One-component nanocomposites based on polymer-grafted cellulose nanocrystals

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Abstract

One-component nanocomposites (OCNs) are a class of materials resulting from the assembly of hairy nanoparticles (NPs) in the solid state ^[1]. In contrast to traditional polymer nanocomposites, which often suffer from macrophase separation at high filler contents, OCNs cannot undergo phase separation of their components as these are covalently linked. As a result, the composition of OCNs can be tuned through a wider range of filler-to-polymer ratios. Additionally, the processing of OCNs is straightforward either via solvent casting of the NPs dispersion or by melt processing the dried material at a temperature above the glass or the melting transition of the grafted polymer.

While the synthesis of cotton cellulose nanocrystal (cCNC)-based hairy NPs has been extensively reported in the context of filler-matrix compatibility ^{[2] [3]}, the solid state properties of such hairy NPs are virtually unexplored. Moreover, most works in the field employ complex controlled polymerization methodologies to graft polymer chains from the surface of CNCs ^[4], which limits the upscaling potential of such routes. We present here the synthesis of cCNC-based OCNs displaying poly(hexyl methacrylate) or poly(methyl methacrylate) grafts through a synthetically undemanding protocol, which involves the surface functionalization of cCNCs with benzophenone moieties as radical photoinitiators (Figure 1), and the subsequent acrylate polymerization upon irradiation with 365 nm UV light. We show that this enables the synthesis of cCNC-based OCNs with cCNC contents of up to 40% and report the mechanical properties of these materials, which strongly depend on the nature of the grafted polymers.

References

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Figure 1. PHMA-OCN containing 20 wt. % of cCNCs.