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Strength improvement in injection-molded jute-fiber-reinforced polylactide green-composites

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ABSTRACT

The mechanical properties of green-composites based on polylactic acid (PLA) with jute fibers were investigated. A long fiber pellet was developed to obtain a high aspect ratio of residual fiber after injection molding. Comparative studies were carried out, where shorter fiber pellets were compounded by different screw configurations using a twin-screw extruder. To interpret the results of our mechanical tests, the fiber geometry, dispersion state, and fiber fracture surfaces after tensile testing were analyzed. We found that the composites made of short fiber pellet (which suffer high compound intensity), exhibited optimal mechanical performance. Although, compounding with a twin-screw extruder decreased the overall aspect ratio of residual fibers, we observed that it significantly facilitated both the dispersion of the jute yarn to jute bundle and the decohesion of jute bundle to elementary fibers. This fiber separation caused by high intensity mixing led to efficient load transfer from matrix to fiber, and improvement of interfacial strength. These findings provide us with an insight into the critical parameters required to develop a high performing jute/PLA composite.

1. Introduction

In recent years, the use of natural products has increased due to its low environmental impact. For example, the energy production of natural fibers such as jute, flax, and hemp is significantly lower than that of conventional reinforcement fiber including glass and carbon. The growth of natural fiber has also contributed to the reduction of carbon dioxide in the atmosphere. In addition, the density of natural fibers (1.3–1.5 g/cm^3) is much lower than that of glass (2.5 g/cm^3). In fact, natural fiber reinforced composites have generated wide interest in various engineering fields, particularly automotive applications, owing to their high specific strength, durability, lightweight (high modulus/density ratio) and low cost production [1].

Systems including polypropylene (PP) and natural fibers are commonly used for such purposes owing to their increased processability and cost performance. Recently, the use of biodegradable materials such as polylactic acid (PLA) has gradually increased due to their apparent ease of disposal [2]. Despite this, the drawbacks of these natural fiber reinforced green-composites include their cost and poor mechanical properties in comparison to composites constructed of glass fibers (GF) and PP [3–5]. Therefore, to extend the use of fully bio-based composites, higher mechanical performance is required.

GF/PP composites are widely used as secondary structures and their mechanical properties are dominated by the length of the fiber [6–9]. The high aspect ratio of these fibers after processing leads to higher tensile and impact strengths. This is achieved through the use of long fiber pellets and appropriate injection techniques which result in fiber lengths of >5 mm, exhibiting tensile strengths of ~100 MPa [10]. The glass fibers can reinforce the matrix efficiently.

In contrast, it is difficult to reinforce the matrix using natural fibers, as they possess hydrophobic surfaces and anisotropic internal structures. To address these issues, the surface state of natural fibers is typically modified by alkalization and/or silanization treatments [11–16]. Use of maleic anhydride-grafted polypropylene (MAPP) as a coupling agent can further improve the interfacial shear strength (IFSS) between a natural fiber and its matrix [17,18]. Earlier studies carried out by Thomason presume that the lower reinforcement efficiency of natural fiber is due to its anisotropic feature [19]. Here, the compression residual stress, which

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primarily affects the IFSS [20], is lower for natural fibers compared to GF, owing to a low transverse and shear modulus.

It is clear, that the structure of natural fiber is significantly different from its glass counterpart. The natural fiber is composed of cellulose as the reinforcement (elementary fibers) and amorphous polymers as an adhesive. This dominates the mechanical properties, by aligning the cellulose fibrils next to the fibers' axis. For instance, jute fiber bundle (diameter = 25–50 μm), which has a lower orientation angle than other natural fibers, displays a high modulus but is brittle in nature [19]. The bundles are produced through a spinning process and supplied in the form of a twisted yarn. In contrast, glass fibers are supplied as bundles using a sizing agent. The fiber structure and product form of natural fiber is completely different from glass fiber. Recently, long fiber pellet of natural fiber is developed, similarly to glass fiber [21–23]. However, the validity of using this technique for natural fiber composite has not been confirmed yet.

Previous studies on tensile strength of natural fiber/PLA composites are summarized in Table 1. Compression molding can fabricate natural fiber/PLA composites without damaging natural fiber during processing [24–27]. Lim et al. examined the effects of fiber loading and surface treatment on the strength of hemp/PLA composite made by compression molding. They found that the composites with 40% volume fraction of alkali treated fiber showed the best tensile strength. The composites showed much higher strength compared to neat PLA. The best data of PLA/natural fiber composites was reported by Plackett et al. [26]. In this study, non-woven jute fiber mats were stacked with several PLA films and subjected to compression molding. The key parameter in their study was the temperature to impregnate the PLA matrix to jute mat without degrading the matrix and fiber at a constant high temperature. As no fiber damage was observed during this process, composites with significantly high strengths could be constructed. However, compression molding technique is time consuming and unsuitable for mass production.

The preferred manufacturing method for mass products is typically injection molding. A major advantage of this technique lies in its high processability, although the fiber is known to degrade during the procedure. Previous work by Yang et al. has focused on the development of long jute fiber pellets and fabrication through injection molding [21]. Upon addition of the jute fiber, they observed an increase in the material modulus; however, the tensile strength was found to decrease. Bax et al. used an injection molding technique to fabricate Cordenka/PLA and flax/PLA composites, and remarkably achieved a strength of 58 MPa for the former composite [28], with a maximum impact strength of 72 kJ/m^2 (4.5 times higher than what has been observed for pure PLA). They concluded that the addition of Cordenka fiber was capable of modifying the brittleness and low impact properties of PLA. However, the most favorable results have been reported by Sawpan et al. [14], where the authors noted an exceptionally high strength value of 75.5 MPa (148%) for PLA combined with 30 wt.% hemp fibers, compared to pure PLA = 51 MPa. By conducting alkali and silane fiber treatments, they found that the alkali treated reinforced fiber composites displayed improved mechanical properties, owing to the exceptional fiber/matrix adhesion. The jute/PLA composites reported by Bledzki et al. and Gunning et al. also illustrated high mechanical performance [29,30]. Gunning et al. investigated the effective process condition for PLA based green-composites via twin-screw extrusion, and found that the composite produced using lower temperature profiles exhibited superior properties to those produced at higher temperature, owing to reduce thermal degradation of natural fiber during compounding. They fabricated PLA based composite using jute, hemp and lyocell fiber with various fiber loading. In their experiment, jute/PLA composite with 50 wt.% fiber loading showed the best mechanical properties. The tensile strength of jute/PLA afforded 82.2 MPa, which was the maximum value, reported for injection molded samples. Although the average residual fiber length was short (approximately 1 mm), due to the high compounding intensity applied by a twin-screw extruder, the composites exhibited enhanced mechanical properties than what has been reported to date.

The purpose of this study is to develop PLA based green-composite materials that exhibit superior mechanical properties. Particular emphasis is placed on the material processing steps. By employing a continuous pultrusion technique, we successfully demonstrated the fabrication of natural long fiber pellets (LFPs). To further investigate the characteristics of these LFPs, we compared our results with a short fiber pellet (SFP), constructed using a twin-screw extruder. Both composites were fabricated by a method known as injection molding. Tensile and Izod impact tests were also performed to measure the overall strength. Furthermore, microscopy techniques were employed to study both the fiber size and distribution after processing, in order to validate the results against their mechanical properties.

2. Experimental

2.1. Materials

The TE-8000 PLA, supplied from Unitika Ltd. was used in this study. A twisted jute yarn (Tex 400) was purchased from Tesac Co. In order to suppress the hydrolysis during processing, LA-1 (Nisshinbo Chemical Inc.) was added to PLA. In addition, the crystallizing agent, Ecopromote (Nissan Chemical Industries), was used to increase the crystallinity of PLA composites. The weight ratio of jute, PLA, LA-1 and crystallization agent was set to 50, 46, 2 and 2 wt.%, respectively.

2.2. Composite processing

LFP (length = 6 mm) was fabricated using a standard long pellet instrument (Kobe Steel, Ltd.) as reported previously [21,23]. All of

Table 1
Comparison of the mechanical properties of PLA based green composite fabricated by various process.

Fiber	Fiber content (wt.%)	Process	Stress (MPa)	Strength in %	Young's modulus (GPa)	Additional information	Source
Flax	30	CM	53	106	8.3	Using extruded pellet	[25]
Hemp	40 (volume)	CM	54.6	156	8.45	Film stacking + surface treatment	[24]
Ramie	30 (volume)	CM	66.8	148	–	Compound pellet + surface treatment	[12]
Jute	40	CM	100.5	182	9.4	Film stacking method	[26]
Jute	35–39	IM	49	80	10.5	Using long fiber pellet	[21]
Cordenka	30	IM	58	130	4.85	Using compound pellet	[28]
Jute	30	IM	81.9	129	9.6	Using compound pellet	[29]
Jute	50	IM	82.2	121	–	Optimal compound process	[30]
Hemp	30	IM	75.5	148	8.18	Surface treatment	[14]

CM: Compression molding, IM: Injection molding.