SUPPORTING INFORMATION

Synthesis and stability of water-in-oil emulsion using partially reduced graphene oxide as a tailored surfactant

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S1. Solubility of partially-reduced Graphene Oxide (prGO)

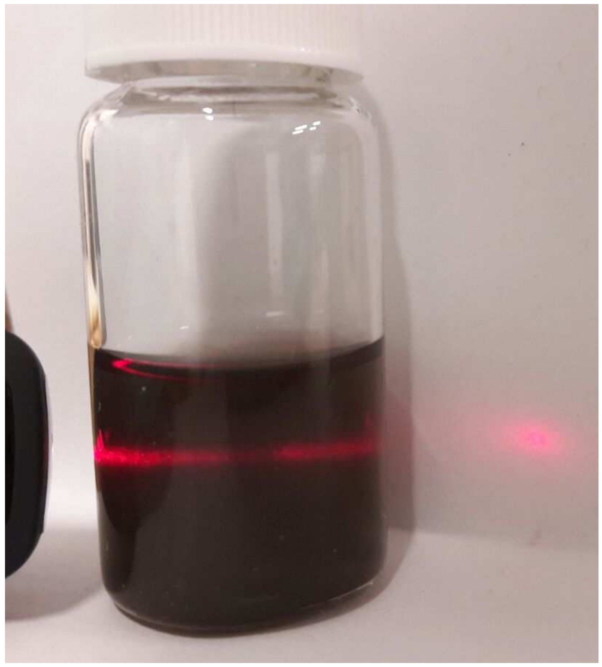

Figure S1 Tyndall effect in prGO-water dispersion

S2. Mean and standard deviation calculations from droplet size distribution curve

Effect of parameters such as prGO concentration and the oil volume fraction on the W/O emulsion

Log-normal, $y = y_0 + \frac{A}{\sqrt{2\pi}\sigma x} e^{\left(\frac{-\left(\ln\left(\frac{x}{x_c}\right)\right)^2}{2\sigma^2}\right)}$ .... (1)

Gaussian, $y = y_0 + \frac{A}{\left(\sigma\sqrt{\left(\frac{\pi}{4\ln(2)}\right)}\right)} e^{\left(\frac{-4\ln(2)(x-x_c)^2}{\sigma^2}\right)}$ ………………………….. (2)

Table S3 Standard deviation 'σ' and mean 'x_c' of the log-normal fit at varying prGO concentration

prGO Concentration (w/w)	σ	x_c
0.005	0.53	9.4
0.01	0.55	9.8
0.05	0.6	11
0.1	0.8	16

Table S4 Standard deviation 'σ' and mean 'x_c' of the log-normal and Gaussian fit at varying oil volume

Oil volume (%)	σ	x_c
40	0.53	9.2
30	0.19	9.4
20	3.0	10.7
15	7.7	11.7

S3. Destabilization analysis using time-dependent confocal microscopy and emulsion photographs

De-stabilization studies and coalescence dynamics of the W/O emulsion

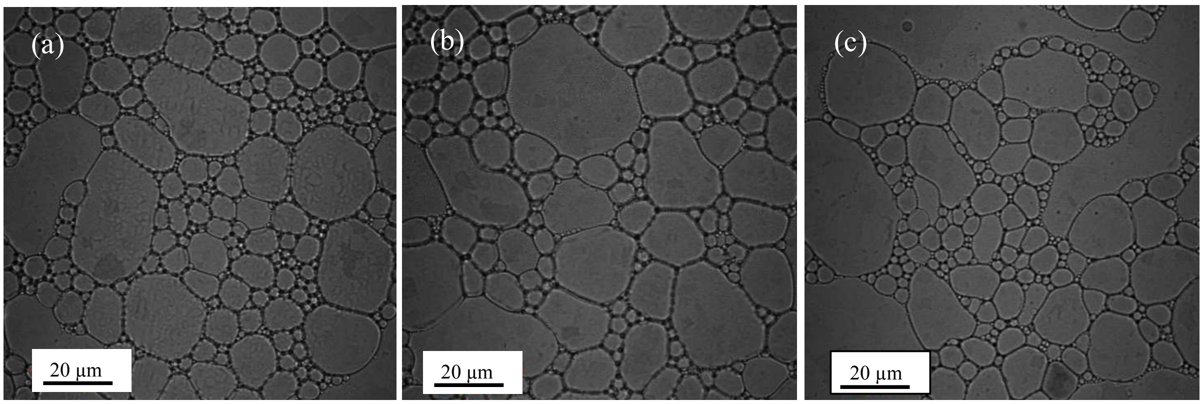

Figure S2 Confocal images of W/O stabilized by prGO taken on (a) 6th day, (b) 12th day and (c) 18th day after formulation.

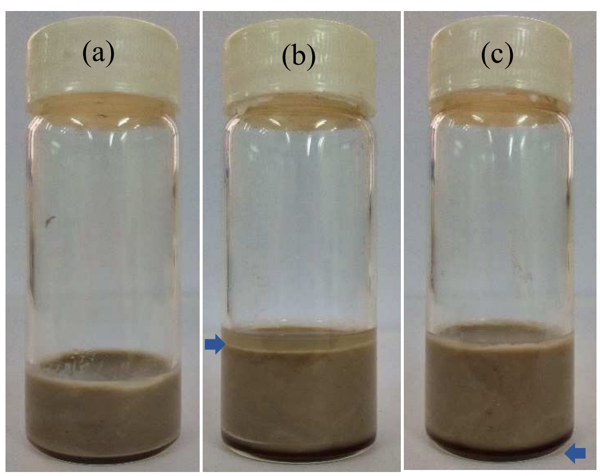

Figure S3 Photographs of W/O stabilized by prGO taken on (a) 1st day, (b) 5th day and (c) 10th day. The emulsion sediments initially separating from the oil phase. After removing oil, the droplets merge and break to form water

To understand the destabilization process and its underlying mechanism, the W/O emulsion was carefully observed during the early stages of destabilization. Figure S2 shows the photographs of the destabilizing emulsion taken on (a) 1st day, (b) 5th day and (c) 10th day. The W/O emulsion contains 75:25 water-to-oil ratio and stabilized by using 0.05% (w/w) prGO. It was observed that 5 days after emulsion formulation, the oil starts separating to form a surface layer. To understand this, it is important to know that the density of water is higher than the density of canola oil. Also, the water droplets are encapsulated by prGO sheets as shown in the microstructure before. As a result, the water droplets with prGO settle downwards in the vial. This could be the reason that oil starts surfacing as water starts settling. This kind of behavior has been observed in emulsions that undergo sedimentation destabilization. Hence, it can be said that the destabilization of W/O emulsions prepared using prGO starts with sedimentation. The sedimentation results in the removal of interstitial oil between the water droplets, which further gives rise to collapse of water droplets by any of mechanism discussed before and consequently droplet coalescence. The droplet coalescence results in formation of a separate water phase and has been indicated by the blue arrow in Figure 9(c). The destabilization mechanism after sedimentation needs to be further identified, and this is explored below.

S4. Variation of Sauter mean diameter with time

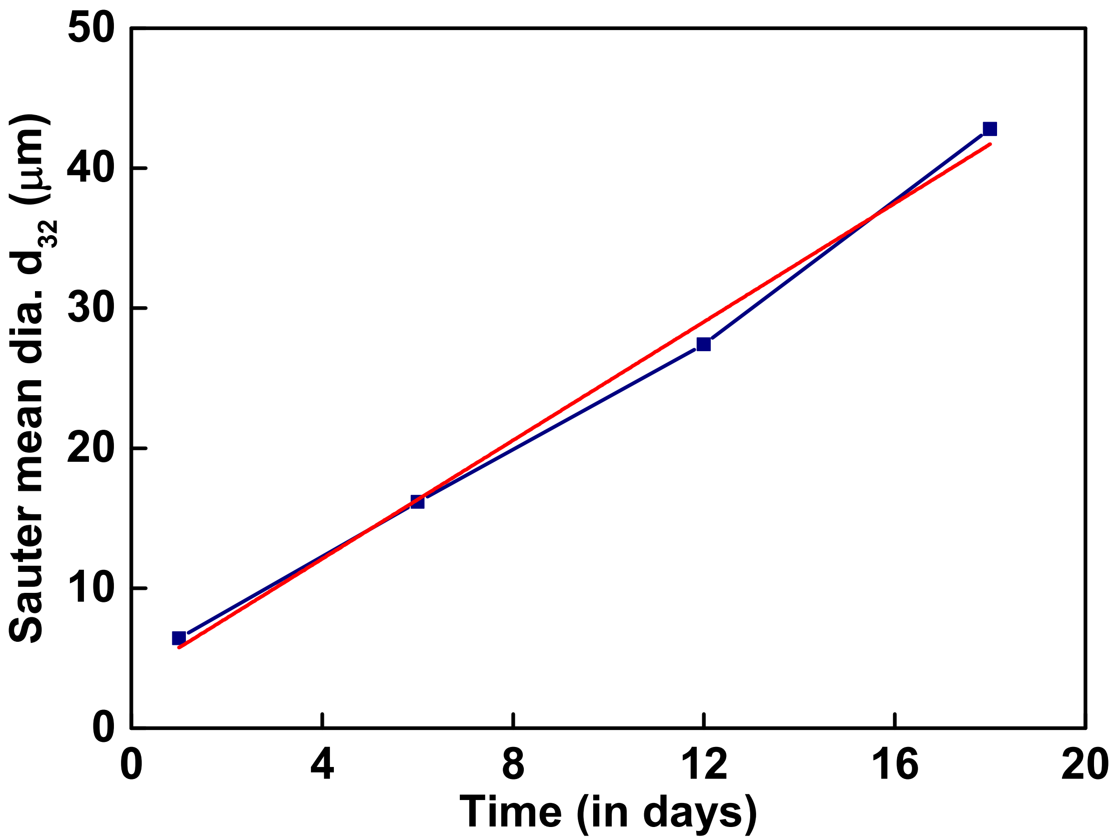

Figure S4. Evolution of Sauter mean diameter with time. The curve is linearly fitted. Due to coalescence, the mean diameter of the prGO stabilized emulsion increases, and the phases separate by the 20th day after preparation

The Sauter mean diameter (mean volume-surface diameter), d_{32} is inversely proportional to the specific surface area of droplets. For each refinement time, the Sauter mean diameter (SMD), d_{32} values are obtained as follows:

$$d_{32} = \frac{\Sigma nidi^3}{\Sigma nidi^2} \quad \ldots\ldots (3)$$

where, n is the number of droplets with diameter d.

Figure S3 shows the evolution of d_{32} with the variation in the average droplet size calculated from the curve fitting to the droplet size distribution. It is observed that the mean diameters increase with time. This rapid increase in d_{32} can be correlated with the argument of coalescence destabilization in the prGO stabilized W/O emulsion as indicated by Figure 9.