Supporting Materials (SM)

Fractals in a eutectogel: *Self-assembly of SDS in a deep eutectic solvent*

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S1. X-ray reflectivity (XRR) at the air-glyceline interface

The use of a lamellar model to fit the SANS profile of the gel phase was rationalised by observations in the X-ray reflectivity (XRR) data at the air-glyceline interface (**Figure S1**). The peaks at $q \sim 0.3$ and 0.6 Å^{-1} can be clearly seen here, relating to $n_\alpha = 1, 2$.

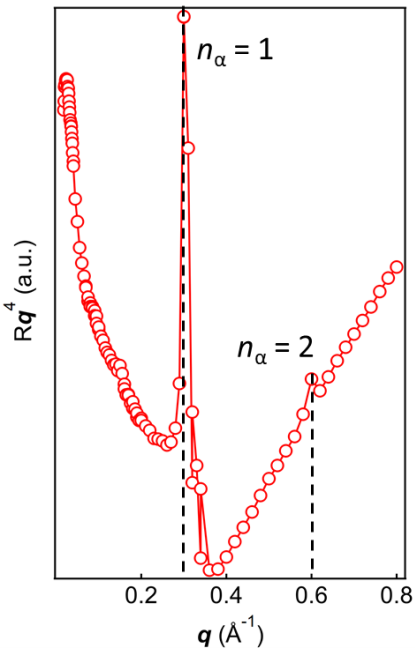

Figure S1 XRR profile of 0.4 wt % SDS in glyceline, plotted as Rq^4 to improve the clarity of the Bragg peaks. Data was taken on the I07 beamline, Diamond Light Source, Oxford, UK.

S2. The effect of surfactant concentration on aggregate formation

The size of the fractal aggregates increased as the surfactant concentration, c_{SDS}, decreased in SANS model analyses, this was also confirmed in PLM imaging (**Figure S2**). The largest aggregate sizes are in the lower concentrations, possibly due to growth in the higher c_{SDS} being hindered by the number of aggregates forming.

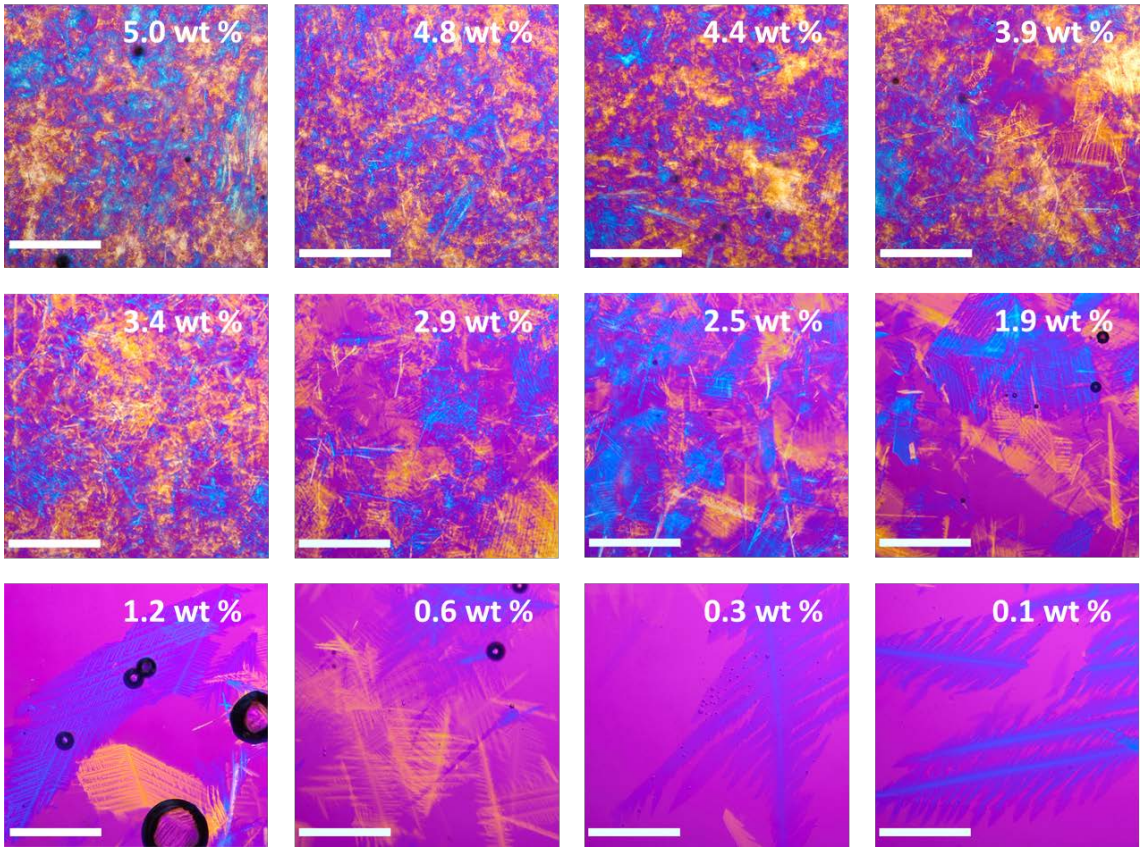

Figure S2 The effect of surfactant concentration, c_{SDS}, on aggregate formation, c_{SDS} are shown top right for each image, and each scale bar represents 200 µm. These images were taken at 10 x magnification with a 530 nm first-order waveplate.

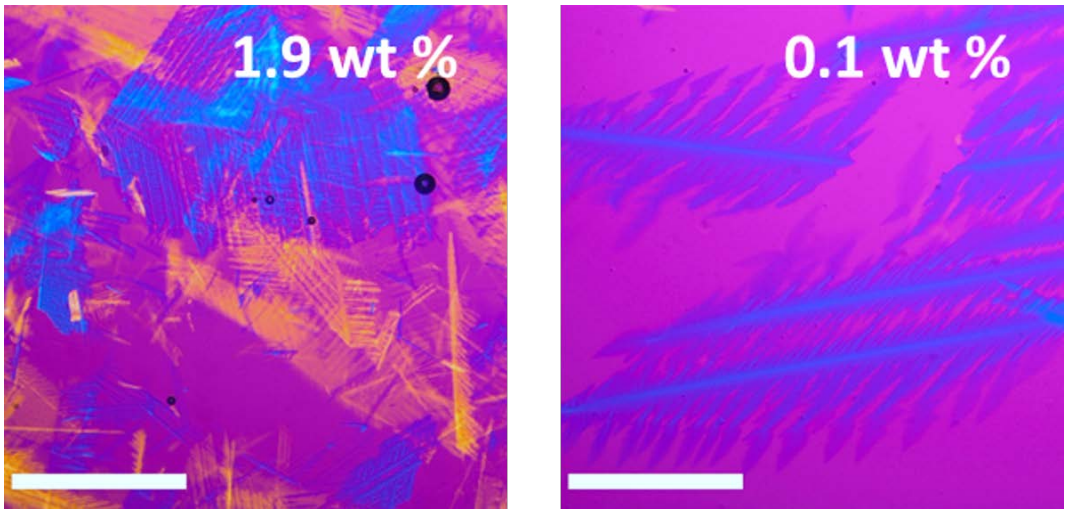

Figure S3 Comparison between higher and lower c_{SDS} fractal aggregates, c_{SDS} are shown top right for each image, and each scale bar represents 200 µm. These images were taken at 10 x magnification with a 530 nm first-order waveplate.

S3. Model refinement for SANS data fitting of the SDS-in-glyceline gel at 25 °C

The PLM imaging of the SDS-in-glyceline gel indicated the formation of fractal aggregates, so the three fractal models available on SasView were trialled to find the most appropriate model for the system (**Figure S4**), with tables summarising the fitting parameters for each (**Table S1** - **Table S5**). Of the three fractal models trialled, the mass fractal model was found to have the best fit with physically sensible values for the fit parameters. A fit to the lamellar paracrystal model and a Debye-Anderson-Brumberger (DAB) model is shown as well, as a lamellar fit was chosen to try and account for the Bragg peak. The DAB model was used to obtain a value for the correlation length, ξ.

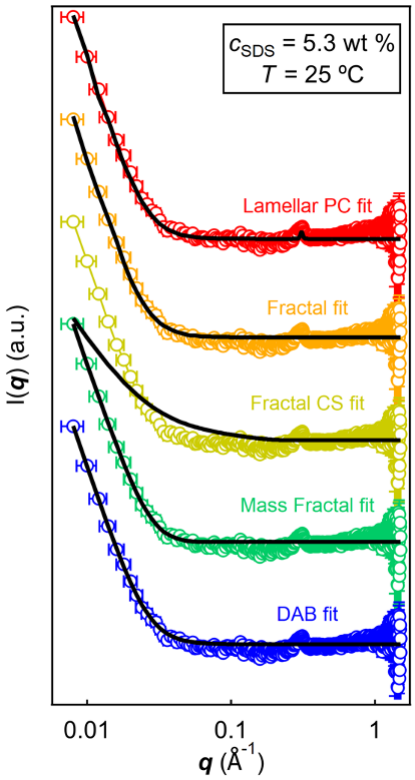

Figure S4 Fitted SANS data for 5.3 wt % SDS in glyceline at 25 °C using: a lamellar paracrystal model (red), a fractal model (orange), a fractal core-shell model (yellow), a mass fractal fit (green), and a Debye-Anderson-Brumberger (DAB) model (blue).

Table S1 Table summarising the fitting parameters for the paracrystalline lamellar stack model used to simulate the data for 5.3 wt % *h*-SDS in *d*-glyceline at 25 °C: SDS bilayer thickness t_L, number of layers in the stack n_{Layers}, *d*-spacing, polydispersity of the *d*-spacing σ_d, scattering length density of SDS ρ_{SDS}, scattering length density of glyceline ρ_{Gly}, polydispersity of the SDS bilayer thickness σ_t, and chi squared value χ^2.

Lamellar Stack Paracrystal Model	5.3 wt %
t_L (Å)	20.0
n_{Layers}	57.7
d-Spacing (Å)	20.4
σ_d (Å)	0.013
ρ_{SDS} (10^{-6} Å^{-2})	0.40
ρ_{Gly} (10^{-6} Å^{-2})	5.50
σ_t	1.0
χ^2	3.8

Table S2 Table summarising the fitting parameters for the fractal model used to simulate the data for 5.3 wt % *h*-SDS in *d*-glyceline at 25 °C: volume fraction φ, radius *r*, fractal dimension D_m, correlation length ξ, scattering length density of SDS ρ_{SDS}, scattering length density of glyceline ρ_{Gly}, polydispersity of the radius σ_r, and chi squared value χ^2.

Fractal Model	5.3 wt %
φ	0.30
r (Å)	4936
D_m	0.19
ξ (Å)	17.2
ρ_{SDS} (10^{-6} Å^{-2})	0.354
ρ_{Gly} (10^{-6} Å^{-2})	5.747
σ_r	0.51
χ^2	3.47

Table S3 Table summarising the fitting parameters for the fractal core-shell model used to simulate the data for 5.3 wt % *h*-SDS in *d*-glyceline at 25 °C: volume fraction φ, radius r, thickness t, fractal dimension D_m, correlation length ξ, scattering length density of the core ρ_{core}, scattering length density of the shell ρ_{shell}, scattering length density of glyceline ρ_{Gly}, polydispersity of the radius σ_r, and chi squared value χ^2.

Fractal Core-Shell Model	5.3 wt %
φ	0.021
r (Å)	15.0
t (Å)	5.0
D_m	2.02
ξ (Å)	6487
ρ_{core} (10^{-6} Å^{-2})	-0.304
ρ_{shell} (10^{-6} Å^{-2})	5.200
ρ_{Gly} (10^{-6} Å^{-2})	5.743
σ_r	0.10
σ_t	0.71
χ^2	14.9

Table S4 Table summarising the fitting parameters for the mass fractal model used to simulate the data for 5.3 wt % *h*-SDS in *d*-glyceline at 25 °C: radius of the fractal aggregate r, fractal dimension D_m, and chi squared value χ^2.

Mass Fractal Model	5.3 wt %
r (Å)	56.5
D_m	2.95
χ^2	3.4

Table S5 Table summarising the fitting parameters for the DAB model used to simulate the data for 5.3 wt % *h*-SDS in *d*-glyceline at 25 °C: correlation length ξ, and chi squared value χ^2.

DAB Model	5.3 wt %
ξ (Å)	484.9
χ^2	3.4

S4. Purity determination of sodium dodecyl sulfate

Figure S5 shows the ^{1}H NMR spectra throughout the progression of the recrystallisation procedure; to demonstrate the purity of SDS at the end of the steps, the final ^{1}H NMR spectrum was analysed.

^{1}H NMR (400 MHz, D_2O) δ (in ppm) 4.64 (D_2O, s), 3.89 (2H, t, **a**), 1.54 (2H, q, **b**), 1.17 (18H, m, **c**), 0.74 (3H, t, **d**)

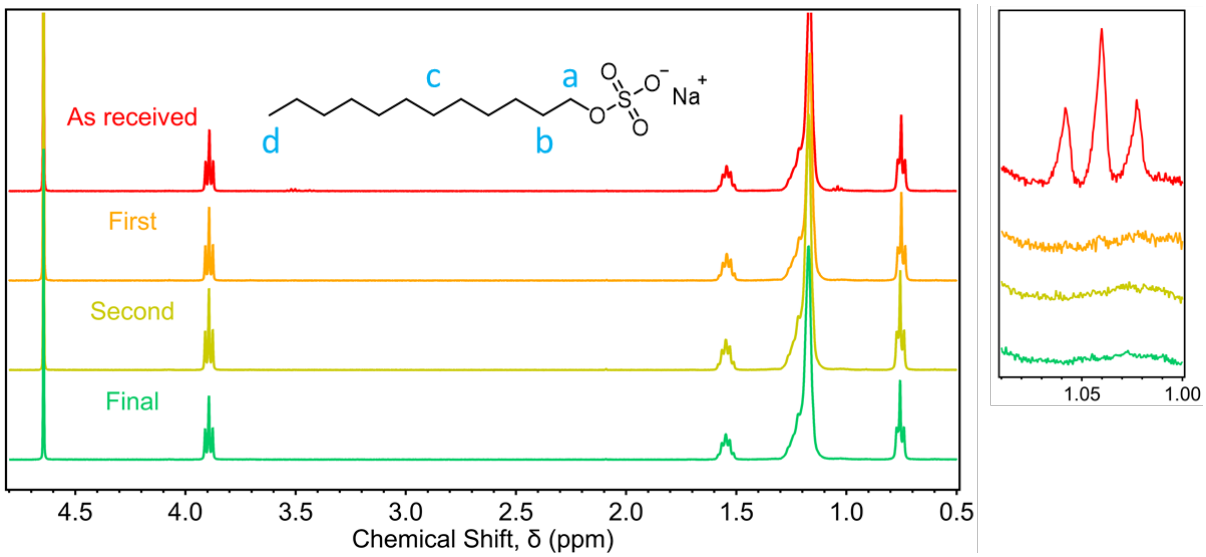

Figure S5 A series of ^{1}H NMR spectra from each step of the recrystallisation of SDS: before recrystallisation (**red**), after one recrystallisation step (**orange**), after two recrystallisation steps (**yellow**), and three recrystallisation steps (**green**). The molecular architecture of SDS is given in the **inset**, with labelled protons according to the analysis, and a zoom of the chemical shift, δ = 1.10 – 1.00 ppm is given (**right**) to show the disappearance of impurity peaks. The D_2O solvent peak is shown at 4.64 ppm.