Understanding interfacial stabilization in one-step W/O/W Pickering double emulsions using cellulose nanofibrils and oleic acid

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Double emulsions are complex systems consisting of emulsion droplets that contain smaller droplets inside, resulting in a three-phase structure with two interfaces. Compared with simple emulsions, double emulsions offer the advantage of encapsulating and controlling the release of both hydrophilic and hydrophobic active compounds. However, due to their high instability, double emulsions are typically prepared using large amounts of synthetic stabilizers and two-step methods.[1] To design more sustainable emulsions, cellulose nanofibrils (CNFs) are attractive candidates due to their renewability and biodegradability, although they typically form only simple O/W Pickering emulsions.[2] In this work, CNFs were combined with oleic acid to enable the one-step production of water-in-oil-in-water ($W_1/O/W_2$) double emulsions, using almond oil as the oil phase. Colloidal stability was monitored over time using optical and confocal microscopy, which showed that cationic and anionic CNFs effectively stabilized the external O/W_2 interface. Experimental results combined with classical molecular dynamics simulations revealed that oleic acid, naturally present in almond oil, stabilizes the internal W_1/O emulsion. Notably, anionic CNFs were more effective in preserving the double-emulsion morphology for over two months under ambient conditions. These findings highlight the potential of functionalized CNFs together with oleic acid as sustainable stabilizers for the development of double emulsions suitable for food, pharmaceutical, and cosmetic applications.

Keywords:
Cationic cellulose nanofibrils, oxidized cellulose nanofibrils, Pickering emulsions, double emulsions, molecular dynamics

References:

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