#### **Supplementary Information**

*In situ* photogelation kinetics of Laponite nanoparticle-based photorheological dispersions

K. Anne Juggernauth<sup>1, 2, \*</sup>, Amy E. Gros<sup>2</sup>, Norman A.K. Meznarich<sup>2</sup>, and Brian J. Love <sup>2,3,4\*\*</sup>

## Dynamic light scattering (DLS)

## **Experimental Procedure**

Dynamic Light Scattering (DLS) experiments were conducted on an ALV compact goniometer system (Model ALV/SP-125, Langen, Germany) using an ALV/SO-SIPD photon detector. Dilute solutions of F127 and Laponite were analyzed to investigate the interactions between the nanoparticles and F127 molecules within the dispersion. An Innova 70C argon ion laser (Coherent Inc., Santa Clara, CA) with a wavelength of 488 nm and a power of 200 mW was used as the light source. Data was collected for one minute per reading after equilibration at 30 °C at a fixed angle of 90°. The ALV fitting software uses a constrained regularized method (CONTIN)<sup>20</sup> to obtain particle size distributions from the autocorrelation functions.

#### <u>Results</u>

Interaction of Laponite with F127 via dynamic light scattering (DLS)

Samples containing 0.72 wt% F127 with varying Laponite concentrations ranging from 0.2 - 0.6 wt% were used to investigate interactions between Laponite nanoparticles and surfactant molecules. A 1.0 wt% Laponite solution without F127 was also prepared to determine the nanoparticle size distribution. The low concentration solutions were used to avoid effects arising from interactions between particles in higher concentration dispersions as well as to minimize absorption of light during the DLS experiments. Fig 2 shows the distribution of the hydrodynamic radius, R<sub>H</sub>, for increasing concentrations of Laponite (0.2 - 0.6 wt%) in the presence of 0.72 wt% F127 as well as 1 wt% Laponite with no F127.



**Fig S1.** Hydrodynamic radii as measured by dynamic light scattering (DLS) at 90° for samples containing 1 wt% Laponite only as well as 0.72 wt% F127 with 0.2 wt%, 0.4 wt% and 0.6 wt% Laponite

A particle size of  $\sim 20$  nm was measured for suspensions containing only 1.0 % Laponite which is consistent with the known size of the disk – like nanoparticles. These values agree closely with prior work by Nelson and Cosgrove <sup>15</sup> as well as Nicolai and Cocard <sup>21</sup> who both reported bare Laponite particle radius on the order of 20 nm as well as adsorbed layer thicknesses as a function of surfactant molecular weight. In the presence of F127, the hydrodynamic radius increases from 20 nm up to 40 nm indicating surface adsorption of F127 onto Laponite disks. The diameter increase confirms the role of the F127 in stabilizing the nanoparticles in the dispersion via surface adsorption. It should also be noted that in all cases, the DLS profiles show a large size distribution. We attribute this breadth to flocculation and possible particle aggregation.

# **Rheological studies**

Figure S2 shows the evolution of the storage and loss modulii of the sample as a function of time for different Laponite concentrations (top row) and different Pluronic concentrations (bottom row). The trends in both G' and G" shown here match closely with the observed trends seen in the complex viscosity evolution shown in Figure 3 in the text and confirm the trends discussed in detailed surrounding the change in the complex viscosity upon UV illumination.



**Fig S2.** Storage modulus, G'((a) and (c)) and loss modulus, G" ((b) and (d)) evolution during *in situ* photogelation experiments for varying Laponite concentrations with 3.6 wt% F127 and 13mM PAG (top row) and varying F127 concentrations with 3 wt% Laponite and 13mM PAG (bottom row)