Effect of Compatibilizer on the Structure-Property Relationships of Kenaf-Fiber/Polypropylene Composites

DAAN FENG

Amoco Polymers, Inc.
Alpharetta, Georgia

D. F. CAULFIELD

USDA Forest Service
Forest products Laboratory
Madison, Wisconsin 53705

and

A. R. SANADI*

Biological Systems Engineering
University of Wisconsin, Madison
Madison, Wisconsin 53705

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 bast fibers. These fibers have excellent specific properties and have potential to be outstanding reinforcing fillers in plastics. This paper reports the structure-property relationships of kenaf fiber reinforced polypropylene (PP) and its impact copolymers. The use of maleated polypropylenes (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 impact copolymer blends with coupling agent have better high temperature moduli and lower creep compliance than the uncoupled systems. The coupling agent also changes the crystallization and melting behavior of these blends. Because of the better adhesion between the polymer molecules and kenaf fibers, the coupled samples have more restricted molecules than the uncoupled blends. As a result, the crystallization of the coupled high molecular weight blends is slower than the uncoupled blends, resulting in a lower crystallization temperature (T_C) and reduced crystallinity. For the lower molecular weight blends, the coupling agent enhances the crystallization of polymer matrix and results in a higher crystallization temperature and increased crystallinity of the coupled blend. The coupled blends also have more defects in the polymer crystals, and the crystallinity of coupled blends is also lower than the uncoupled blends. This could explain the lower melting temperatures of the coupled samples as compared to uncoupled samples.

Parts of this manuscript were presented and published at the Fourth International Conference on Woodfiber-plastic Composites, Madison, WI, USA, 1997, and the 2nd International Wood and Natural Fibre Composites Symposium, Kassel, Germany, June 1999.
The Forest Products Laboratory is maintained in cooperation with the University of Wisconsin. The article was written, in part, by U.S. Government employees on Official time, and is therefore in the public domain and not subject to copyright.
*Corresponding author: arsanadi@facstaff.wisc.edu

INTRODUCTION

Lignocellulosic based thermoplastic composites have been used commercially for a number of years. Particulate fillers such wood flour and natural fibers such as jute and flax are commonly used by the automotive industry for interior applications. Research is now under way on using ramie, one of the strongest cellulosic fibers available, as a replacement for glass fibers for interior panels for Airbus planes (1). The usual method of making the finished automotive interior product is by compression molding of non-woven mats of both natural and polypropylene fibers. Blended pellets of lignocellulosic-plastics for injection molding have only recently been available in substantial quantities in the market. This will greatly enhance the use of these renewable particles/fibers by the plastics industry for high volume, low cost applications.

The use of kenaf, a natural fiber, as a reinforcement for polyolefins has been studied in detail and reported elsewhere (2, 3). 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 composites of predominantly cellulose, lignin and hemicellulose. Crystalline cellulose is the main reinforcing element, while lignin and the hemicellulose can be the considered as the binding matrix. The properties of the fibers and filaments can vary according to the source, age, extraction techniques, and history of the fiber. Although fiber properties can vary a great deal, earlier work has shown that fibers from the same batch with similar histories results in composite properties with a remarkably low variation in properties (3).

The specific stiffness of 50% by weight kenaf-PP is comparable to that of 40% by weight glass-PP composites; details of the comparison of properties are published elsewhere (2, 3). The properties of kenaf fiber composites compare well with other similar bast fibers such as jute. This is because the crystalline cellulose content and microfibril angles of the two fibers are fairly similar. Fibers such as hemp and ramie that have higher crystalline cellulose contents and smaller microfibril angles should result in higher strengths in injection molded tests. However, we have noticed that in the case of bast fibers, kenaf flows much easier than jute and flax fibers (in PP), and therefore, higher fiber contents can be achieved.

There are major challenges to the wide use of kenaf fibers in composites. The quality of natural fibers varies according to the type, soil, cultivation and fiber separation methods and standardizing the quality is important. The cost of kenaf currently varies with quality and cleanliness from about $ 0.33 to $ 0.88 per kg, but could decrease with better seed quality resulting in higher yields, improved separation techniques and higher demand. It is also important to have a steady supply of high quality kenaf available to the industry, with enough supply to last in case of crop damage. Moisture uptake of the composites is definitely a concern and means to design around this is necessary. There are chemical methods to decrease fiber water absoprtion and thereby reduce water uptake in the composite. However, the use of such techniques depends on whether the economics are favorable, which at present does not appear to be so.

The inherent differences between the highly polar lignocellulosic fibers and the non-polar polypropylene (PP) can result in difficulties in the dispersion of fibers along with poor fiber-matrix interactions. Dalvag *et al.* (4) investigated the use of additives other than coupling agents in composites of HDPE or PP. Additives included stearic acid, paraffin and polyethylene waxes, and mineral oil. Several of the additives (stearic acid and paraffin) resulted in prevention of filler agglomeration and led to improved impact strength and elongation at rupture. The additives did not affect tensile modulus, and a decrease in strength was attributed to possible plasticizing effects. It was also noted that addition of paraffin wax did not significantly change the viscosity of the composite blends. Raj *et al.* (5) and Kokta *et al.* (6) investigated the use of a number of additives in composites of wood flour (often with pretreatment) and a variety of polyolefins. Additives included stearic acid, mineral oil, and sodium silicate. Dispersion was investigated (5, 7) by visual inspection under microscope or by measurement of the concentration of aggregates in each sample. In general, the additives resulted in improvements in dispersion, tensile modulus and strength, and impact energy.

Many groups have investigated the use of coupling agents to improve the compatibility between the fiber and plastic matrix. Several groups have also reported the efficiency of MAPP, and cited literature goes back to the late 1970s. Recent work (8, 9) suggests that the use of maleated polypropylene (MAPP) with sufficiently high molecular weight and anhydride content significantly improve 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.* (10).

Earlier work on 50% by weight of kenaf in PP composites indicated that the specific moduli of these composites were comparable to that of 40% by weight of glass fiber-PP, although the toughness of the kenaf composites were lower than the glass composites (2). In this study, the properties of PP homopolymers were compared to that of PP impact copolymers. The effect of coupling agent on the mechanical properties and dynamic mechanical behavior are reported. The crystallization and melting behavior of the kenaf composites were compared using a differential scanning calorimeter. Some creep data comparing the effect of improved adhesion were also obtained.

Table 1. Composition of PP/Kenaf Fiber Blends.

Sample ID	Polymer Type	Polymer MFR g/10 min	Polymer wt%	Kenaf Fiber wt%	Coupling Agent wt%
HP	Solvay 1602	12	50	50	0
HP-MA	Solvay 1602	12	47	50	3
ICP1	Amoco 3541	20	50	50	0
ICP1-MA	Amoco 3541	20	47	50	3
ICP2	Amoco 3143	2.5	50	50	0
ICP2-MA	Amoco 3143	2.5	47	50	3

EXPERIMENTAL

Materials

Kenaf filaments, about 15 cm to 20 cm long, harvested from mature plants were cut into lengths of about 1 cm. The fibers were not dried to remove the moisture present, and the moisture content of the fibers varied from 6% to about 9% by weight. In all our experiments the weight and volume percent reported was the amount of dry fiber present in the blend. A polypropylene homopolymer, PP, and two impact ethylene-propylene copolymers were blended with kenaf fibers. The PP homopolymer was Solvay 1602. The ethylene-propylene impact copolymers were Amoco 3541 and Amoco 3143. The coupling agent was a maleated polypropylene.

Batch blending of formulations was performed in a one-liter thermokinetic mixer (K-Mixer, Synergistics Ltd., Canada). The short fibers, PP and MAPP (if used) were compounded in a high intensity kinetic mixer where the only source of heat is generated through the kinetic energy of rotating blades. The blending was accomplished at 4600 rpm, which resulted in a blade tip speed of about 30 m/s. The blended mass was then automatically discharged at 190°C. A total weight of 150 g was used for each batch, and about 1.5 kg of blended material was prepared for each set of experiments. The total residence time of the blending averaged about 2 min.

The discharged mass was cooled by pressing in a cold press and then granulated. The granules were injection molded into ASTM standard specimens using a 33 ton Cincinnati Milacron injection molding machine at 190°C. The specimens were then stored at 20% humidity and 32°C for at least three days before testing. Specimen dimensions were according to the respective ASTM standards.

All composites had 50% by weight of the kenaf fibers and the uncoupled composites were designated as HP for homopolymer, ICP1 (lower molecular weight-Amoco 3541) and ICP2 (higher molecular weight-Amoco 3143) 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. Details of the polymer-kenaf blends are listed in Table 1.

Measurements

All mechanical tests were performed according to ASTM standard testing methods. The tensile properties were tested according to ASTM 638-90 at a crosshead speed of 5 mm/min. The Izod impact properties were tested according to ASTM 256-90 method.

Differential scanning calorimetry (DSC) measurements of 10 mg samples were performed on a TA DSC2910 and two specimens were tested for each composite type. The DSC sample was first heated from 25°C to 230°C at 2O°C/minute rate to erase the previous thermal history. The sample was then cooled at 10°C/minute rate to −100°C to recrystallize the sample and measure the crystallization temperature (T_C). After cooling, the sample was reheated from −100°C to 230°C at 20°C/minute to measure the melting temperature (Tm) and heat of fusion.

Dynamic mechanical properties of these samples were measured on a TA DMA983 instrument under nitrogen purge. The measurements were carried out at temperatures ranging from −100°C to 180°C at a heating rate of 2°C/minute. The test frequency was 1 Hz and the sample size was 45 mm × 12.5 mm × 3 mm.

The creep properties of these samples were also measured on the TA DMA983. The creep tests were carried out at temperatures ranging from −20°C to 100°C at 10°C increments. At each temperature, the sample was first loaded for 10 minutes and then followed by a 15-minute relaxation. The initial displacement was 0.3 mm. Master curves were generated from the creep data by using the WFL equation at a reference temperature of 71°C.

RESULTS AND DISCUSSION

Mechanical Properties

The mechanical properties of the PP/kenaf fiber blends are reported in Table 2. The potential covalent bonding between the anhydride group with the hydroxyl groups of the kenaf, along with chain entanglement between the MAPP and PP chains creates a good stress transfer interface. As expected, the failure strains of the coupled systems were significantly higher than the uncoupled composites. The ethylene content of the impact copolymers permits higher extensions as compared to the homopolymer composites. In contrast, the

Table 2. Mechanical Properties of PP/Kenaf Fiber Blends.

Sample ID	Izod (J/m) Notched	UN-notch	Tensile Strength (MPa)	Tensile Modulus (GPa)	Failure Strain YO
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

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

In all cases the tensile strengths of the coupled composites were about twice as much as those of the uncoupled systems. There is significant improvement in the tensile strengths of the coupled composites. A large proportion of the fibers present in the composites, both coupled and uncoupled, are still in their filament form, which results in low aspect ratios. Separation into their individual fibers would not only increase their aspect ratios, but also greatly increase the interface area between the fibrils and matrix. Both factors could dramatically increase the strength properties of the composites.

The blending in the thermokinetic mixer can cause severe fiber attrition. However, the advantage is that the fibers are well distributed in the matrix and no significant fiber agglomeration was observed. Surface material was probably stripped from the fibers and this was clearly indicated by the dark brown tinge of the matrix after blending. Earlier work indicated that there was a wide distribution of fiber lengths in the composite blend after injection molding. The median fiber length was around 0.5 mm, with both shorter and longer lengths present in the blend. Assuming a kenaf fiber strength of about 500 MPa and a interfacial shear strength equal to that of matrix shear strength (17 MPa, estimated from the Von-Mises criterion using polymer tensile strength), one obtains a fiber critical length of about 0.36 mm. In other words, the composite strength can be improved if the longer fiber lengths can be maintained.

It is interesting to note the higher tensile moduli of the uncoupled blends. This behavior has also been observed for flexural data. This could be due to differences in the polymer morphology. In general, transcrystalline zones are formed parallel to the flow direction where the fibers are predominantly oriented during injection moldings (11).

Thermal Properties

DSC results of HP and HP-MA are shown in Figs. I and *2*, and *Table 3*. The HP-MA has a broader crystallization peak than HP. The crystallization temperature (T_C) of HPMA is also lower than the HP sample indicating that the HPMA blend crystallizes slower than HP (*Fig. 1*). The melting behavior of the two samples is shown in *Fig. 2*. The results show that the HP-MA sample has a slightly broader melting peak with lower peak temperature. The low melting temperature indicates that the HP-MA sample has more defects in PP crystals than the HP sample.

DSC results of ICP blends are shown in Figs. 3 and 4 and Table 3. The results (*Fig. 3*) indicate that the crystallization behavior strongly depends on the molecular weight of the impact copolymer. In the case of the lower molecular weight (higher melt flow index, MFI) composites, the coupled system (ICP1MA) had a much higher T_C than the sample without the coupling agent, and the difference is considerable. In the case of the high molecular weight ICP2 composites, the sample with coupling agent (ICP2MA) had a lower T_C than the sample without coupling agent. It is not possible to determine the percent crystallinity with any accuracy, since the exact fraction of polymer in the 10 mg DSC sample is not known. The lower molecular weight copolymer composites with coupling agent (ICP1MA) have lower T_m than the samples without coupling agent (Fig. 4)—this is due to the larger number of

Table 3. Thermal Properties of PP/Kenaf Fiber Blends.

Sample	Tc °C	Onset Tc °C	Hc J/gm	Tm °C	Onset Tm °C	Hf J/gm
HP	120.7	125.0	46.4	167.3	154.3	47.3
HP-MA	120.0	125.6	48.0	166.5	153.1	46.2
ICP1	119.9	124.2	41.5	169.3	158.5	39.2
ICP1-MA	124.7	130.1	41.5	167.6	151.9	37.5
ICP2	123.6	127.9	39.6	169.5	155.2	37.4
ICP2-MA	121.4	126.6	40.7	168.4	155.8	38.6

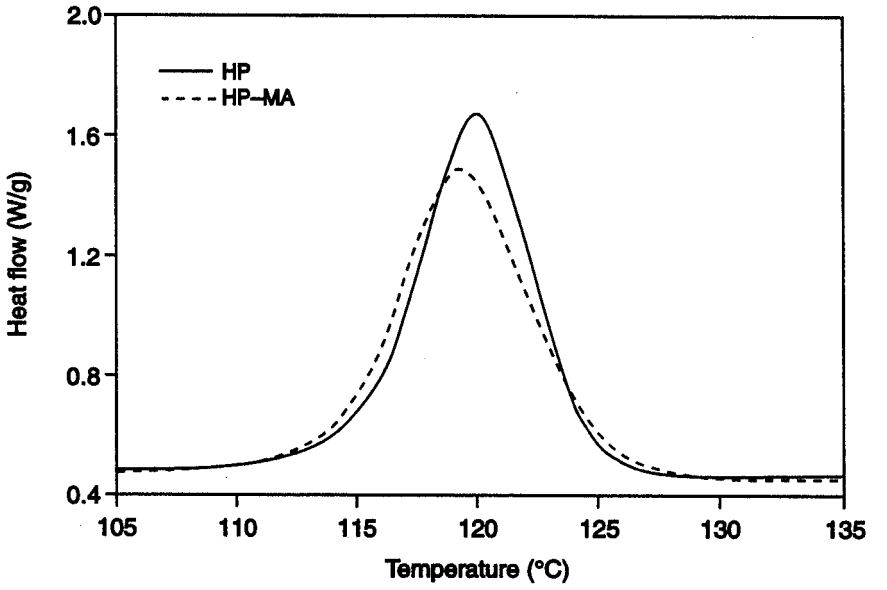

Fig. 1. DSC cooling scans of HP and HP-MA blends.

defects in the crystals. In case of the higher molecular weight composites with coupling agent (ICP2MA). the T_m is slightly more than ICP2: in this case, the low molecular weight MAPP could enhance mobility of the polymer chains and enhance crystal quality.

Several factors must be considered to understand the crystallization and melting behavior of these blends. (i) The presence of a low molecular weight polymer, which is the coupling agent, and its lubricating effect on the higher molecular weight polymers. (ii) The presence of coupling agent that results in interactions, covalent and acid-base, between the anhydride (AN) and the hydroxyl (–OH) groups on the lignocellulosic surface. (iii) The interaction between the AN groups among themselves, within the same molecule or between neighboring coupling agent molecules (12). Thermodynamic segregation of the MAPP to the lignocellulosic surface could play a major role in determining the morphology around the fiber. Good interaction between the MAPP and the lignocellulosic surface restricts the mobility of the molecules. Furthermore. the anydride groups themselves disrupt the regularity of the PP chains, which could hinder crystallization or create defects in the crystals. On the other hand, the low molecular weight of the MAPP and the interactions between AN groups could favor crystallization

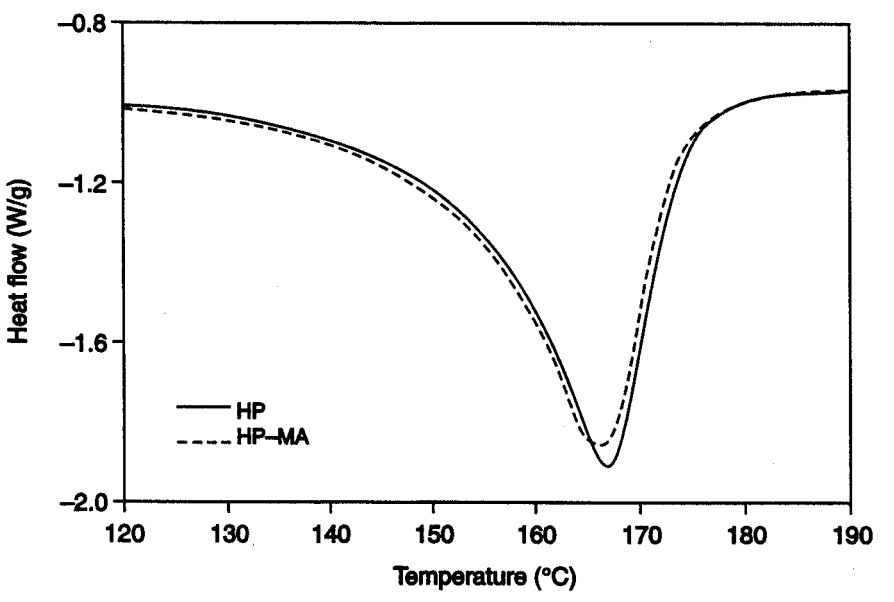

Fig. 2. DSC heating scans of HP and HP-MA blends.

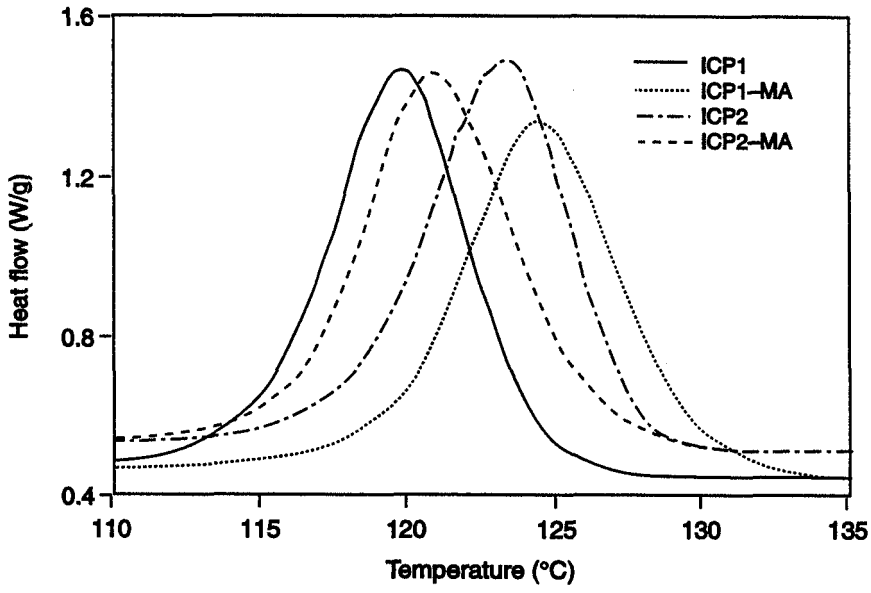

Fig. 3. DSC cooling scans of ICP and ICP-MA blends.

(12). The glass-rubber transition of the system would thus depend on which of above mentioned factors are dominant.

A fourth important factor that has previously been neglected is that during blending, some of the surface matrix material of the kenaf fibers themselves are sheared off from the fiber and blend in with the matrix. This sheared material could have lignin and amorphous cellulose. and oils and other surface extractives that are inherently present in the fiber. This can be clearly seen as the dark brown tinge of the polymer matrix after blending. In other words. the matrix material is a blend of PP and several constituents that are contributed by the fiber. These matrix additives may also contribute to the reduced T_g. However, in our blends, since we are comparing systems that have the same amount of fibers, we can assume that the effect of the sheared material in modifying the crystallization and melting behavior for the same polymer is equivalent. This will be discussed in a future paper.

In the case of HPMA and ICP1MA, the crystalline defects created with the presence of the MAPP could be dominant in reducing T_m. On the other hand, for ICP2MA the presence of the lower molecular weight MAPP enhances the molecular mobility and allows better packing in the crystals. This could decrease the

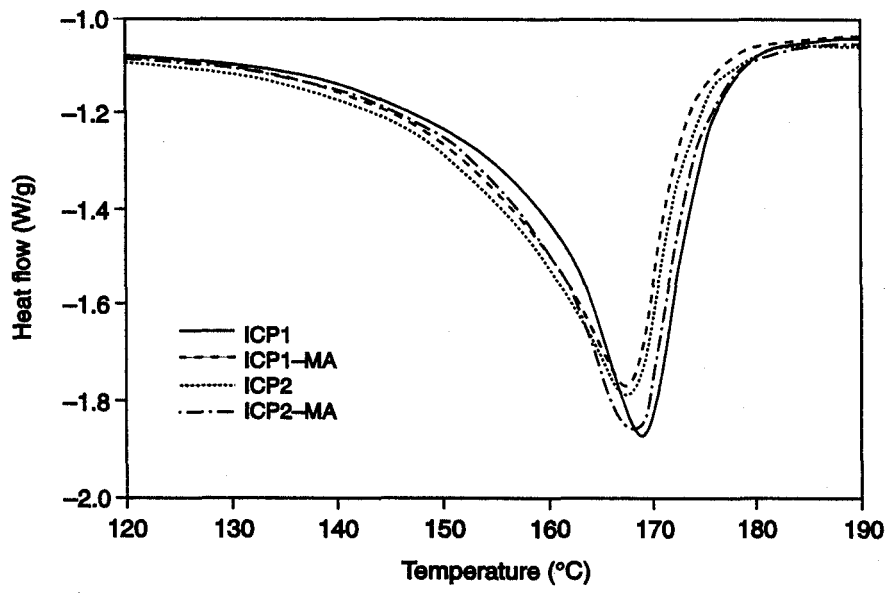

Fig. 4. DSC heating scans of ICP and ICP-MA blends.

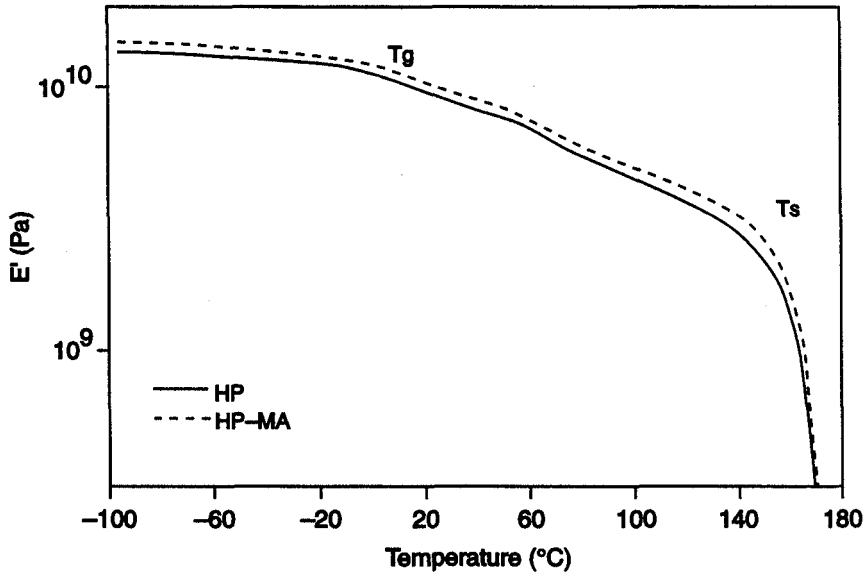

Fig. 5. Storage modulus spectra of HP and HP-MA blends.

number of defects in the crystalline region, and thereby enhance T_m. The presence of the -OH (on the fiber surface) and MA interactions and the lower MW of the MAPP (lubricant effect) could help in increasing the crystallinity of the higher molecular weight impact copolymer thereby causing a slight increase in T_m.

Dynamic Mechanical Behavior

Hompolymer/Kenaf Fiber Blends

DMA spectra of these PP/kenaf fiber composites are shown in *Figs. 5 to 10*. *Figure 5* 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 drop since the glassy amorphous molecules turns to a rubbery phase. At T_s, the polymer starts to melt, and the Sample starts to become soft and the modulus decreases. The coupled blend, HP-MA, has a higher E´ than the uncoupled HP sample for entire temperature range. The HP-MA also has a higher softening temperature (T_s) than the HP sample (Table 4). The higher E´ and T_s suggest that HP-MA blend has a better adhesion between the PP matrix and kenaf fibers. However, it is confusing why the coupled blends (HP-MA) had a higher dynamic storage modulus, while

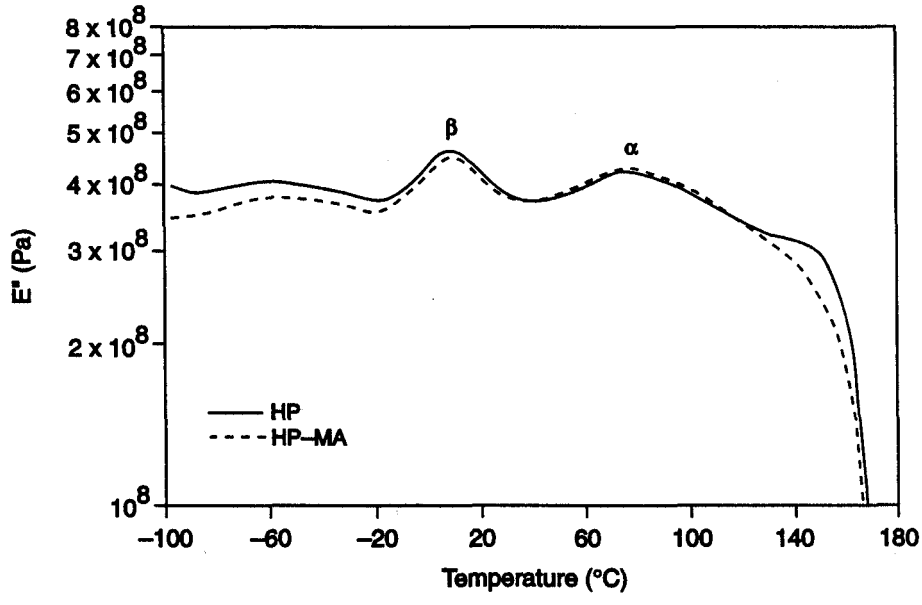

Fig. 6. Loss modulus spectra of HP and Hp-MA blends.

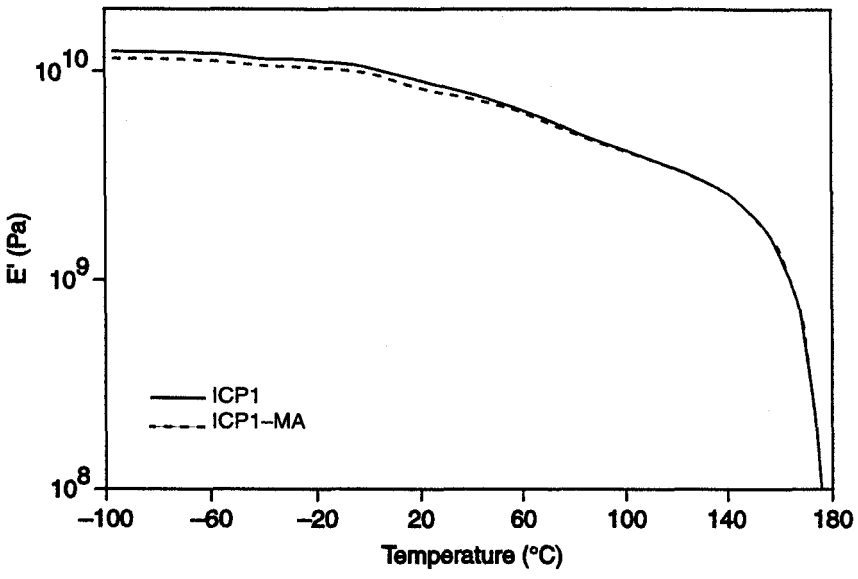

Fig. 7. Storage modulus spectra of ICP-1 and ICP1-MA blends.

the static Young's modulus of the uncoupled (HP) was higher. As mentioned earlier, the behavior for higher moduli for uncoupled blends was also observed during flexural testing.

The loss modulus spectra (E") of both HP and HP-MA are show in Fig. 6. Two transitions, a and b, are observed around 80°C and 10°C. The a transition is related to the relaxation of restricted PP amorphous chains in the crystalline phase and the b transition is related to unrestricted amorphous PP (13–16). *Figure 6* shows that both the peak temperature and the peak amplitude of the a peak of HP-MA are higher than that of HP. The HP-MA blend also has a higher peak temperature of the b peak than the HP blend. But the b peak amplitude of HP-MA is lower than the b peak amplitude of HP. In general, the increase in peak temperature indicates a decrease in chain mobility, and the E″ peak amplitude is related to the amount of amorphous PP chains involved in the transition. The lower b peak amplitude of HP-MA indicates that the HP-MA blend has a lower amorphous content than the HP sample. The higher a peak amplitude of HP-MA sample indicates that the HP-MA blend has more rigid amorphous molecules in the crystalline phase (crystal defects) than the HP sample. This suggests that the crystallinity of HP-MA is slightly higher than the HP blend. The higher peak temperatures of both a and b peaks of HP-MA blend indicate that the PP molecules are more restricted in HP-MA blend than that in HP sample. Because of better adhesion, the polymer chains have more restriction, which results in an increase in b transition temperatures.

Table 4. DMA Transition Temperatures of PP/Kenaf Fiber Blends.

Sample ID	E″ Transition Temperatures (°C) g	b	a	Ts (°C)
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

Impact Copolymer/Kenaf Fiber Blends

The DMA spectra of ICP/kenaf fiber blends are shown in *Figs.* 7 to 10. Figures 7 and 8 show the DMA spectra of lower 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, ICP1 (*Fig.* 7). At temperatures above 100°C. the E' of ICP1-MA is slightly higher than that of ICP1.

The loss modulus (E″) spectra of ICP1 and ICP1-MA are shown in *Fig. 8*. Three transition peaks, a, b, and g, are observed. The a transition is related to the relaxation of "rigid amorphous" PP chains in the crystalline phase. The b transition is due to the relaxation of unrestricted amorphous PP. The g transition around –50°C is related to the relaxation of amorphous ethylene segments of the polymer chains. As *Fig. 8* shows, the b peak temperature of the coupled system is higher than the uncoupled system. This suggests that the ICP1-MA has more restricted amorphous polypropylene molecules, possibly near the fiber surface. Because of better adhesion, the polymer molecules are more restricted in motion, resulting in less distinct and broader transition peaks. The results also show that the uncoupled blend, ICP1, has a bigger a transition peak and a broader transition than the coupled blend ICP1-MA.

The DMA spectra of high molecular weight ICP2/kenaf fiber blends are shown in Figs. 9 and 10. Figure 9 shows the E′ spectra of ICP2 and ICP2-MA blends. The

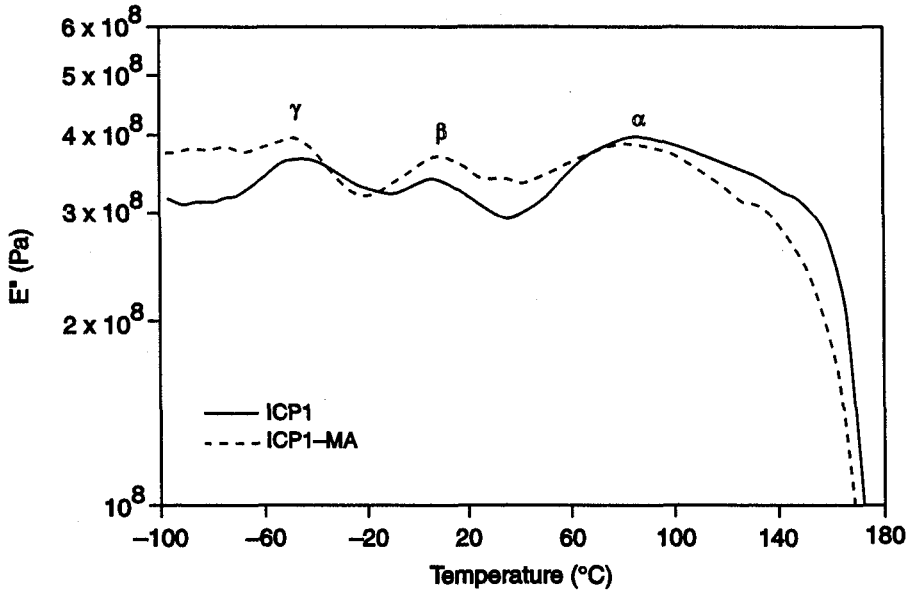

Fig. 8. Loss modulus spectra of ICP1 and ICP1-MA blends.

ICP2-MA E′ spectrum has a smaller slope than that of ICP2. The ICP2-MA is more flexible at low temperatures (below 40°C—indicating higher toughness) and a higher storage modulus at temperature above 80°C. Three transition peaks, a, b, and g, are observed (*Fig.* 10). The ICP2-MA has a lower b peak temperature than the ICP2 blend. This could indicate that the effect of the fiber-MA interaction at the interface is overcome by the lubricant effect of lower molecular weight MAPP, which lowers the b peak temperature. The same observation can be seen in the b onset temperature, which is lower for the coupled system than the ICP2 b onset. This lower b onset temperature for the coupled systems is also observed for both the HP and ICP1 blends, an indication that the lower molecular weight does affect the systems. However, in the HPMA and ICP1MA systems, the fiber-MA interactions at the interface are dominant over the lubrication effect, and results in higher b peak temperatures. On the other hand, the peak amplitude of the ICP2 a transition is much higher for the coupled system. This indicates more defects in the ICP2MA than the ICP2 composites.

Dynamic Creep Behavior

The dynamic creep curves of these PP/kenaf fiber blends are shown in *Figs.* 11 to 13. *Figure* 11 shows the creep compliance of HP and HP-MA blends as a

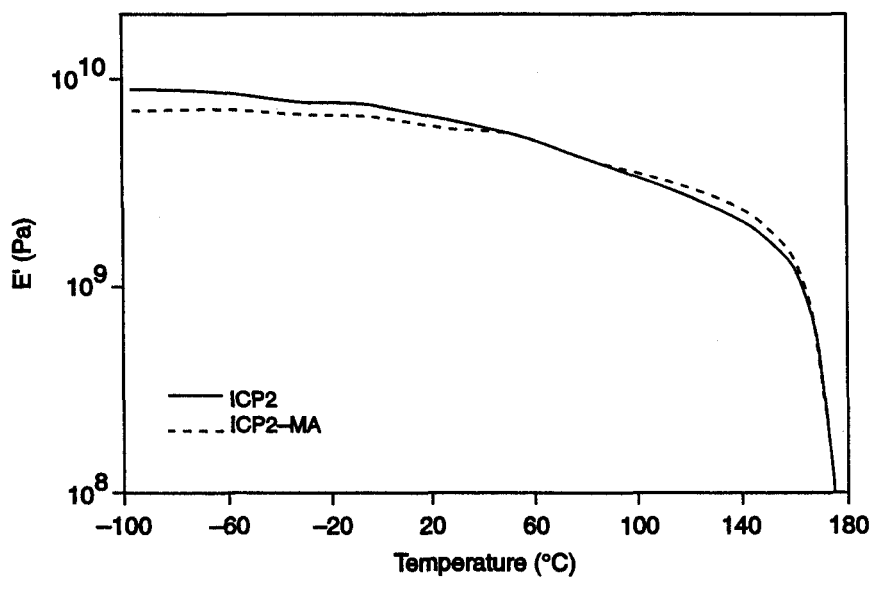

Fig. 9. Storage modulus spectra of ICP2 and ICP2-MA blends

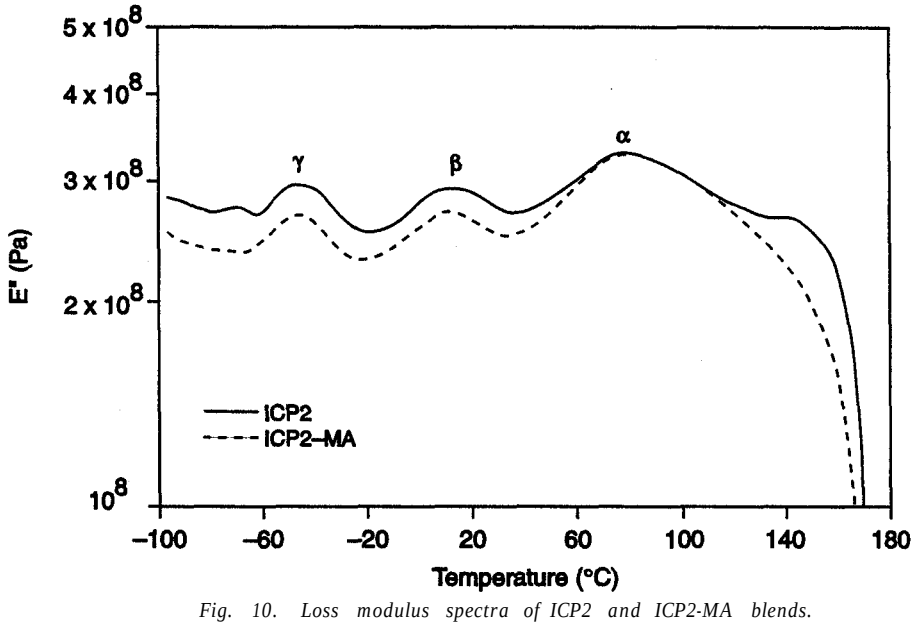

Fig. 10. Loss modulus spectra of ICP2 and ICP2-MA blends.

function of time. Surprisingly, there was little difference in the compliance for the uncoupled and coupled homopolymer composites. At shorter times, the compliance of the coupled was lower, while at longer times the compliance was higher than the uncoupled composites.

On the other hand, the molecular weights of the copolymers have a strong influence on the creep behavior of the blends. For the low molecular weight impact copolymer ICP1, the coupling agent improves the creep behavior significantly (*Fig.* 12). For the high molecular weight impact copolymer ICP2, the influence of the coupling agent is smaller (*Fig.* 13). The creep behavior of the ICP/kenaf blends, or for any composite, is dependent on several factors including crystallinity, adhesion between the fiber and polymer matrix, and polymer chain entanglements. The high molecular weight copolymer has a higher entanglement density than the lower molecular weight polymers, and the effect of the coupling agent on the creep of the high molecular weight copolymers is much less pronounced.

CONCLUSION

The results show that the coupling agent enhanced the adhesion between the PP matrix and kenaf fibers

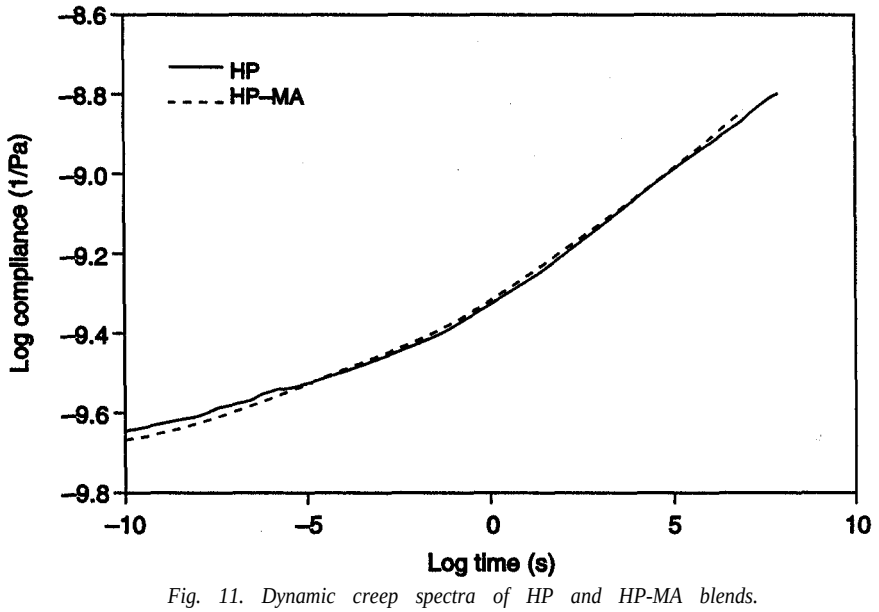

Fig. 11. Dynamic creep spectra of HP and HP-MA blends.

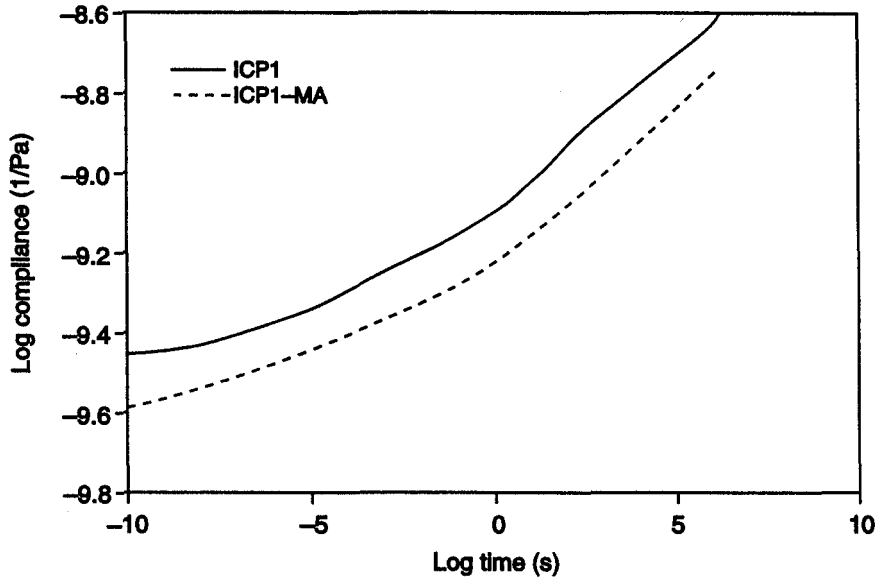

Fig. 12. Dynamic creep spectra of ICP1 and ICP1-MA blends.

and resulted in the blends with coupling agent having higher tensile and impact strengths. In the case of the Young's moduli, the uncoupled blends were higher than the coupled systems and this difference was very significant for the impact copolymers. Future papers will deal with this phenomenon.

The coupling agent also changed the crystallization and melting behavior of these blends. Owing to the better interaction/adhesion between the polymer molecules and kenaf fibers, the coupled samples had more restricted molecules than the uncoupled blends. As a result, the crystallization of the coupled low molecular weight blends, HP-MA and ICP1-MA, is slower than the uncoupled samples. The coupled blends also have more defects in the polymer crystals. Thus, the melting temperatures of coupled samples are lower than that of uncoupled samples.

The dynamic mechanical properties are also affected by coupling agents. Better adhesion between the polymer matrix and kenaf results in the coupled blends' having better high temperature modulus and higher softening temperatures than the uncoupled blends. The creep properties also improved with the use of the coupling agent for the impact copolymers.

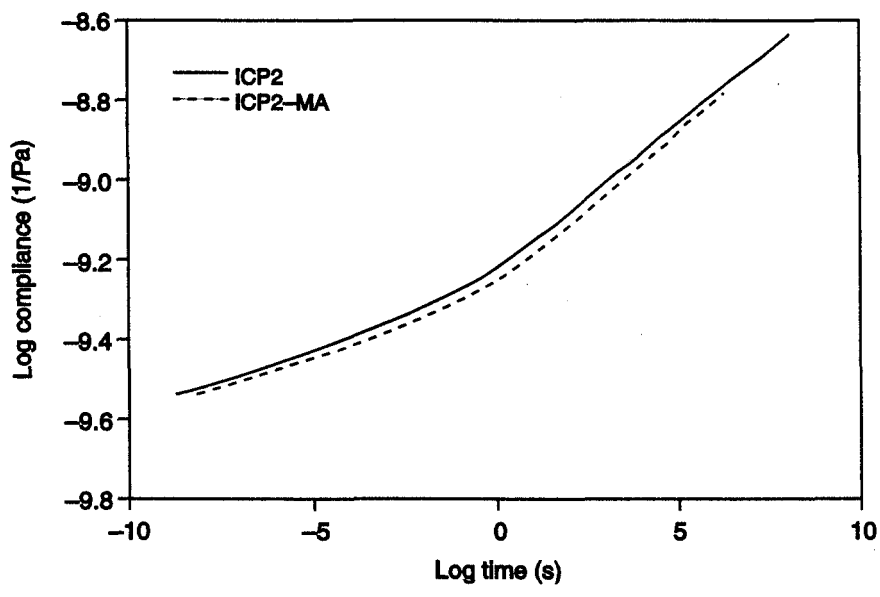

Fig. 13. Dynamic creep spectra of ICP2 and ICP2-MA blends.

ACKNOWLEDGMENTS

The authors would like to acknowledge funding from USDA NRI/CSRS (#9701817). One of the authors (ARS) would like to thank the Forest Product Laboratory for funding, facilities and space provided during the course of the study.

REFERENCES

1. Daimler-Benz, *Daimler-Benz High Tech Report*, News from Research, Technology and Environment, 9 (1995).
2. A. R. Sanadi, D. F. Caulfield, and R. M. Rowell, *Plast. Eng.*, April, 1994, p. 27.
3. A. R. Sanadi, D. F. Caulfield, R. E. Jacobson, and R. M. Rowell, *Ind. Eng. Chem. Res.*, **34**, 1889 (1995).
4. H. Dalvag, C. Klason, and H. E. Stromvall, *Int. J. of Polymeric Materials*, 11 (1985).
5. R. G. Raj and B. V. Kokta, *J. Appl. Polym. Sci.*, **18** (1989).
6. B. V. Kokta, R. G. Raj, and C. Daneault, *Polym.-Plast. Technol. Eng.*, **28**, 247 (1989).
7. A. R. Sanadi, C. Clemons, R. M. Rowell, and R. A. Young, *J. Reinforced Plast. Compos.*, **13**, 54 (1994).
8. J. H. Felix and P. Gatenholm, *J. Appl. Polym. Sci.*, **50**, 699 (1993).
9. P. Zadorecki and P. Flodin, *J. of Appl. Polym. Sci.*, **30**, 10 (1985).
10. P. Gatenholm, J. Felix, C. Klason, and J. Kubat, *Cellulose-Polymer Composites with Improved Properties, In Contemporary Topics in Polymer Science*, **7**, J. C. Salamone, ed., Plenum Press, New York (1992).
11. B. Moginger, U. Muller, and P. Eyerer, *Composites*, **22**, 432 (1991).
12. P. Jarvela, S. Li, and P. Jarvela, *J. Appl. Polym. Sci.*, **26**, 813 (1996).
13. G. McCrum, B. E. Read, and G. Williams, *Anelastic and Dielectric Effects in Polymer Solids*, pp. 377–87, John Wiley & Sons Ltd., London (1967).
14. R. P. Chartoff, *The Application of Dynamic Mechanical Methods to Tg Determination in Polymers: An Overview, in Assignment of the Glass Transition*, J. R. Seyler, ed., ASTM STP 1249 (1994).
15. B. Wunderlich, *The Nature of Glass Transition and its Determination by Thermal Analysis, in Assignment of the Glass Transition*, R. J. Seyler, ed., ASTM STP 1249 (1994).
16. H. Boyd, *Polymer*, **26**, 323 (1985).