Stabilization of water-in-water emulsions by macromolecular complexation at the interface

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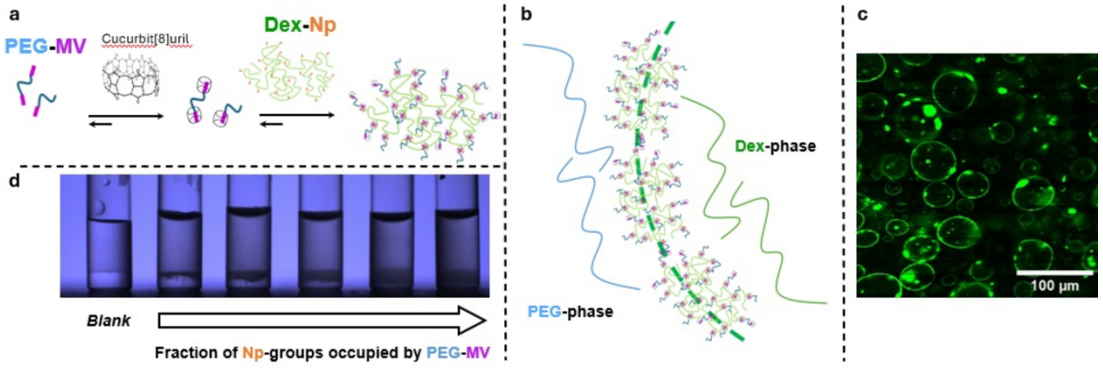
Aqueous biphasic dispersed systems, referred as water-in-water emulsions, constitute a distinct type of colloidal systems in which two immiscible aqueous phases coexist. These systems are formed thanks to the incompatibility between two polymers dissolved in water, such as dextran (Dex) and poly(ethylene glycol) (PEG), arising from their low entropy of mixing. The capacity to encapsulate active ingredients and regulate substance release, while being composed of fully biocompatible components, holds great potential for application of such systems in food science, encapsulation and drug delivery [1]. Stabilizing these unconventional emulsions constitutes a great challenge because their interfaces are ill-defined in terms of hydrophilic/hydrophobic domains and are thicker than in common oil/water emulsions [2]. Consequently, classical surfactants are generally ineffective, and stabilization mainly relies on solid particles, although the underlying mechanisms remain unclear [3].

In this context, the project deals with the development of a novel strategy to stabilize such water/water (W/W) interfaces by using macromolecular stabilizers, that possess an affinity for both the PEG- and Dex-rich phases, and therefore are located at the droplet surface similar to ordinary surfactants at water/oil interfaces. This strategy is based on the formation of complexes between PEG and Dex chains bearing methyl viologen (MV) and naphtol (Np) guest moieties, correspondingly, *via* host-guest inclusions in the presence of a host, *i.e.* cucurbit[8]uril (CB[8]) [4] (Figures a-b). The proposed method allows to tune the structure of such amphiphile-like stabilizers, and therefore, their affinity to the W/W interface (Figure d). By combining confocal microscopy (Figure c), small-angle neutron scattering, and isothermal titration calorimetry, we access the structure of the formed complexes in the bulk and at W/W interface, and demonstrate their efficiency and robustness in stabilization of water-in-water emulsions.

Keywords:
Water-in-water emulsions, bis-hydrophilic stabilizer, interfacial host-guests complexation



Figures a) Formation of the host-guest macromolecular complex b) sketch of the interfacial complexation occurring in W/W emulsions; c) confocal microscopy image of such W/W emulsions highlighting the positioning of the complex at the interface; d) W/W emulsions at t = 2 h as a function of the ratio

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