# Decoupling Interfacial and Bulk Stabilization in Natural Wax Pickering Emulgels using Cellulose Nanofibers

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In this work, we present the stabilization of natural wax-in-water Pickering emulgel systems using TEMPO-oxidized cellulose nanofibers (TOCNFs). Stable colloidal emulgels were obtained at exceptionally low nanofiber concentrations, with the nanofibers' degree of carboxylation exerting a minor effect on the critical stabilization concentration. We report that the formation of these systems is driven by high-energy emulsification above the wax melting point, where the resulting colloidal stability is maintained during the in situ crystallization of the dispersed phase, with the resulting particle size dependent on the initial energy imput.

The stabilization mechanism is attributed by a dual effect: the interfacial assembly of TOCNFs providing surface coverage, which was proven by confocal and electron microscopy, and the development of a hierarchical entangled fibrillar network in the continuous phase. This TOCNF network acts as a supramolecular cage, imparting a macroscopic yield stress and a solid-like viscoelastic behavior. Rheological analysis confirms that these systems behave as structurally arrested soft glassy materials, where the storage modulus is governed by both physical entanglement of the nanofibers and the jamming of the TOCNF-coated wax particles. Thermal characterization reveals a reduction in both the melting temperature and enthalpy compared to bulk wax, indicating a crystallization depression attributed to confinement and fractionated nucleation within the TOCNF shells. Finally, when applied to paper substrates, these emulgels produce high-performance, bio-based highly hydrophobic water barriers.

**Keywords:**
cellulose nanofibers; natural waxes; Pickering emulgel; confined crystallization; supramolecular network; hydrophobic coating

**References:**

Lombardo, S. et al. Decoupling interfacial and bulk stabilization in beeswax-in-water Pickering emulgels using carboxylated cellulose nanofibers
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