Towards a mechanistic understanding of water-in-water emulsion stabilization by bio-based polymers

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Water-in-water (W/W) emulsions are dispersions of two polymer-rich aqueous phases, immiscible above a given concentration. Due to the absence of organic solvents, they are increasingly considered as attractive systems for the delivery of active compounds, particularly because of their biocompatibility and low environmental impact. However, their large-scale development must overcome the challenges of stabilization related to the high interface thickness and the ultralow interfacial tension, which strongly reduce the efficiency of conventional surfactants (1). The aim of this work is to provide a better understanding of the stabilization mechanisms of W/W emulsions using bio-based polymers, by establishing the relationships between interfacial properties, emulsion microstructure, and stability. Three different strategies are investigated: nanoparticle-based stabilization, described in this abstract, lignin-chitosan complexation, and lignin-derived block copolymers. A range of bio-based polymers derived from lignin, chitin, and cellulose were considered, with particular attention given to lignin nanoparticles (LNPs) extracted via enzymatic hydrolysis (LNPs-EH) and soda processes (LNPs-SP). W/W emulsions were formulated using a model aqueous two-phase system composed of dextran and poly(ethylene oxide) (PEO). The phase diagram of dextran/PEO system was experimentally determined by turbidimetric titration using a cloud/clear point approach (2). The morphology and surface properties of the bio-based nanoparticles were characterized by transmission and scanning electron microscopy (TEM and SEM), Fourier-transform infrared (FTIR) spectroscopy, and zeta potential measurements. Wettability was assessed by contact angle measurements, while emulsion stability was monitored over time using optical and confocal microscopy, bottle tests, and static multiple light scattering. Wettability tests on glass slides show that LNPs-SP are more hydrophilic than LNPs-EH, and in lyophilized pellet-form, these display lower contact angles with dextran- and PEO-rich phases, indicating a higher affinity for both phases. This difference leads to distinct interfacial behaviors: LNPs-SP exhibit a greater tendency to adsorb at the water–water interface, while LNPs-EH remain predominantly in the bulk phase (Figure 1). Despite the adsorption of LNPs-SP onto the droplet surface, the emulsion coalesces gradually. This discrepancy in stabilization mode can be due to variations in surface chemistry induced by the extraction processes. Moreover, LNPs-SP improve emulsion stability compared to LNPs-EH, highlighting the key role of wettability in controlling interfacial adsorption and relative stabilization in W/W emulsions, in Pickering-like mechanism. Future research will investigate the combined use of different bio-based nanoparticles to improve interfacial structuring and emulsion stability.

Keywords:
water-in-water emulsion, bio-based stabilizers, lignin, interfacial adsorption, emulsion stability

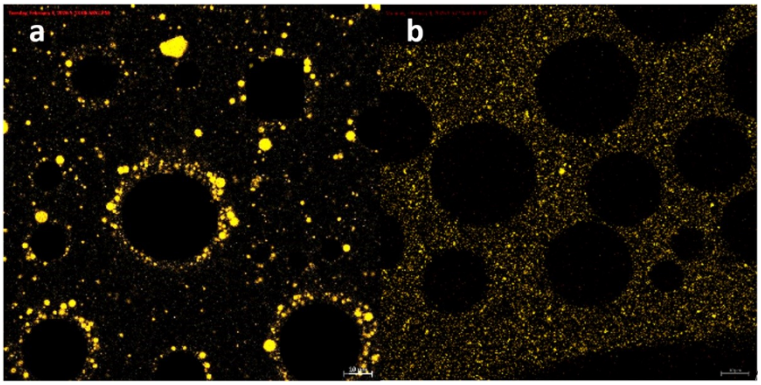

Figure 1 : Confocal fluorescence microscopy images of W/W emulsions acquired 3h after formulation using a ×63 immersion objective: (a) emulsion formulated with LNPs-SP; (b) emulsion formulated with LNPs-EH. LNPs appear in orange.

References:

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