A smart combination of nanocellulose with a biodegradable polyester: Interfacial design, application, and end-life in aquatic environments

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Utilizing biodegradable material alternatives plays a crucial role in addressing the environmental consequences of fossil-based plastic pollution in marine environments. Natural and biodegradable polymers like cellulose have shown great potential to replace plastics. However, its susceptibility to water and moisture poses challenges when blending with hydrophobic polymers, thereby limiting its widespread use. To address these problems, we innovatively introduced the Pickering emulsion as a versatile template for combining hydrophilic nanocellulose with hydrophobic polyesters such as polycaprolactone (PCL) with the help of lignin nanoparticles (LNPs) as an interfacial compatibilizer. Combining LNPs with nanocellulose enabled the production of an ultra-stable PCL emulsion with small droplet sizes. After drying the emulsion, free-standing nanocomposite films with an even dispersion of PCL onto the surface of the nanocellulose network were achieved. The nanocomposites exhibited unprecedented mechanical properties in both dry and wet states (tensile strength up to 200MPa and 87MPa, respectively) due to the favorable intercomponent interactions. This interfacial design conferred additional properties to nanocellulose such as great protection against water vapor, UV, and oxidation, making it outstanding for the packaging application. Furthermore, the biodegradation rate and the kinetics of composite films were determined under aerobic conditions in simulated freshwater and real seawater environments. The changes in morphology and structural properties of the composite films during biodegradation were also systematically studied. Despite PCL having an aquatic ultimate biodegradation rate of 75%–80% and lignin under these conditions having no aquatic biodegradation, the smart combination of CNFs with PCL and lignin resulted in composite films with excellent overall biodegradability between 85% and 97% depending on the composite

formulations. All in all, our strategy offers a facile route to design multifunctional cellulose-based composites using only lignocellulosic and biodegradable polymers without any chemical modifications that harness the maximum benefit of each individual constituent.