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# Impact toughness, viscoelastic behavior, and morphology of polypropylene-jute-viscose hybrid composites

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**ABSTRACT:** In this investigation, we studied the impact toughness and viscoelastic behavior of polypropylene (PP)–jute composites. In this study, we used viscose fiber as an impact modifier and maleated PP as a compatibilizer. The toughness of the composites was studied with conventional Charpy and instrumental falling-weight impact tests. The composites' viscoelastic properties were studied with dynamic mechanical analysis. The results show that the incorporation of viscose fibers improved the impact strength and toughness to 134 and 65% compared to those of the PP–jute composites. The tan  $\delta$  peak amplitude also increased with the addition of the impact modifier and indicated a greater degree of molecular mobility. The thermal stability of the composites was evaluated with thermogravimetric analysis. The addition of 2 wt % maleated polypropylene (MAPP) to the impact-modified composite improved the impact strength and toughness to 144 and 93%, respectively. The fiber–matrix morphology of the fracture surface and the Fourier transform infrared spectra were also studied to ascertain the existence of the type of interfacial bonds. Microstructural analysis showed the retention of viscose fibers in the composites compared to the more separated jute fibers. © 2015 Wiley Periodicals, Inc. J. Appl. Polym. Sci. **2016**, *133*, 42981.

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#### INTRODUCTION

In recent days, long-fiber-reinforced thermoplastics have been gaining broad attention, particularly in automotive applications.<sup>1</sup> Oksman et al.<sup>2</sup> developed high-performance naturalfiber-reinforced thermoplastic composites using a long-fiber thermoplastic extrusion technique. Ganster et al.3 also adopted a pultrusion technique using a conventional corotating twinscrew extruder for the preparation of thermoplastic composites. The development of high-performance composite materials has mainly focused on the achievement of a high modulus and strength, but from the perspective of the automotive application, a higher strength is not sufficient; the material's abilities to absorb energy and resist impact loading should also be important criteria.4,5 Oksman et al.'s2 study reported that long-jutefiber-reinforced polypropylene (PP) composites offered a higher stiffness compared to sisal-, banana-, and flax-reinforced PP composites. Sarkhel and Choudhury<sup>6</sup> also reported that jute fiber composites possesses high stiffness- and strength-to-weight ratios but had very low energy absorption capabilities.

Extensive research concerning the energy absorption of natural fiber composites is being carried out by many researchers. Oksman

and Clemon<sup>7</sup> investigated the effects of an elastomer–compatibilizer combination on the morphology and mechanical properties of PP–wood flour composites. They found that the combination of an elastomeric impact modifier and maleated polypropylene (MAPP) as a compatibilizer resulted in an increased toughness. Our earlier study<sup>8</sup> showed that the addition of 10 wt % viscose fibers with 2 wt % MAPP in PP–J30 (30 wt % jute fiber reinforced PP) composites achieved optimized mechanical properties, including toughness. The improvement of the composite energy absorption and toughness could be achieved with matrix modification,<sup>9,10</sup> the addition of impact modifier,<sup>11</sup> hybridization with tougher fibers,<sup>12</sup> optimization of the interface between the fibers and matrix with a coupling agent,<sup>13–16</sup> optimization of the fiber content,<sup>17</sup> and control of the fiber length and fiber orientation.<sup>18</sup>

Regenerated cellulose is a cellulose derivative that is usually based on wood.<sup>19</sup> Regenerated cellulose fibers are suitable for structural composites because of their quality and performance. Table I demonstrates the mechanical properties of different regenerated cellulose fibers.<sup>20–24</sup> Viscose fibers have a lower strength and stiffness but a higher elongation at break compared to, for example, Lyocell fibers. Therefore, it is expected that the

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Regenerated fibers	Density (g/cm <sup>3</sup> )	Tensile strength (MPa)	Young's modulus (GPa)	Elongation at break (%)	Reference	
Cordenka	1.8	770-890	19.0-21.0	11.0-15.0	Ganster et al. (2013)	20
Cotton	1.51	287-597	5.5-12.6	7.0-8.0	Ataollahi <i>et al</i> . (2011)	21
Lyocell	1.3	790-1400	30.5-36.0	6.0-10.3	Carrillo et <i>al</i> . (2010) and Ozcelik Kayseri et <i>al</i> . (2010)	22, 23
Modal	1.3	368-506	11.0-15.4	8.6-12.2	Ozcelik Kayseri et al. (2010) and Adusumali et al. (2006)	23, 24
Viscose <sup>a</sup>	1.3	293-323	7.0-15.0	23.0-25.0	Ganster et al. (2013) and Ozcelik Kayseri et al. (2010)	20, 23

Table I. Physical and Mechanical Properties of the Regenerated Cellulose Fibers

<sup>a</sup> For the comparison of viscose fiber properties with other regenerated cellulose fibers.

hybridization of PP-jute composites with higher extendable viscose fibers would increase the composites' strain to failure and enhance the energy absorption of the composites.

The energy absorption or toughness of composites has been evaluated with various techniques, including unnotched and notched Izod and Charpy tests and instrumental falling-weight (IFW) impact tests. Only a few studies on IFW testing of natural-fiber-reinforced composites have been done.<sup>25</sup> It has been reported that jute-fiber-reinforced thermoplastics have an inadequate impact strength.<sup>26</sup> The evaluation of the fallingweight impact properties is usually based on the hysteresis cycle of load versus deflection. For a fiber-reinforced plastic material, it is likely that the impact behavior is time-dependent, that is, dependent on the velocity of the hammer when it strikes the specimen.<sup>4</sup> As the results of tests are shown as the load time and absorbed energy-time data, the history of failure can be studied in greater detail compared to the more commonly used Izod or Charpy tests. The time variation of the peak impact load is strongly dependent on the stiffness of the composite. From the IFW information, loading force, velocity of the impactor, and impact time, the composite failure mechanisms, such as crack initiation, crack opening at the fiber-matrix interface, fiber breakage, debonding, and pullout of fibers, can be inspected.<sup>27,28</sup> The high value of the elongation at failure is expected to lead to improved ductile behavior and result in a good value of the specific energy absorption compared with that of unmodified jute-reinforced PP composites.

In this study, we attempted to investigate the amount of energy required to break jute fiber–thermoplastic composites and how this was affected by modification with an impact modifier (viscose fibers) and MAPP compatibilizer. The modified and unmodified jute–PP composites' viscoelastic properties were also characterized with dynamic mechanical analysis. In addition, the postimpact damage and failure mechanism of the fractured specimens were assessed, and the fiber dispersion of the jute and viscose fibers in the PP matrix were studied with scanning electron microscopy (SEM).

#### EXPERIMENTAL

#### Materials

**Matrix.** The PP used in this study was the homopolymer Propel 1350YG (extrusion grade, Indian Oil Corp., Ltd., Chennai, India), with a melt flow index of 35 g/10 min (230°C, 2.16 kg).

**Reinforcement.** The jute fibers used in this study were in the form of continuous roving, which was connected together manually according to our earlier study.<sup>7</sup> The fibers were procured from Chrandra Prakash & Co. Pvt., Ltd. (Jaipur, India). The diameter of a single fiber ranged from 20 to 25  $\mu$ m, and the density of the jute fibers was 1.45 g/cm<sup>3</sup>.

**Impact Modifier.** The viscose fibers were procured from Cheran Spinning Mills (Erode, India). The viscose fibers used as the impact modifier were also used in the continuous roving form. The appearance of the fibers was soft and silky. The fiber length and density were 38 mm and 1.3 g/cm<sup>3</sup>, respectively.

**Coupling Agent.** A PP-grafted maleic anhydride, Epolene E-43 (Sigma Aldrich), with a weight-average molecular weight of about 9100, was used as the coupling agent.

Processing of the Composite Material. Long-fiber thermoplastics processing with a high-performance corotating twin-screw extruder (model ZE-25, Berstorff Maschinenban GmbH, D-3000, Hannover, Germany) was used and has been reported elsewhere.8 The continuous fiber roving was incorporated into a side feeder of the extruder, which fed it directly into the polymer melt. The composites were prepared by the variation of the impact modifier concentration according to our earlier study, and the jute fiber content was kept constant at 30 wt %.8 Material formulations and denotations used for comparison are shown in Table II. The extrudates were compression-molded into rectangular sheets  $250 \times 125 \text{ mm}^2$  with a thickness of 3 mm with a conventional compression mold from Hindustan Hydraulics (India) with a capacity of 150 tons. The samples for impact testing were prepared from the compressed sheet according to ASTM standards.



Table II. Material Formulations and Denotations

Material	PP (wt %)	Jute fibers (wt %)	Viscose fibers (wt %)	MAPP (wt %)
PP-J30	70	30	0	0
PP-J30-M2	68	30	0	2
PP-J30-V10	60	30	10	0
PP-J30-V10-M2	58	30	10	2

#### Testing and Characterization

The Charpy impact test was performed as per ASTM D 256 (notched) with a pendulum-type impact tester. The impact test was carried out from the normal direction perpendicular to the orientation of the fibers; the specimens were cut from the compressed sheets with a geometry of  $75 \times 10 \times 3$  mm<sup>3</sup> with a notch depth of 2.54 mm and a notch angle of  $45^{\circ}$ . The impact energies were measured for at least 10 test samples of every composition, and the average values are reported.

The low-velocity falling-weight impact test was performed according to ASTM D 7136 on a CEAST 9350 Fractovis Plus IFW apparatus (Pianezza, Italy). The instrument was equipped with a hemispherical impactor 20 mm in diameter and fitted at a height of 458.9 mm, and the total impact mass was 3.6 kg with a velocity of 3 m/s; this gave an incident energy of 16.3 J. Test specimens measuring  $60 \times 60 \times 3$  mm<sup>3</sup> were attached to the supporting ring. The impact striker fell to the specimen and produced damage up to penetration. The resistance force was measured by a load cell with respect to time. The parameters force–deformation, energy absorption, and velocity were calculated with Ceast software. At least five specimens were tested for each category, and the average values are reported.

The morphology of the composites was studied with SEM. The fractured cross sections of the IFW specimens were sputtercoated with gold and studied in an SEM instrument at an acceleration voltage of 20 kV. The fiber dispersion of the microtomed cross section of the composite samples was studied.

Dynamic mechanical analysis was performed with a TA Instruments DMA Q800 (Delaware). The measurement was conducted in three-point bending mode at a frequency of 1 Hz under a nitrogen atmosphere. The dynamic storage modulus, loss modulus, and loss factor values of the composites were determined as a function of temperature, which ranged from -25 to  $100^{\circ}$ C.

The thermal stability of the composites was measured with thermogravimetric analysis (Pyris, PerkinElmer). The samples were heated from 0 to 600°C at a rate of 20°C/min under nitrogen flow (50 mL/min).

Fourier transform infrared (FTIR) spectra of the PP/jute composites modified with an impact modifier and MAPP were recorded with an Agilent Cary 630 FTIR spectrometer. The spectrometer was used in transmission mode with a resolution of 4 cm<sup>-1</sup> in the range 500–4000 cm<sup>-1</sup>.

#### **RESULTS AND DISCUSSION**

#### Impact Toughness

The notched Charpy impact and IFW properties of the PP-J30 and modified composites are shown in Table III. The impact strength was improved with the addition of viscose fibers to the PP-J30 composite. The improvement in the impact strength may have been due to several factors: the properties of the impact modifier, the matrix, the fiber length, the adhesion between the matrix and fiber, and so on. The results in Table III show that the addition of 10 wt % impact modifier to PP-J30 improved the impact strength from 3.2 to 7.5 kJ/m<sup>2</sup>; this was an improvement of 134% and indicated the ability of the viscose fibers to dissipate the energy along the length of fiber. The major mechanism for increasing the energy absorption of the composites accounted for by this study was the pullout behavior of fibers. We showed in our earlier investigation that debonding and fiber pullout resulted in a greater energy absorption capability on the jute-fiber-reinforced impact-modified composite compared to the unmodified one. The addition of MAPP on the impact-modified composites formed a weak interface, which increased the fiber pullout length and thus increased the energy dissipation.

Typical load-time and force-deformation curves from the IFW impact test are presented in Figure 1, and the initial peak, maximum load, and time are presented in Table III. The initial peak value of the impact force represents the starting point of the damage, the maximum load reveals the higher load-bearing capability, and the contact duration is the time taken for damage initiation and propagation. The peak load values for the PP-J30 and PP-J30-M2 (2 wt % of maleated polypropylene) composites were 1175 and 1407 N, respectively. The PP-J30 composite exhibited a lower peak load compared PP-J30-M2; this indicated stress transfer from the matrix to the fibers. The incorporation of 10 wt % viscose fibers (impact modifier) increased the area under the curve, whereas the peak load reduced the impact force to 1065 N, as shown in Figure 1(a). This indicated that the damage propagation extended beyond the peak load; this was attributed to the adsorption of energy until complete perforation of the specimen. This is also clearly noted in Figure 3 (shown later), that the impact-modified composites did not exhibit complete perforation on the back surface, whereas the unmodified composites showed complete damage.

We also noted that the contact duration of the PP–J30–V10 (10 wt % of viscose fiber) composite was 4.2 ms higher than that of the PP–J30 composites; the time elapsed from damage initiation to penetration on impact-modified composite indicated the increase in energy absorption. The addition of MAPP on impact-modified composites further increased the contact duration to 14.3 ms indicates the difficulty in the complete perforation of composites shown in Figure 1(b).

The amount of energy absorbed was equal to the area of the impact hysteresis cycle (force vs deflection).<sup>29</sup> When a material is subjected to any kind of impact loading, it absorbs energy by deflection/deformation. Deflection is usually accompanied by damage initiation and/or propagation in the form of fiber breakage, matrix cracking, and fiber/matrix debonding. As



	Charpy impact	IFW					
Composition	strength (kJ/m <sup>2</sup> )	Impact energy (J)	Load (N)	Deformation (mm)	Time (ms)	Velocity (m/s)	
PP-J30	3.2 (0.2)	8.0 (0.7)	1175.6	18.6	9.7	1.7	
PP-J30-V10	7.5 (0.2)	12.9 (1.1)	1065.8	23.4	13.4	0.9	
PP-J30-M2	3.1 (0.1)	6.8 (1.4)	1407.2	17.5	10.1	1.8	
PP-J30-V10-M2	7.8 (0.2)	15.4 (0.6)	1187.0	24.8	14.3	0.1	

Table III. Charpy and IFW Impact Results of the PP-J30 Composites Modified with Viscose Fibers and MAPP

The values in parentheses are standard deviations.

shown in Figure 1(c), the energy required to initiate the crack in the impact-modified composites was lower than the unmodified PP–J30 composites. The unmodified composite exhibited brittle fracture with little propagation energy; the samples fragmented after they were subjected to the incident energy. Figure 1(c) clearly shows a sudden drop in the force without much deformation. Thus, the total energy absorption of the composites was reduced because of their failure behavior. However, changes in the damage mechanism of the composites were observed with the introduction of viscose fibers to the jutefiber-reinforced thermoplastic composites. A drastic increase in the deflection with a slight decline in the damage initiation load was observed in the impact-modified composites. This clearly indicated that specimens modified with the impact modifier required a higher penetrative energy; this enhanced the total energy absorption of the composite. The improved damage mechanism with higher plastic deformation could be explained by the higher elongation behavior of the viscose fibers. In addition to the two different composites, shown in Figure 1(d), PP–J30–V10 modified with MAPP exhibited a higher energy absorption; this revealed that the MAPP compatibilizer together with the viscose fibers formed weak interfacial adhesion.

Figure 2 shows the variation in the energy absorption and velocity with respect to time of composites modified with



Figure 1. Typical load-time and force-deformation curves for the PP-J30 composites modified with viscose fibers and MAPP.



Figure 2. Typical energy absorption-time and velocity-time curves for the PP-J30 composites modified with viscose fibers and MAPP.

viscose fibers and MAPP. There was an increase in the total energy absorption when viscose fibers were present. The energy used for the impact test was 16.2 J, whereas the PP–J30–V10 samples absorbed a total energy of 12.9 J, as shown in Figure 2(a). When the composites were modified with MAPP, a notice-able change in the energy absorption compared to the unmodified composites was observed, as shown in Figure 2(b). Our previous study<sup>7</sup> also showed that the mechanical properties and the impact strength increased in all of the modified composites.

In general, the area under the velocity–time curve represents the displacement. The impact velocity affects the energy dissipation of composites during impact.<sup>30</sup> The full penetration of the impactor on the PP–J30 samples was observed, as depicted in Figure 3, and the residual velocity was 1.7 m/s. The possible reason may have been greater fiber breakage or poor adhesion between the matrix and fiber. Figure 2(c) shows that the residual velocity of PP–J30–V10 was found to be 0.9 m/s; this showed that the addition of the impact modifier drove the residual velocity to a minimum value. Dhakal *et al.*<sup>27</sup> noticed the zero residual velocity for four- and five-layered nonwovenhemp-fiber-reinforced polyester composites. They believed that the effect was due to the higher impact energy dissipation. In our study, it was obvious that the impact resistance increased compared to that of the unmodified fiber-reinforced composite;

this may have been due to the better adhesion of PP/viscose compared to that of PP/jute. The better adhesion did not allow the crack to easily propagate through the interface or to easily debond from the matrix; this led to an increased energy absorption of composites. According to Kim *et al.*,<sup>31</sup> PP/rayon displayed better adhesion between the PP/pineapple fibers because of the high cellulose content; this resulted in a higher energy absorption. Furthermore, the addition of MAPP on the impact modifier resulted in zero residual velocity because of the weak interfacial adhesion between the fiber and the matrix.

#### Microstructure

Figure 4 shows the influence of the impact modifier and coupling agent on the microstructure of the PP–J30 composites. Figure 4(a) shows more fiber pullouts, which left a void in the matrix phase because of worse bonding between the matrix and jute fibers.<sup>32,33</sup> This behavior led to composites with less plastic deformation after the maximum load was reached. The addition of the impact modifier on the PPJ-30 composite microstructure [Figure 4(b)] showed less fiber pullouts compared to PP–J30; the fiber pullout paths were recognized; this proved that the viscose fiber induced the plastic deformation after the maximum load and thereby contributed to the energy absorption of the composites by promoting debonding and pullout of fibers from the matrix. Johnson *et al.*<sup>33</sup> reported a PP/wood/Lyocell-fiber-





Figure 3. Fragmentation characteristics of the IFW test on the PP–J30 composites modified with viscose fibers and MAPP. FS = front side; BS = back side.

reinforced hybrid composite; they found that the Lyocell fiber had fewer pullouts during failure and favored a higher elongation at break. Similarly, Kim *et al.*<sup>31</sup> also noticed better fiber-matrix adhesion with rayon-reinforced composites, and they found improved toughness in the composites compared to pineapple-wood-fiber-reinforced thermoplastic composites.

The addition of MAPP resulted in improved interfacial adhesion, and the composite exhibited fiber breakage before the complete pullout of the fibers, as shown in Figure 4(c). The effect was also observed in the load–deformation curve; that is, a higher load with less deformation was observed; this indicated that the strong interaction between the fiber and matrix led to brittle failure. These findings were in agreement with results reported by Goriparthi *et al.*<sup>34</sup> They found a decrease in the impact strength along with the energy absorption of the compatibilized PLA/Jute composites; this was due to the fiber breakage before debonding. However, the failure mechanism was changed on the compatibilized impact-modified PP–J30 composite, as demonstrated in Figure 4(d). This composite recorded the maximum energy absorption; this was attributed to the formation of weak interfacial adhesion between the fiber and the matrix. This proved that the jute fiber in the composites enhanced the interaction with the matrix, whereas the viscose fiber did not show much effect on the interfacial adhesion with MAPP.

#### **Viscoelastic Properties**

The storage modulus as a function of the temperature for all of the composites is represented in Figure 5(a,b) and tabulated in Table IV. As reported by Doan et al.,<sup>35</sup> a general trend of improvement in the thermomechanical properties with the addition of jute fibers in to the PP matrix was observed; this indicated improved stiffness of the composites. It was evident that the addition of a small amount of impact modifier caused a gradual softening of the PP-J30 composite. A possible explanation was that the incorporation of viscose fibers induced flexibility in the composite because of the lower modulus values. This behavior was also confirmed by static mechanical testing. Adekunle et al.<sup>36</sup> noted a similar finding in flax-fiber-reinforced biobased resins with Lyocell as an impact modifier. The Lyocell hybridization was reported to exhibit a slight reduction in the storage modulus compared to those in biobased resin/flax composites; the authors believed that the effect was due to the Lyocell hybridization. Oksman and Clemon's<sup>25</sup> study showed that the addition of 10% styrene-ethylene/ butylene-styrene (SEBS)-g-maleic anhydride (MAH) reduced the storage modulus; they believed that the flexible SEBS was the reason why the storage modulus was lowered. However, their results revealed an improvement in the toughness of the composites. The PP-J30-V10-M2 sample showed a higher storage modulus, and the same composite offered a higher energy absorption during impact loading, as shown in Figure 5(b). The possible reason could have been that the jute fiber in the composites enhanced its interaction with the matrix, whereas the viscose fiber had a substantially lesser effect with MAPP.

The loss modulus values of the composites modified with an impact modifier and MAPP was compared with PP-J30, as shown in Figure 5(c,d); their values are shown in Table IV. The PP-J30-V10-M2 composite showed a higher value of loss of modulus compared to PP-J30 and the impact-modified composites. The higher loss peak clearly indicated the improved impact energy absorption of the composite.37 The magnitude of the loss modulus peak variation resulted in a severe decline in the storage modulus.<sup>38</sup> A broadening of the loss modulus peak was observed with the impact-modified composites compared to that of PP-J30. This indicated an increase in the energy absorption caused by the viscose fibers.<sup>39</sup> The hybridization of the viscose fibers with the PP/jute composite was found to affect the properties of the system. The incorporation of long viscose roving in the PP-J30 composite lowered the loss modulus. This could have been considerable because of the property mismatch of the fibers. The addition of MAPP to the impactmodified composite improved the loss modulus corresponding to the viscous dissipation.<sup>40</sup> The improvement in the modulus was due to the reduced flexibility of the composite. These





Figure 4. SEM images of the fractured specimen from IFW testing of the composites modified with viscose fibers and MAPP.

constraints on the segmental mobility were believed to be due to the enhanced interaction of the jute fiber with the matrix in the presence of the compatibilizer.

Figure 5(e,f) and Table IV compare the tan  $\delta$  value of the composites as a function of the temperature. It was evident from the curve that the incorporation of viscose fibers increased the damping performance of the PP-J30 composite. The damping peak in the composites indicated that once the deformation was induced in a material, the material would not recover its original shape.37 This study showed that the peak amplitude increased with the addition of the impact modifier; this indicated a greater degree of molecular mobility, but the position of the peak shifted to 3°C upward. This indicated some kind of interaction between the viscose fiber and the matrix. As shown in Figure 5(e), it was evident that the addition of MAPP to the PP–J30 composites shifted the tan  $\delta$  position about 4°C upward; this indicated enhanced interaction of the fibers with the matrix because of the addition of MAPP. Oksman and Clemon<sup>25</sup> noted similar findings; when wood flour and MAPP were used with the PP/Ethylene/propylene/diene/terpolymer (EPDM) system the tan  $\delta$  peak shifted 4°C upward. However, the addition of MAPP to the impact-modified composite resulted in peak broadening along with the decrease in the tan  $\delta$  peak from 12 to 11°C. This might have been due to the formation of weak interfacial adhesion between the fiber and the matrix.

#### **Interfacial Properties**

The FTIR spectra of the PP-jute composite modified with viscose fibers and MAPP is shown in Figure 6. The spectra of the PP-J30 composite showed a weak intense peak in the range 3200-3600 cm<sup>-1</sup>; this was attributed to the O-H stretching groups of jute fiber. CH<sub>2</sub> symmetric and asymmetric peaks, due to the alkyl chain of PP, appeared at 2958 and 2870 cm<sup>-1</sup>. The peak at 1455 cm<sup>-1</sup> was attributed to the stretching of C-H bonds in the cellulosic structure. The peak at 1382 cm<sup>-1</sup> was attributed to the O-H bending vibrations, and the strong band at 1040 cm<sup>-1</sup> was attributed to the characteristic C-O-C stretching. In case of the PP-J30-M2 composites, the intensity of the peak at 3200-3600 cm<sup>-1</sup> corresponding to the O-H frequencies appeared to be high compared to the PP-J30 composite. This was due to the hydrogen-bond interaction between the ester group of the maleic anhydride and the hydroxyl group of the cellulose fiber; this enhanced the interactions with the matrix. The impactmodified composites showed a much broader peak; this indicated the formation of interactions between the sulfide group of the regenerated cellulose (viscose fiber) and the O-H group of the cellulose (jute) fibers. The compatibilized impactmodified jute-reinforced composites showed a reduced O-H intensity compared to the PP-J30-V10 composites because of the formation of weak hydrogen bonding; this deteriorated the



Figure 5. Storage modulus, loss modulus, and tan  $\delta$  as a function of the temperature for the PP–J30 composites modified with viscose fibers and MAPP.

interactions with the matrix (i.e., the viscose fiber had a substantially lesser effect with MAPP).

#### **Thermal Properties**

The thermogravimetric curves of PP–J30, PP–J30–M2, PP–J30–V10, and PP–J30–V10–M2 are shown in Figure 7. The PP/jute composites prepared at 30 wt % fiber loading showed an initial

peak between 339 and 386°C, which corresponded to the thermal degradation of cellulose.<sup>31</sup> The second decomposition occurred between 378 and 414°C; this was primarily attributed to aromatization and involved a dehydration reaction.<sup>29</sup> As shown in Figure 7, the impact-modified composites exhibited a slightly lower thermal stability than the PP–J30 composites. This may have been

Composition	Storage modulus (Pa)	Tan $\delta$ peak (°C)	Loss modulus (Pa)	Tan $\delta$ peak height
PP-J30	$1.1 \times 10^{9}$	9	$8.5 \times 10^{7}$	0.07
PP-J30-V10	$6.0 \times 10^{8}$	11	$5.0 \times 10^{7}$	0.08
PP-J30-M2	$2.6 \times 10^{10}$	12	$1.4 \times 10^{9}$	0.05
PP-J30-V10-M2	$2.4 \times 10^{10}$	11	$1.6 \times 10^{9}$	0.07

Table IV. Dynamic Mechanical Properties of the PP-J30 Composites Modified with Viscose Fibers and MAPP

Values with respect to the tan  $\delta$  peak temperature.

attributed to the lower thermal stability of the viscose fiber. The modification of the PP–J30 composites with 2 wt % MAPP influenced the thermal stability of the composites. This effect might have been due to the stronger interaction between the fiber and matrix caused by the formation of the covalent bonds at the interface.<sup>32</sup> The addition of MAPP on the impact-modified composites caused an improvement in the thermal stability of the PP–J30–M2 composites because of presence of the weak interfacial adhesion between the fiber and the matrix.

#### Fiber Dispersion and Orientation

Figure 8 shows the SEM micrographs of the microtome PP–J30 composites modified with viscose fibers and compatibilized with MAPP. The cross sections of the pure jute and hybrid composites were examined with respect to the fiber dispersion and orientation. In general, the good dispersion and distribution of fibers is essential for achieving high mechanical properties of composites.<sup>2</sup> Figure 8(a) shows that the jute fibers were well dispersed in the polymeric matrix without any aggregation. It was obvious that fibers with stiff structure and nontangling behavior showed better homogeneity in fiber distribution along the sample sections.<sup>41</sup> Furthermore, the jute fibers tended to oriented predominantly parallel to the flow direction. Figure 8(b) shows nonhomogeneous dispersion on impact-modified composites; the viscose fibers were observed in bundles. The



Figure 6. FTIR spectra of the PP–J30 composites modified with viscose fibers and MAPP. [Color figure can be viewed in the online issue, which is available at wileyonlinelibrary.com.]

possible reason for the poor dispersion of viscose fibers in the composites was the soft and tough nature because soft and tough fiber bundles are more difficult to separate during processing. The effect was agreed well with Oksman *et al.*'s<sup>2</sup> report; their results show that the toughest fibers were retained the fiber bundles in the composite. Several further studies have demonstrated that it is difficult to disperse properly cellulose fibers in polyolefin polymers because of strong hydrogen bonding between the fibers.<sup>18,42–44</sup> Figure 8(b) also demonstrates that the viscose fiber bundles were aligned mostly in the same direction as those in the jute fibers. It is well known that the energy absorption is the function of the plane orientation.<sup>4</sup> Thus, the influence of the energy absorption with the incorporation of viscose fibers clearly indicated that the fiber bundles were oriented in the direction of the extrusion.

However, the agglomeration of fiber bundles could be reduced with the addition of surface modifiers. Raj *et al*: $s^{45}$  study detailed the importance of using surface modifiers to improve the fiber dispersion in cellulose fiber/PP composites. The addition of MAPP on the impact-modified and unmodified composite showed a slight improvement in the quality of the dispersion in the polymeric matrix, as shown in Figures 6(d) and 8(c). Therefore, the observation of our study predicted that the further improvement in the fiber dispersion without agglomeration in the composite led to the achievement of better impact properties along with other mechanical properties.



Figure 7. Thermogravimetric analysis of the PP–J30 composites modified with viscose fibers and MAPP.

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Figure 8. SEM micrographs of the microtome-PP-J30 composites modified with viscose fibers and MAPP.

### CONCLUSIONS

This study was about the impact toughness, energy absorption, viscoelastic properties, and fiber dispersion of the modified long-jute-fiber-reinforced thermoplastic composites. The prepared composites were studied with conventional Charpy impact and IFW impact test methods.

The results show that the addition of viscose fibers improved the propagation energies because of the higher energy dissipation. The Charpy and IFW tests showed 134 and 61% improvements in the impact strength and energy absorption, respectively.

The microscopy analysis showed fiber pullout paths in the impact-modified composites that were not observed in the PP–J30 composites. This indicated that the toughness of the composites was influenced by the viscose fibers, whereas the SEM images showed a negative effect on the homogeneity of fiber distribution on the impact-modified composites. The study proved that the incorporation of tough viscose fibers retained the fiber bundles in the composites.

The impact modification of the PP-jute composites was found to also affect the material's viscoelastic properties. A slight negative effect was observed on the storage modulus, whereas the peak amplitude increased; this indicated a greater degree of molecular mobility.

Furthermore, the addition of MAPP in PP–J30 resulted in improved interfacial adhesion between the fibers and matrix; thus, the composite failed catastrophically during the impact event and, thereby, reduced the energy absorption of the composites.

The fractography micrographs of the surface-modified composites indicated the clear debonding and pullout of the fibers because of the weak interfacial adhesion developed between the fibers and the matrix. The SEM images also showed a slight improvement in the quality of dispersion on both the PP–J30– M2 and PP–J30–V10–M2 composites.

Similarly, the shift in the tan  $\delta$  position about 4°C upward indicated enhanced the interaction of the fibers with the matrix. However, the compatibilized impact-modified composite showed enhanced toughness with the formation of weak interfacial adhesion; in case of dynamic mechanical analysis, a broadening peak along with the decrease in the peak position to 1°C was observed when compared to that of the PP–J30–M2



composites. The FTIR spectra confirmed the weak interfacial adhesion of the PP–J30–V10–M2 composites, whereas the thermogravimetric analysis thermograms displayed a lower thermal stability when compared to that of the PP–J30–M2 composites.

In this study, we concluded that the improvement in the impact performance was achieved with the incorporation of viscose fibers, and when the efficiency of the viscose fibers was improved, dispersion could further improve the impact properties along with other mechanical properties.

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