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Structure and rheology of nanocellulose interfacial layers controlling digestion

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The use of particles such as nanocelluloses, i.e. cellulose nanocrystals (CNC) and nanofibrils (CNF) received increasing attention for the Pickering stabilization of fluid interfaces [1]. The adsorption of nanocellulose and nanocellulose-protein composites at oil-water or air-water interfaces facilitates the formation of stable and biocompatible emulsions and foams but depends heavily on the particles' surface properties. In this contribution, we review the structure of differently designed adsorption layers by neutron reflectivity and interfacial rheology measurements as a function of physico-chemical boundaries conditions (pH, salts, enzymes) [2, 3], surface properties of the cellulose crystals (natural, methylation, esterification) [4, 5], and protein or polysaccharide addition [6]. Native unmodified CNC (hydrophilic, negatively charged, and anisotropic nanoparticles) showed negligible viscoelasticity that could be increased by charge screening due to a shift from repulsive to attractive CNC interactions. Methylated CNCs formed dense monolayers with higher dynamic moduli compared to native CNCs and could be thermo-gelled into multilayers. The esterified CNCs formed aggregated clusters at the interface, resulting in a Maxwellian frequency behavior with distinctive relaxation times, a rarely observed phenomenon for interfacial layers. Scattering length density profiles obtained from neutron reflectivity measurements are used to elucidate the thickness and roughness of the adsorption layer, and in case of nanocellulose-protein composites, their spatial composition. Supported by *in vivo* digestion experiments in humans we rationalize the design principles of nanocellulose-stabilized emulsions and foams for food and drug delivery vehicles [7-9].

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