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Effect of fiber orientation on tensile behavior of biocomposites prepared from nettle and poly(lactic acid) fibers: Modeling & experiment

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ABSTRACT

In this work, a micromechanical model of tensile behavior of fibrous composites was formulated by taking into account of the stress-strain behavior of fiber and matrix, the directional arrangement of fibers in the composites, and the relative space occupied by the constituent fibers and matrix in the composites. A simple and easy-to-use expression was obtained for predicting the tensile stress of composites as a function of tensile stresses of fiber and matrix, fiber and matrix volume fractions, and fiber orientation factor. The model was validated with a set of biocomposites prepared by using nettle and poly(lactic acid) fibers and employing compression moudling technology. The model was also compared with the well-known "rule of mixtures" equation using the experimental results. While the "rule of mixtures" equation was found to highly overestimate the tensile stress, the model predicted the experimental results satisfactorily.

1. Introduction

The mechanical behavior of fiber reinforced composites has always been an interesting theme of research. In the recent days, there is a resurgence of scientific interest to investigate the tensile behavior of the composites with a view to structural engineering applications [1–15]. It is revealed that the tensile properties of the composites are decided by the tensile characteristics of the fibers and matrix, their mutual interaction, and the interactions between composites and outer influences. The internal structure of composites plays a key role in deciding their tensile behavior. A few attempts were made to examine the effect of fiber orientation in determining the tensile properties of composites. It was reported that if all the reinforcing fibers were lying parallel to the longitudinal axis (direction of applied load) of the composites then the tensile stress developed in the composites along this axis was given by $\sigma_c = \sigma_f \nu_f + \sigma_m \nu_m$, where σ_c, σ_f, and σ_m denote the tensile stresses of composite, fiber, and matrix, respectively, and ν_f and ν_m indicate the volume fractions of fiber and matrix, respectively, in composites. This expression is often referred to as "rule of mixtures" equation. In the past, a few attempts were made to estimate the tensile properties of fiber reinforced composites by using this expression. Gomes et al. [16] reported that according to the rule of mixtures, the estimated tensile strength of curaua fiber reinforced cornstarch-based biocomposite was about 630 MPa, while the experimental data showed tensile strength around 327 MPa. Cao et al. [17] observed that the tensile strengths of bagasse fiber reinforced aliphatic polyester composites were lower than those estimated from the rule of mixtures by 22% and 46% at 20% and 50% fiber content, respectively. Furthermore, Herrera-Franko & Valadez-Gonzalez [18] found that the contribution of henequen fibers to the longitudinal stiffness of high density polyethylene composite was much lower than that estimated from the rules of mixtures. Similarly, Ku et al. [19] reported a poor correspondence between the experimental readings of tensile properties of natural fibers (hemp. Hardwood, and rice hulls) reinforced high-density polyethylene composites and those estimated ones from the rule of mixture. The poor correspondence between the theoretical and experimental readings was probably due to the fact that the "rule of mixtures" equation is not valid if the fibers are not lying parallel to the longitudinal axis of the composites. If the fibers are oriented to a finite number of directions, or they display a continuous distribution of directions, then Cox [20] suggested to modify the "rule of mixtures" equation as follows: $\sigma_c = \eta_0 \sigma_f \nu_f + \sigma_m \nu_m$, where η_0 is called orientation efficiency factor. Krenchel [21] reported a method for estimating the orientation efficiency factor. According to him, a group of parallel fibers with a total cross-sectional area Δa_f lying at an angle ϑ to the direction of applied load are equivalent to a group of fibers of an area $\Delta a'_f$ aligned in the direction of the applied load, where $\Delta a'_f = \Delta a_f \cos^4 \vartheta$. For groups of differently oriented fibres, the equivalent area of the total reinforcement is $a'_f = \sum \Delta a_f \cos^4 \vartheta$ and the cross-sectional area of the total reinforcement is $a_f = \sum \Delta a_f$. Then, the orientation efficiency factor η_0 can

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be calculated as follows: $\eta_0 = a'_f/a_f = \sum \Delta a_f \cos^4 \vartheta / \sum \Delta a_f$. Clearly, if all the fibers are lying parallel to the longitudinal axis (direction of applied load) of the composite then $\eta_0 = 1$, hence $\sigma_c = \sigma_f \nu_f + \sigma_m \nu_m$ ("rule of mixtures" equation). In literature, a few experimental studies were reported to use Cox's equation in conjunction with Krenchel's methodology to examine the effect of fiber orientation on the tensile behavior of fiber reinforced composites. Serrano et al. [22] found that the fiber orientation factor and mean fiber orientation angle significantly affected the tensile properties of composites prepared from old newspaper fibers and polypropylene matrix. However, there is hardly any theory available for scientifically explaining the role of fiber orientation on the tensile behavior of fiber reinforced composites.

In this work, an attempt was made to develop a model of fiber orientation induced tensile behavior of fiber reinforced composites and validate it with the experimental results obtained on fibrous biocomposites prepared from nettle and poly(lactic acid) fibers. The nettle/PLA biocomposites are relatively new in the field of fibre reinforced composites. They were not much studied earlier despite the fact that nettle fibers were reported to exhibit in principle the same potential as other bast fibers to act as reinforcement for poly(lactic acid) matrix [23]. But, PLA was earlier suitably used with other bast fibers like flax, jute, hemp, ramie, kenaf [24–38]. Such green composites made up of natural fibers as reinforcement and biodegradable resin as matrix are generally used for automotive applications [39]. In fact, in one of the recent studies [4], it was found that the biocomposites prepared with equal weight proportion of nettle and poly(lactic acid) exhibited high potential for automotive dashboard panel application because of their excellent mechanical properties.

2. Materials and methods

2.1. Materials

In this work, the Himalayan nettle (*Girardinia diversifolia*) filaments of 1 m–2 m length, procured from Uttarakhand Bamboo & Fiber Development Board in India, were used. They are displayed in Fig. 1. Poly(lactic acid) fibers of 55 mm cut length and 6 denier (6.67×10^{-4} g/m) fineness were also used in this study. Further, easily obtainable EMPARTA® Sodium hydroxide pellets (low chloride) with assay (NaOH) more than 97% and EMPLURA® Acetic acid were used for alkali treatment of nettle fibers.

2.2. Methods

2.2.1. Alkali treatment on nettle fibers

The nettle filaments as procured were cut into staple fibers of 55 mm ± 1 mm length, followed by cleaning them mechanically and

Fig. 1. Nettle filaments.

sorting them manually. The cleaned staple nettle fibers were treated with alkaline (NaOH) solution in a laboratory-based batch process. The optimum alkali treatment conditions, reported elsewhere [40], were followed for alkali treatment. In one batch, 40 g of cleaned nettle fibers were immersed into 2000 cm^3 of alkaline solution with an alkali concentration of 10%. The fibers were treated at 61.5 °C temperature for 1800 s. The treated fibers were extracted from the solution, squeezed, washed with distilled water and neutralized with 10% acetic acid. The fibers were then washed with distilled water and squeezed to remove excess water. Afterwards, they were manually opened to single fiber stage. This was followed by drying in a hot air oven at a temperature of 30 °C for 1.728×10^5 s. The dried fibers were kept in standard testing atmospheric conditions as stipulated in ASTM D1776–04 standard.

2.2.2. Development of biocomposites and PLA film

The alkali-treated nettle fibers were mixed with poly(lactic acid) fibers as homogeneously as possible by manually in seven different blend proportions (w/w) (10/90, 25/75, 30/70, 35/65, 40/60, 45/55, 50/50). The fiber blends were fed to a laboratory-based miniature carding machine (Make: Trytex, India). The machine had a feed roll running at 0.0173 rad/s, cylinder at 5 rad/s, and doffer at 0.0345 rad/s. It delivered parallel-laid fiberwebs consisting of nettle and poly(lactic acid) fibers with seven different blend proportions (Fig. 2a). Afterwards, the fiberwebs were processed through a laboratory-based compression moulding machine (Make: Carver, USA), keeping temperature at 160 °C and pressure at 0.6 MPa. This is displayed in Fig. 2b. In this way, a set of biocomposites with seven different blend proportions of nettle and poly(lactic acid) was developed (Fig. 2c). In addition to these seven biocomposites, a film was prepared from 100% poly(lactic acid) fibers following the same method as stated above. The nominal basis weight and thickness of the biocomposites as well as the film were 900 g/m^2 and 1 mm, respectively.

2.2.3. Characterization of nettle fibers

The alkali-treated nettle fibers were tested for density and fineness. The density of nettle fiber was determined using Archimedes method in accordance with ASTM D3800-9 standard. A weighted quantity of fibers was fully immersed in water and the volumetric displacement of water was observed. The density of fiber was determined by the ratio of the weight of fiber to the volume of water displaced. In order to measure the fineness of alkali-treated nettle fibers, the single fiber weighing method (ASTM D1577-07, Reapproved 2012) was followed. For this, nettle fibers of 100 mm staple length were weighted individually using an electronic micro-weighing balance (Model: AND GR-201). An average of fifty readings was taken. Further, the alkali-treated nettle fibers were tested for tensile properties on Instron tensile tester according to single fiber tensile test as prescribed by ASTM D3822-07 standard. The gauge length and crosshead speed were kept constant at 25 mm and 0.0417 mm/s, respectively. Fifty fibers, each of 55 mm cut length, were randomly selected for this test. Each of fifty fibers was first weighed using the above-mentioned electronic micro-weighting balance. Then, each fiber was attached to a paper window and subsequently to the jaws of the tensile tester. The raw tensile data were used and recalculated to obtain the average stress-strain curve of the fiber. Subsequently, using linear interpolation method, the stresses were calculated for the given strains with a step of 0.1% fiber strain. The average of fifty readings was taken to determine the mean strength and mean strain-at-break.

2.2.4. Characterization of biocomposites and PLA film

The biocomposites were tested for fiber orientation characteristics and tensile properties. To determine the orientation of fibers, the images of the biocomposites were taken using a scanning electron microscope (Zeiss, Model EVO50) at 50× magnification level. The images of the fibers were digitized in the form of coordinates of many points using an image processing system. The observed fibres were then