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In this research, cellulose nanofibres (CNFs) were extracted and dispersed through a combination of alkali-acetylation treatments followed by soft mechanical treatments. Thereafter, PLA-based nanocomposites with CNF, nanocrystalline cellulose (CNC) and/or commercially available nanoclay (Cloisite™ 30B) were prepared and evaluated for use in food packaging. It was determined that composites with CNF or CNC and clay led to a great reduction in the oxygen transmission rate (OTR) and the water vapour transmission rate (WVTR) (up to a 90% reduction in the OTR and 76% in the WVTR for PLA/CNF 5%/C30B 5%). A significant increase in thermomechanical resistance was obtained (the storage modulus of PLA/CNF 5%/C30B 5% at 85°C, was 3.7 times higher than for neat PLA) and increased crystallisation kinetics (the PLA/CNF 1%/C30B 1% showed an 81% reduction in half-crystallisation time compared with neat PLA) without a significant reduction in optical properties at moderate nanoparticle loading.

Furthermore, it was found that solvent casting at a low temperature induced sub-micron-sized spherulites, which had little influence on water diffusion and transparency decline. On the other hand, high temperature processing led to larger spherulite sizes, which had a more significant impact on water diffusion and transparency reduction but also showed an increased water sorption. Finally, it was found that cellulose nanofibers reduced water diffusion to an extent similar to C30B (21% vs. 27%), while hybrid composites showed 49% decrease, albeit CNF based composites showed increased water sorption (7% for PLA/CNF 1% composite and 9% for PLA/CNF 1%/C30B 1% when compared with neat PLA). The reduced diffusivity of the hybrid nanocomposites suggested that the material was promising for active packaging, since low diffusivity leads to the slower release of active compounds such as essential oils. On top of that the CNF was surface modified with hydrocinnamic acid (gCNF), with the aim of reducing even more the release rate of carvacrol and to enhance the CNF dispersion with the PLA matrix. Consequently, carvacrol-loaded PLA/CNF, PLA/gCNF and hybrid PLA/CNF/C30B composites were prepared and evaluated on controlled release applications. It was established that the surface modification of CNF greatly enhanced the dispersion of the gCNF and that carvacrol-loaded hybrid composites showed a decreased release rate, high ductility and a reduced WVTR which made those composites promising material for food packaging films.

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