

Supplementary Information

1. Nanoparticle size distribution in the 8 h sample determined by TEM

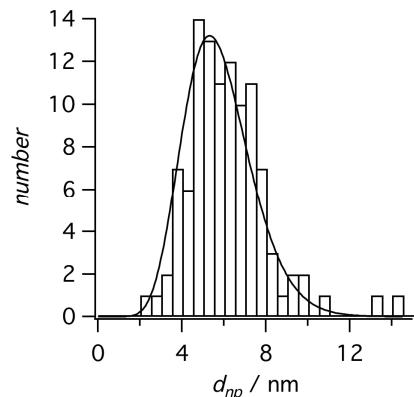


Figure SI-1. Histogram of nanoparticle diameters in 8 h sample. Solid line: Schultz distribution fit to the data.

On the basis of the TEM image in Fig. 2b of the manuscript, a histogram of the nanoparticle diameters was determined. The nanoparticles were clearly size-polydisperse. A fit of a Schultz distribution to the histogram yielded an average diameter of 6.0 ± 1.9 nm.

2. Comparison of SAXS models for nanoparticles

The contribution of nanoparticles to the SAXS patterns were fitted using five different form factor models, *i.e.* monodisperse uniform spheres, uniform oblate ellipsoids and core-shell oblate ellipsoids, and polydisperse Schultz distributed uniform spheres and core-shell spheres (Figure SI-1). The models were evaluated for the unheated (0 h) and 8 h heated sols. For the 0 h sol, an electrostatic structure factor contribution was included. For the 8h sol, the contribution of ellipsoidal crystals was first fitted and subtracted. The resulting fitting parameters and reduced Chi square values are summarized in Tables SI-1 (0 h sample) and SI-2 (8 h).

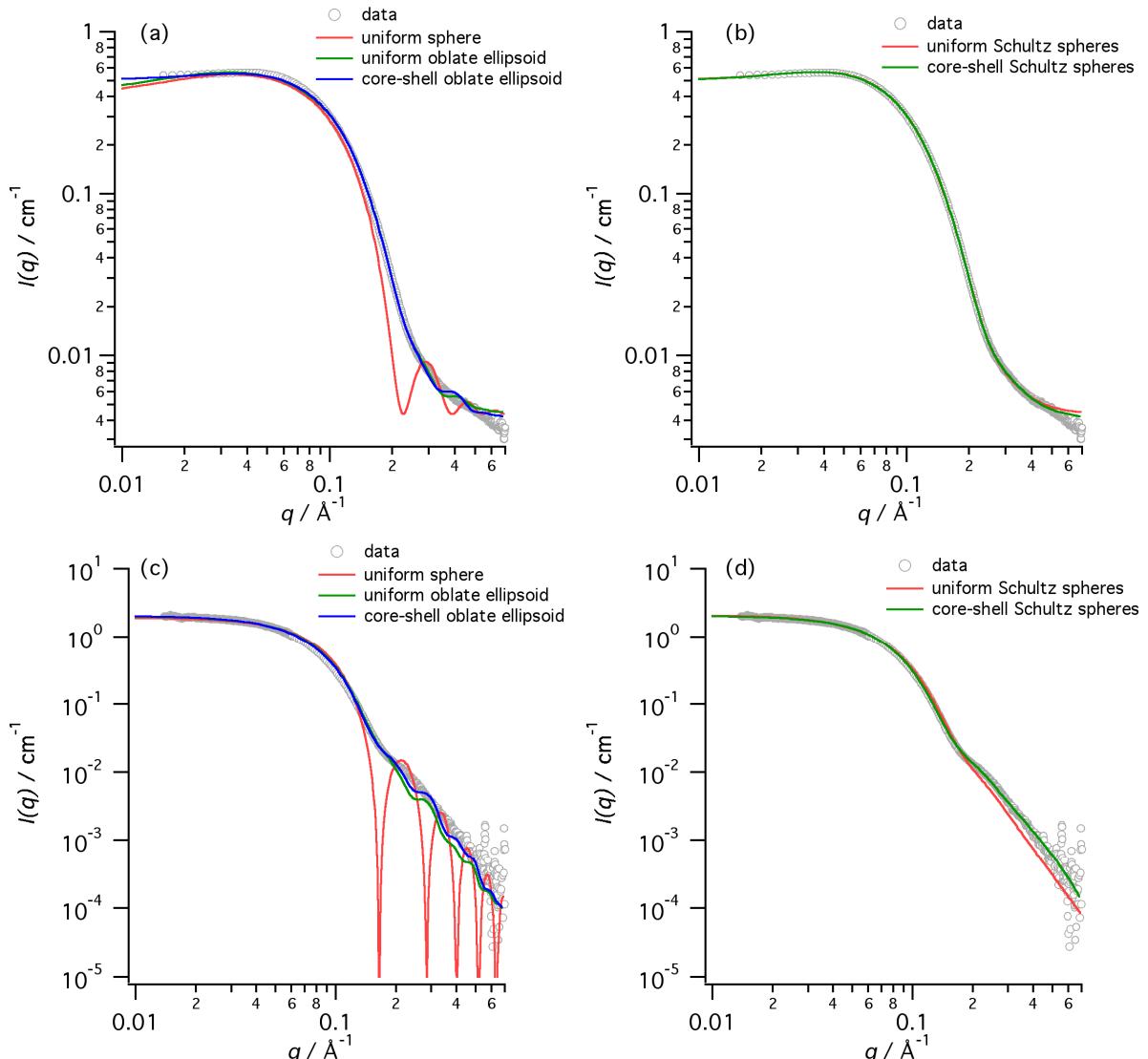


Figure SI-2. Comparison of different models for the nanoparticle population. (a, b) Unheated (0 h) sample. (c,d) Sample heated 8 h.

Table SI-1. Fit parameters obtained from form factor model comparison of 0 h sol. The structure factor was the Hayter-Penfold model in all cases

| model | $\sqrt{\frac{\chi^2}{N}}$ (a) | $\phi_{core+shell}$ | r, r_a (Å) | r_b (Å) | pdi | t, t_a (Å) | t_b (Å) | charge (e ⁻) | [monovalent salt] (mM) |
|--------------------------------|----------------------------------|---------------------|-----------------|--------------|------|-----------------|--------------|-----------------------------|------------------------|
| sphere | 30.46 | 0.0056 | 19.9 | - | - | - | - | 3.3 | 2.1 |
| oblate ellipsoid | 4.45 | 0.0075 | 12.0 | 22.6 | - | - | - | 2.4 | 1.6 |
| core-shell oblate ellipsoid | 2.17 | 0.0153 | 14.1 | 25.4 | - | 22.3 | 28.0 | 4.7 | 11.4 |
| schultz spheres | 1.12 | 0.0075 | 14.8 | - | 0.27 | - | - | 4.0 | 9.9 |
| schultz core- shell spheres | 0.76 | 0.0128 | 15.7 | - | 0.25 | 2.6 | - | 3.7 | 8.7 |

(a) Reduced χ^2 value, where N is the number of fitted data points.

Table SI-2. Model comparison of nanoparticle contribution of 8 h sol. Crystals were oblate ellipsoids (volume fraction = 1.58×10^{-4} , $r_a = 49$ nm, $r_b = 94$ nm). Different form factors are listed. The structure factor was set equal to 1

| model | $\sqrt{\frac{\chi^2}{N}}$ | $\phi_{core+shell}$ | r, r_a (Å) | r_b (Å) | pdi | t, t_a (Å) | t_b (Å) |
|-----------------------------|---------------------------|---------------------|-----------------|--------------|-------|-----------------|--------------|
| sphere | 17.00 | 0.0069 | 27.1 | - | - | - | - |
| oblate ellipsoid | 7.86 | 0.0072 | 18.3 | 33.2 | - | - | - |
| core-shell oblate ellipsoid | 6.89 | 0.0108 | 33.5 | 19.0 | - | 38.0 | 20.5 |
| schultz spheres | 6.84 | 0.0072 | 22.0 | - | 0.26 | - | - |
| schultz core-shell spheres | 2.13 | 0.0142 | 25.2 | - | 0.23 | 6.1 | - |

3. Bragg region (cf. Figure 1)

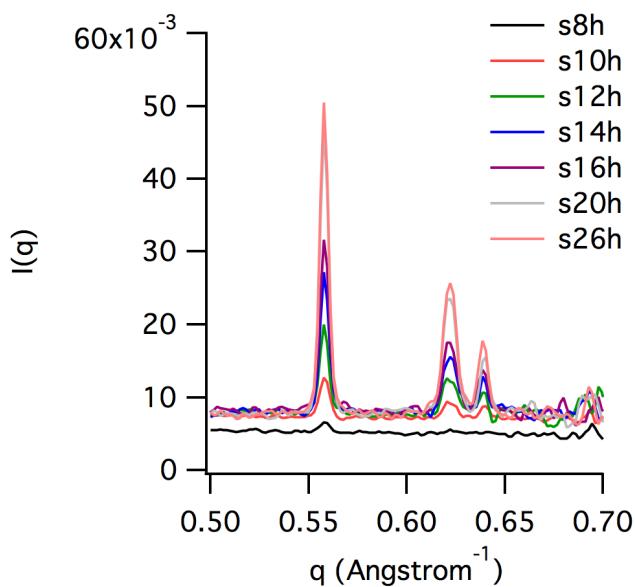


Figure SI-3. Region of the MFI-type Bragg reflections in the SAXS patterns, for samples heated 8 – 26 h.