MELT RHEOLOGICAL PROPERTIES OF NATURAL FIBER-REINFORCED POLYPROPYLENE

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Abstract

The melt viscosities and mechanical properties of 3 different natural fiber-polypropylene composites were investigated. Coir (coconut), jute, and kenaf fibers were compounded with polypropylene at 30% by weight content. A capillary rheometer was used to evaluate melt viscosity. The power-law model parameters are reported over a shear rate range between 100 to 1000 s^{-1}. Effects on melt viscosity with the use of a coupling agent and different fiber types were also evaluated.

Introduction

The use of natural fiber-reinforced thermoplastic composites is gaining popularity in automotive, cosmetic, and plastic lumber applications [1-3], Natural fibers offer economical and environmental advantages over traditional inorganic reinforcements and fillers. Economical benefits include lower cost than most glass fibers and mineral fillers and lower processing temperatures. Environmental advantages include being derived from annual growth renewable resources and by reduction of petroleum based products.

The improvement in mechanical properties with the addition of natural fibers is well documented in literature [4-7]. The papers present a wide ranger of fibers, matrix materials, compatibilzers, and compounding techniques. The investigation of melt rheological properties of natural fiber-reinforced thermoplastic is scarce in literature [8-11]. Work has been done to determine the effects of fiber content and melt temperature on viscosity of natural fiber-reinforced polypropylene [8,9]. Understanding the melt viscosity aids engineers in the design of extrusion dies and injection molds by being able to calculate flow rates and lengths, pressure drops across restrictions, and shear stresses.

In this study, coir, jute, and kenaf fibers were compounded with polypropylene. The melt viscosity was evaluated as a function of shear rate at 170°C. The viscosity dependence on shear rate was evaluated with the power law viscosity model. The effects of coupling agent and fiber type on the power-law parameters were investigated. A table of mechanical properties is provided as reference material.

Experimental

Materials

The polypropylene (PP) used was a 12 melt flow index (MFI), general-purpose homopolymer suitable for injection molding and extrusion. Coupling agent used was a high molecular weight, maleic anhydride-grafted-polypropylene (MA-g-PP). The coir (coconut), jute, and kenaf fibers were supplied by various industrial sources. The fibers were granulated through an 8.0-mm screen to increase their bulk density.

Compounding

The fibers were dried at 90°C for 24+ hr in a steam-heated oven to expel moisture. The fibers, polypropylene, and MA-g-PP were dry-blended prior to compounding. The fiber content of the composite was 30%, by weight. Polypropylene content was 68%. The coupling agent was added at 2%. A jute composite was prepared with no addition of a coupling agent with 30% fiber content and 70% PP. Table 1 lists all composites formulations.

The composites were compounded on a 32-mm, intermeshing, co-rotating twin-screw extruder. Screw speeds were between 170 and 200 rpm. Melt temperature for all compounding was maintained between 188 and 193°C. Output ranged from 10 to 15 kg/hr with melt pressures between 1000 and 1700 kPa. The extrudates were water cooled and pelletized.

Viscosity Measurements

A capillary rheometer attached to a 19-mm, single screw extruder was used to measure melt viscosity. The capillaries measured 2-mm in diameter, with lengths of 20- and 30-mm (L/D=10 & 20). The viscosity was measured at apparent shear rates ($\dot{\gamma}_{aw}$) between 100 and 750 s^{-1}. The capillaries for apparent shear rates between 10 and 75 s^{-1} were 3-mm in diameter with L/D's equal to 10 and 15. The metering section and die temperatures of the extruder were maintained at 170°C. The mass output ($\dot{m}$) and pressure drop (ΔP) across the die was measured to determine the apparent shear rates and shear stresses, respectively. Melt flow index (MFI) tests were performed

according to the standard ASTM D1238. The tests were performed at 190°C to prevent fiber degradation.

Results and Discussion

Figure 1 shows the relationship between shear stress (τ_w) and apparent shear rate ($\dot{\gamma}_{aw}$) for pure polypropylene (100 PP) and 30% Jute-70% PP (30 Jute-0) at 170°C.

The shear stress (τ_w) at the wall of the capillary was calculated using [12]:

$$\tau_w = \frac{R\Delta P}{2L} \qquad (1)$$

where L is the length and R is the radius of the capillary. The pressure drop, DP, was measure across the die length.

The apparent shear rate ($\dot{\gamma}_{aw}$) was calculated as [12]:

$$\dot{\gamma}_{aw} = \frac{4Q}{\pi R^3} \qquad (2)$$

where Q is the volumetric flow rate and R is the radius of the capillary.

To calculate the shear rate ($\dot{\gamma}_w$) at the wall, the Rabinowitsch correction [12] was applied to the apparent shear ($\dot{\gamma}_{aw}$) rate values. Shear stresses were corrected using the Bagley plot technique to compensate for die entrance effects caused by flow contraction [12]. Viscosity is calculated as [12]:

$$\eta = \frac{\tau_w}{\dot{\gamma}_w} \qquad (3)$$

Figure 2 illustrates the shear rate dependence on viscosity of molten polypropylene and jute fiber-reinforced PP composites. The observed linearity between viscosity (η) and shear rate ($\dot{\gamma}_w$) on a log-log scale is described as a power-law model. Following the form [12]:

$$\eta = m\dot{\gamma}_w^{n-1} \qquad (4)$$

where n is the power-law index and m is the consistency index, with units of Pa-s^n.

Low Shear Rate Viscosity

The power law can accurately represent the shear-thinning behavior of viscosity with respect to shear rate [13]. However, molten polypropylenes exhibit a Newtonian plateau, which is not taken into account with the power-law model. The power law model over predicts the viscosity at low shear rates. Figure 3 illustrates this point with viscosity-shear rate plots of pure polypropylene and jute composite between shear rates of $10-1000\ s^{-1}$. An alternative model that fits the entire curve is the Carreau model. This model incorporates 4 parameters and is solved numerically.

Effect of MA-g-PP on Melt Viscosity

Maleic anhydride-grafted-polypropylene (MA-g-PP) is used as a compatibilizer to chemically bond the hydrophilic fibers to the hydrophobic polypropylene matrix. Sanadi et al. [4] reported that small amounts, 2%-3%, of MA-g-PP added to the composite significantly increases the mechanical properties. The improved bonding promotes more efficient stress transfer from the matrix to the fibers. A 40% increase in tensile strength was observed in the jute composites by adding 2% MA-g-PP to the formulation. This is illustrated in the tensile stress-strain response of the composites in Fig. 4.

Li et al. [14] studied the effects of MA-g-PP on the melt viscosity of polypropylene. An approximate 15% reduction in melt viscosity was reported for an addition of 5% of MA-g-PP at 200°C. The reduction in viscosity continued to increase with increasing MA-g-PP content. Also reported was the power-law index decreased changed slightly with the addition of 5% MA-g-PP. Figure 5 shows the effect of MA-g-PP on the melt viscosity of the 30 Jute-2 compared to 30 Jute-0. No significant effects on viscosity were observed, contrary to Li et a1 [14].

Fiber Influence on Melt Viscosity

Shown in Fig. 6 is the power law region for the coir, jute, and kenaf composites. Table 2 lists the power-law parameters for the composites. Both jute composites exhibited a large increase in the consistency index (m) indicating the greatest increase in viscosity of all composites. Coir fibers influenced the power law parameters the least. The changes in the power-law index (n) values between the composites may indicate a greater difference in low shear rate viscosity. These trends are supported by the melt flow indices (MFI) of the composites, presented in Table 2.

Future Work

Work is underway to evaluate the rheological properties of two additional natural fibers. Flax fibers, used as reinforcement for automotive plastics [15], and wood flour, a natural filler used to reduce cost, have been selected. Low shear rate ($\dot{\gamma}_w$) viscosities are being measured for all composites. The shear rate dependence of viscosity will be fitted to the Carreau Model to compensate for the Newtonian plateau, which the power-law model over predicts. Melt viscosities at 160°C and 180°C will be measured. This information will allow for the Carreau-Arrhenius model constants to be calculated to determine temperature dependence on viscosity.

Effect of fiber type on die swell is to be analyzed. Fiber damage as a function of shear rates and shear stresses is being investigated. Lastly, the thermal properties of a few selected natural fiber-polypropylene composites are to be determine so the presented information may be used in computer modeling and simulation of polymer processing.

Conclusions

Coir (coconut), jute, and kenaf fibers were compounded with polypropylene at 30% by weight. The melt viscosities were evaluated by capillary rheometry and compared to pure polypropylene. The viscosity (η), as a function of shear rate ($\dot{\gamma}_w$), was fitted to the power-law model. However, the power-law model over predicts the viscosity at low shear rates. Comparison of the consistency indices (m) shows that jute had the greatest influence on viscosity, followed by kenaf and coir. Comparison of the power-law indices (n) shows that jute exhibited the greatest amount of shear thinning. Power-law indices ranged from 0.32 for jute to 0.37 for polypropylene. Little effect on viscosity was seen with the use of a maleic anhydride-grafted-polypropylene (MA-g-PP) coupling agent. The benefits of the compatibilizer are seen in the improvements in tensile properties of the jute composites.

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Keywords

Natural fibers, Polypropylene, Viscosity, Power-law model.

Tables

Composite	Fiber[1]	Matrix[1]	Additive[1]
100 PP		100 PP	
30 Coir-2	30 Coir	68 PP	2 MAPP
30 Jute-2	30 Jute	68 PP	2 MAPP
30 Jute-0	30 Jute	70 PP	
30 Kenaf-2	30 Kenaf	68 PP	2 MAPP

[1] % by weight

Composite	m (Pa-s^n)	n (-)	M F I [2] (g/10 min)
100 PP	5918	0.37	5.20
30 Coir-2	8379	0.36	1.78
30 Jute-2	11497	0.32	1.54
30 Jute-0	11967	0.32	1.18
30 Kenaf-2	10049	0.34	1.71

[2]Test performed at 190°C

Table 3: Selected Mechanical Properties of Natural Fiber-Reinforced Polypropylene

Composite	Strength *(MPa)*	Modulus *(GPa)*	Strain *(%)*	Strength *(MPa)*	Modulus *(GPa)*	Strain *(%)*	Un-notched *(J/m)*
100 PP	34.9	1.91	3.66	32.2	1.73	7.41	561
30 Coir-2	54.5	3.17	3.52	39.9	2.67	4.26	104
30 Jute-2	61.3	4.00	3.50	46.9	4.08	3.81	214
30 Jute-0	52.5	4.55	2.83	33.7	5.18	2.51	121
30 Kenaf-2	60.2	3.97	3.43	44.9	4.09	3.35	180

Tensile: ASTM D 638
Flexural: ASTM D 790
Izod Impact: ASTM D 257

Figures

Figure 1: Shear Stress (τ_w) vs. Shear Rate (γ_w) at 170°C for 100 PP and 30 Jute-0

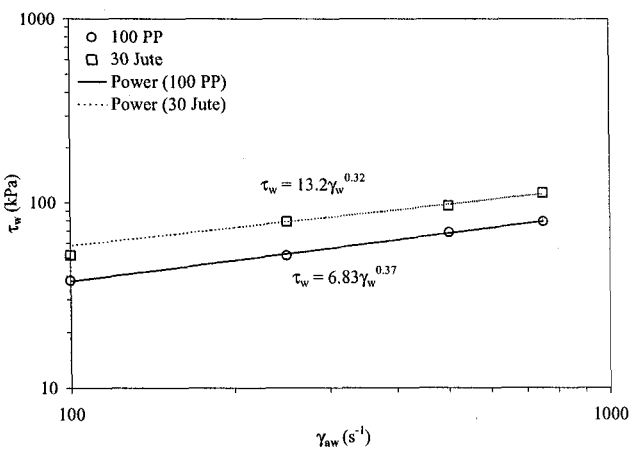


Figure 2: Viscosity (η) vs. Corrected Shear Rate (γ_w) for 100 PP and 30 Jute-0

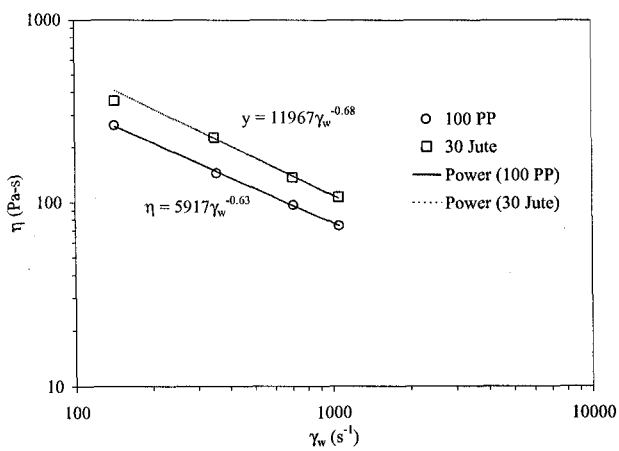


Figure 3: Power-Law Model over Predicting Low Shear Rate Viscosity for 100 PP and 30 Jute-0

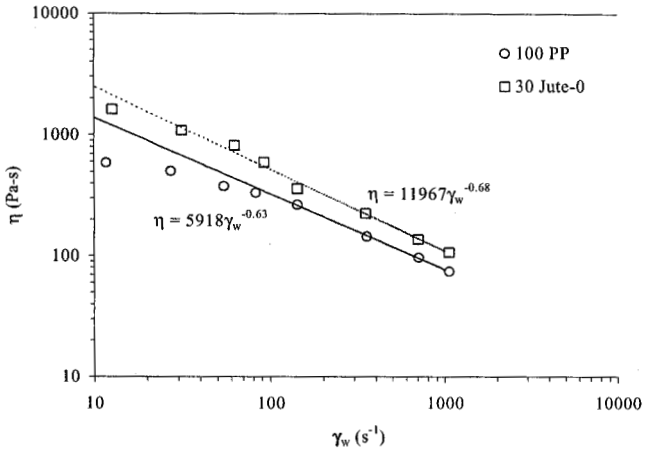


Figure 4: Effects of MA-g-PP on the Tensile Stress-Strain Response of 30% Jute-Polypropylene.

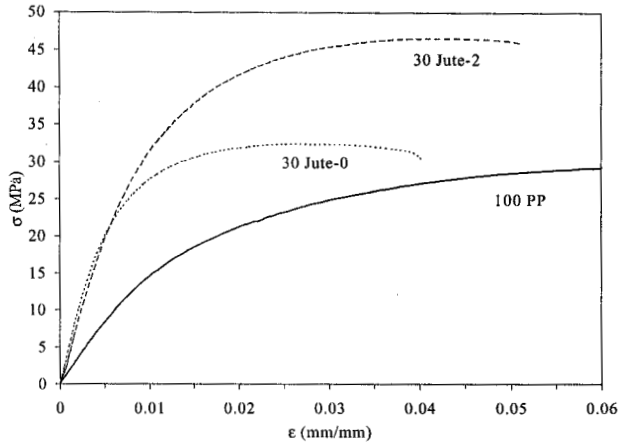


Figure 5: Effects of MA-g-PP on the Melt Viscosity of 30 Jute-Polypropylene

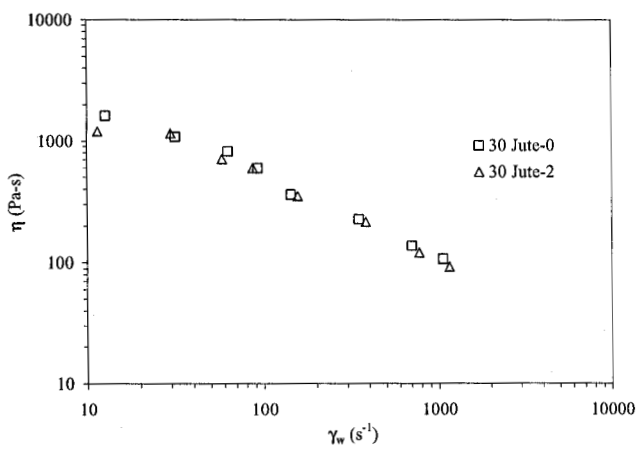
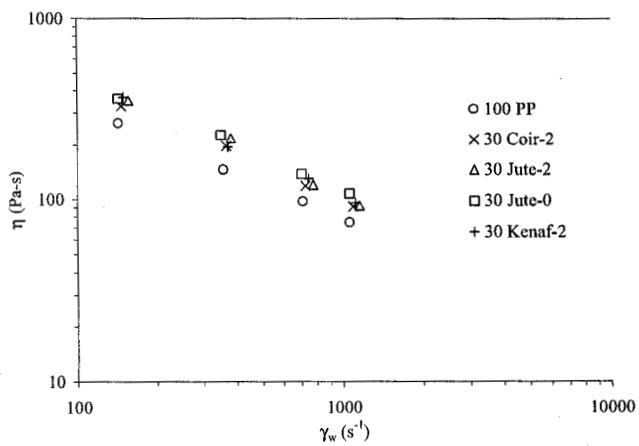


Figure 6: Effect of Fiber Type on Melt Viscosity for the Shear –Thinning Region



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