

Surface engineering of cellulose nanofibers toward nanocomposite materials

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Abstract

Cellulose nanofiber (CNF), a naturally occurring nanomaterial, is a promising building block for future bio-based material. The CNFs have a unique structure, arising from their crystal structure, where cellulose chains are assembled into nanofibrils. Because of the crystal structure, the CNFs show excellent mechanical properties, such as high strength and high modulus. Therefore, the CNFs are expected to be used as reinforcing agents for plastics. However, it is generally hard to mix CNFs and plastics, and achieving the reinforcing potential in plastics has been a challenge.

Here, we have developed a surface engineering method for CNFs to achieve the reinforcing potential in plastics. Surface modification of ultrafine CNFs (~3 nm in width) was carried out by selectively modifying carboxy groups of 2,2,6,6-tetramethylpiperidiny1-1-oxyl-oxidized CNFs. The carboxy groups were selectively and stoichiometrically modified by the method, and the surface-modified CNFs were individually dispersed not only in water but also in organic solvents with low polarity such as chloroform and toluene.

CNF/plastic nanocomposite was successfully prepared by casting and drying the mixture of the CNF dispersion and plastic solution. The CNFs were individually dispersed in the plastic matrix and the composite showed high optical transparency. The well-dispersed CNFs efficiently enhanced the mechanical properties. Both the tensile strength and elastic modulus were increased by the addition of the CNFs. When the surface-grafted chains were appropriately selected, the increase in Young's modulus was in good agreement with theoretically predicted values. Furthermore, not only the modulus and strength, but also work of fracture of the nanocomposite increased. This is likely due to that efficient stress-transfer at the interfaces. In addition, the CNFs formed rigid and thermally stable interfibrillar network. Because of the network structures, storage modulus of the composite dramatically increased especially above glass-transition region of polymers. The increase in the modulus was in good agreement with values predicted by percolation theory. Thermal dimensional stability of the composites was dramatically increased due to the strong interfibrillar networks. The techniques pave the way toward novel green nanocomposite production.