

## Nanocellulose: The Sustainable Reinforcement Revolutionizing Polymer Nanocomposites

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### SUMMARY

In recent years, there has been a significant research interest in nanocellulosic materials as a potential nanofiller for reinforcing polymer matrices. This is due to their renewable nature, ready availability, biocompatibility, affordability, excellent physical properties, and customizable surface properties. This review aims to provide an overview of the various methods for fabricating nanocellulose-reinforced polymer nanocomposites and their properties and applications. The article focuses on the reinforcement of nanocellulose in various polymer matrices, including hydrophilic and hydrophobic matrices. Nanocellulose-reinforced polymer nanocomposites have enormous potential across various biomedical, packaging, electronic, and environmental applications.

### INTRODUCTION

Nanocellulose, which includes cellulose nanocrystals (CNCs) and cellulose nanofibers (CNFs), is a popular research topic due to its properties. It is used as reinforcement in nanocomposites in various industries. However, its high moisture absorption, poor wettability, and processing limitations are drawbacks. Composites have improved properties compared to the matrix phase, with discontinuous reinforcement phases. Natural composites like wood and bone serve as benchmarks for designing novel composites. Nanotechnology enables the control of materials at the nano-scale dimensions, with nanocellulose being a renewable biomaterial resource with novel properties for advanced material applications. Nanocomposites are an exciting class of materials where nanomaterials are reinforced, and nanocellulose as a reinforcing phase has gained significant attention due to its excellent properties. With a Young's modulus in the range of 100-130 GPa and a large specific surface area of several hundred m<sup>2</sup>.g<sup>-1</sup>, nanocelluloses can reinforce various polymer matrices with very low filler loadings to achieve targeted properties. Nanocellulose-reinforced nanocomposites with thermoplastic and thermoset polymers as matrices could be used in diverse fields such as packaging products, construction, automotive, furniture, electronics, membranes, etc. Renewable and biodegradable materials are gaining attention due to ecological threats caused by petroleum-based polymer products. This paper reviews the use of nanocellulose as a potential reinforcing component in polymer nanocomposites. It discusses different methods of preparing polymer nanocomposites, their properties, and potential applications. The review concludes by highlighting some of the challenges associated with using nanocellulose as filler in polymeric composites.

### Nanofillers Unleashed: Discovering the Power of Polymer Nanocomposites and Nanocelluloses:

Polymer nanocomposites are composite materials in which the polymer domain is the continuous phase. The reinforcing phase has at least one dimension, typically less than or equal to 100 nm. Due to nano-sized fillers, nanocomposites possess excellent properties that micro and macro composites cannot achieve. Nanoreinforcing materials with large specific surface areas and high aspect ratios (length by breadth ratio) exhibit better mechanical strength than their micro-sized counterparts. They require less quantity than micro fillers to reinforce the polymer matrix effectively. Therefore, some of the properties of the pristine polymer, such as density, remain unaffected in the resultant nanocomposites. The properties of nanocomposites depend on three major factors: the properties of the polymer matrix and nano reinforcing phase, together with interfacial interaction between filler surface and matrix polymer. For a specific nano reinforcement phase and matrix polymer, the properties of polymeric nanocomposites are highly dependent on the dispersion and distribution of nanomaterials in the continuous matrix phase. Nanocellulose, a common name for cellulosic nanomaterials,

typically has at least one dimension of about 100 nm. It includes bacterial nanocellulose (BNC) produced by certain strains of bacteria, cellulose nanofibres (CNF), and cellulose nanocrystals (CNC) extracted from lignocellulosic biomasses. Nanocellulose can be prepared from lignocellulosic biomass in two stages: pretreatment of raw cellulosic biomass and transformation of pretreated biomass into cellulose nanomaterials by mechanical, chemical, enzymatic, or a combination of more than one method. In the first stage, pretreatment is used to purify the biomass to obtain individual cellulosic fibres. The primary process, transformation, uses various methods such as mechanical, chemical, a combination of mechanical and chemical, and enzymatic processes to disintegrate cellulose nanocrystals from the individual cellulose fibre matrices. Acid hydrolysis is the most common chemical method for extracting nanocellulose from lignocellulosic biomasses. In the acid hydrolysis process, acid attacks intermolecular and intramolecular bonds among cellulose molecules, hemicellulose, and lignin in the biomass hierarchy structure to break down the rigid structure of lignocellulosic material. The morphology and dimension of nano cellulose depend on the source of natural resources when the same preparation method is employed.

Cellulose nanocrystal has several excellent characteristics that make it an ideal nano-dimensional material for the reinforcing phase in the polymer matrix. It has a higher aspect ratio than micro cellulosic materials, enabling good stress transfer between fillers and polymer matrix. It also has a high specific contact surface area due to the nano-sized dimensions. Hence, low concentration is required to disperse and distribute throughout the matrix and achieve targeted properties. The enormous hydroxyl groups on the surface of the cellulosic nanofiller could form hydrogen bonding/participate in the chemical reaction with the matrix for better reinforcement in the matrix. Plenty of hydroxyl groups also provide an opportunity for the functionalization of nanocellulose with appropriate molecules for effective nanocellulose reinforcement into various types of matrix polymer, viz. polar/non-polar. Cellulose nano fillers are environmentally friendly and renewable nanofillers.

### **Exploring the Potential of Nanocellulose and Polymer Composites:**

Polymer nanocomposite processing can be divided into physical and chemical methods. Physical methods include solution processing, melt mixing, and electrospinning, while in-situ polymerization is chemical. Nanocomposites based on cellulosic nanomaterials can be prepared with non-biodegradable and biodegradable polymers as potential matrices. However, hydroxyl groups on the nanocellulose surface make it difficult to disperse in nonpolar polymeric matrices. Nanocellulose aggregation in the polymer matrix increases as the size of the nanocellulose decreases due to an increase in the specific surface area. Nanocellulose surface modification with appropriate molecules such as esterification, silylation, cationization, and polymer grafting with polar/nonpolar molecules can control the aggregation. Solution processing is a simple approach to preparing polymer nanocomposites. Nanofillers are dispersed in the base polymer or pre-polymer solution by solution processing method. Nanofillers and polymers can be agitated physically to obtain a homogeneous mixture, such as mechanical stirring, ultrasonic methods, etc. In melt mixing, nanomaterials are dispersed into the polymer matrix at melt conditions, and extrusion can obtain polymer nanocomposites. However, uniform dispersion and distribution of nanocellulose in the polymer matrix are essential for improving the properties of resultant nanocomposites. Before the polymerization, nanofillers could be impregnated in the monomer solution; then, polymerization can be carried out for the in situ processing of nanocellulose polymer nanocomposite.

### **Properties of Polymer-Nanocellulose Nanocomposites**

#### **Mechanical properties**

Nanocellulose polymer nanocomposites can improve mechanical properties by ensuring uniform dispersion of nanomaterials in the polymer matrix and good interfacial interaction between fillers and matrix. An optimum concentration is required to improve mechanical properties. Beyond this limit, further increases in nanomaterials can decrease mechanical properties due to agglomeration in the polymer matrix. High aspect ratio creates entanglements in the nanocomposite domain structure, further improving mechanical properties. However, aggregation of nanofillers can promote local stress concentrations, resulting in mechanical failure of the polymer nanocomposites. The interaction between the nanocellulose fillers and polymer matrix may be reduced resulting in a decrease in the tensile strength of the composite films.

**Thermal properties**

Nanocellulose polymer nanocomposites have crucial thermal properties, which are characterized by measuring  $T_m$ ,  $T_g$ , crystallization temperature, and thermal degradation properties. The incorporation of nanocellulose leads to an increase in  $T_g$  due to strong hydrogen bond formation between the PVA matrices and nanocellulose. However, the thermal degradation properties do not exhibit any clear trend concerning the filler concentration. The nanocomposite films' thermal degradation properties increase with the increase in nanocellulose filler loading.

**Barrier properties**

Nanocellulose polymer nanocomposites have poor permeability due to their crystalline nature and the percolating network formed with the matrix through hydrogen bonding. The network structure decreases penetrant diffusivity, making these films suitable for barrier membranes for organic solvents. The interaction between the nanofillers and matrix increases tortuosity and reduces transport rate. The water absorption capacity of nanocomposites decreases due to the network structure between nanocellulose and the polymer matrix. Nanocellulose material is hygroscopic because of its abundance of hydroxyl groups. Beyond the threshold limit of filler concentration, water absorption increases with increasing nanocellulose filler in the matrix.

**Other properties**

The response property of hydrogels is crucial for their applications in various fields. Hydrogels that contain nanocrystalline cellulose exhibit a rapid deswelling rate because of the interconnected micropores formed by the nanocrystals and pristine hydrogel through extensive hydrogen bonding. The swelling ratio of nanocomposite hydrogel decreases with increasing nanocrystal, as the more vital interaction of the nanocrystal through hydrogen bonding in the gels reduces the swelling. The advantage of using nanofillers instead of microfilters in the polymer matrix is preserving transparency for the matrix polymers while achieving other functional properties of the nanocomposite materials. The transparency of the nanocellulose polymer composite will be preserved when nanofillers have homogeneous dispersion and distribution in the polymer matrix. However, the transparency of the nanocomposite films may decrease with increasing nanomaterial concentration in the polymer matrix due to possible agglomeration. Acrylic nanocellulose nanocomposite films preserved good transparency when prepared using a coupling agent in situ mini-emulsion polymerization technique. This ensures improved dispersion and prevents aggregation during the polymerization and film formation.

**Applications of Polymer-Nanocellulose Composites**

Renewable resource-based materials are being explored for packaging applications. Proper food packaging aims to preserve food quality and safety. Nanocellulose, with features such as high crystalline nature, nontoxic properties, and hydrogen bonding ability, can be used as a filler for packaging materials. Reinforcing nanocellulose with nanoclay in the poly(lactic acid) matrix significantly improves water and oxygen barrier properties. Nanocellulose can also be used as a reinforcement phase in biobased films, coatings, and foam. Algae nanocellulose/polypyrrole composite provides large catalytic oxidation currents and enables storing of the charge. Polyaniline nanocellulose composite films have enhanced conductivity and flexibility and can find applications in electronic fields.

**CONCLUSION**

Nanocellulose has potential for improving matrix polymer properties. However, challenges need to be addressed before polymer-nanocellulose nanocomposites can be produced and applied. Specifically, the influence of nanofiller size, shape, and surface chemistry on domain structure and properties of resultant polymer nanocomposites needs to be explored. Also, the compatibility of nanocellulose in different polymer matrices is important. Poor dispersion and distribution of nanofillers in the polymeric domain structure is a major problem, but surface modification of nanocellulose can improve interfacial interaction. More research is needed to develop polymer-nanocellulose nanocomposites for diverse applications.

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