Cellulose diacetate reinforced with electrospun cellulose fiber: A new route to prepare an all cellulose-based composite

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**Abstract**

An all cellulose-based composite was produced by applying cellulose diacetate (CDA) and electrospun cellulose fibers (ECFs) as a matrix and reinforcing fibers, respectively. The ECFs were prepared by electrospinning CDA followed by alkali treatment. The alkali treatment of the electrospun CDA fibers removes acetyl groups and creates a new crystalline domain similar to cellulose fibers, which may result in increased mechanical properties. The physical properties of ECFs are highly correlated with the acetyl content. The mechanical properties of the CDA/ECF composites were evaluated by measuring the tensile strength and Young's modulus as a function of the hydrolysis time. The results showed that the integration of the CDA matrix with 15% weight/weight (w/w) ECF, which was alkali-treated for 7 h, was optimal, resulting in an improved tensile strength and Young's modulus by factors of 1.7 and 2.2, respectively. This study demonstrates that the increased mechanical properties are attributed to the reinforcement of ECFs as well as good interfacial adhesion.

**Keywords:**

- A. Polymer–matrix composites
- B. Strength
- D. Mechanical testing

**1. Introduction**

The development of cellulose nanofibers has drawn significant research interest in the last few decades due to their unique characteristics such as a large surface area, high mechanical strength, and low thermal expansion coefficient [1]. Numerous researchers have used this concept to prepare a series of polymer composites [2–5]. However, due to strong inter- and intra-molecular hydrogen bonding, such short hydrophilic cellulose nanofibers have strong intrinsic interactions. As a result, it is very difficult to achieve homogeneous dispersion in a polymer matrix [6,7]. In addition, nano-fibrillations of various materials, such as wood, cotton, ramie, and bacterial and tunicate celluloses, are still limited to the laboratory level [8]. Their time- and energy-consuming manufacturing processes further limit the practical application of cellulose fibers in industry.

As an alternative but effective method, cellulose fibers can be produced by the use of cellulose diacetate (CDA), in which CDA is electrospun to prepare CDA fiber mats followed by hydrolysis to transform the cellulose fibers [9,10]. Electrospinning is a simple way to fabricate nanofibers in large quantities for their practical application in composite materials. The physical properties of the electrospun cellulose fibers (ECFs) are highly correlated with the acetyl content. The removal of acetyl groups creates new crystalline domains which increase the mechanical properties of the fibers.

Some researchers have reported a novel all cellulose-based composite structure possessing strong mechanical features by using selective dissolution of the cellulose fiber surface [11–14]. In this study, different types of all cellulose-based composites were produced by applying CDA and ECFs as a matrix and reinforcing fibers, respectively.

In the preparation of the ECFs, the degree of substitution was controlled in order to maintain the balance between the mechanical properties and compatibility with the CDA matrix. The hydrolysis of electrospun CDA fibers removes acetyl groups and creates a cellulose-like crystalline domain which enhances the mechanical strength of the CDA/ECF composite. On the other hand, the hydroxyl groups produced by hydrolysis decreases the chemical compatibility with the hydrophobic CDA matrix. Therefore, the acetyl groups on the ECFs should be partially preserved in order to maintain a balance between these two opposing effects.

We attempted to achieve an all-cellulose based composite by incorporating 15% (w/w) ECFs, which were alkali-treated for various times, into a CDA matrix using a film casting method. The alkali-treatment of the ECFs was characterized by attenuated total reflection-infrared (ATR–FTIR), and the acetyl content was determined by titration. The crystalline structure of the ECFs was determined by X-ray diffraction (XRD). The physical properties of the CDA/ECF composites were characterized by their tensile and thermal stability properties, and the interfacial adhesion between...