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Composites Science and Technology 66 (2006) 952–963

COMPOSITES SCIENCE AND TECHNOLOGY

www.elsevier.com/locate/compscitech

Jute/polypropylene composites I. Effect of matrix modification

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Received 16 November 2004; received in revised form 15 August 2005; accepted 20 August 2005
Available online 4 October 2005

Abstract

Natural fibre-reinforced polymers can exhibit very different mechanical performances and environmental aging resistances depending on their interphase properties, but most studies have been focused on fibre surface treatment. Here, investigations of the effect of maleic anhydride grafted polypropylene (MAHgPP) coupling agents on the properties of jute fibre/polypropylene (PP) composites have been considered with two kinds of matrices (PP1 and PP2). Both mechanical behaviour of random short fibre composites and micro-mechanical properties of single fibre model composites were examined. Taking into account interfacial properties, a modified rule of mixture (ROM) theory is formulated which fits well to the experimental results. The addition of 2 wt% MAHgPP to polypropylene matrices can significantly improve the adhesion strength with jute fibres and in turn the mechanical properties of composites. We found that the intrinsic tensile properties of jute fibre are proportional to the fibre's cross-sectional area, which is associated with its perfect circle shape, suggesting the jute fibre's special statistical tensile properties. We also characterised the hydrophilic character of natural fibres and, moreover, humidity environmental aging effects. The theoretical results are found to coincide fairly well with the experimental data and the major reason of composite tensile strength increase in humidity aging conditions can be attributed to both improved polymer–matrix and interfacial adhesion strength.

Keywords: A. Polymer–matrix composites; B. Interface, strength, mechanical behaviour; D. Micro-mechanical tests; E. Injection moulding

1. Introduction

In recent years, the use of natural fibres as reinforcement is increasingly replacing the conventional inorganic fibres in polymer matrix composites. Especially, natural fibre-reinforced thermoplastics have a good potential in the future as a substitute for wood-based material in many applications. The development of environment-friendly 'green' materials is because of natural fibre's biodegradability, light weight, low cost, high specific strength compared to glass and carbon, recycling and renewing natural sources of plants, such as bast, seed, leaf, wood and fruit. The jute fibre is an important bast fibre and comprises bundled ultimate cells, each containing spirally oriented microfibrils bound together, which has similar structures of other natural fibres like hemp, flax, sisal, etc. The primary component of the fibre is cellulose, which is a linear condensation polymer consisting of D-anhydro-glucopyranose units joined together by β-1,4-glucosidic bonds [1]. The major part of the cellulose consists of a micro-crystalline structure with high order of crystalline regions. Generally, higher cellulose content leads to higher stiffness, in turn the cellulose content and microfibril angle have a major influence on the mechanical properties of the resultant composites. Other components of the jute fibre are hemi-cellulose, lignin, pectin, waxy and water soluble substances. Because of the structural features, the high level of moisture absorption and poor wettability of the natural fibre material results in insufficient adhesion between fibres and polymer matrices leading to debonding during use and aging [2,3], which continue to be a topic of current research.

Previously, selective removal of non-cellulosic compounds constitutes the main objective of the chemical treatments of natural fibres to improve the performance

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0266-3538/$ - see front matter
doi:10.1016/j.compscitech.2005.08.009

of fibre-reinforced composites. Various fibre surface treatments have been reported, such as: alkali [4–8], silane [9–13], combination of alkali and silane [14,15], monomer grafting under UV radiation [16–18], maleic anhydride grafted polypropylene [19–22], and others [23–28]. Among those commercial coupling agent maleic anhydride grafted polypropylene (MAHgPP) has been found to be the most efficient in improving interfacial adhesion of natural fibres and a PP matrix [29–38]. Kamaker et al. [39–41] reported that using 3 wt% MAHgPP (type G-3002, with an average molecular weight of 40,000 and containing 6 wt% of maleic anhydride, from Eastman Chemical products, Kingston, TN) as coupling agent in jute/PP composite increases composite mechanical properties. The tensile strength is doubled from 29.82 to 59.13 MPa and the bending strength increases from 49.97 to 87.66 MPa in composite with 50 wt% fibre content. Similarly, Gassan and Bledzki [19,20] showed that tensile, flexural and dynamic strength increase up to 50% but impact energy decreases due to the lower energy absorption in the interface of jute/PP composite when jute fibres are treated by 0.1 wt% MAHgPP solution in toluene for 5 min at 100 °C. In a comparable system, the tensile strength increases from 40 to 69 MPa for a viscose/PP composite using 6 wt% Exxelor PO1015 as a coupling agent [42]. Besides the fibre tensile strength, moisture absorption of fibre plays a very important role in the reinforcement. During service, composites are subjected to a variety of environmental conditions, which can influence the mechanical properties. One major issue caused by aging during use is moisture. The rate and total amount of moisture absorbed by a composite depends on fibre type, matrix, fibre/matrix interface, temperature and relative humidity. Overall, the MAHgPP's ability to enhance the composite properties depends on many factors, such as type of MAHgPP (graft level, random or block copolymer, molecular weight), miscibility of MAHgPP with the PP matrix, PP grade, composite processing conditions, etc.

Theories for both strength and modulus of composites have been well developed. The rule of mixtures (ROM) concept has been most widely used for strength and modulus predictions although it is not completely adequate for composites containing short, randomly oriented fibres [43]. A large number of research interest was dedicated to theoretical and numerical models with varying degrees of success. However, agreement with the experimental data of randomly oriented short fibre-reinforced composites cannot be accurately predicted with these models, since these models disregard the existence of an interphase [44]. Therefore, an extended approach is developed in our recent work based on the established procedures for getting reliable information on properties of the interphase [45].

The objective of this study is to find out how matrix modification based on MAHgPP affects the interfacial adhesion and mechanical properties of jute/polypropylene composites. Both micro-mechanical methods and mechanical composite testing are used to evaluate the introduced properties changes in two kinds of polypropylenes PP1 (low melt flow rate) and PP2 (high melt flow rate) matrixes. Interfacial fracture morphology is studied by atomic force microscopy (AFM). In addition, single fibre tensile tests, moisture absorption, and fibre length/diameter distribution analysis were also used to assess the specific behaviour of jute fibres in PP-composites.

2. Experimental

2.1. Materials and composite processing

Jute fibre, PP, and coupling agent MAHgPP are used in this study. Jute yarn of 1100 tex yarn fineness was obtained from Spinnerij Blancquaert NV (Lokeren, Belgium). PP1 was supplied by Borealis A/S as commercial grade HD 120 M and PP2 was provided by Basell (Frankfurt, Germany) as commercial grade Purell 570 U. The commercial grade Exxelor PO 1020 (Exxon Mobil Corp., USA) was selected on base of comparison with different commercial MAHgPP grades performed previously [46]. The material properties are listed in Table 1 in terms of densities, average molecular weight, melt flow rate (MFR), melting points, prices and maleic anhydride graft level of MAHgPP.

Composite samples of jute yarn, PP granules, and MAHgPP granules were compounded on a co-rotating twin-screw extruder ZSK 30 (Werner & Pfleiderer, Stuttgart, Germany). Fig. 1 shows the schematic diagram of the screws, mass temperatures in the extruder barrel zones, and dosage of PP, MAHgPP, as well as input of jute yarn into the melt to prevent fibre damage. The extruded strands were cooled by immersion in a water bath and pelletized. From compound granules, the dog-bone shaped specimens ($160 \times 10 \times 4$ mm, according to DIN 53455, specimen No. 3, ISO 527.2) and plates ($80 \times 80 \times 2$ mm) were made using an injection moulding machine Ergotech 100 (Demag Ergotech Wiehe GmbH).

2.2. Fibre property characterisation

2.2.1. Single fibre tensile test

The tensile strength of the filaments is derived from force–displacement curves. This test is in accordance with EN ISO 53812 and ASTM D 1577. The measurement is conducted in accordance with EN ISO 5079 using the Fafegraph ME testing device (Fa. Textechno) equipped with a 10 N force cell after having determined the fineness of each selected fibre using a vibration approach in a Vibromat testing equipment (Fa. Textechno). Assuming a circular cross-section, a relation between the resonant frequency ν and the fineness T_t at known pre-load F_v and gauge length L is defined as

$$T_t = \frac{F_v}{4 \cdot \nu^2 \cdot L^2}. \quad (1)$$

The tensile test is conducted with a cross velocity of 0.5 mm/min using a gauge length of 5–20 mm, at 65% RH and 20 °C temperature. The results of 50 single fibre