Short Jute Fiber-Reinforced Polypropylene Composites: Effect of Compatibilizer

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Received 7 June 1996; accepted 29 December 1997

ABSTRACT: Jute fibers were chopped to approximately 100 mm in length and then processed through a granulator having an 8-mm screen. Final fiber lengths were up to 10 mm maximum. These fibers along with polypropylene granules and a compatibilizer were mixed in a K-mixer at a fixed rpm, 5500, and dumped at a fixed temperature, 390°F, following single-stage procedure. The fiber loadings were 30, 40, 50, and 60 wt %, and at each fiber loading, compatibilizer doses were 0, 1, 2, 3 and 4 wt %. The K-mix samples were pressed and granulated. Finally, ASTM test specimens were molded using a Cincinnati injection molding machine. At 60% by weight of fiber loading, the use of the compatibilizer improved the flexural strength as high as 100%, tensile strength to 120%, and impact strength (unnotched) by 175%. Remarkable improvements were attained even with 1% compatibilizer only. Interface studies were carried out by SEM to investigate the fiber surface morphology, fiber pull-out, and fiber–polymer interface.
 J Appl Polym Sci 69: 329–338, 1998

Key words: jute; polypropylene; maleated polypropylene; compatibilizer; composite

INTRODUCTION

The art of making composites is restricted mainly to synthetic polymer and synthetic reinforcing fibers like glass, carbon, and nylon. The potentiality of natural fiber-based composites using cellulose, wood, jute, kenaf, hemp, sisal, pineapple, coir, etc., as reinforcing fiber in a thermosetting resin matrix has received considerable attention among scientists all over the world[1–5] for their excellent specific properties. Some of them have already been used as industrial products. Composites, based on thermoplastic resins, are now becoming popular due to their processing advantages.[6,7] The mechanical properties of thermoplastics are often inadequate for specific applications, and for this, different man-made fibers, mainly glass or other mineral fillers like china clay, talc, and $CaCO_3$, are mixed. On the other hand, jute is an annually regenerative, lignocellulosic self-composite biopolymer bast fiber.[8] It is nonabrasive, has low density and high strength and is abundantly available in tropical countries and is, therefore, of particular interest for use as a reinforcing fiber in thermoplastic composites.

Among commodity thermoplastics, polypropylene (PP) possesses outstanding properties like low density, high vicat softening point, good flex life, sterilizability, good surface hardness, scratch resistance, very good abrasion resistance, and excellent electrical properties.[9] Jute is predominantly polar due to the presence of various polar groups on its backbone.[10] On the contrary, PP is

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Contract grant sponsor: UNDP; contract grant number: IND/92/302.

Journal of Applied Polymer Science, Vol. 69, 329–338 (1998)
 CCC 0021-8995/98/020329-10

nonpolar.[11] So, for better interaction between the two otherwise incompatible surfaces, the presence of a compatibilizer was envisaged.[12] The grafting of dicarboxylic anhydrides onto polyolefins and their use as compatibilizers have appeared in the literature.[13–16] Comparative rhelogical studies on jute fiber and glass fiber-filled PP composite melts[17] revealed that both follow the power law relationship; however, little additional power is required to mold the filled materials. Work on Coir–PVC/PP[18] reported no remarkable increase in mechanical properties. The development of high-density polyethylene-impregnated jute cloth boards resulted in a 44–54% increase in tensile strength.[19] As jute is abundantly available in India, it is worthwhile to study jute–PP composites with an aim to achieve comparable properties of other filled PP composites. The objectives of this work were (1) to obtain various mechanical properties of the composites at different fiber loadings and (2) to determine the optimum dose of the compatibilizer for the jute–PP system.

EXPERIMENTAL

Materials

Jute fiber was grade W-2, Chorchorus Capsularies); the compatibilizer, Epolene, G-3002, Eastman (maleated polypropylene); and the polypropylene homopolymer (PP), Fortilene PP, 1602 (MFI 12.0, Solvay Polymers).

Methods

Jute fibers were first chopped to a length of approximately 100 mm by a guillotine machine. These were then fed to the granulator having an 8-mm screen. The granulator reduced the feed up to a length of 10 mm maximum. A calculated quantity of granulated jute fiber (on a dry basis), the compatibilizer, and PP were fed into the K-mixer. Jute fiber loadings were 30, 40, 50, and 60 wt % and the compatibilizer dose varied from 0, 1, 2, 3, and 4 wt %. Throughout the experiments, the parameters of the K-mixer of the dump temperature, 390°F; rpm, 5500; mixing time, 45–60 s; and batch size, 125/150g were retained. Immediately after dumping from the K-mixer, the dough was pressed into a cold hydraulic press so as to increase the surface area for fast cooling to avoid fiber burning. These jute fiber-encapsulated PP cakes were then cut into small pieces by a band saw and fed into the granulator for size reduction.

These jute–PP granules were dried at 105°C for 4 h and then molded using a Cincinnati Milacron 33T injection-molding machine to mold the ASTM test pieces for Izod impact, flexural strength, and tensile strength. The testing speed for flexural and tensile strengths was 5 mm/min. This testing was using an Instron 4303. The micrographs were taken by a scanning electron microscope (SEM), Leica S 440.

Nomenclature

$J\ XYZ$, where J = Jute fiber, XY = wt % of dry jute fiber, and Z = wt % G-3002. Values for XY = 30, 40, 50, and 60, and values for Z = 0, 1, 2, 3, and 4.

RESULTS AND DISCUSSION

During mixing and molding, no significant processing problem was noticed. An all around remarkable improvement in the mechanical properties of the resultant composites using the compatibilizer was observed.

Specific Gravity

The specific gravity of the composites varied from 1.02 (for J 300 series) to 1.10 for (J 600 series) as compared to 0.89 for PP, 1.23 for 40% glass, and 1.25 for 40% $CaCO_3$-loaded PP composites.[20] Due to the low specific gravity of jute–PP composites, their specific mechanical properties became comparable even to glass–PP composites.[20]

Water Absorption

Water absorption values for a 24 h cold soak and 2 h in boiling water, are shown in Table I. Water-

Table I Percent Water Absorption (Based on Initial Weight)

Conditions	PP	J300	J302	J400	J402	J500	J502	J600	J602
2 h in boiling water	0.1	0.93	0.77	1.5	1.16	2.12	1.47	3.06	2.22
24 h in cold water	0.03	0.56	0.42	0.61	0.44	1.07	0.6	1.86	0.91

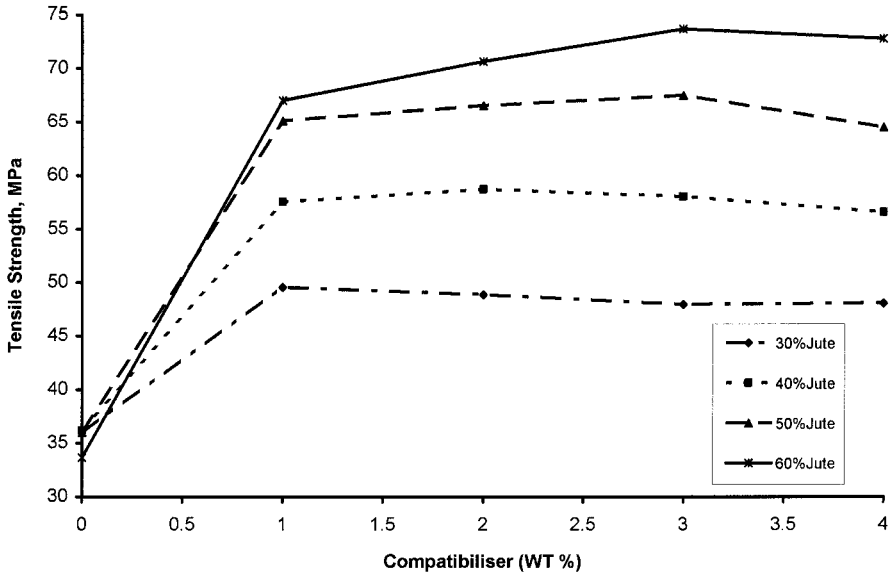

Figure 1 Tensile strength versus compatibilizer percent.

absorption values were found increased with increase in fiber loading. Use of the compatibilizer, however, decreased water absorption at the same fiber loading, which might be attributed to some of the hydrophilic –OH groups reacting with acid anhydride to form ester linkages and thereby giving lower water-absorption values. Similar observations were also observed by Rowell et al.[21] during acetylation of wood and Hedenberg and Gatenholm[15] during conversion of plastic cellulose waste into composites.

Tensile Strength and Modulus

Figure 1 shows the improvements in tensile strength with the compatibilizer. It was observed that at 0% compatibilizer the tensile strengths of the composites were 33.65–36.17 MPa at a fiber

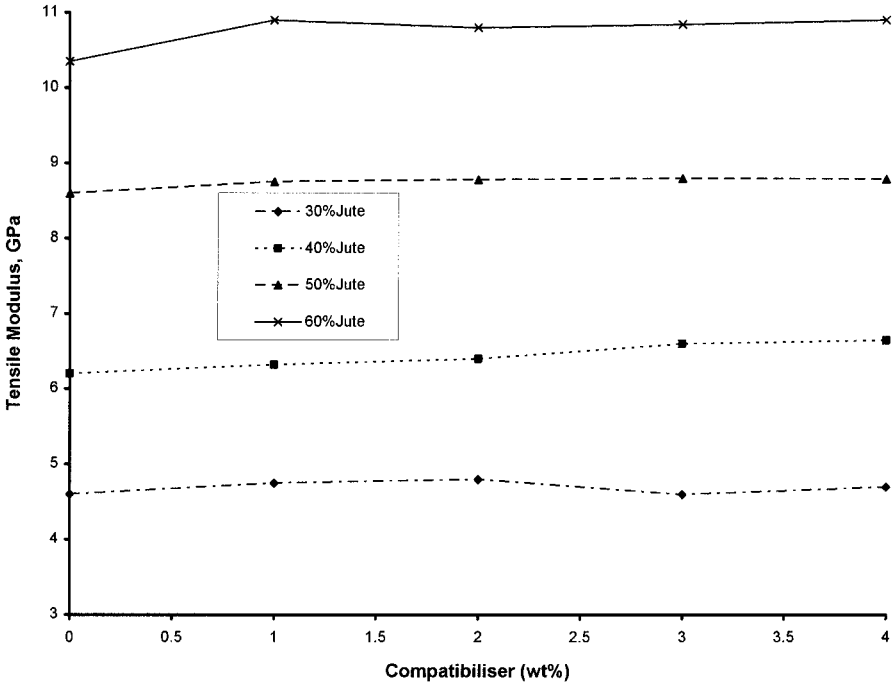

Figure 2 Tensile modulus versus compatibilizer percent.

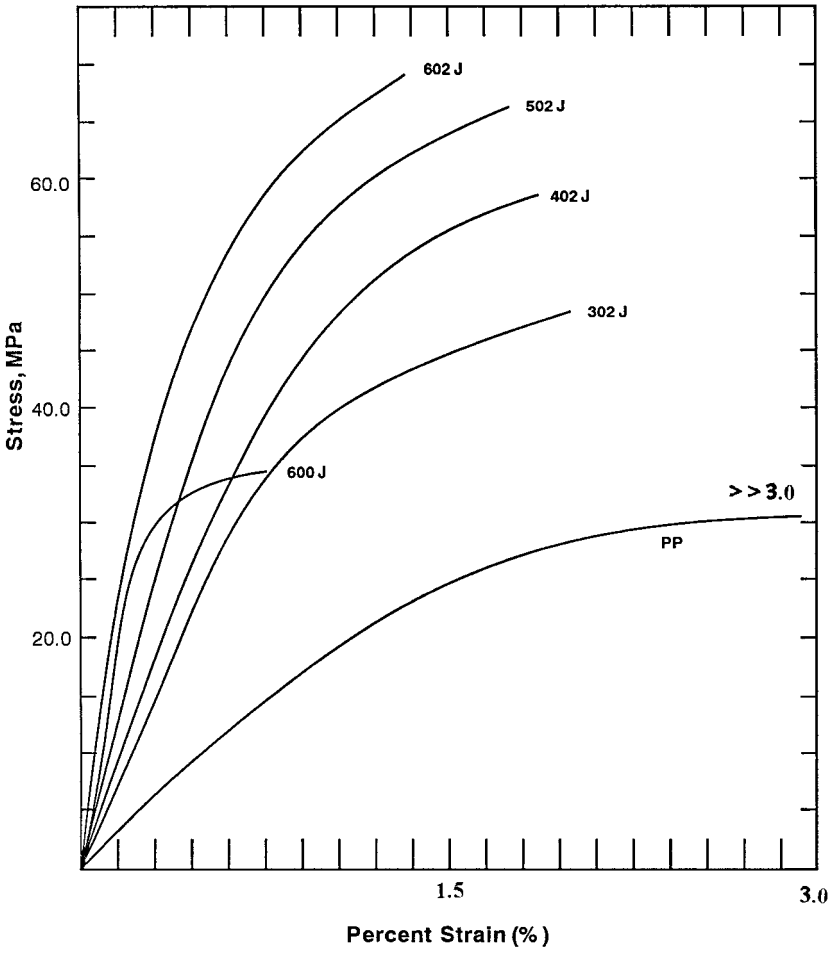

Figure 3 Stress versus strain curves of PP, jute–PP, compatibilized, and uncompatibilized systems.

loading of 30–60%, suggesting that there was little stress transfer from the matrix to the fibers irrespective of the amount of fiber present. When only 1% compatibilizer was added, the strengths increased phenomenally. Further increase in the compatibilizer beyond 1% had virtually little effect. The effect was greater in composites having more fiber contents up to 73.71 MPa for 60% fiber loading. The percent maximum increase in the tensile strengths for the J 300, J 400, J 500, and J 600 series were 50.15, 62.34, 87.48, and 119.05, respectively. When the compatibilizer was added, the sharp rise in strength was due to stress transfer from the matrix to the fiber via the compatibilizer.[22] The increase in fiber content restricted the mobility of the PP chain and this was reflected in the tensile moduli values, although addition of the compatibilizer in the same series did not alter the values appreciably. The moduli for the J 300 to J 600 series were 4.60, 6.2, 8.6, and 10.35 GPa, respectively.

The tensile modulus versus the compatibilizer percent graph is shown in Figure 2. From the figure, it is evident that, on average, 2–3% compatibilizer was optimum. Typical comparative stress versus strain curves of PP, jute–PP (compatibilized and uncompatibilized systems) are shown in Figure 3, where a decrease in failure strain was observed with an increase in fiber content due to restrictions posed by the fiber. However, addition of the compatibilizer helped in stress transferring from the matrix to the fiber and is observed from the figure.

Flexural Strength and Flexural Modulus

The change in flexural strength and flexural modulus, with the compatibilizer, are shown in Figures 4 and 5, respectively. A significant increase in these values was observed. The percent maximum increase in flexural strengths for the J 300 to J 600 series were 31.66, 44.09, 64.30, and 90.88,

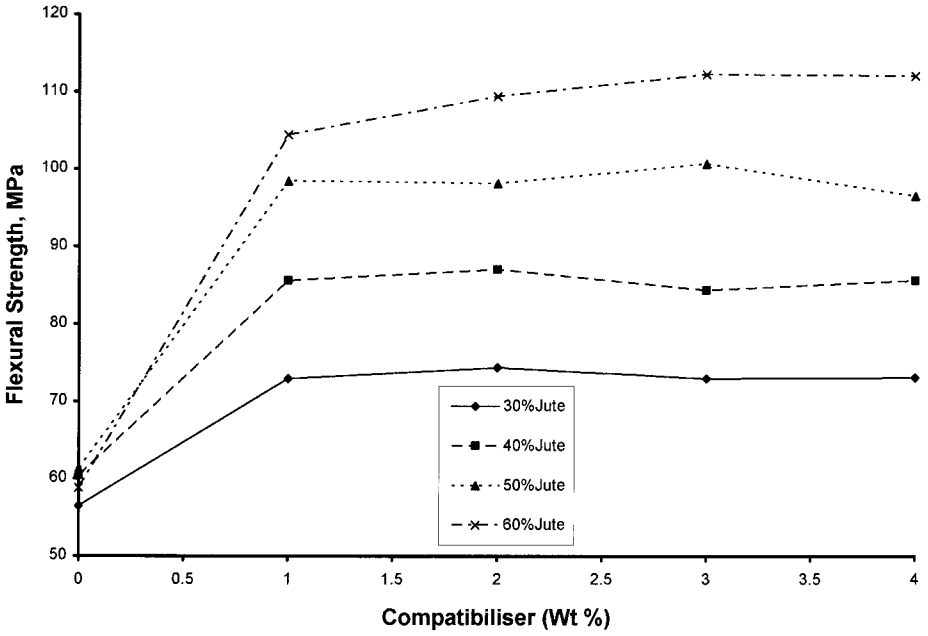

Figure 4 Flexural strength versus compatibilizer percent.

respectively. At 0% compatibilizer, there was virtually no change in the flexural strength (56.48–61.29 MPa) with increased fiber loading from 30 to 60%, indicating, therefore, that there was little interfacial bonding for the transfer of the load from the matrix to the fibers. Yet, when the compatibilizer was added, sufficient interfacial bonding had occurred between the fibers and the matrix. From the values, an optimal dose of 2–3% compatibilizer was observed. The trend of the flexural modulus was identical to the tensile modulus described earlier where the compatibilizer had no effect. However, the effect of the fiber content was obvious.

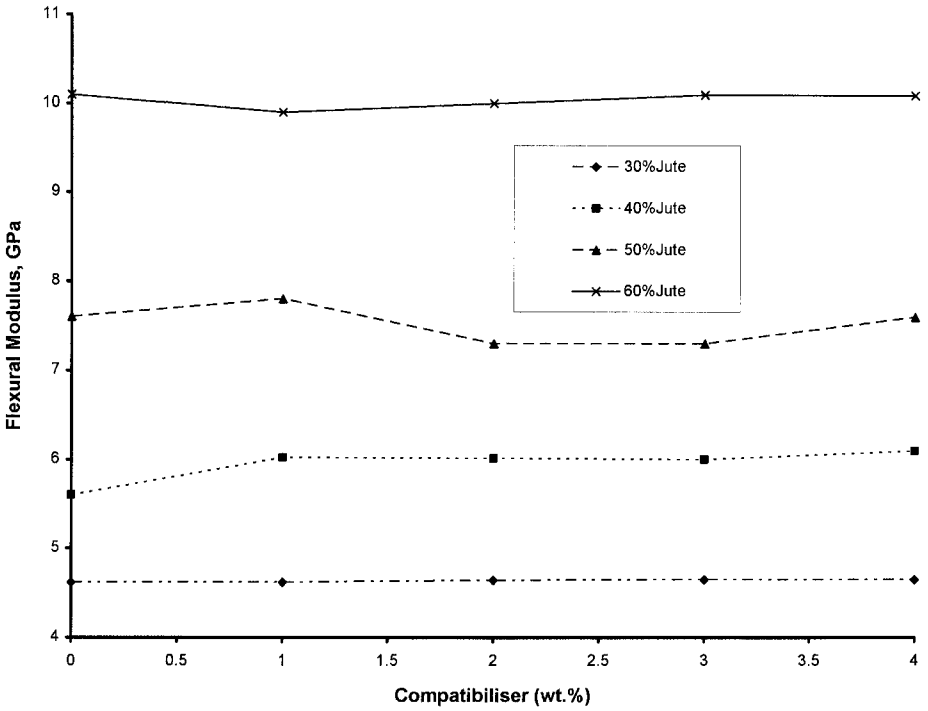

Figure 5 Flexural modulus versus compatibilizer percent.

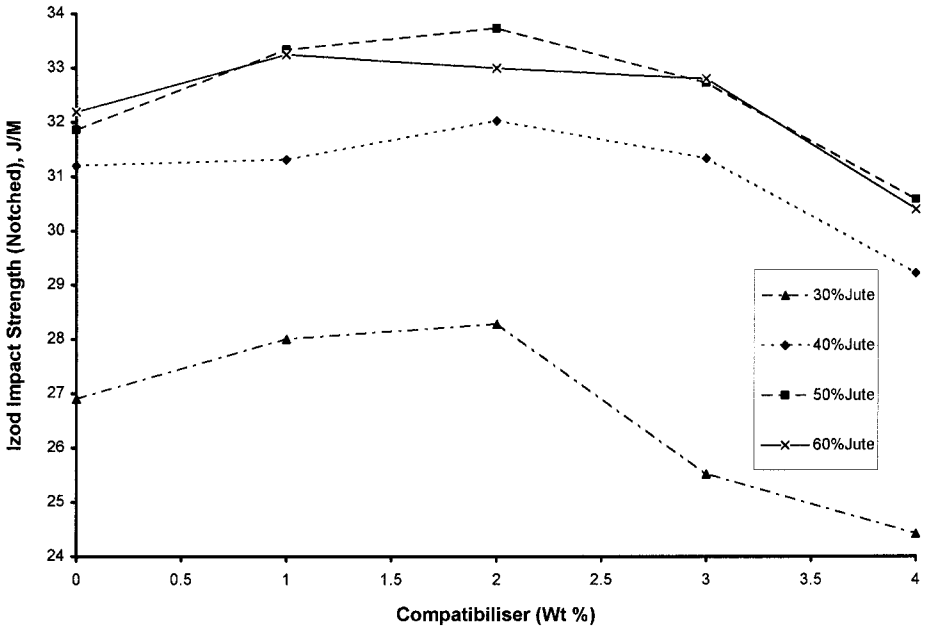

Figure 6 Izod impact strength (notched) versus compatibilizer percent.

Impact Strength

The effect of the compatibilizer on the impact strength is shown in Figures 6 and 7 for notched and unnotched samples, respectively. The notched values were found to increase from 26.9 J/m (for 30% fiber) to 33.73 J/m (for 60% fiber). The effect of the percent compatibilizer on the notched impact strengths was negligible with marginal improvement up to 3–4%, beyond which the deterioration in this property was observed having its effect more pronounced at 30% fiber content. This could be attributed to the migration of too much compatibilizer around the fibers, causing self-entanglement among the compatibilizers rather than the polymer matrix, resulting in slippage.[22,23]

The effect was quite different in the case of the unnotched samples. The values at 0% compatibilizer were 70.7 J/m (60% fibers) and 195.6 J/m (30% fiber). The low value at high fiber content might be due to the presence of too many fiber ends within the body of the composites, which could cause crack initiation and, hence, potential composite failure.[24] The addition of the fibers also increased the probability of fiber agglomeration[23] which created regions of stress concentrations that required less energy to initiate a crack. Similar observations were also reported elsewhere.[22,24] However, in the presence of the compatibilizer, the fibers themselves could modify the microstructure of the surrounding matrix and, thus, there was a sharp increase in the unnotched impact strength. It was apparent that even with 30% fiber content and 1% compatibilizer, the unnotched impact strength was more or less equivalent to 60% fiber content. Unlike notched specimens, no deterioration was observed beyond 3% compatibilizer addition. In this case, the compatibilizer migration around the fibers acted as a damper to the shock wave, imparted during the impact that was transmitted onto the fibers evenly.

Tensile Energy Absorption (TEA)

TEA is the area under the stress–strain curve up to failure. The effect of the compatibilizer on the TEA at different fiber and compatibilizer loadings is shown in Figure 8. The TEA was found to decrease with increase in fiber loading. Increase in fiber loading restricted the mobility of the polymer chains and this caused a decrease in the failure strain and TEA also.[22] However, with the addition of the compatibilizer, a sharp rise in the TEA was also observed. This might be due to the better interaction at the jute–PP interface in the presence of the compatibilizer. This was observed up to 3% compatibilizer. At 4% compatibilizer, the engineering trend was found to be downward. This was probably due to the migration of too much of the compatibilizer around the fiber surfaces, which caused slippage as described earlier.

Interface Study

Interface studies were carried out to investigate the fiber surface morphology, fiber pull-out, and

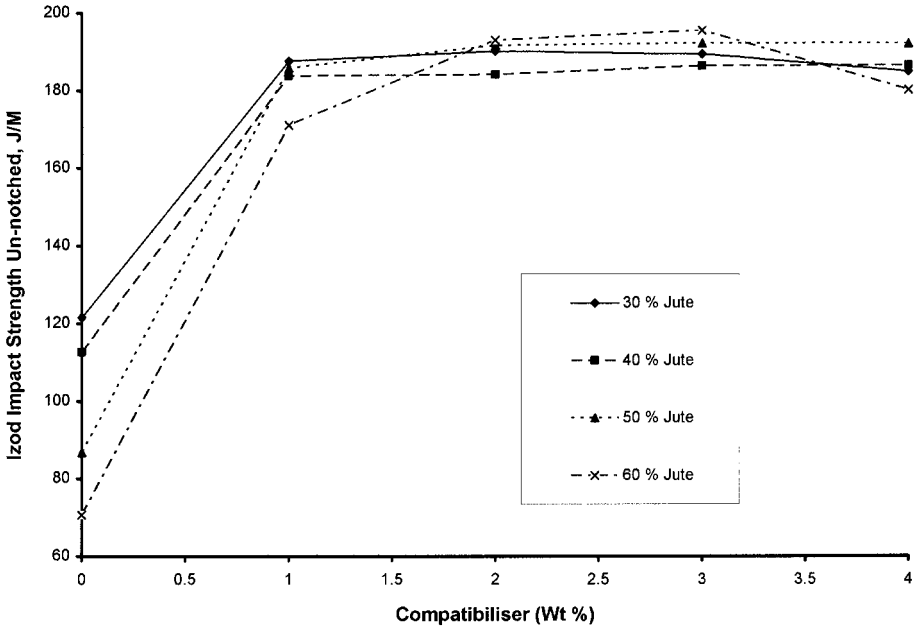

Figure 7 Izod impact strength (unnotched) versus compatibilizer percent.

fiber–polymer interface by an SEM. Figures 9 and 10 show the micrographs of the tensile-fractured specimens of uncompatibilized and compatibilized systems, respectively. In Figure 9, a large number of holes resulting from the fiber pull-out from the matrix is evident. In Figure 10, a considerably fewer number of such holes and many broken fiber ends, embedded in the polymer matrix, is evident. Figures 11 and 12 show the surface morphology of the pull-out fiber. From the figures, it is clear that the adhesion between the fiber and matrix was better for the compatibilized system.

Correlation of Mechanical Properties with Other Factors

For understanding the stiffness and strength of short-fiber-reinforced thermoplastics between the

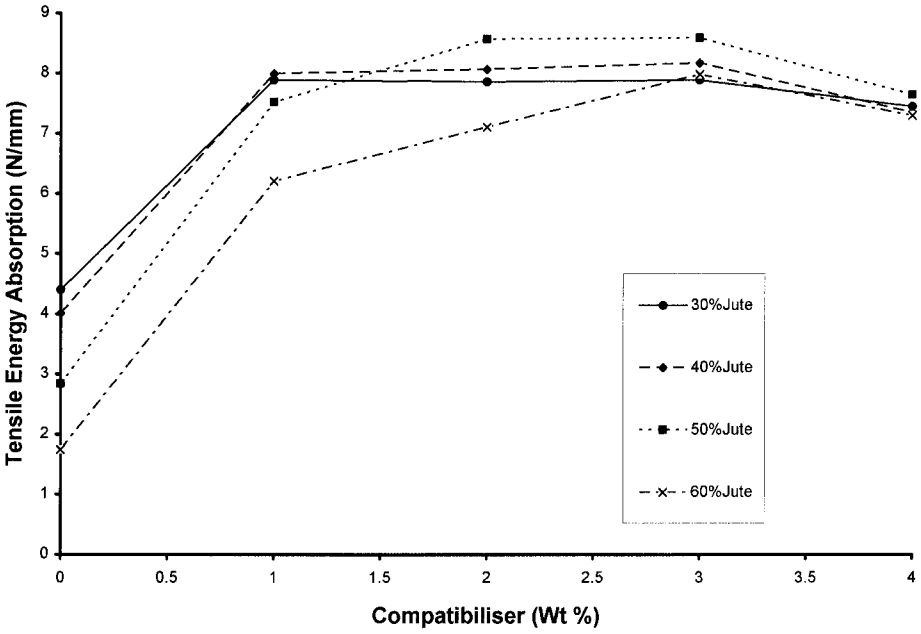

Figure 8 TEA versus compatibilizer percent.

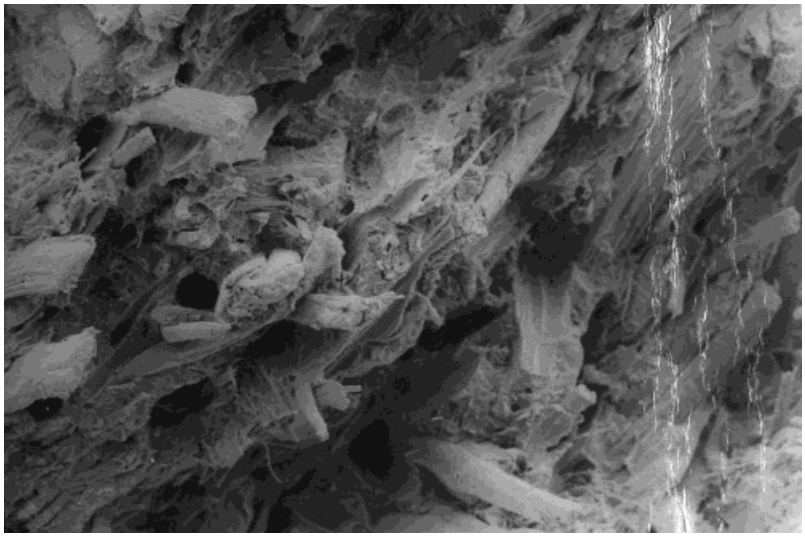

Figure 9 SEM micrograph of tensile fractured specimen of uncompatibilized system.

non-polar PP and polar jute fiber, several factors are very influential. These are dispersion, adhesion, fiber breakage and agglomeration, fiber distribution and orientation, the volume/weight fraction of the fibers, microstructure, etc. To improve dispersion and adhesion, use of maleic anhydride-grafted polyolefins have been reported by earlier workers.[15,25,26] The formation of covalent linkages between maleic anhydride and hydroxyl groups of cellulose was indicated by Hedenberg and Gatenholm[15] through IR and ESCA analysis. For better stress-transfer efficiency of the interface, sufficient maleic anhydride groups should be present in the compatibilizer for better interaction with the —OH group present on the fiber surface and the compatibilizer also should have a sufficient tail length for better entanglement with the matrix.[27] The K-mixer, used for mixing, caused substantial fiber breakage due to very high shear. The strengths obtained in our composites were, thus, limited by this fiber breakage. Use of the compatibilizer did not alter the moduli values substantially, probably due to the formation of a transcrystalline zone forming around the fiber in the case of the uncompatibilized system also. The complex fiber distribution and orientation also contributed to the high values of moduli and the value increased with the fiber percentage. At high fiber loading, the possibility of fiber agglomeration hindered the plastic flow process and the re-

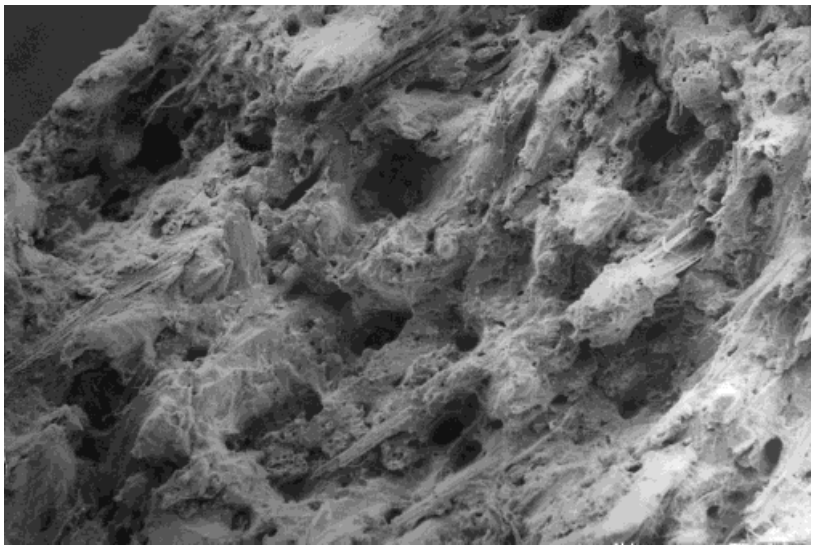

Figure 10 SEM micrograph of tensile fractured specimen of compatibilized system.

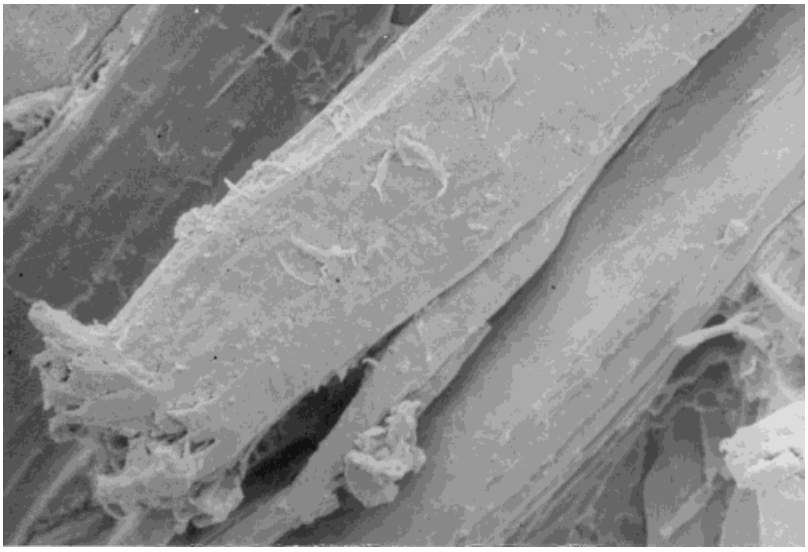

Figure 11 SEM micrograph of pull-out fiber of uncompatibilized system.

sultant composite had less TEA, as evident in Figure 8. In the presence of the compatibilizer, the fibers themselves could modify the microstructure of the surrounding matrix as evident from the same studies.

CONCLUSION

The following conclusions may be drawn from the above studies:

1. The sharp increase in mechanical properties and decrease in water absorption values after addition of the compatibilizer might be attributed to linkage between the hydrophobic hydroxyl groups of jute and the carboxyl groups of the compatibilizer.
2. All these results justify that the role of jute fiber was not as a filler fiber but as a reinforcing fiber in a properly compatibilized system.
3. This system produced a new range of low-energy, low-cost composites having interesting properties and should be given priority over costly and high-energy synthetic reinforcing fiber wherever possible.
4. The drawback of this system was that it had low impact strength as compared to 40%

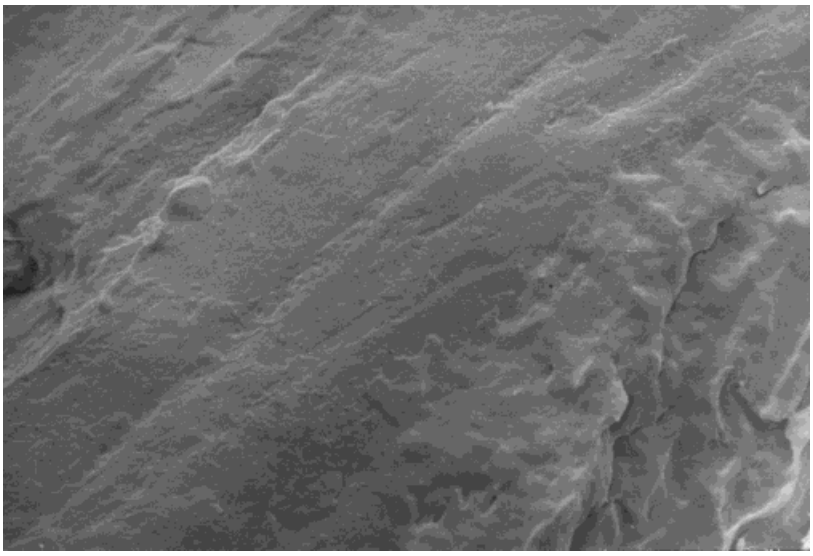

Figure 12 SEM micrograph of pull-out fiber of compatibilized system.

glass-filled PP,[20] but there was immense scope for improvement by using an impact modifier.

5. The jute–PP system might not be recommended for electrical applications due to high water absorption values, as compared to a glass-, mica-, $CaCO_3$-, or talc-loaded system.

The authors acknowledge UNDP grant (Project No. IND/92/302) for support of this research work. Prof. K. Jayachandran, Director, IJIRA, is also acknowledged for his kind permission to publish this article.

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