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HIGHLY FILLED LIGNOCELLULOSIC REINFORCED
THERMOPLASTICS: EFFECT OF INTERPHASE
MODIFICATION

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ABSTRACT

Although lignocellulosic fiber-thermoplastics composites have been used for several decades, recent economic and environmental advantages have resulted in significant commercial interest in the use of these fibers for several applications. Kenaf is a fast growing annual growth plant that is harvested for its best fibers. These fibers have excellent specific properties and have potential to be outstanding reinforcing fillers in plastics. High volume fillings are possible with natural fibers due to their non-abrasive nature and lower densities. This paper reports the structure-property relationships of using compatibilizers and PP impact copolymers in lignocellulose-PP composites. The use of maleated polypropylene (MAPP) is important to improve the compatibility between the fiber and matrix. A significant improvement in impact strengths was observed when the MAPP was used in the composites. Results also indicate that the samples with coupling agent have a better high temperature modulus and higher softening temperatures than uncoupled systems. This is possibly due to the better adhesion/interaction between the polymer molecules and kenaf fibers, the coupled samples have more restricted molecules than the uncoupled blends.

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1. INTRODUCTION

The use of kenaf, a natural fiber, as a reinforcement for polyolefins has been studied in detail and reported elsewhere (Sanadi, et al., 1994, 1995). Kenaf, a bast fiber, is extracted from the plant *Hibiscus cannabinus*. Extraction results in long filament and lengths greater than 1 m are common. The filaments themselves consist of individual fibers (2 to 6 mm long) which are themselves composites of predominantly cellulose, lignin and hemicellulose. Cellulose is the main reinforcing element of the fiber, while lignin and the hemicellulose can be considered as the binding matrix in the composite. The properties of the fibers and filaments can vary according to the source, age, extraction techniques, and history of the fiber.

There are several advantages of using natural fibers in plastics apart from their relatively low cost of the fibers: these include lower specific gravity, relative non-abrasive and low cost. The main disadvantages of these fibers in composites are the low processing temperatures allowable and the high moisture absorption of the fibers and composites. Recent work (Felix et al. 1993; Sanadi et al. 1994) suggests that the use of maleated polypropylene (MAPP) with sufficiently high molecular weight and anhydride content significantly improves the fiber-matrix bonding. The formation of covalent linkages between the MA and the hydroxyl groups of the cellulose has been indicated through IR and ESCA by Gatenholm et al. (1992).

2. EXPERIMENTAL

2.1 .Materials. Kenaf filaments, about 15 cm to 20 cm long, harvested from mature plants were cut into lengths of about 1 cm. A polypropylene homopolymer and two impact ethylene-propylene copolymers were blended with kenaf fibers. The homopolymer was Solvay 1602 (12 MFR). The ethylene-propylene impact copolymers were Amoco 3541 (20 MFR) and Amoco 3143 (2.5 MFR). The coupling agent was a maleated polypropylene and designated as MAPP. Batch blending of formulations was performed in a 1 liter thermokinetic mixer, granulated and then injection molded. All composites had 50 % by weight of the kenaf fibers and the uncoupled composites were designated as HP for homopolymer, ICP2 (Amoco 3541-high melt index and lower molecular weight) and ICP1 (Amoco 3143- low melt index and higher molecular weight) for the ethylene-propylene copolymers. The blends with MAPP were designated as HP-MA, ICP1 -MA, and ICP2-MA the amount of MAPP was kept constant at 3 % by weight of the composite. Dynamic mechanical properties of these samples were measured on a TA DMA983 instrument under nitrogen purge. The measurements were carried out at temperature ranging from - 100°C to 180°C at a heating rate of 2°C/min and the test frequency was 1 Hz.

3. RESULTS AND DISCUSSION

3.1. Mechanical Properties. The mechanical properties of the PP/kenaf fiber blends are reported in Table II. In all cases the tensile strength of the coupled composites were about twice as much as the uncoupled systems. As expected the failure strains of the coupled systems were significantly higher than the uncoupled composites. The ethylene content of the impact copolymers permit higher extensions as compared to the homopolymer composites. In contrast, the tensile modulus of all the coupled composites were lower than the uncoupled

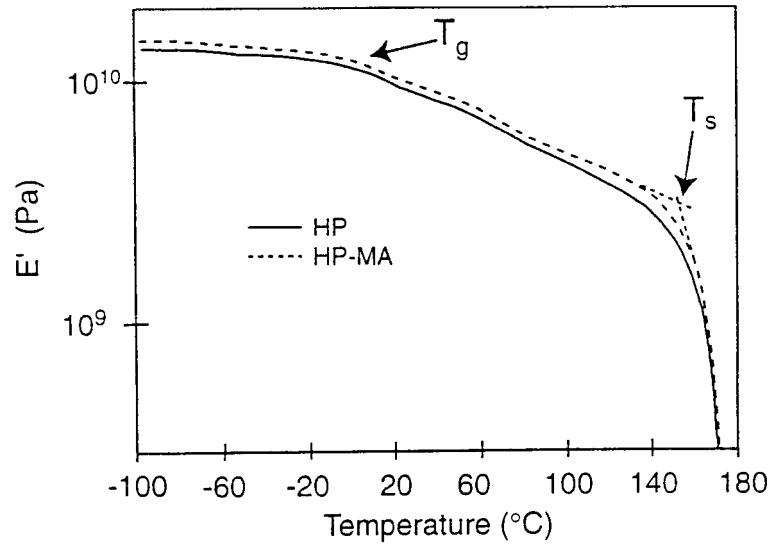


Figure 1: DMA E' of HP/Kenaf Blends

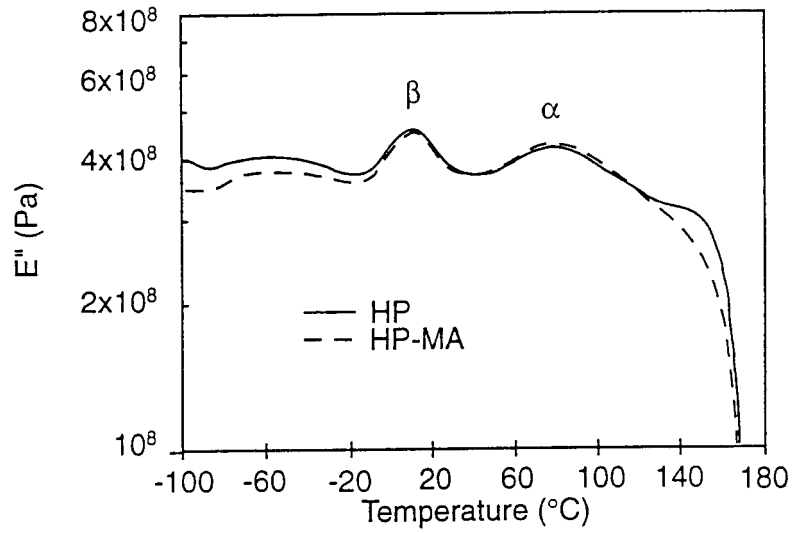
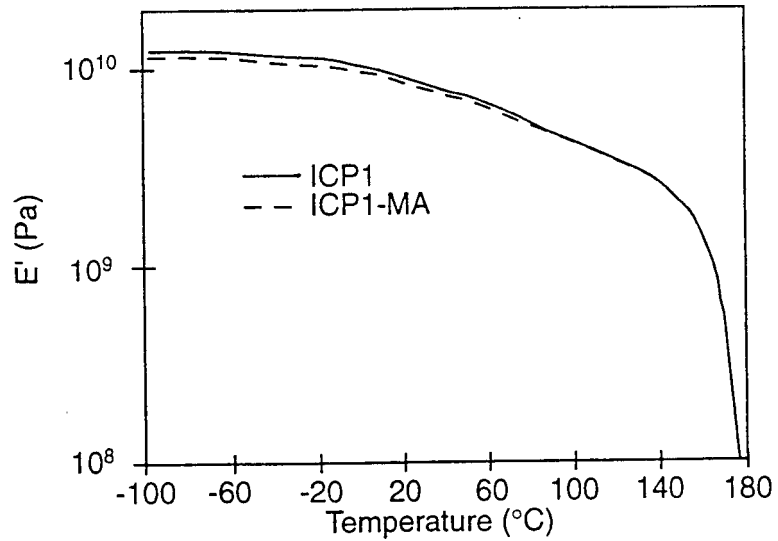


Figure 2: DMA E'' of HP/Kenaf Blends



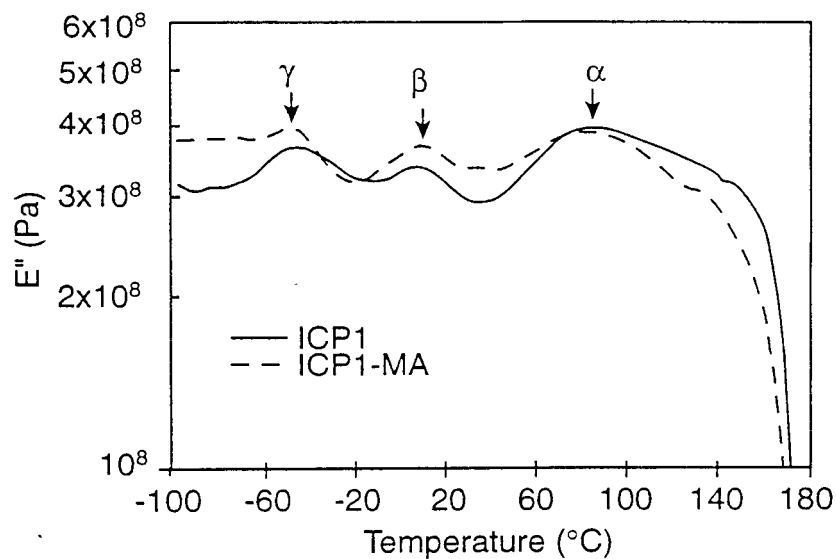


Figure 4: DMA E'' of ICP1/Kenaf Blends

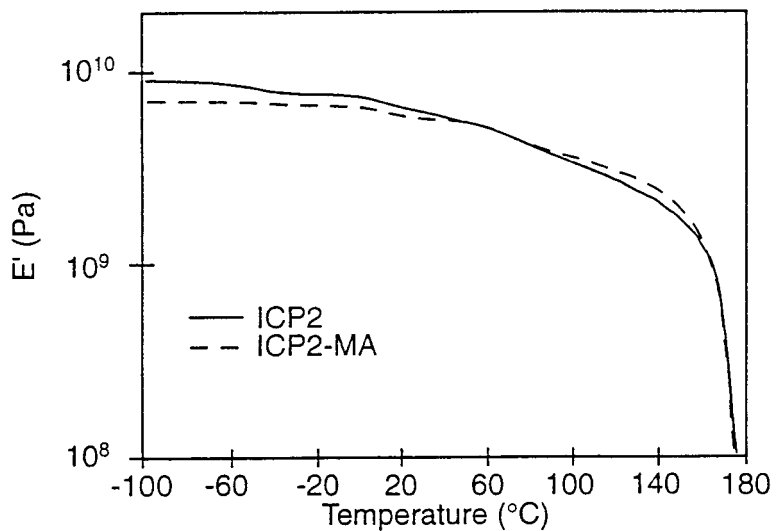


Figure 5: DMA E' of ICP2/Kenaf Blends

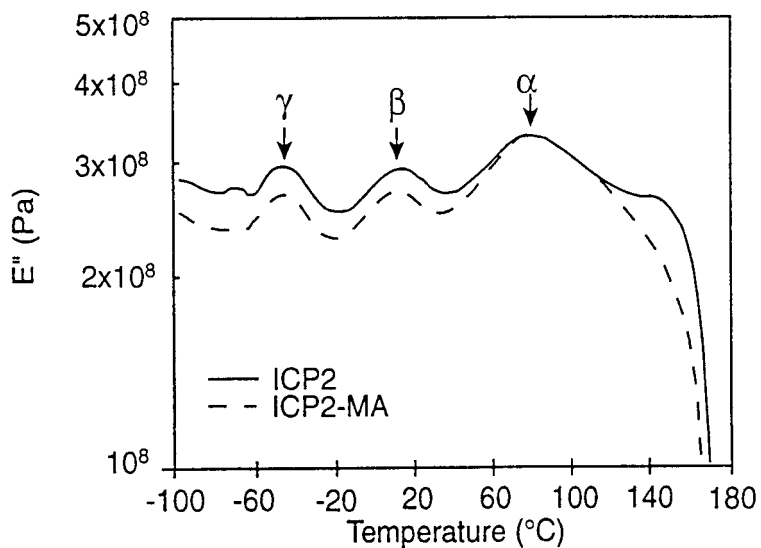


Figure 6: DMA E'' of ICP2/Kenaf Blends

systems. In all cases, the Izod notched and un-notched impact properties of the coupled systems were higher than the uncoupled composites. As expected the tensile strength of the impact polymers were less than homopolymer in both coupled and uncoupled composites.

Table I. Mechanical properties of PP/Kenaf fiber blends.

Sample ID	Izod (J/m)		Tensile	Tensile	Failure
	Notched	UN-notch	Strength	Modulus	Strain
			(MPa)	(GPa)	%
HP	33.90	87.60	33.30	9.30	1.10
HP-MA	36.70	167.50	65.80	8.30	1.82
ICP1	41.30	76.30	25.60	12.99	0.94
ICP1-MA	74.30	211.95	52.50	7.45	2.48
ICP2	46.76	118.83	23.34	11.39	0.90
ICP2-MA	70.54	220.14	50.13	7.91	2.40

3.2 Dynamic Mechanical behavior. DMA spectra of these PP/kenaf fiber composites are shown in Figures 1 to 2. Figure 1 shows the storage modulus (E') spectra of HP and HP-MA blends. Two transitions are observed on the E' spectra, T_g and T_s , pointed by arrows. At the T_g transition, the E' shows a sudden decrease because of the glass transition. At T_s , the polymer starts to melt, and the sample starts to become soft and the modulus drastically decreases. The results show that the coupled blend, HP-MA, has a higher E' than the uncoupled HP sample for entire temperature range (Fig. 1). The HP-MA also has a higher softening temperature (T_s) than the HP sample (Fig. 1 and Table II). The higher E' and T_s suggest that HP-MA blend has a better adhesion between the PP matrix and kenaf fibers. The loss moduli spectra (E'') of both HP and HP-MA are show in Fig. 2. Two major transitions, α and β , are observed around 80°C and 10°C . The α transition is related to the relaxation of restricted PP amorphous chains in the crystalline phase. The β transition is related to unrestricted amorphous PP (McCrum et al. 1967; Boyd 1985; Jourdan. et al. 1989; Jarvela et al. 1996).

The DMA spectra of ICP/kenaf fiber blends are shown in Figures 3 to 6. Figures 3 and 4 show the DMA spectra of low molecular weight ICP blends, ICP1 and ICP1 -MA. At temperature below 100°C , the storage E' of the coupled blend, ICP1 -MA is lower than that of the uncoupled blend, ICP 1 (Fig. 3). At temperatures above 100°C . the E' of ICP1 -MA is slightly higher than that of ICP1. The T_s of ICP1-MA is also slightly higher than the uncoupled blend. The loss modulus (E'') spectra of ICP1 and ICP1-MA are shown in Figure 4. Three transition peaks, α , β , and γ , are observed. The α transition is related to the relaxation of restricted PP amorphous chains in the crystalline phase. The β transition is due to the relaxation of unrestricted amorphous PP. The γ transition around -50°C is related to the relaxation of amorphous ethylene segments of the polymer chains. The DMA spectra of high molecular weight ICP2/kenaf fiber blends are shown in figures 5 and 6. Figure 5 shows the E' spectra of ICP2 and ICP2-MA blends. The ICP2-MA E' spectrum is flatter than that of ICP2 indicating that the ICP2-MA is more flexible at low temperatures (below 40°C), and the modulus is

higher at temperatures above 50°C. The E'' spectra of ICP2 and ICP2-MA are shown in Fig. 6. Three transition peaks, α , β , and γ , are observed. The ICP2-MA has a higher α peak and

Table II. DMA transition temperatures of PP/Kenaf fiber blends.

Sample ID	E'' transition temperatures (C)			Ts (C)
	γ	β	α	
HP		9.3	76.6	161.6
HP-MA		9.6	79.3	162.4
ICP1	-47.0	8.6	80.5	163.3
ICP1-MA	-49.3	8.9	79.2	162.1
ICP2	-46.0	11.4	76.8	164.1
ICP2-MA	-46.0	10.8	79.6	162.9

lower β peak than ICP2 blend. This indicates that the ICP2-MA blend has a higher crystallinity than the ICP1 blend.

In conclusion, the results show that the coupling agent enhanced the adhesion between the PP matrix and kenaf fibers. Due to the better adhesion, the blends with coupling agent have better mechanical properties. The dynamic mechanical properties are also affected by coupling agents. Due to the better adhesion between the polymer matrix and kenaf fibers, the coupled blends have better high temperature modulus and higher softening temperatures than the uncoupled blends.

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