



Mechanical properties of sisal fiber reinforced high density polyethylene composites: Effect of fiber content, interfacial compatibilization, and manufacturing process



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ARTICLE INFO

Article history:

Received 11 December 2013

Received in revised form 6 June 2014

Accepted 26 June 2014

Available online 3 July 2014

Keywords:

A. Polymer–matrix composites (PMCs)

B. Creep

D. Mechanical testing

ABSTRACT

In this paper, we investigated the effect of fiber content, interfacial compatibilization, and manufacturing process on the mechanical properties (tensile, impact and creep) of sisal fiber (SF) reinforced high-density polyethylene (HDPE) composites. The increase of fiber content and interfacial compatibilization with maleic anhydride grafted HDPE (MAPE) were found to improve the mechanical properties of the composites. Compared with simultaneous blending, a pre-impregnation process with the compatibilizer, namely MAPE, improved the interfacial bonding between the fibers and the matrix, which in turn improved the mechanical properties of the composites. The General Power-Law equation was used to model the creep behavior of the composites. The identified material parameters based on the creep data were used to predict the creep-recovery behavior of the composites, and good agreement was achieved between the predicted and experimental creep-recovery responses.

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1. Introduction

In the last several decades, natural fiber-reinforced polymer composites have received substantial attention [1,2]. Natural fibers are harvested from renewable resources and readily available at low prices. Their specific properties are comparable to synthetic fibers (e.g., glass fibers) that are traditionally used as reinforcing phases in polymer based composite materials [1,3]. Furthermore, natural fibers are nonabrasive and exert less wear on processing machine compared to inorganic fibers. Sisal fiber, one of the most widely used natural fibers, has an annual output about 4.5 million tons all over the world [4]. Among the available cellulosic fibers, sisal fiber possesses moderately high specific strength and stiffness and is a good candidate as reinforcing material [5,6,2,1,3]. Sisal fiber reinforced thermoplastics composites have gained tremendous interest because of their easy processing, low cost and recyclability [2]. Among them, polyolefin is the most widely used thermoplastics matrices [7–18].

Concerning the mechanical properties of natural fiber reinforced polymer composites, the primary issue is the poor

interfacial adhesion between the hydrophilic fibers and the hydrophobic polymeric matrix due to the polarity difference between them [17]. Interfacial modification through wetting agent can improve the adhesion between the fibers and the matrix, which in turn can enhance the mechanical properties of the composites. Maleated polyolefin is an effective compatibilizer to enhance the coupling and interaction between sisal fiber and polyolefin matrix [8–10,14,16]. In one of our previous works [8], a pre-impregnation process was carried out on the single screw extruder equipped with a special die assembly to pre-coat sisal fibers with maleated polypropylene (PP) before blending sisal fibers with the PP matrix, which substantially improved the interfacial adhesion between sisal fibers and the PP matrix. Li et al. [17] studied two types of fiber surface treatment methods, i.e., chemical bonding and oxidation, to improve the interfacial bonding between sisal fibers and HDPE matrix. Their single fiber pull-out tests and microstructural observations demonstrated that oxidants, namely permanganate (KMnO₄) and dicumyl peroxide (DCP), can roughen the fiber surface and introduce mechanical interlocking with the HDPE matrix. They also found that van der Waals force can be setup between gamma-methacryloxypropyltrimethoxy silane treated sisal fibers and the HDPE matrix, whereas 3-aminopropyltriethoxy silane can only react with the sisal fibers but not with HDPE.

Creep is a mechanical behavior of materials that deformation accumulates as time evolves under constant loading, temperature

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and humidity. It is an important physical property that should be taken into consideration for the development of composite materials which undergo long-term loading, especially at high temperature and humidity to ensure long-term stability of strength and structural integrity. Park and Balatinecz [19] demonstrated improved creep property for wood-fiber reinforced polypropylene composites with increasing wood-fiber content and fiber–matrix interfacial modification with a wetting agent. Cyras et al. [20] conducted flexural creep tests on sisal-fiber-PCL (polycaprolactone)-starch composites at different temperatures. The creep compliance increased with the increase of temperature and with the decrease of the fiber content. They utilized a four-parameter Burgers model to quantify the creep behavior of the composites. Alvarez et al. [21] investigated the effect of fiber content on the flexural creep property of sisal fiber reinforced cellulose derivatives/starch composites, and they found a significant improvement of the creep resistance of the polymeric matrix due to the addition of sisal fibers to the composites. They also applied various mathematical models to model the creep property of the composites, which included the Norton equation, a creep power law and a four-parameter Burgers model.

2. Method

2.1. Materials

Blow-molding grade high-density polyethylene (HDPE, LADENE B 5429) with a density of 0.954 g/cm^3 and melt flow index of 29.0 g/10 min (Saudi Basic Industries Corporation (SABIC), Saudi Arabia) was used as the matrix material.

Maleic anhydride grafted HDPE (MAPE), supplied by Chenguang Chemical Research Institute, China, was used as the compatibilizer in this study. Sisal fibers (SF) were obtained from Huari Natural Fibre Products Co., Ltd., Guangdong, China. The fibers with diameter ranging from 100 to 200 μm were supplied as a roll of sisal fiber yarns.

2.2. Mechanical characterization of single sisal fibers

The mechanical properties (tensile strength, modulus and elongation at break) of single sisal fibers were characterized according to the standard of ASTM D 3822-01 on an Instron model 5567 universal testing machine (Instron Corp., High Wycombe Bucks, UK). Fig. 1(A) shows a schematic illustration of the specimen and the corresponding value of the geometric parameters.

Single sisal fibers were randomly extracted from the fiber bundle with tweezers. They were then cut into $\sim 30 \text{ mm}$ long segments and dried in an air-circulating oven at 60°C for 48 h. Subsequently, 10 randomly selected fibers were glued onto the pre-cut cardboard frame shown in Fig. 1(A). The specimens were then dried for another 12 h at 60°C to remove the extra moisture that might be absorbed by the fiber during the preparation of the specimen. After completely dried, the diameter of the fiber was measured by using a micrometer attached to an optical microscope at five locations along the fiber length and averaged. Subsequently, the specimen was mounted onto the tensile machine, and the two thin cardboard strips parallel to the fiber were cut in order to let the fiber bear the tensile loading during the tensile test. The crosshead speed was set to 2.5 mm/min according to experimental standard.

2.3. Fabrication of the composites

Two methods were adopted to fabricate the PE/SF or PE/SF/MAPE composites. In the first method, the sisal fibers, HDPE (and MAPE if necessary) were melt-blended simultaneously with a twin-screw extruder (Brabender GmbH & Co., Duisburg, Germany).

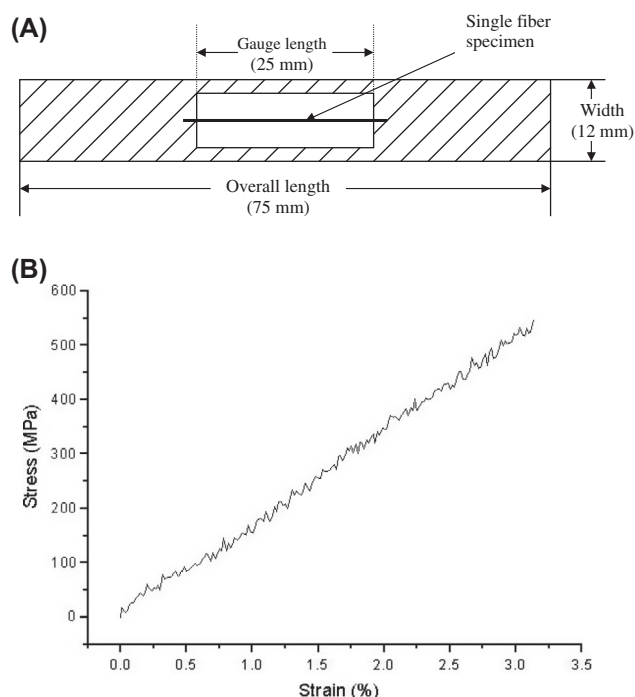


Fig. 1. (A) Schematic illustration of the specimen for the single fiber tensile test; (B) a representative stress–strain curve of a single sisal fiber.

Hereafter this fabrication method will be referred to as *simultaneous blending*. Before melt-blending, sisal fibers were chopped into $\sim 5 \text{ mm}$ long short fibers and then dried in an air-circulating oven at 60°C for 48 h. The melt-mixing temperature and speed were 220°C and 30 rpm, respectively. The composites without the MAPE compatibilizer were also prepared for comparison. In the composites with compatibilization, the weight percent of MAPE relative to the sisal fibers was kept constant at 50%. Therefore, in the final composites, a 20% fiber content implied a 10% MAPE content with respect to the total weight of the composite material. Specifically, the following composites were fabricated: (1) PE/10% SF, (2) PE/20% SF, (3) PE/10% SF/5% MAPE, and (4) PE/20% SF/10% MAPE. 20% was the highest fiber content at which we could reach due to the difficulty to blend the constituents simultaneously at higher fiber content, which might be caused by the high viscosity of MAPE.

In the second method, a pre-impregnation technique was utilized to fabricate MAPE compatibilized PE/SF composites [8]. To facilitate the interfacial wetting between the high viscosity MAPE and sisal fibers, MAPE and HDPE at 1:1 weight ratio were melt-blended by the twin-screw extruder to reduce the viscosity of MAPE before the pre-impregnation process. The sisal fibers were also dried in an air-circulating oven at 60°C for 48 h. During the impregnation process, the extruding temperature and rotation speed of the screw were 175°C and 1 rpm, respectively. The obtained MAPE/SF pre-pregs were cut into $\sim 5 \text{ mm}$ long pellets for further processing. The weight of polymeric component in the resulted pre-pregs was determined by subtracting the weight of sisal fibers from the weight of MAPE/SF pre-pregs. Subsequently, the pre-preg pellets were further melt-blended with pure HDPE to dilute fiber content to the desired level. Melt-blending was conducted with a single-screw extruder. This fabrication method will be referred to as *pre-impregnation* in the sequel. The extruder temperature and screw speed were set to 175°C and 50 rpm, respectively. In the final composites obtained through the pre-impregnation process, the weight percent of MAPE relative to the sisal fibers was kept constant at 50%. Therefore, in the final

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