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Feature article

Recent developments on nanocellulose reinforced polymer nanocomposites: A review

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ABSTRACT

The development of nanocellulose and nanocellulose-based composites and materials has attracted significant interest in recent decades because they show unique and potentially useful features, including abundance, renewability, high strength and stiffness, eco-friendliness, and low weight. This review addresses critical factors in the manufacturing of nanocellulose composites, followed by introducing and comprehensively discussing various nanocellulose composite processing techniques. The review also provides advances on rubber and thermoset polymer matrices, such as unsaturated polyester resin, formaldehyde resins, and polyethylene terephthalate, used to reinforce cellulose nanocrystals (CNCs) or cellulose nanofibers (CNFs). The paper concludes with new findings and cutting-edge studies on electrospun nanocellulose composites. Different aspects, including preparation methods, morphology, mechanical behavior, thermal properties, and barrier action, as well as comparisons of CNC- and CNF-reinforced rubbers or thermoset polymers and electrospun composites, are investigated.

Contents

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1. Introduction

Presently, renewable and biodegradable materials are receiving extensive attention from both scientific and industrial communities, because the use of conventional petroleum-based polymer products has created ecological threats such as global warming and plastic pollution. Cellulose is the most abundant natural polymer, and has been used to propose rational solutions for these issues. In recent decades, the interest in nanostructured celluloses, or nanocellulose, has sharply increased because of the specific chemical and physical properties of these materials. Nanocelluloses are typically distinguished into two main groups: i) those obtained by acid treatment, referred to as cellulose nanocrystals (CNCs), and ii) those produced mainly by mechanical disintegration, called cellulose nanofibers (CNFs). The final chemical and physical properties of nanocellulose depend directly on the source and preparation conditions of the cellulose. According to their different properties, CNCs and CNFs are used for different applications.

The use of nanocellulose as reinforcement in nanocomposites has become a popular research topic. In addition to the numerous advantages of nanocellulose, such as low cost, low density, renewability, low energy consumption, high specific properties, biodegradability, and relatively good surface reactivity, it shows better properties as a reinforcing phase in nanocomposites as compared to micro- or macro-cellulose composites. The tailorability, design flexibility, and processability of nanocellulose–polymer composites permit extensive utilization in the automotive, packaging, electronics, and biotechnology industries, among others. However, some disadvantages are associated with nanocellulose use as reinforcing material, particularly its high moisture absorption, poor wettability, incompatibility with most polymeric matrices, and limitations in processing temperature. These drawbacks have encouraged scientists to focus on these issues; various methods, by the modification of nanocellulose or polymer matrices or by all-new processing techniques, have been proposed to produce high-performance nanocellulose-reinforced composites with good properties.

Although many publications have reported on the use of nanocellulose-reinforced polymer composites, few review papers have discussed this topic [1–9]. Certain recent advances and findings have not been addressed satisfactorily in previous publications. Most prior publications focused on the use of either CNC or CNF, while here, we provide a comparative study on the effects of CNC versus CNF on the properties of composites incorporating them.

The purpose of this review is to combine all recent research to provide readers with a comprehensive overview of the advanced science and engineering of nanostructured cellulose composites. It provides an in-depth look at nanocellulose types of CNC and CNF, composite processing, and the effects of nanocellulose on the mechanical, thermal, crystalline, barrier and other properties of matrix materials, including rubber and thermoset polymers such as epoxy and unsaturated polyester. Following this review, we extensively discuss electrospun nanocellulose composites, their preparation, and their properties.

2. Nanocellulose composite processing

Nanocellulose particles and related materials can be prepared by different processing techniques that directly influence the final material properties. Particle organization, degradation, and interaction with the matrix are some examples discussed further in this section.

First, it is important to emphasize that, independent of the selected processing method, the major challenge in nanocomposite preparation is the quality of particle dispersion. Despite relevant advances in recent decades, overcoming agglomeration remains difficult. The achievement of a completely homogeneous dispersion of individual nanoparticles has proven remarkably difficult [10]. This is because of the natural tendency to agglomerate that nanoparticles present, and is not exclusive to cellulose nanoparticles. Certain physical characteristics promote the intrinsic tendency of nanoparticles to agglomerate. In one nanoparticle, almost 50% of the constituent atoms can be on the particle surface, which has a high specific surface area and surface energy. When dried, cellulosic materials pass through the simultaneous processes of irreversible agglomeration, reorganization, and co-crystallization caused by the attractive forces present on the cellulose surface [11,12]. In nanocellulose, these forces arise from the presence of surface hydroxyl groups, which can reach areal concentrations approaching 7.2 groups/nm^2 in CNCs. An estimated energy of 7.5×10^{-16} J is attributed to the hydrogen bonds (H-bonds) between two parallel CNCs in contact. This value is almost two orders