Short Jute Fiber-Reinforced Polypropylene Composites: Dynamic Mechanical Study

A. K. RANA,¹ B. C. MITRA,^{1,*} A. N. BANERJEE²

¹ Indian Jute Industries' Research Association, 17 Taratola Road, Calcutta-700 088, India

² Department of Polymer Science and Technology, University of Calcutta, 92 A.P.C. Road, Calcutta-700 009, India

Received 27 January 1998; accepted 18 April 1998

ABSTRACT: Short jute fiber-reinforced polypropylene (PP) composites were prepared using a high-speed thermokinetic mixer. A compatibilizer was used to improve the molecular interaction between jute and PP. Both the percent weight fraction of the jute fiber and compatibilizer were varied to study the dynamic mechanical thermal (DMT) properties. Dynamic parameters such as storage flexural modulus (E'), loss flexural modulus (E'), storage shear modulus (G'), loss shear modulus (G''), and loss factor or damping efficiency (tan δ) were determined in a resonant frequency mode. The transition peak nature, amplitude, and temperature of E', E'', G', G'', and tan δ of different compositions were shown to indicate possible improvements of molecular interaction in the presence of a compatibilizer. The modulus retention term, a plot of the reduced modulus with the weight fraction of the jute fiber, also indicate its improvement. © 1999 John Wiley & Sons, Inc. J Appl Polym Sci 71: 531–539, 1999

Key words: jute thermoplastics composite; polypropylene; maleated polypropylene; compatibilizer; dynamic mechanical thermal analysis

INTRODUCTION

The art of making composites is restricted mainly to synthetic polymers and synthetic reinforcing fibers like glass, carbon, and nylon. The potential of natural fiber-based composites using cellulose, wood, jute, kenaf, hemp, sisal, pineapple, coir, etc., as reinforcing fibers 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 in 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, of low density and high strength, and 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

Correspondence to: A. N. Banerjee.

^{*} Present address: NIRJAFT, 12 Regent Park, Calcutta-700 040, India.

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

Journal of Applied Polymer Science, Vol. 71, 531-539 (1999)

^{© 1999} John Wiley & Sons, Inc. CCC 0021-8995/99/040531-09

Serial No.	Sample ID	PP (% by Weight)	Jute Fibre (% by Weight)	Compatibilizer (% by Weight)
1	РР	100	_	_
2	J300	70	30	_
3	J302	68	30	2
4	J400	60	40	_
5	J402	58	40	2
6	J500	50	50	_
7	J501	49	50	1
8	J502	48	50	2
9	J503	47	50	3
10	J504	46	50	4
11	J600	40	60	_
12	J602	38	60	2

Table I Compositions of the Various Jute-PP Composites

nonpolar.¹¹ So for better interaction between the two otherwise incompatible surfaces, the presence of a compatibilizer was envisaged.¹² Grafting of dicarboxylic anhydrides onto polyolefins and their use as compatibilizers have appeared in the literature.^{13–16}

With the advent of different agro-based thermoplastic composites for different applications for which dynamic data are directly relevant (e.g., machinery, transport, buildings, domestic appliances, and acoustic devices), it is worthwhile to study the polymer composite structure to investigate the dynamic mechanical thermal properties, particularly the dynamic modulus and internal friction, over a wide range of temperature.^{17–20}

As it is known from the DMTA principle when internal molecular motion occurs, the material response in a viscoelastic manner and the strain response lags behind the stress. The tangent of this phase lag is expressed as tan δ and is also known as the damping factor. The damping properties of a material represent its capacity to reduce the transmission of vibration caused by mechanical disturbances to a structure. The DMT properties are mostly dependent upon the amount of fiber,^{21,22} the presence of additives like the compatibilizer, filler, and impact modifier,²³ fiber orientation,^{24,25} and mode of testing. Over a wide range of temperature polymeric materials usually exhibit more than one relaxation region or socalled transition.¹⁹ In this study, the fibers were randomly placed and our aim was to compare different dynamic parameters at different levels of the fiber within the experimental range. The effect of a compatibilizer was also studied to obtain fundamental information regarding molecular interaction using the viscoelastic properties in a dynamic condition.

EXPERIMENTAL

Materials

The materials used are as follows: (1) jute fiber (Grade W-2, Chorchorus Capsularies); (2) compatibilizer—Epolene, G-3002 ($M_n = 20,000; M_w = 60,000$), Eastman (maleated polypropylene); (3) polypropylene homopolymer (PP)—Fortilene PP, 1602 (MFI 12.0, Solvay Polymers).

Methods

Jute fibers were granulated using a Ball and Jewell granulator (Sterling Inc., Milwaukee, WI) having a 4-mm screen. The output was reduced to a maximum length of 5 mm. The calculated quantity of the granulated jute fiber (on a dry basis), compatibilizer, and PP were mixed in a K-mixer. The jute fiber loadings were 30, 40, 50, and 60 wt % and the compatibilizer doses were 0, 1, 2, 3, and 4 wt %.

Throughout the experiments, the following parameters of the K-mixer were retained: dump temperature = 199° C, rpm = 5500, mixing time 45-60 s, and batch size 125/150 g. Immediately after dumping from the K-mixer, the dough was pressed in the cold hydraulic press so as to increase the surface area for fast cooling to avoid fiber burning. These jute-PP cakes were then cut into small pieces by a band saw and then fed to



Figure 1 Variation of storage flexural modulus of jute–PP composite with temperature at different jute fiber content.



Figure 2 Variation of storage shear modulus of jute–PP composite with temperature at different jute fiber content.



Figure 3 Variation of storage flexural modulus of jute–PP composite with temperature at different compatibilizer levels and at 50% by weight jute fiber content.



Figure 4 Variation of storage shear modulus of jute–PP composite with temperature at different compatibilizer levels and at 50% by weight jute fiber content.





Figure 5 Variation of storage flexural modulus of jute–PP composite (control and 2% compatibilized).

the granulator for size reduction to form granules suitable for injection molding.

The jute–PP granules were dried at 105°C for 4 h before molding using a Cincinnati Milacron 33T injection-molding machine to mold a specimen bar of a dimension of $50 \times 12.5 \times 3.0$ mm at 190°C and a 1200 psi clamping pressure. The compositions of the various jute/PP composites are shown in Table I. The dynamic mechanical properties were measured by using a DuPont DMA 983. The testing parameters were as follows: mode, resonant frequency; sample size, 12.5 \times 3.0 mm; clamping distance, 10 \times thickness; oscillation amplitude, 0.2 mm; heatng rate, 5°C/ min; and temperature range, 35–160°C. In all cases, four specimens were tested and the average values are reported.

RESULTS AND DISCUSSION

In Figure 1, the variation of E' at different weight fractions of the fiber is shown as a function of temperature. It can be seen from the figure that the E' of PP is much lower than that of the jute-PP system. With a gradual addition of the

Figure 6 Variation of reduced modulus of jute–PP composite with jute fiber content.

jute fiber, the E' values are also increased sharply and gradually. This is due to the reinforcement imparted by the fiber that allows stress transfer from the matrix to the fiber. With the addition of 2% compatibilizer, the E' value is found to increase more than for the uncompatibilized system (J500 and J502). This phenomenon was observed uniformly throughout the experi-

Table IIVariation of Modulus Retentionwith Weight Fraction of Jute Fiber(Control and 2% Compatibilized)

	Jute Fiber (% Wt)	Modulus Retention	
Sample ID		${E_{110}''}^{\circ}/{E_{35}'}^{\circ} imes 100$	${E'_{160}}^{\circ}/{E'_{35}}^{\circ} imes 100$
J600	60	53.50	18.94
J602	60	57.12	20.16
J500	50	59.80	16.08
J502	50	53.49	17.07
J400	40	49.50	14.28
J402	40	50.70	15.81
J300	30	43.55	13.74
J302	30	43.68	13.85
PP	—	25.03	3.77



Figure 7 Variation of tan δ of jute-PP composite with temperature at different jute fiber content.



Figure 8 Variation of tan δ of jute-PP composite with temperature at different compatibilizer levels and at 50% by weight jute fiber content.

ment. This may be due to the migration of the compatibilizer to the fiber surface and the linking with the matrix by its tail, resulting in a stiffer combination. Our studies on the mechanical properties was also in agreement with this.²⁶ At 160°C, PP lost its E' to a very low level, whereas compounds having jute fiber reinforcement have some strength depending on the fiber content (E' of J600 and PP at 160°C are 0.9677 and 0.06201 GPa, respectively).

Figure 2 shows the variation of G' with temperature at different weight fractions of the fiber. The nature of the curves are found to be very similar to that of Figure 1 and the same explanation is true for this case also.

In Figure 3, the variation of E' at different compatibilizer levels, 0-4%, is shown at a 50% weight fraction of the fiber content. Earlier work on cellulose/lignocellulose thermoplastic composites^{15,27,28} revealed that during compounding the compatibilizer migrates to the fiber to form a bond and the tail of the compatibilizer became entangled in the PP matrix. In the figure, the same phenomenon is observed. With increase in the compatibilizer percent, the E' values are increased. This may be due to increase in the bulkiness of the fiber due to formation of the bond between the compatibilizer and the fiber. For the same reason, the plot of G' versus temperature (Fig. 4) at different compatibilizer levels also shows the same trend.

The better interaction between the fiber and polymer in the presence of the compatibilizer can also be understood from Figure 5, where E' is plotted against the weight fraction of the jute fiber at 35 and 110°C. It shows that with increase in the fiber content the graphs become steeper for the compatibilized system. The modulus enhancement of both the control and the compatibilized system with the fiber weight fraction is also demonstrated by the relative flexural storage modulus (E'_c/E'_m) , where E'_c and E'_m are the E's of the composite and matrix, respectively) in Figure 6. At high fiber content, the compatibilized system shows the higher value at a particular fiber content. At 110°C, this is more prominent than at 35°C. This may be because the poor fiber polymer interaction in the absence of the compatibilizer is exaggerated at higher temperature.

Table II shows the variation of the modulus retention term²⁹ of the composites with the weight fraction of the fiber. It is evident from the table that with increase in fiber content the mod-



Figure 9 Variation of loss flexural modulus of jute–PP composite with temperature at different jute fiber content.

ulus retention values increase and further increase with further addition of the compatibilizer.

Figures 7 and 8 show the variation of tan δ with temperature for various weight fractions of the fiber and compatibilizer. In Figure 7, it is seen that the introduction of fibers has reduced the peak height that is, tan δ is lowered with increase in the fiber content. This phenomenon also was observed by earlier workers.^{30,31} The reason may be due to the restriction of the mobility of the polymer chains by the fiber.

The other prominent effect is broadening of the transition region. This may be due to the inhibition of the relaxation process within the composites upon incorporation of the fibers. In Figure 8, no appreciable change in the tan δ values are observed at different compatibilizer levels.

Figures 9 and 10 show the variation of E'' with temperature for different fiber contents and for different compatibilizer levels. If the applied mechanical energy (work) is not stored elastically, it must be lost—converted to heat through molecular friction, that is, viscous dissipation—within the material. This is, precisely, the loss modulus.³² It is seen that with gradual introduction of



Figure 10 Variation of loss flexural modulus of jute–PP composite with temperature at different compatibilizer level and at 50% by weight jute fiber content.

the fibers the transitional peak is also gradually shifted to a higher region (Fig. 9). From J300 to J600, a shift of 10°C is observed. In the experimental range, a tiny hump at 80°C for PP and a broad hump for J600 at 110°C are also observed. This may be explained as movement toward the T_g of the dry cellulose of the jute fiber which is about 200°C.³³ In Figure 10, between J500 and J504, the shift in the transition peak temperature is only 4°C. However, in both cases, there is an increase in the peak height with the addition of the fiber and compatibilizer.

CONCLUSIONS

The following conclusions may be drawn from the above studies for PP reinforced by jute fiber:

- 1. The E' increases with increase in fiber content. With addition of the compatibilizer, a further increase in values is also observed.
- 2. The transition, that is, the tan δ peak, is shifted to higher temperature as the amount of fiber loading increases.

- 3. Marginal shifting of the transition is observed with increase in the compatibilizer dose at the same level of fiber.
- 4. Efficiency of the compatibilized system increases with increase in the amount of fiber as compared to the uncompatibilized system. This is shown by a reduced modulus and a modulus retention term (Fig. 6 and Table II).

Future work on this subject may be extended to some advanced grade of PP, polyolefin elastomers, etc.

Samples were prepared at the Forest Products Laboratory, USA. The UNDP grant (Project No. IND/92/302) for carrying out this work is highly acknowledged. Prof. K. Jayachandran, Director, IJIRA is also acknowledged for his kind permission to publish this article.

REFERENCES

- Mitra, B. C.; Basak, R. K.; Sharkar, M. J Appl Polym Sci 1998, 67, 1093.
- Wells, H; Bowen, D. H.; Macphail, I; Pal, P. K. in 35th Annual Technical Conference 1980, Reinforced Plastics/Composites Institute, SPI, Sect I-F, 1980.
- 3. Pavithran, C.; Mukherjee, P. S.; Brahmakumar, M. J Reinf Plast Compos 1991, 10, 91.
- 4. Verma, I. K.; Ananthakrishnan, S. R.; Krishnamoorthy, S. Composites 1989, 20, 383.
- Ranganathan, S. R.; Pal, P. K.; Rana, A. K.; Mitra, B. C. Indian Pat. 170,305, 1990.
- Kodokian, G. K. A.; Kinloch, A. J. J Mater Sci Lett 1988, 7, 625.
- Hoggat, J. T.; Oken, S.; House, E. E. U.S. Air Force Report AFWAL-TR-80-3023, April 1980.
- Atkinson, R. R. Jute Fibre to Yarn, Temple, London, 1964, p. 31.
- Brydson, J. A. Plastic Materials, 3rd ed., Newnes Butterworths, London, 1975, Chap. 11.
- Lee, M.; Ed., International Encyclopedia of Composites, VCH, Vol. 4, Chap. on natural fiber composites.
- Encyclopedia of Polymer Science and Technology, Vol. 11, Propylene Polymers, Wiley-Interscience, New York, 1969.
- Stepek, J.; Daoust, H. Additives for Plastics (Polymers-5), Springer-Verlag, 1983, Chap. 4.
- Samay, G.; Nagy, T.; White, J. L. J Appl Polym Sci 1995, 56, 1423.
- Hyndryckx, F.; Dubois, P. H.; Patin, M.; Jerome, R.; Teyssie, P. H.; Garcia Marti, M. J Appl Polym Sci 1995, 56, 1093.

- Hedenberg, P.; Gatenholm, P. J Appl Polym Sci 1995, 56, 641.
- Sanadi, A. R.; Clemons, C.; Rowell, R. M.; Young, R. A. J Reinf Plast Compos 1994, 13, 54.
- Ferry, J. D. Viscoelastic Properties of Polymers, Wiley, New York, 1961.
- Murayama, T. Dyanamic Mechanical Analysis of Polymeric Materials, 2nd ed., Elsevier, Amsterdam, 1978.
- Read, B. F.; Dean, G. D. The Determination of Dynamic Properties of Polymers and Composites, Wiley, New York, 1978.
- Neilsen, L. E. Mechanical Properties of Polymers and Composites, Marcel Dekker, New York, 1975.
- Tsai, S. W.; Halpin, H. T. Introduction to Composite Materials, Technomic, Westport, CT, 1980.
- Adams, R. D.; Short, D. F. J Phys D Appl Phys 1973, 6, 1032.
- Kolarik, F.; Lednicky, F.; Pukanszky, B. in International Conference on Composite Materials, Vol. 6, London, 1987, pp. 452–461.

- Adams, R. D. Damping Properties Analysis of Composites, Vol. 1, Engineered Materials Handbook, ASTM International, Metals Park, OH, 1987, pp. 207–209.
- 25. Adams, D. F.; Doner, D. R. J Compos Mater 1967, 1, 4.
- Rana, A. K.; Mandal, A.; Mitra, B. C.; Jacobson, R.; Rowell, R.; Bannerjee, A. N. J Appl Polym Sci to appear.
- Felix, J. M.; Gatenholm, P. J Appl Polym Sci 1991, 42, 609.
- Raj, R. G.; Kokta, B. V.; Maldas, D.; Daneault, C. J Appl Polym Sci 1989, 37, 1089.
- Khanna, Y. P.; Wenner, W. M.; Kumar, R.; Kavesh, S. J Appl Polym Sci 1989, 38, 571.
- Saha, N.; Bannerjee, A. N.; Mitra, B. C. J Appl Polym Sci 1996, 60, 657.
- Saha, N.; Bannerjee, A. N. J Appl Polym Sci 1996, 62, 1199.
- Rosen, S. L. Fundamental Principles of Polymeric Materials, Wiley, New York, 1982.
- Alfthan, E.; de Ruvo, A.; Brown, W. Polymer 1973, 14, 329.