DNA Driven Colloidal Aggregation at a Liquid-Liquid Interface

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Liquid-liquid interfaces are in principle ideal substrates for the controlled assembly of colloidal patterns, if colloids could be deposited on them in a reversible manner. Techniques such as in Pickering emulsions or Langmuir trough allow placing colloids at a liquid-liquid or liquid-air interface, however, the adsorption is effectively irreversible. Here we present a new way to decorate liquid-liquid interfaces with colloids, exploiting the selective binding of complementary DNA strands grafted to the colloid and the liquid interface respectively[1]. For this we used surfactant-stabilized oil-droplets functionalised with single-stranded (ss)DNA, mixed with small polystyrene colloids grafted with complementary ssDNA. We made two observations: Firstly the degree of colloidal adsorption can be tuned reversibly. Secondly, the surface-bound colloids are fully mobile, thus allowing the formation of well-equilibrated target structures. Interestingly, we were able to tune the aggregation behaviour of the surface-bound colloids. At low bulk surfactant concentrations, the interface-bound colloids form a 2D fluid, but as the surfactant concentration approaches the critical micelle concentration, the surface-bound colloids organise into finite-sized crystalline domains due to micelle-induced depletion forces. Our experimental findings are supported by simulations studies. Our approach may allow the creation of complex, multi-component patterns of surface-bound colloids.

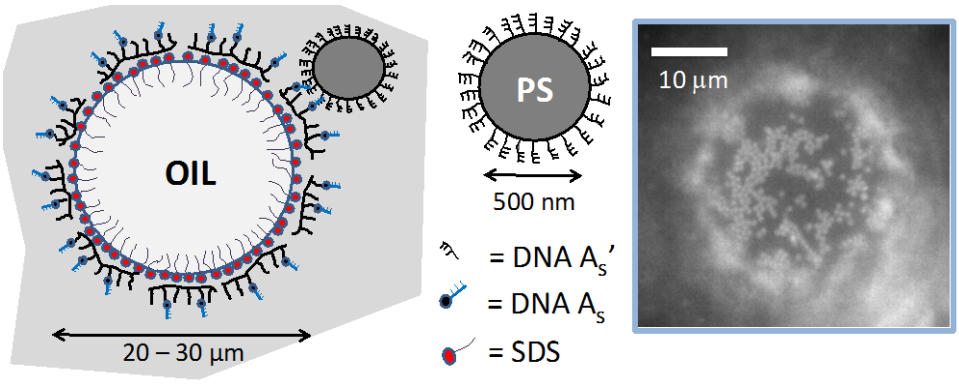

Figure: (*left*) Schematic representation of the DNA-functionalized oil droplets (ODs) to which polystyrene (PS) particles coated with the complementary DNA can bind from solution. The ODs are stabilized with sodium dodecyl sulphate (SDS) and the DNA is attached to the ODs via a positively charged comb-like polymer. (*right*) Fluorescent microscopy image showing fluorescently labled PS particles attached to the ODs. Here we focused on the 'south pole' of the OD.

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[1] Joshi et al. 'Kinetic control of the coverage of oil droplets by DNA-functionalised colloids', *Science Advances,* **2**, e1600881 (2016).