Shifts: 9	**Local contact(s)**: Hermida Merino Daniel	*Received at ESRF:*

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Scientific background. The experiment aimed to study the effect of shear on transient nano- and micro-scale networks of nanosized fat crystal platelets dispersed in oil. Understanding behaviour of fat crystal dispersions in oil under dynamic processing conditions (shear) is of both industrial and fundamental interest. This impedes rational design and engineering of fat-based food materials with enhanced shelf-life stability and sensorial quality. Moreover, this will provide the necessary means to develop the relationship between processing and the growth of the multi-length scale structure of fat network. While knowledge on the crystal polymorphism exists, information about how nanoscale fat crystallites form hierarchical multiscale networks is not understood [1-5]. Preliminary rheological, rheo-MRI and SAXS observations on network formation process under shear showed aggregation and growth of platelet thickness at early stage of network formation. Once a network has formed (ageing), shear results in its irreversible disruption. The mechanisms that underlie network formation and disruption under shear are however unknown. Based on *ex situ* ageing experiments hypotheses on shear-induced fat crystal alignment and clustering have been proposed. Their verification will close the current knowledge gap on nanoscale fat crystal network behavior under shear and thus enable evidence-based recommendations to improve current processing routes of fat-based food products in the foods industry.

Samples and experimental techniques. In the experiments we used Rheo-SAXS to study structural changes of fat crystal dispersions in static and under shear conditions. The experimental setup allowed to probe length scales in the order of 4-210 nm. This length region covers well the cross-sectional dimension of Average Crystallite Thickness (ACT) of fat nanoplatelets which are in the order of 10-100 nm and would allow to observe the presence of liquid-crystalline structure if such should appear. A horizontal Couette shear-cell was designed for this experiment, which allowed to point the X-ray beam along vorticity direction of the flow as well as probing the mesostructure at different positions across the gap between the moving and the static surface of the cell. In order to have a homogeneous flow profile across the gap, a gap of only 1 mm was used with a 38 mm outer radius. Shear rates were varied in a wide range from 0.1 to 140 s^{-1}. The beam cross section at the sample position was about 120 μm, which allowed to measure five different positions across the gap without overlap. We used the Pilatus detector with the pixel size 172x172 microns square positioned at distance of 1400 mm away from sample for the SAXS measurements.

We used two type of oil dispersions that contained different concentrations of ultrarapid crystallized fat crystal nanoplatelets. The nanoplatelets were dispersed in vegetable oil at ambient temperature. In this manner fat crystallisation and crystal network formation were largely decoupled. After preparation, fat crystal dispersions were rapidly frozen at -20 C, which is a known procedure to stall crystal growth/ dissolution.We studied: 1) *Ex situ* aged dispersions under different shear rates; 2) Effect of *in situ* ageing under fixed shear rates on “fresh” fat dispersions; 3) Effect of *ex situ* ageing under mild shear. As a control, experiments in static condition was performed on each dispersion (without applied shear).

Results.

The 2D SAXS patterns shown in Figure 1 were taken from an oil dispersion containing 20% of fat crystalls at static conditions and at shear rates 5 and 70 s^{-1}. The first SAXS pattern for fat crystal dispersions at static conditions (Figure 1a) clearly shows an isotropic diffraction peak characteristic of unoriented material, while the SAXS patterns, that were obtained under the shear, demonstrate visible anisotropy. Thus radial averaging of the patterns have shown the anisotropy for the wide range of shear-rates (1-140 s^{-1}). With rising shear rates the intensity of anisotropical peak is increased which indicate the partly destruction of fat crystal microstructure. Moreover first results have shown that orientation of the platelets slightly changes with different shear rates. The scattering patterns for each sample taken at static conditions after all the shear manipulations were different to the ones taken in the beginning of the experiment which indicates irreversible shear-induced disruption of the fat crystal network.

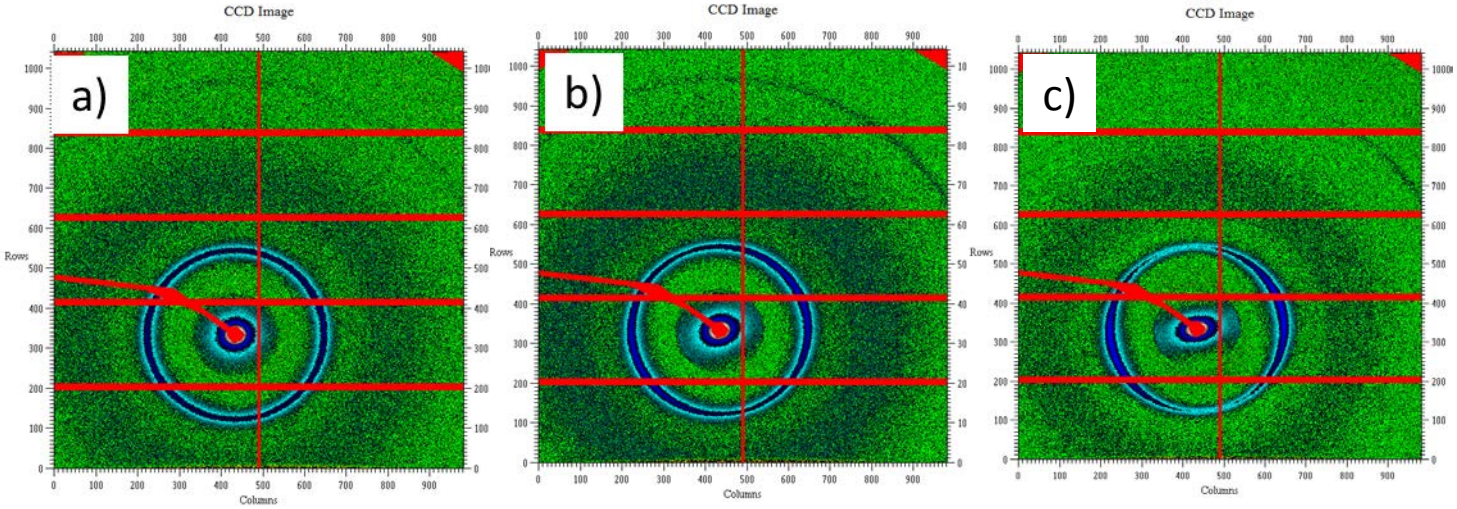

Figure 1. *Ex situ* aged dispersions under different shear rates. 2D SAXS pattern for 20% fat crystal dispersion in oil at shear rates 0, 5 and 70 s^{-1} respectively.

Figure 2 shows results of axially averaged Q-dependencies of the scattered intensity for fat crystal dispersion in oil in 1 min, 4h and 8h after starting the shear the sample at 0.1s^{-1}. The width of the diffraction peak changes with aging, indicating that fat crystal plates become thicker due to stacking and/or Ostwald ripening.

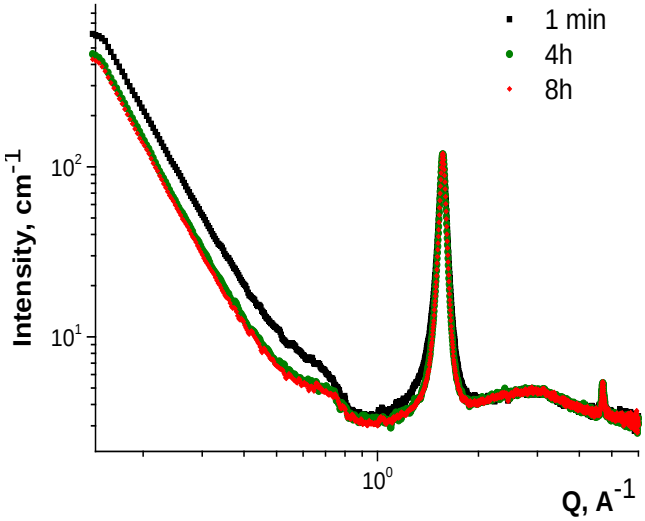

Figure 2. *In situ* ageing experiment under fixed shear rate 0.1s^{-1} on "fresh" fat dispersions. Q-dependencies of the scattered intensity for fat crystal dispersion in oil in 1min, 4h and 8h after starting the experiment.

Conclusions and Perspectives

Our preliminary analysis of the data indicates that shear irreversible disrupts the network of flocculated fat crystal platelets. In the shear field, we observe strong alignment of fat crystal platelets, this make it unlikely that shear only results in flow of isotropic tumbling flocs. The *in situ* ageing experiments results show that network formation is preceded by growth in platelet thickness. We will now proceed with quantification of our hitherto qualitative observations.

References

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