Polymer 52 (2011) 180–190

Contents lists available at ScienceDirect

Polymer

journal homepage: www.elsevier.com/locate/polymer

Morphology and mechanical properties of glass fiber reinforced Nylon 6 nanocomposites

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ARTICLE INFO

Article history:
Received 25 August 2010
Received in revised form
27 October 2010
Accepted 30 October 2010
Available online 5 November 2010

Keywords:
Nanocomposite
Glass fibers
Nylon 6

ABSTRACT

Nylon 6 composites containing both an organoclay and glass fibers as fillers were prepared by melt processing. The aspect ratios of the glass fibers and the clay platelets were determined by electron microscopy techniques. The aspect ratio of each type of filler decreased as filler loading increased. A two particle population model for the tensile modulus was constructed based on the Mori–Tanaka composite theory. The experimental levels of reinforcement appear to be reasonably consistent with model predictions when changes in particle aspect ratios are accounted for. The tensile strength increases and elongation at break decreases as the content of either filler increases according to expected trends. Izod impact strength increased with glass fiber content but decreased with clay content.

1. Introduction

Fiber reinforced polymer composites have been widely used for applications requiring high stiffness and strength, e.g., aerospace, automotive, marine, and sporting goods [1–3]. In most cases, rather high loadings of glass fibers are required to achieve the desired performance; this leads to an undesirable increase in specific gravity, decreased melt flow, and increased brittleness [4].

In recent years, polymer nanocomposites have attracted great interest due to the considerable enhancement in stiffness realized at very low filler loadings [5–12]. Substantial improvements in mechanical [13,14], barrier [15,16], thermal [17,18], and flammability [19,20] properties have been reported while maintaining similar density and optical properties to those of the neat polymer matrix. Among these, nanocomposites based on polyamides have received attention due to their excellent compatibility with specific organoclays [13,21–23].

Although polymer nanocomposites reinforced by montmorillonite (MMT) have exhibited improved thermal and mechanical properties at very low filler contents; loadings of more than 10 wt% MMT lead to poor dispersion and processing characteristics [9,10,12]. On the other hand, glass fiber loadings of 30 to 50 wt% are quite common [4]. It is of interest to explore whether combining these two fillers would give the desired performance at low to intermediate filler loadings. Such materials could be viewed as a polymer matrix containing two different types of fillers of rather different size scales. Alternatively, one might view these materials as a nanocomposite matrix filled with glass fibers because the clay platelets are so much smaller than the glass fibers. Although there have been extensive reports on polymer nanocomposites and micro-composites, only a few preliminary studies have been reported on the structure and properties of glass fiber reinforced polymer composites where the nanocomposite plays the role of the matrix as illustrated by the conceptual vision shown in Fig. 1.

Recently, several efforts have been made to investigate the combined effects of fillers at two different size scales, i.e., micro and nano. Akkapeddi [4] reported that the nature of the increase in properties of polyamide 6 composites reinforced by MMT and short glass fibers is not completely additive, but there is clearly an increase over single filler based composites. Similar enhanced mechanical properties are reported for the composites based on polyamide 6 [24–28], and some thermosetting polymers [29–32]. The slight enhancement of tensile properties by two fillers were reported for glass fiber reinforced polypropylene nanocomposites using glass fiber mat [33,34]. More recently, Isitman et al. [35] observed a synergistic flame retardancy effect of organoclays for glass fiber reinforced nylon 6 with conventional flame retardants.

Because of our experience with nylon 6 nanocomposites [13,21,22,36–41] and glass fiber composites [42–46], nylon 6 was chosen as the polymer for this study. The purpose of this work is to explore the morphology and mechanical property changes upon the incorporation of short glass fibers into nylon 6/MMT

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doi:10.1016/j.polymer.2010.10.059

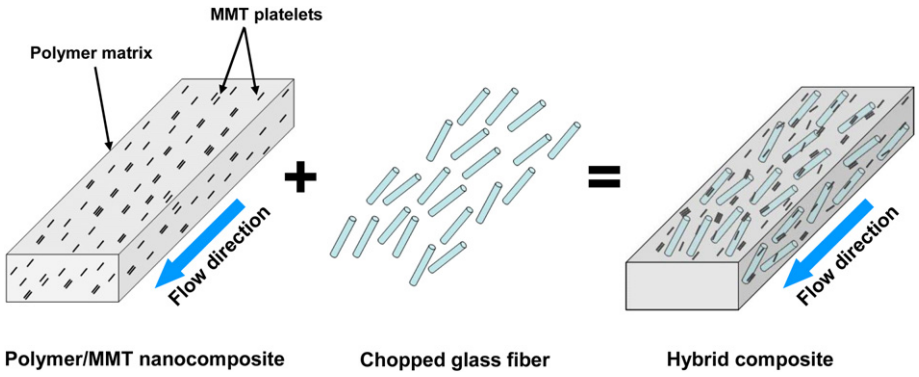

Fig. 1. Hybrid composites based on a polymer/MMT nanocomposite and short glass fibers.

nanocomposites. In other words, this study investigates the reinforcement from two fillers of very different size scales: glass fibers with diameters on the order of 10 μm and MMT platelets with thickness of 1 nm.

2. Experimental section

2.1. Materials

Table 1 summarizes the materials used in this study. A commercial grade of nylon 6 was chosen that is commonly used for injection molding and extrusion applications. The organoclay was formed by cation exchange reaction between sodium montmorillonite and trimethyl hydrogenated-tallow ammonium chloride, designated here as $M_3(HT)_1$, and was obtained from Southern Clay Products. The data below are reported in terms of weight percent montmorillonite, wt% MMT, in the composite rather than the amount of organoclay, since the silicate is the reinforcing component. The chopped strand glass fibers (fiber diameter = 13 μm, length = 4 mm) with a proprietary surface treatment intended for use in polyamide matrices was provided by Owens Corning. Prior to melt processing, all materials employed were dried for a minimum of 24 h in a vacuum oven at 80 °C.

Table 1
Materials used in this study.

Material (designation in this paper)	Commercial designation	Specifications	Supplier
Polymer			
Nylon 6[a]	Aegis® H73WP	$[COOH]/[NH_2] = 0.9$ $T_g = 55$ °C, $\overline{M}_n = 22{,}000$ Brabender torque = 6.37 N·m[b]	Honeywell
Fillers			
Organoclay	$M_3(HT)_1$,[c] Trimethyl hydrogenated-tallow ammonium montmorillonite	Organic loading = 95 mequiv/100 g clay Organic content = 29.6 wt % d_{001} spacing = 18.0 Å[d]	Southern clay products
Glass fiber	CS 995-13P, 4 mm	Chopped strand glass fiber with unknown sizing[e] $D = 13$ μm $L = 4$ mm	Owens Corning

[a] Referred to as a medium molecular weight grade in prior studies from this laboratory [39,47,48].
[b] Data from reference [48].
[c] The selected organoclay is designated as $M_3(HT)_1$ in this study, where M = methyl and HT = hydrogenated tallow. Tallow is a natural product composed predominantly (63%) of saturated and unsaturated C18 chains. HT is the saturated form yet still contains a small fraction of double bonds.
[d] The basal spacing corresponds to the characteristic Bragg reflection peak d_{001} obtained from a powder WAXS scan of the organoclay.
[e] Chopped strand glass fiber coated with a proprietary sizing intended for use in polyamide matrices.

2.2. Melt processing

The nanocomposites of nylon 6 and the organoclay were made in a Haake co-rotating, intermeshing twin-screw extruder (diameter = 30 mm, $L/D = 10$) operating at a barrel temperature of 235–240 °C, a feed rate of ~1 kg/h, and a screw speed of 280 rpm. Extruded nanocomposite pellets were dry mixed with various amounts of glass fiber and injection molded to prepare standard 3.18 mm thick tensile (ASTM D638) and Izod bars (ASTM D256) using an Arburg All-rounder 305-210-700 injection molding machine with an injection pressure of 70 bar and a holding pressure of 35 bar. The temperature of the feed zone of the injection molder was set at 240 °C and was ramped up to 270 °C at the nozzle. The mold temperature was held at 80 °C. After molding, the specimens were immediately sealed in a polyethylene bag and placed in a vacuum desiccator for a minimum of 24 h prior to mechanical testing.

In the early stages of this work, another processing procedure was considered where the nanocomposite was formed as described above, but the glass fibers were compounded into the nanocomposite using a single screw extruder. This single screw extruder causes far less fiber breakage than would have been the case if the glass fibers were compounded into the material using the twin screw extruder. The resulting glass fiber reinforced nanocomposite was injection molded into test bars. However, the resulting mechanical properties obtained by this method were, within the margin of error, the same as obtained in the first described method. Thus, this simpler processing method was used for the remainder of this work because it facilitated sample preparation and minimized, but did not eliminate, glass fiber attrition.

2.3. Morphology characterization

Fracture surfaces of the composites were examined with a LEO 1530 Scanning Electron Microscope (SEM) operating at an accelerating voltage of 10 kV. SEM specimens were sputter-coated with gold prior to observation. For analysis of the glass fiber length, the polymer was burned off from the injection molded samples in a furnace at 500 °C for 4 h to isolate the short glass fibers. The glass fibers obtained were dispersed in water with a small amount of detergent to reduce surface tension. Using a pipette, a small amount of solution with fibers was placed on a glass slide and then observed by SEM. The morphology of MMT particles was