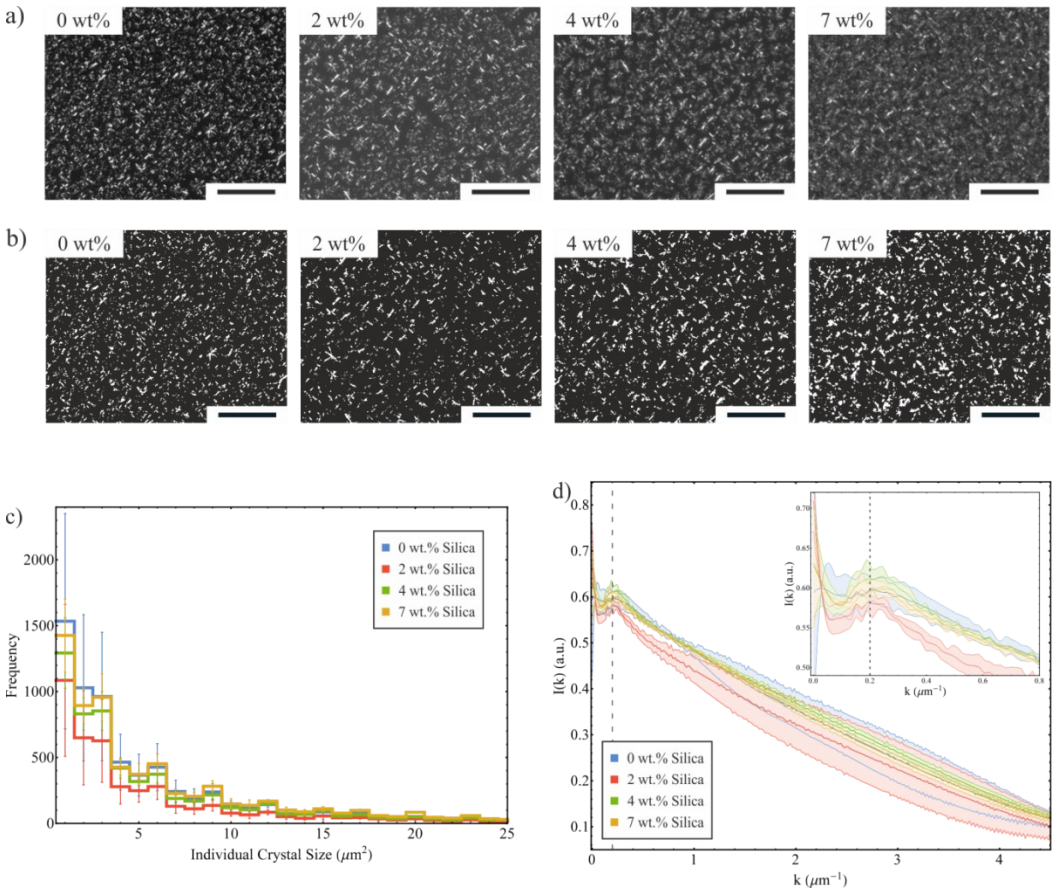
Electronic Supplementary Material (ESI) for RSC Advances.

Exploring Concentration, Surface Area and Surface Chemistry Effects of Colloidal Aggregates on Fat Crystal Networks – Supplementary Information

Polarised Light Microscopy Results

Silica Concentration Results

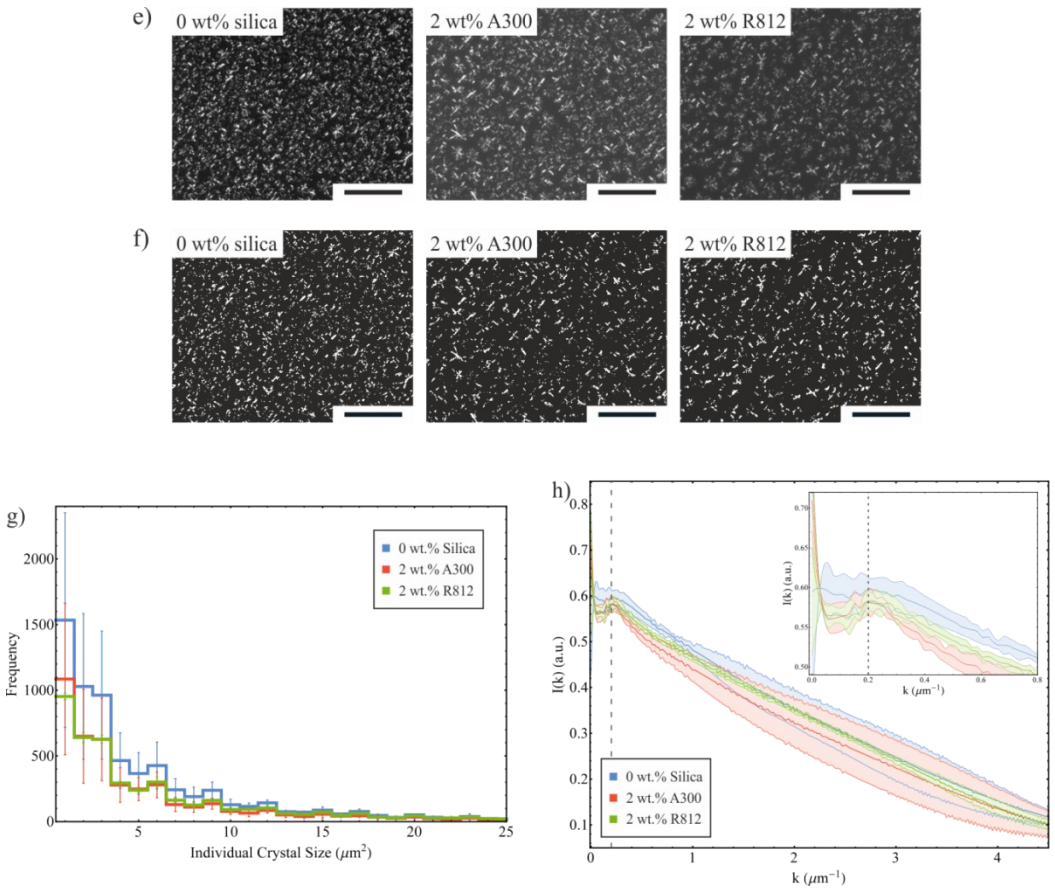


a) Greyscale images for 10 wt.% fat samples with different concentrations of A300 hydrophilic silica. The scale bars represent 100μm. b) The corresponding binarized images. c) Histograms of individual crystal areas for the different silica concentrations. d) Structure factors for the different silica concentrations.

Figures a and b show the greyscale and binarized polarised light microscopy images respectively for samples containing different concentrations of hydrophilic A300 silica and 10

wt.% fat. From this data we do not observe any trends with silica concentration. This is quantified by the histograms of individual crystal sizes for different silica concentrations which all coincide within experimental error. In the structure factors we see a distinct peak at approximately 0.2 μm^{-1}, corresponding to a typical lengthscale of 30 µm, but this peak does not significantly vary with the amount of silica present.

Silica Surface Chemistry Results



e) Greyscale images for 10 wt.% fat samples with different types of silica; no silica, 2 wt.% A300 hydrophilic silica, 2wt.% R812 hydrophobic silica. The scale bars represent 100 µm. f) The corresponding binarized images. g) Histograms of individual crystal areas for the different silica types. h) Structure factors for the different silica types.

Figures e and f shows samples containing 2 wt.% hydrophilic A300 silica and 2 wt.% hydrophobic R812 silica. We find it difficult to extract any obvious changes with the silica

surface chemistry here. We do measure a slight drop in the number of small crystals when silica is present, shown in figure g, however this result is within experimental error and so it is difficult to make any conclusions here. In the structure factors we see a similar peak at 0.2 μm^{-1} and all of the structure factors overlap within error.

These results are consistent with the composite gel network model from our previous work (Chauhan, Raamanand R., et al. "The effect of colloidal aggregates on fat crystal networks." Food & Function (2017). In our model system, we begin with a pre-melting step to destroy any fat crystal memory, which leaves only a silica network at high temperatures. This network is relatively weak and so as the fat crystallizes it is able to move and rearrange the silica aggregates. This results in a layer of silica aggregates on the surface of the fat crystal cluster chains forming a composite gel network.

Rheological data presented in our previous work showed that the G' value within the LVR for the silica network is roughly two orders of magnitude weaker that the fat network, and we proposed that the fat crystal networks grow unhindered by the silica present. Therefore, we do not expect to see a significant difference in the microscopy data.

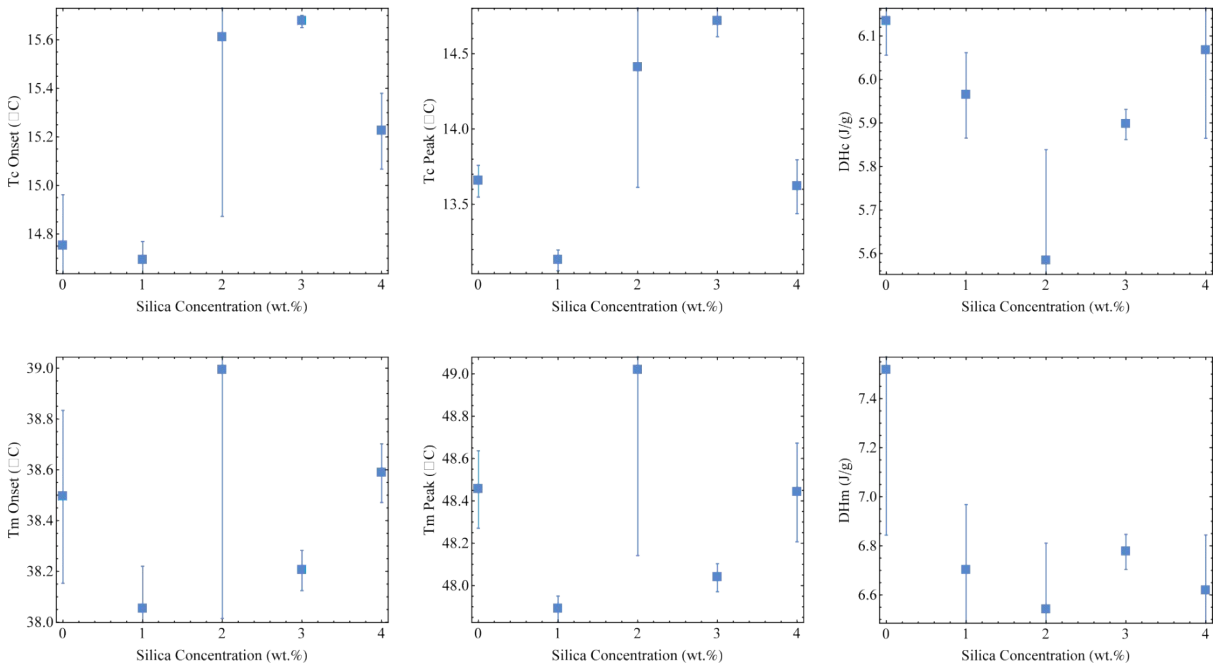
Differential Scanning Calorimetry Analysis

Here, we further analyse the DSC data and in the following plots we show the crystallization onset temperatures (Tc Onset), crystallization peak temperatures (Tc Peak), crystallization enthalpy (DHc), melting onset temperatures (Tm Onset), melting peak temperatures (Tm Peak) and melting enthalpy (DHm) for the four sets of samples discussed in the main paper: silica concentration, silica surface area and two sets of data looking at different silica surface chemistries.

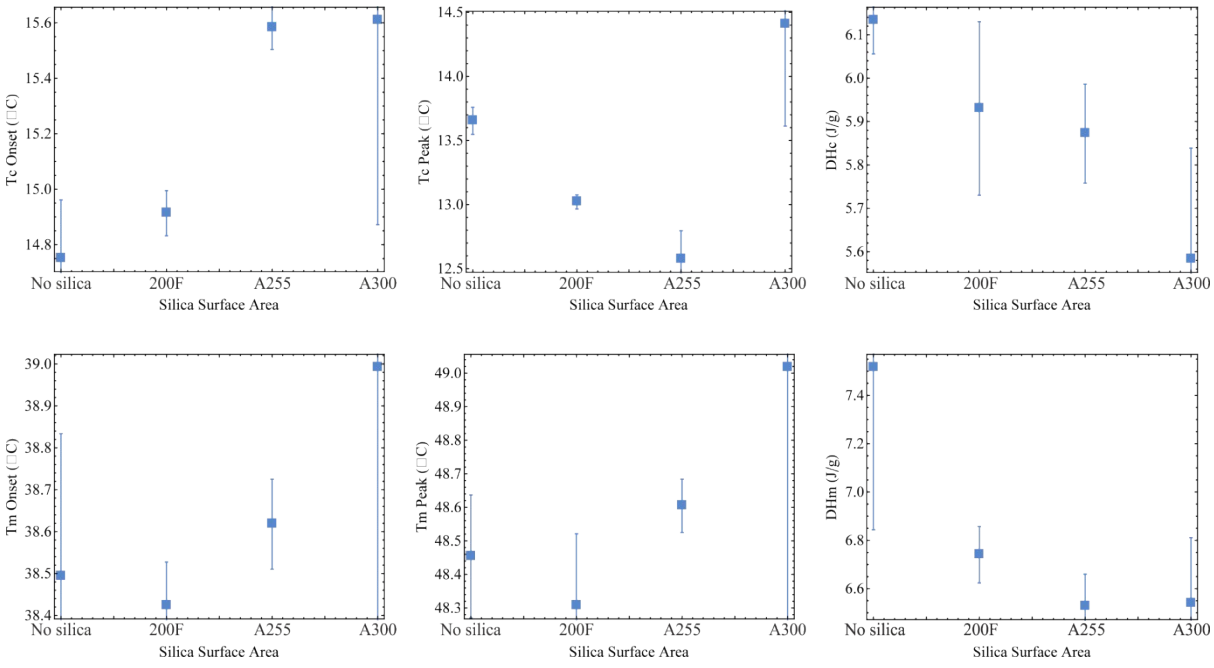
We do see some small trends in the presence of silica, such as a decrease in the crystallization enthalpy with increasing silica surface area. However, these are very small effects and are within experimental error. We also note that we see similar amounts of variation in the melting curves which are much more similar than the crystallization curves in terms of

shape and magnitude. Some of this noise is due to the analysis of the peaks since it is difficult to accurately integrate under the DSC curves when the baseline is not flat and the limits are not well defined. Therefore, we chose to focus on the key temperatures, since they are much more clearly defined.

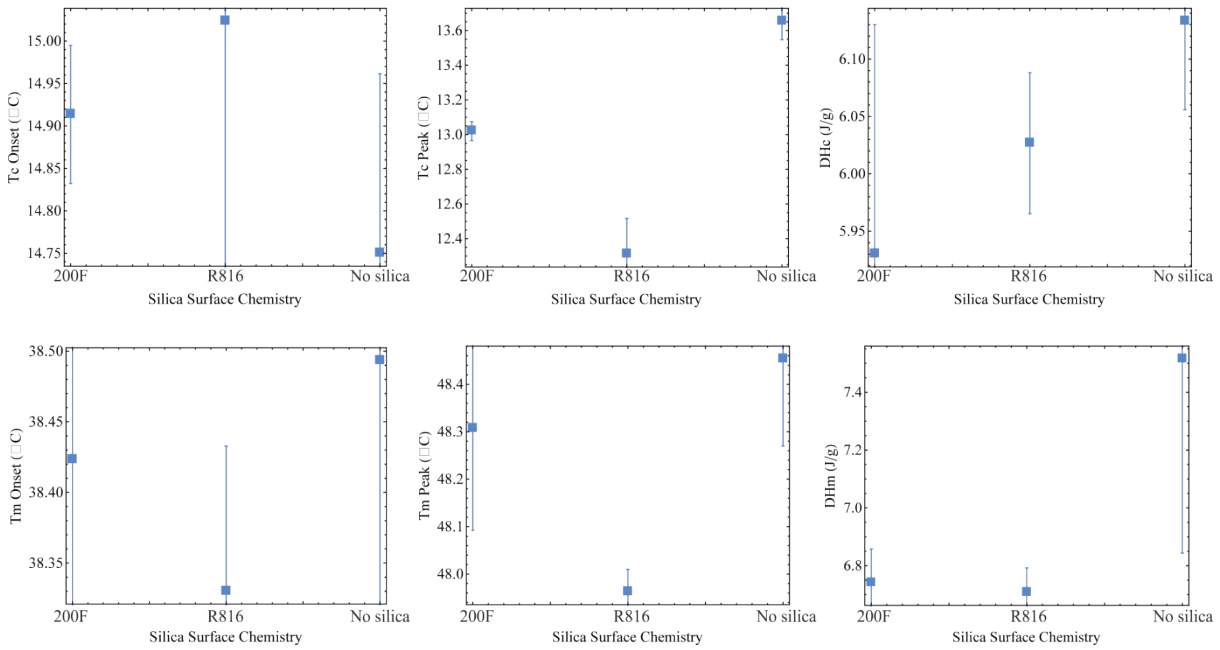
Silica Concentration Samples – 5 wt.% fat, varying amounts of A300 hydrophilic silica



Silica Surface Area – 5 wt.% fat, 2 wt.% silica



Silica Surface Chemistry Pair 1 (200F & R816) – 5 wt.% fat, 2 wt.% silica



Silica Surface Chemistry Pair 2 (A255 & R812) – 5 wt.% fat, 2 wt.% silica