Static and Dynamic Mechanical Properties of a Kenaf Fiber–Wood Flour/Polypropylene Hybrid Composite

Mehdi Tajvidi

Department of Wood and Paper Science and Technology, College of Natural Resources, University of Tehran, P.O. Box 31585-4314, Karaj, Iran

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ABSTRACT: A natural fiber hybrid composite containing equal proportions of kenaf fibers (KFs) and wood flour (WF) as the reinforcements and polypropylene (PP) as the polymer matrix was prepared, and its static and dynamic mechanical properties were compared with KF/PP and WF/PP composites. Static tensile and flexural tests and dynamic mechanical analysis (DMA) were carried out. The hybrid composite exhibited tensile and flexural moduli and strength values closer to those of the KF composite, which indicated a higher reinforcing efficiency of KFs compared with WF. DMA revealed that although the glass-transition temperature remained unchanged by the replacement of half of the WF by KFs, the α-transition temperature of the hybrid composite was identical to that of WF composite. The magnitudes of both the α and β (glass) transitions of the hybrid composite were comparable to that of the WF/PP composite. © 2005 Wiley Periodicals, Inc. J Appl Polym Sci 98: 665–672, 2005

Key words: composites; fibers; poly(propylene); mechanical properties; transitions

INTRODUCTION

Natural fiber thermoplastic composites are finding applications in many industries and have consequently been of great interest to many researchers and research laboratories in recent years. Kenaf fiber (KF), wood flour (WF), rice hulls, newsprint, pulp fibers, and cellulose fibers are the main natural fibers that have been used as reinforcements. Polyethylene (high and low density), polypropylene (PP), and poly(vinyl chloride) are also often used as the polymer matrix.[1] A lower density, a higher specific strength and modulus, relative nonabrasiveness, an ease of fiber surface modification, wide availability, a lower price, and renewability are among the main advantages of natural fibers over synthetic fibers. On the other hand, the main disadvantages of natural fibers in composites are their lower allowable processing temperatures, incompatibility between the hydrophilic natural fibers and hydrophobic polymers, and the potential moisture absorption of the fibers and, in turn, the manufactured composite.[2,3]

A *hybrid composite* is generally defined as a composite with more than one reinforcing phase. Although the production of hybrid composites of glass fiber/wollastonite and engineering thermoplastics has commercially been of interest, hybrid composites of natural fiber/thermoplastic composites have not been studied extensively, and the bulk of existing studies have focused on the hybrid composites of one natural and one nonnatural fiber.[4] Jacobson and Caulfield[5] reported the results of a study on a cellulose fiber–wollastonite nylon 6 composite, where their aim was the development of a technique to replace glass fibers with lower density cellulose fibers in such composites.

Natural reinforcements in the form of fibers (e.g., KFs) with a relatively high aspect ratio normally result in better performance in comparison with particulate reinforcements (e.g., WF). Hence, an improvement in the mechanical properties of WF–thermoplastic composites could be expected if long KFs are added to the system. The question then would be whether the hybrid composite produced in this way would behave more like the WF or KF composite. Fu et al.[6] discussed the application of the rule of hybrid mixtures and the laminate analogy approach for the prediction of the modulus of hybrid composites containing short fibers and particulate fillers as reinforcement. The authors reported that the reinforcing efficiency of the fibers played an important role in determining the final composite's mechanical characteristics. The tensile strength of the polyester/hybrid ramie cotton fabric composites was studied by Paiva Júnior et al.[7] The tensile behavior was dominated by the volume fraction of the ramie fibers aligned with the test direction. Mishra et al.[8] reported that the addition of a relatively small amount of glass fiber to the pineapple leaf fiber and sisal fiber-reinforced polyester matrix enhanced

Correspondence to: M. Tajvidi (mtajvidi@ut.ac.ir).

TABLE I
Composition of the Evaluated Formulations[1]

Formulation	Code	Fiber content (wt %)	Resin content (wt %)	Compatibilizer content (wt %)
WF	PP–WF	25	74	1
KF	PP–KF	25	74	1
Hybrid	PP–KF–WF	WF = 12.5 KF = 12.5	74	1

the mechanical properties of the resulting hybrid composites. The resistance of a bamboo-fiber-reinforced PP composite and bamboo/glass-fiber-reinforced PP hybrid composite to hygrothermal aging and their fatigue behavior under cyclic tensile load were studied by Thwe and Liao.[9] A preliminary study on the tensile and flexural properties of PP/oil palm empty fruit bunch–glass fiber hybrid composites was also reported by Rozman et al.[10]

Dynamic mechanical analysis (DMA) or dynamic mechanical thermal analysis is a sensitive technique that characterizes the mechanical responses of materials by monitoring property changes with respect to the temperature and/or frequency of oscillation. The technique separates the dynamic response of materials into two distinct parts: an elastic part [storage modulus (E')] and a viscous or damping component [loss modulus (E'')]. The elastic process describes the energy stored in the system, and the viscous component describes the energy dissipated during the process.[11] I have been unable to find any literature on the dynamic mechanical characterization of hybrid composites consisting of two or more natural fibers as the reinforcement phase.

The objective of this study was to evaluate the mechanical performance and dynamic mechanical properties of a KF–WF/PP hybrid composite.

EXPERIMENTAL

Materials

PP, a Basell Pro-fax PD702 homopolymer (Hoofddorp, the Netherlands) with a melt flow index of 35 g/10 min (230°C, 2.16 Kg) and a density of 0.902 g/cm^3, was used as the polymer matrix in this study. WF (40-mesh maple) was supplied by American Wood Fibers, Inc. (Schofield, WI). KFs were supplied by Kengro Corp. (Charleston, MS). Maleic anhydride PP was UNITE(R) MP and was supplied by Aristech Chemical Corp. (Pittsburgh, PA).

Methods

Composite preparation

Polymer, WF, and the compatibilizer were weighed and bagged according to the various fiber contents indicated in Table I. They were then mixed in the proprietary mixing equipment of Teel Global Resources, Inc. (Baraboo, WI). The compounded materials were then ground with a pilot scale grinder to prepare the granules.

Specimen preparation

The granules of the three composite formulations were injection-molded to produce standard ASTM specimens. Injection molding was performed with a 33-ton Cincinnati Milacron 32-mm reciprocating screw injection molder (Cincinnati, OH) with a length/diameter ratio of 20:1. The mold temperature was 37.8°C, and the barrel and nozzle temperatures were set to 187.8°C. Specimens for DMA testing were cut from the impact specimens with a table saw. They were further machined down to a nominal thickness of 2 mm with a knee-type Bridgeport vertical milling machine (Shelton, CT). A fly cutter with a carbide insert tool was used. Care was taken to obtain the specimens from the same area of the impact specimens. Each side of the specimen was machined to produce a balanced DMA specimen at the desired thickness. The final specimens measured 52 × 8 × 2 mm.

Conditioning

After preparation, all specimens were conditioned in a humidity-controlled room at 23°C and 50% relative humidity for at least 40 h before the tests were performed.

Tensile testing

Tensile tests were performed according to ASTM D 638M-89. A type M-I dog-bone specimen was selected. Tensile tests were performed with an Instron 5566 testing machine (Norwood, MA) with computer data acquisition system and a load cell capacity of 908 kgf. The crosshead speed was 5.08 mm/min, and strain measurements were performed with an MTS strain gauge (Eden Prairie, MN) mounted on the samples. The gauge length was 2.54 cm.

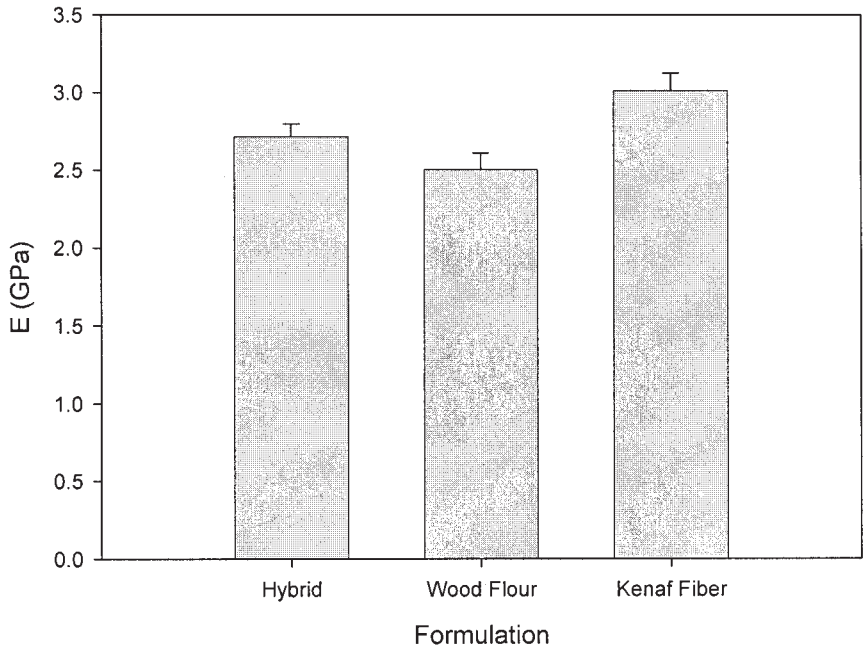

Figure 1 Tensile moduli of various composites.

Three-point bending

Bending tests were carried out on the specimens according to ASTM D 790-90, test method I. The specimens' nominal dimensions were 130 × 13 × 3.2 mm. The span was 100 mm, which resulted in a span-to-depth ratio of 32. The crosshead speed was 5.08 mm/min. Three-point bending tests were carried out with an Instron 5544 testing machine with a data acquisition system.

DMA

Temperature scans was performed with a Rheometric Scientific DMTA V analyzer (Piscataway, NJ). A dual cantilever mode was selected, and the specimens were scanned over a temperature range of −60 to +120°C. The frequency of the oscillations were fixed at 1 Hz, and the strain amplitude was 0.1%, which was well within the linear viscoelastic region. The heating rate was 2°C/min for all temperature scan tests. E', E'', and the mechanical loss factor (tan δ) were collected during the test and were plotted versus temperature. The curves presented are the average of two runs.

Data analysis

Ten tensile and 10 bending specimens of each formulation were tested, and the results were analyzed in a completely randomized design; Duncan's multiple-range test (DMRT) was performed on the group means. SPSS statistical software was used to run the statistical tests. All comparisons were made at a 95% confidence level.

RESULTS AND DISCUSSION

Figure 1 shows the tensile modulus values of the three composite formulations. The KF composite had the highest modulus, and the WF composite had the lowest value. The modulus value for the hybrid composite fell within these two values. DMRT showed that all three means were significantly different at the 95% confidence limit (Table II). However, the tensile modulus of the hybrid composite was closer in value to that of the WF composite. This was interesting because it is usually expected that fiber-like reinforcements such as KFs have a higher reinforcing effect when used alone in a polymer matrix.[6] In a hybrid composite, the properties of the composite are mainly dependent on the modulus and percentage elongation at break of the individual reinforcing fibers. The modulus of KFs is comparatively higher than that of the WF.[1] Therefore, it seemed that the interaction between

TABLE II
Tensile Modulus Means grouped by DMRT

		Subset for α = .05		
Group	*N*	1	2	3
WF	10	2.5027		
Hybrid	10		2.7166	
KF	10			3.0131

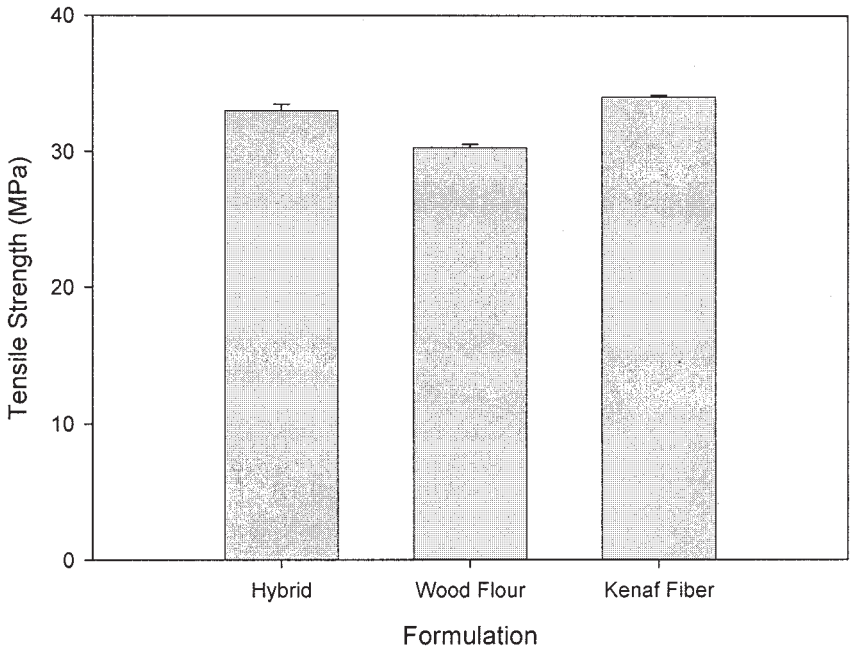

Figure 2 Tensile strengths of various composites.

the two reinforcing agents resulted in more complicated effects than that expected by a simple rule of mixture law.

The tensile strength values of the three formulations are presented in Figure 2, where the same trend as that of the tensile modulus is shown. The replacement of half of the WF by KFs considerably improved the tensile strength of the composite. However, the tensile modulus value of the hybrid composite was close to that of the KF composite (Table III). This was expected because KFs have a higher reinforcing efficiency than wood fibers. This indicated that the presence of KFs improved the stress transfer from the polymer matrix to the fibers so that more stress was borne by the stronger KFs. DMRT showed significant differences between the three composites at the 95% confidence level.

Figure 3 shows the flexural modulus values of the three studied composites. Although DMRT indicated significant differences between all three groups (Table IV) at the 95% confidence limit, the hybrid composite had a flexural modulus that was precisely the average of the other two composites. Similar behavior was observed for tensile modulus, which indicated that the weight fraction of the reinforcement played an important role in determining the moduli of the hybrid composites. In flexural testing, various mechanisms such as tension, compression, and shearing take place simultaneously. In a three-point flexural test, failure occurs due to bending failure and shear failure. Therefore, an increase in the flexural modulus by the addition of KFs indicated an improvement in the various mechanisms involved.

TABLE III
Tensile Strength Means Grouped by DMRT

		Subset for α = .05		
Group	*N*	1	2	3
WF	10	30.2821		
Hybrid	10		33.0033	
KF	10			34.0079

A very interesting behavior was observed when the flexural strength values of the three composites were studied. As shown in Table V, the flexural strength of the hybrid composite was very close in value to that of KF composite so that they were not significantly different from each other. However, the hybrid composite still was between the other two (Fig. 4).

The E' spectra of the composites are presented in Figure 5. A general falling trend was easily observed for all formulations. A clear transition was seen at temperatures around 0°C. This is glass (β) transition, which as it was seen, was the major transition in the composites. Below the glass-transition temperature, the E' values dropped as the temperature increased. In the vicinity of the glass-transition temperature, a very considerable drop was observed, which indicated that the materials were going through a glass/rubber transition. It should be noted that it was difficult to pinpoint a single glass-transition temperature for all formulations because the transition actually occurred over a range of temperature. This is typical in semicrystalline polymers. It is also important to keep in

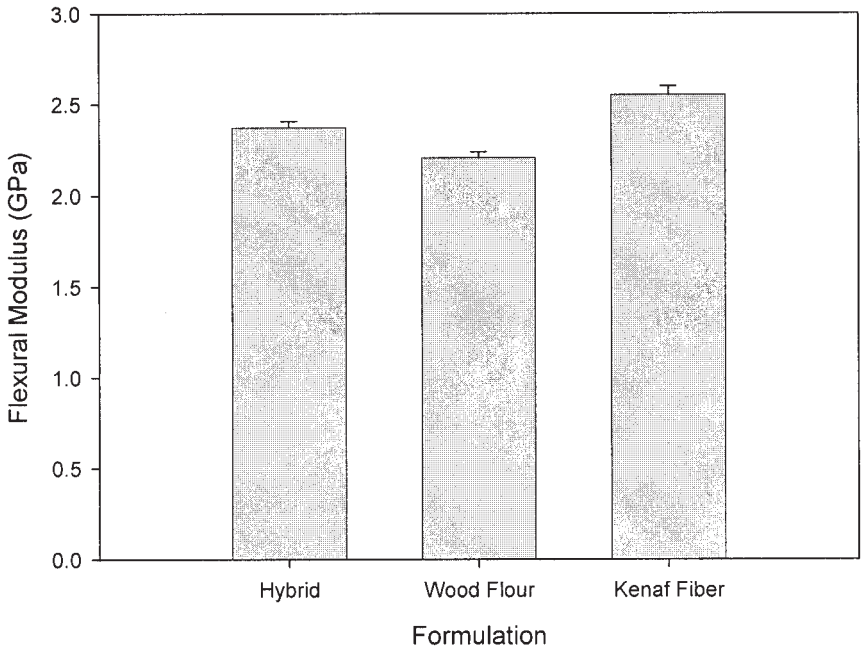

Figure 3 Flexural moduli of various composites.

mind that the glass transition determined from the E' spectrum generally differs from what is determined from E'' spectrum because they actually show two different stages in transition.[12] In the former, the onset of the transition is observed, whereas in the latter the peak point is considered.

As clearly shown in Figure 5, before the glass-transition temperature was reached, the hybrid composite had the lowest E' whereas the curve for the KF composite stayed on top. Just after this point, a different behavior was observed and the hybrid and WF composite curves overlaid for the remainder of the temperature range. This showed that the E' of the hybrid composite was much closer to that of WF composite in the rubbery stage.

Figure 6 shows the loss moduli spectra of the three formulations. Two major transitions were easily observed. The first transition, which was at around 10°C, was the glass transition. As mentioned previously, the glass-transition temperature observed in the E'' curve was about 10°C higher than what was determined from the E' curve. No shifting to higher or lower temperatures was seen in case of the glass transition. Below the onset of the glass transition, the hybrid composite curve was much closer to the KF composite, whereas after this point, it overlaid the WF composite curve. The glass-transition intensity (height of the curve at peak) was identical to the WF curve as well. The second transition was seen in the range 60–80°C. This was the α transition. As shown, in the case of KF composite, this transition was shifted to higher temperatures. Again, the intensity of this transition was higher for the KF composite.

Figure 7 shows the tan δ spectra of the three composites. Below glass-transition temperature, the hybrid composite possessed higher damping, which indicated that more energy was dissipated when two different fibers were present. This parameter is independent of a material's stiffness and, hence, is a very good parameter when the differences in viscoelastic response of the material are sought. At the glass-transition temperature, the KF composite showed a significantly higher tan δ whereas at higher temperatures, the WF composite and the hybrid composite exhibited the highest damping values. Generally, the hybrid composite's damping curve was much more similar to the WF composite in the rubbery stage.

TABLE IV
Flexural Modulus Means Grouped by DMRT

		Subset for α = .05		
Group	*N*	1	2	3
WF	10	2.2060		
Hybrid	10		2.3716	
KF	10			2.5510

TABLE V
Flexural Strength Means Grouped by DMRT

		Subset for α = .05	
Group	*N*	1	2
WF	10	40.1192	
	10		43.4938
KF	10		43.8198

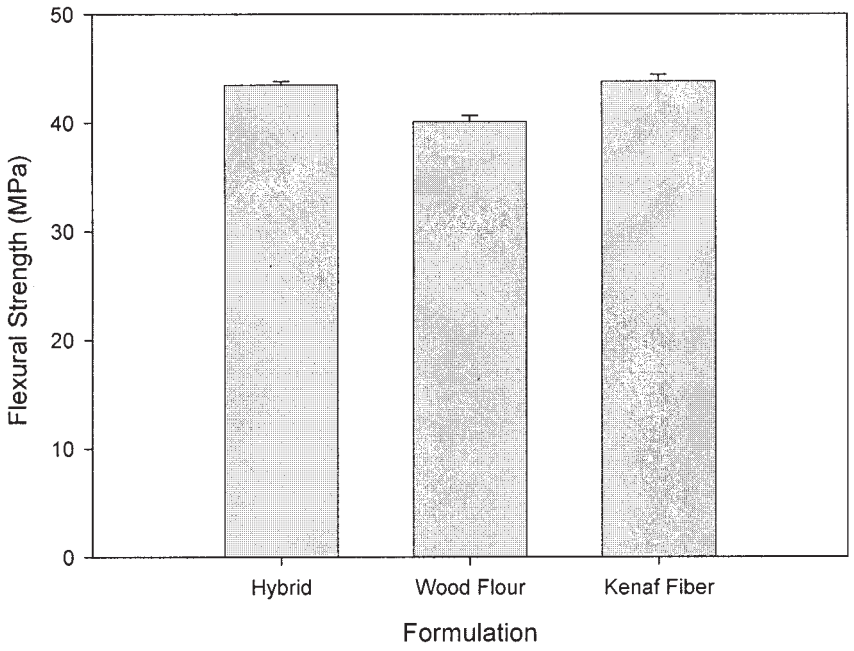

Figure 4 Flexural strengths of various composites.

The β transition, related to the glass–rubbery transition, is due to the molecular motions associated with unrestricted amorphous PP.[12,13] The intensity of the α transition, related to molecular mobility associated with the presence of crystals, was also found to be proportional to the fiber volume fraction.[14] This could explain the higher intensity in case of the KF composites, for these fibers were bulkier and, hence, had a higher volume fraction at a given weight content.

The α transition in semicrystalline polymers is related to the relaxation of restricted amorphous chains in the crystalline phase (defects), also known as *rigid amorphous molecules.*[12,13] The higher intensity of the α transition in the KF composite in comparison with the other two formulations indicated that the number of these defects was higher. Sanadi and Caulfield[14] suggested that the defects that caused the α transition for kenaf–PP composites were predominantly near the

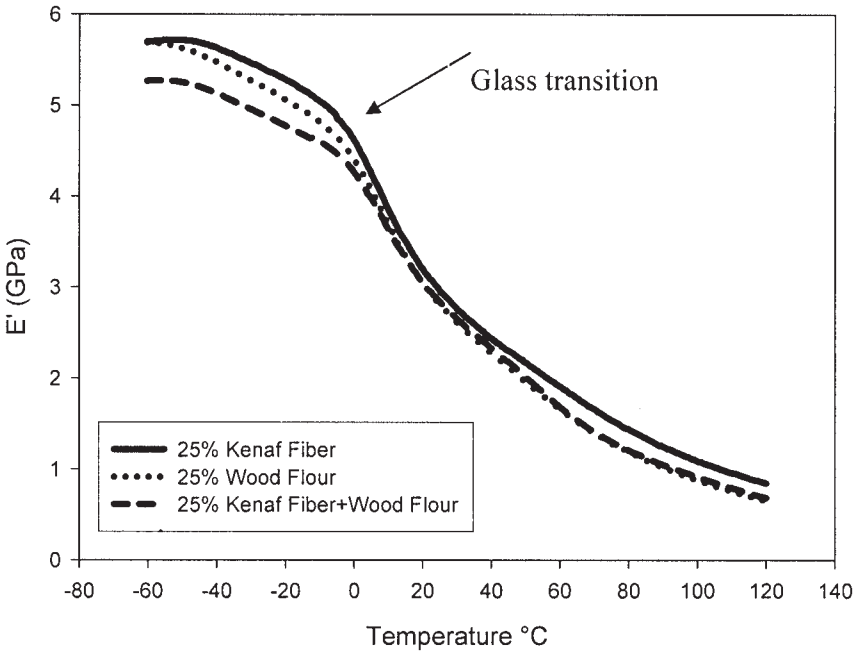

Figure 5 E' spectra of various composites.

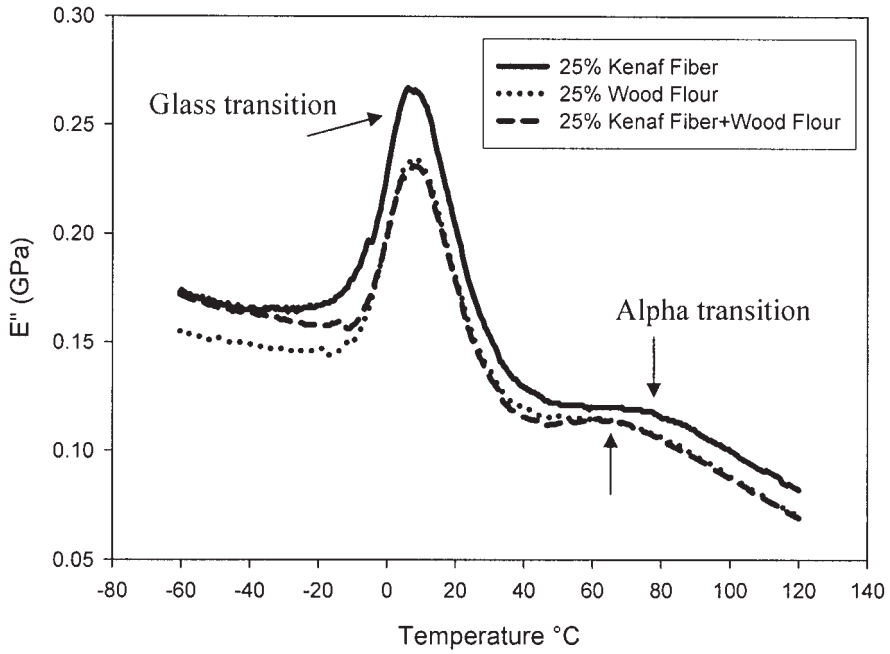

Figure 6 *E″* spectra of various composites.

fiber–matrix interface and existed in the transcrystalline zone. A shift to higher temperatures in the α transition means that the process in which this transition occurs is delayed by the fibers so that more energy is required for the α transition to happen. Therefore, we can conclude that the presence of more fibers restricts the mobility of the chains in the crystalline zone and, hence, shifts the α transition to higher temperatures as seen in the case of the KF composite.

CONCLUSIONS

Static tensile and flexural tests and DMA were carried out to study the mechanical behavior of a WF–KF/PP hybrid composite. Generally, the hybrid composite exhibited properties that were the average of the pure WF and KF composites. However, the flexural strength of the hybrid composite was very close to that of the KF composite. DMA revealed that no change in

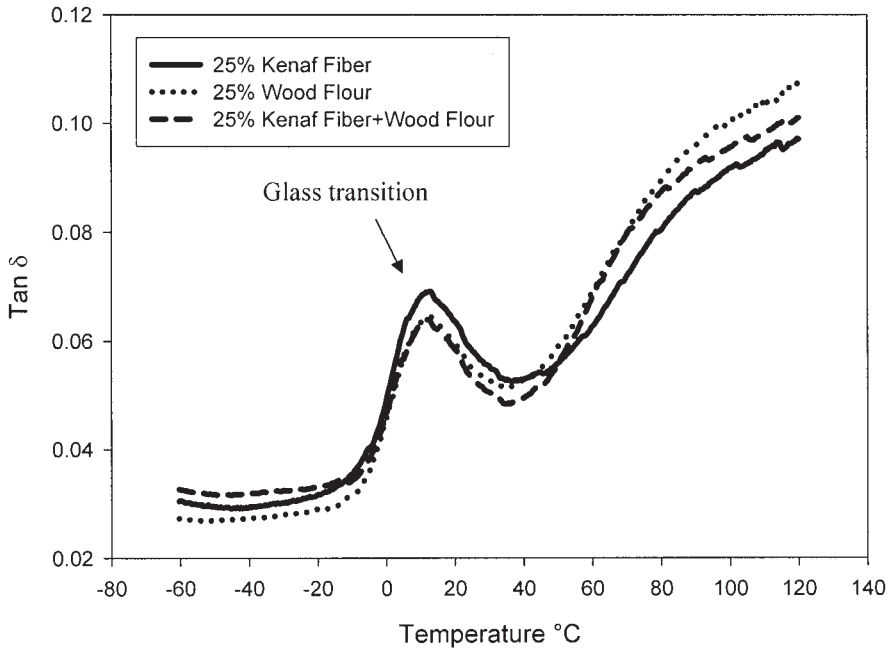

Figure 7 Tan δ spectra of various composites.

the glass-transition temperature was detectable due to the replacement of half of the WF with KFs. The glass-transition intensity (height of the curve at peak) of the hybrid composite was identical to that of the WF composite. The α transition occurred at a relatively higher temperature in the case of KFs, and its intensity was higher as well. This was explained by the bulkier nature and, hence, the higher volume fraction at a given weight content of KFs.

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