Title: Supracolloidal architectures self assembled in microdroplets

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Abstract: We demonstrate a novel method for the formation of a library of structured colloidal assemblies by exploiting the supramolecular heteroternary host-guest interaction between cucurbit[8]uril (CB[8]) and methyl viologen- and naphthalene-functionalised particles. The approach is dependent upon compartmentalization in microdroplets generated by a microfluidic platform. While the distribution of colloidal particles encapsulated within each microdroplet followed a Poisson distribution, tuning the concentration of the initial colloidal particle suspensions provided some level of control over the structure of the formed colloidal assemblies. This ability to direct the assembly of complementarily-functionalised colloids through a supramolecular interaction, without the need for complex modification of the colloidal surface or external stimuli, presents an exciting new approach towards the design of structured colloidal materials with the potential to produce many challenging structures.

Summary of available data

Original data is provided in support of the article "Supracolloidal architectures self assembled in microdroplets". The data is structured into six folders, each correlating to a figure where processed data was presented, in either the published article (Figures 2-4) or the electronic supporting information (Figure S5-6). The folders names are cross-linked to the published figures.

Folder 2: Figure 2

The microscopic bright-field micrographs showing the colloidal particle assemblies captured at **F-C**$_V$ and **R-C**$_{Np}$ particle suspension ratios of 0.05 : 0.05 mg/mL and 0.80 : 0.05 mg/mL. The spreadsheet (two types of particle.xlsx) showing statistical distribution of colloidal particles upon the formation of microdroplets at an oil flow rate of 600 μL/h and aqueous flow rate of 200 μL/h with **F-C**$_V$ and **R-C**$_{Np}$ particle suspension ratios of 0.05 : 0.05 mg/mL, 0.10 : 0.05 mg/mL, 0.20:0.05 mg/mL, 0.40 : 0.05

mg/mL and 0.80:0.05 mg/mL, respectively. The distribution that plotted in graph can be viewed in the file 2.jpg.

Folder 3: Figure 3

Microscopic bright-field, fluorescent and SEM micrographs of colloidal assemblies (n=1-6).

Folder 4: Figure 4

Sequential bright-field micrographs of a Janus colloidal particle assembly encapsulated inside a microdroplet in the presence of CB[8], in the absence of cucurbit[n]uril and in presence of CB[7].

Folder 5: Figure S5

Spreadsheet and plot demonstrate the encapsulation of one type of colloidal particle (**F-C**$_V$) in microdroplet, with particle suspension of 5 mg/mL, 0.5 mg/mL and 0.05 mg/mL respectively.

Folder 6: Figure S6

Spreadsheet and plot showing morphological and size variation for microdroplets that accommodate four and five colloidal particles when the colloidal suspension concentration was increased from 0.05: 0.15 mg/mL to 0.15: 0.15 mg/mL.