Chapter 4

Cellulose Nanofibrils Composite Films

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ABSTRACT

Nanocellulose derived from biomass is a sustainable, lightweight, and mechanically strong material. Extensive research has been directed towards applying nanocellulose for advanced applications. Nanocellulose-based films are novel and unique for designing functional materials considering their combination of sustainability with new properties. Nanocellulose films can be fabricated through simple strategies and make it easy to access various applications, such as barriers, sensors, energy storage, and so on. In this chapter, we summarized the preparation methods of nanocellulose-based films while focusing on nanocellulose as the film matrix and highlighting some representative applications. Given the sustainability of the nanocellulose and ease of introducing functional groups, we believe that nanocellulose-based films promise great potential for future advanced applications.

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INTRODUCTION

Nanocellulose has received significant attention among the research community owing to its unique properties (Isogai, 2020; Lamm et al., 2021; Li, Clarkson, et al., 2021), such as sustainability, low density, good mechanical strength, and biodegradability (Copenhaver et al., 2022; Lamm et al., 2021; Mohammed et al., 2018; Sinquefield et al., 2020; Yanamala et al., 2014). Nanocellulose can be separated from various plants, algae, and other biological materials, which makes them a green and sustainable material (Abdul Khalil et al., 2012). Nanocellulose is comprised of both individual and bundles of nanoscale fibrils of polymeric repeating β-(1-4 linked), D-glucose linkages, held together tightly through hydrogen-bonding of the surface hydroxyl groups. There are mainly three kinds of nanocellulose, including cellulose nanocrystals (CNCs), cellulose nanofibrils (CNFs), and bacterial cellulose (BC). CNCs are rigid, rod-shaped particles with dimensions of 100–200 nm long and 5–20 nm wide (Thomas et al., 2018), and they are also referred to as "cellulose nanowhiskers" or "nanocrystalline cellulose". CNFs contain fibril structures with a length of >1 μm and a width of 5–200 nm (Copenhaver et al., 2021; Thomas et al., 2018). CNCs and CNFs are mainly isolated from plants, such as wood and bamboo, but can also be obtained from some animal tissues, such as tunicates (Thomas et al., 2018; L. Wang et al., 2021). BC is generated by bacteria (e.g., *Acetobacter xylinum*) through biosynthesis, and it has a length up to several micrometers and a width around 25–86 nm (Zhu et al., 2016).

CNFs can be isolated through mechanical, chemical, and biological approaches, and mechanical treatments (L. Wang et al., 2021), such as high-pressure homogenization, cryocrushing, and grinding, are the most commonly used. Depend on the raw materials source, chemical treatment is used to remove the lignin and hemicellulose. CNFs have a long aspect ratio and strong mechanical properties, which make them a good candidate for fiber reinforced composites. Compared to CNFs, which consist of significant amorphous regions, CNCs have a higher crystallinity of 54 - 88%. CNCs are produced through enzymatic and strong acid hydrolysis. CNCs have high axial stiffness (105−168 GPa), high Young's modulus (20−50 GPa), high tensile strength (~9 GPa), low coefficient of thermal expansion (~0.1 ppm/K), low density (1.5−1.6 g/cm^3), lyotropic liquid crystalline behavior, and shear thinning rheology (Iwamoto et al., 2011; Iwamoto et al., 2009; Lahiji et al., 2010; Thomas et al., 2018). BC are produced through cultivating bacteria for a few days in an aqueous culture media containing nutrition. BCs are composed of ribbon-shaped cellulose fibrils. The fibrils are fairly straight and continuous and have low size distribution. BCs have high crystallinity of 84−89%) and their elastic modulus can reach 78 GPa.

Nanocellulose has been processed into films, fibers, aerogels, and hydrogels (K. Li et al., 2022; Zhao et al., 2023) based on the type of the applications, ranging from nanocomposites (Hamedi et al., 2014), gas separation (Matsumoto & Kitaoka, 2016), flexible electronics (Hoeng et al., 2016), and fuel cells (Lu et al., 2015). Nanocellulose based film, where individual CNFs are stacked together to form lightweight and strong materials, is one important material platform for utilizing nanocellulose. To produce nanocellulose based films and to expand their application base, generally simple and versatile processes processing strategies have been adopted, such as solution casting and vacuum filtration. Nanocellulose based films with desired functional materials can be fabricated by introduction of functional additives in the process, or through surface modification of the nanocellulose with desired functional groups. These nanocellulose based films have unique properties, such as flexible, strong mechanical properties, good barrier properties, low thermal conductivities, and others.

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