Long Fiber Extrusion and Improved Energy Absorption Behaviour of Hybrid Thermoplastic Composite

by

Nalini Ranganathan

A thesis submitted in conformity with the requirements for the degree of Doctor of Philosophy
Faculty of Forestry
University of Toronto

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Abstract

The aim of this study was to develop thermoplastic biocomposites of polypropylene (PP) and jute fibers (J) with high energy absorption and without decline of strength and stiffness properties. The preparation of thermoplastic composite is a challenging task; this research presents manufacturing thermoplastic biocomposite using direct long fiber thermoplastic (LFT) extrusion technique. The soft and tough viscose fibers were used as impact modifier. The energy absorption of the composites was studied using notched izod, charpy and instrumental falling weight (IFW) impact tests. In addition fiber breakage during the extrusion process, heat deflection temperature (HDT), viscoelastic behaviour, fracture toughness, fatigue life, interfacial shear (ISS) strength and fiber dispersion were experimentally evaluated.

The optimum mechanical properties of the PP-J composite including toughness was found with the addition of 10 wt% of viscose fibers and 2 wt% maleated polypropylene (MAPP) as compatibilizer. Heat deflection temperature values reduced with addition of viscose fibers whereas the MAPP addition improved the values from 140 to 145°C. Consecutively, the composites viscoelastic behavior was studied using dynamic mechanical analysis (DMA), the
study showed broadening of tan δ peak without change in the peak position on the compatibilized impact modified composite. The higher fracture toughness and fatigue life was also recorded on MAPP modified jute-viscose-PP-composite due to increased work energy by hindering the propagation of crack. The fractography micrographs of surface modified composites indicated clear debonding and pullout of fibers due to the flexible interphase developed between the fiber and matrix. The fiber dispersion seen in SEM images showed that toughest viscose fibers retained as larger bundles in the composites compared to more individual separated jute fibers. It is difficult to disperse viscose fibers due to their soft and tough nature. However to utilize potential of the fibers in the composite, the improvement in fiber dispersion was achieved by increasing the shear during extrusion process.

This work demonstrated that it is possible to use the long viscose fibers as impact modifier to produce LFT composites. The addition of tough viscose fiber improved the energy absorption of long jute reinforced thermoplastic composites through toughening mechanism without much sacrifice in strength and stiffness.
Acknowledgments

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List of Symbols

LFT Long fiber thermoplastics
°C Degree celsius
HDT Heat deflection temperature
SEM Scanning electron microscopy
MAPP Maleated polypropylene
PP Polypropylene
DMA Dynamic mechanical analysis
Ozs Ounces
Wt% Weight percentage
g Gram
cm³ Cubic centimeter
µ Microns
MPa Megapascal
GPa Gigapascal
% Percentage
σ Stress
ρ Density
min Minutes
J Joules
m meter
mm Millimeter
µm Micrometer
LFT Long fiber technique
Rpm Rotation per minute
L/D Length to Diameter
σf Flexural strength
Pmax Maximum load
N Newton
Ef Flexural modulus
L Span length
<table>
<thead>
<tr>
<th>Symbol</th>
<th>Definition</th>
</tr>
</thead>
<tbody>
<tr>
<td>b</td>
<td>Width</td>
</tr>
<tr>
<td>h</td>
<td>Thickness</td>
</tr>
<tr>
<td>UTM</td>
<td>Universal testing machine</td>
</tr>
<tr>
<td>Psi</td>
<td>Parts per square inch</td>
</tr>
<tr>
<td>kV</td>
<td>Kilovolt</td>
</tr>
<tr>
<td>VF</td>
<td>Viscose fiber</td>
</tr>
<tr>
<td>(\tau_{\text{app}})</td>
<td>Apparent interfacial shear strength</td>
</tr>
<tr>
<td>IFSS</td>
<td>Interfacial shear strength</td>
</tr>
<tr>
<td>X</td>
<td>Magnification</td>
</tr>
<tr>
<td>IFW</td>
<td>Falling weight impact</td>
</tr>
<tr>
<td>MFI</td>
<td>Melt flow index</td>
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<tr>
<td>Kg</td>
<td>Kilogram</td>
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<tr>
<td>Mw</td>
<td>Molecular weight</td>
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<tr>
<td>ASTM</td>
<td>American society for testing materials</td>
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<td>ms</td>
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<td>OM</td>
<td>Optical microscopy</td>
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1 Background

LFT compounding is currently of considerable interest in the automotive industry in Europe and USA and is one of the most important trends in the plastics industry at present. LFT is an in-line compounding and direct compression moulding process developed by the extruder manufacturer Coperion Werner & Pfleiderer in the year of 2001. Continuous glass fiber rovings were fed directly into the polymer melt during compounding. The screw speed is determining the rate at which the glass fibers are fed into the extruder. LFT process offer better mechanical performance due to high aspect ratio of the fibers and are tough due to the energy dissipating mechanisms of fiber debonding and pullout (Henning 2005).

Laterally this technique was adapted by many researchers (Knights 2003, Krause et al., 2003); Oksman et al developed the PLA/ flax fibers thermoplastic composites using LFT technique to investigate, if PLA can be used as matrix in composite system (Oksman et al., 2003). The natural fiber reinforced polypropylene also was manufactured using LFT extrusion and the fibers used were sisal, banana, jute and flax and the matrix was a polypropylene (Oksman et al., 2009). Amongst the natural fibers, jute fiber reinforced thermoplastics has fairly larger amount of work. This is because of the good mechanical properties of jute fibers when compared with other natural fibers such as sisal, coir or ramie (Mohanty et al., 2005). But some drawbacks exist with the jute, when used as reinforcement for thermoplastics; such as low impact strength, low fracture toughness, poor bonding between fiber and matrix and poor thermal stability (Khan et al., 2009; Kumar et al., 2012). These drawbacks restrict the applicability of jute fiber reinforced thermoplastics especially for automotive applications. As reported by many researchers, the reason for the poor resistance towards impact strength is due to the very low extensibility of jute fiber (Kabir et al., 2010) and other reason is that the inherent incompatibility between
hydrophilic lignocellulosic fiber and hydrophobic polymeric matrices (Zhang et al., 2011). In addition the toughness of fiber reinforced composites is affected by a number of factors such as matrix intrinsic properties, fiber properties, fiber volume fraction, and interfacial bond strength between the fiber and the matrix, fiber aspect ratio as well as fiber orientation. The toughness can be improved by several ways: 1) hybridize with tough fibers (Mallick et al., 1977); 2) achieving fiber pullout against interfacial shear stress (Xia et al., 1994); 3) optimizing the fiber content (Kim et al., 2008) and dispersion (Paunikallio et al., 2003); 4) controlling the aspect ratio such as fiber length and orientation (Baillif et al., 2009). Several researchers have carried out various methods to improve toughness and energy capability of the composite (Tam et al., 2000; Khalili et al., 2011).

The use of two types of fibers in a suitable combination can potentially result in performance synergy, whose hybrid performance exceeds the sum of individual fiber performances. Many researchers have reported that the hybridizing using high strain to failure fiber will be an efficient way to improve the impact strength of low strain to fiber composites (Tonoli et al., 2011; Samal et al., 2007; Kalaprasad et al., 1996; Mallick and Broutman 1997). Fu et al. showed the enhancement of failure strain with the use of short glass fiber (SGF) in PP/short carbon fiber (SCF) composites, the work reported the increase of fracture toughness due to the positive hybrid (synergistic) effect (Fu et al., 1996). Adekunle et al. used Lyocell fibers (regenerated cellulose) as an impact modifier in jute fiber reinforced thermoset composite and reported an increased impact resistance and long pullout lengths of Lyocell fibers (Adekunle et al., 2011).
The fiber pullout is desired to increase the toughness or resistance to fracture of the composites. The energy absorption at the interface are affected in two different ways: 1) If the interface bonding is too strong then the matrix crack propagates into fibers without leaving intact fibers to produce pullout and crack bridging occurs along the fracture path 2) If the interface bonding is very weak or poor then the stress transfer from matrix to fiber is not possible; thus the above two criteria results in no toughening of the composites (John et al., 2008). Therefore, flexible and weak interface would be preferable, which allows the fibers to debond along the interface and yields excessive fiber pullout to achieve good toughness in the thermoplastic composites (Sakai et al., 1991). The failure mechanism can be changed by controlling the adhesion between fiber/matrix by using a coupling agent. In addition, the use of coupling agent can improve the dispersion of the cellulose fibers. Raj and Kokta indicated the importance of using surface modifiers to improve dispersion of cellulose fiber in PP matrix (Raj and Kokta 1989). Many authors have also used coupling agents in natural fiber composites, to achieve fiber dispersion along with the better interfacial adhesion between fiber and matrix (Baillif et al., 2009; Woodhams et al., 1984). Oksman et al. reported improvement in strength and good toughness using 2% MAPP coupling agent due to better facilitation of stress transfer between the matrix and fibers (Oksman et al., 2009).

A small physical variation of reinforcement varies the overall mechanical performance of the composites. The fiber volume fraction is probably the single most important factor, with most mechanical properties increases with an increase in volume fraction up to a certain point (Saravana bavan et al., 2010). Oksman et al. evaluated the mechanical performance and the fiber microstructure of different concentration of long natural fibers in polypropylene matrix (Oksman et al, 2009). They reported the increase of stiffness and strength with increase in fiber volume
fraction. Zampaloni et al, optimized the fiber content based on the mechanical performance of the composites; they used kenaf, jute, hemp and coir as reinforcement in PP matrix (Zampaloni M et al, 2007).

The fiber pullout process requires additional energy; the toughening from fiber pullout can also be enhanced by increasing the length of the fibers (Xia et al., 1994). In composites prepared using LFT, the length of the fiber during processing can be controlled by reducing the shear forces and by reducing the processing time. This process also tends to orient the fibers along the flow direction, which leads to improved mechanical properties along the flow direction. Several authors have noticed that the increased shear energy causes more fiber breakage, and found that the average fiber length depends on the shear rate controlled by adjusting the screw speed and/or feeding rate (Vaidya et al., 2008; Baillif and Oksman 2009). The long fiber carry significantly higher fraction of load compared to short fibers. Yang et al made long glass fiber reinforced polyamide 6,6 composites and reported that an increase of the initial fiber length enhances the impact strength of the composites. The influence of impact strength caused due to the increased energy dissipation along the length of fiber (Yang et al., 2012).

The main goal of the work was to develop thermoplastic composites of jute fibers with high energy absorption without losing the other mechanical properties. This goal was achieved by developing the suitable combination of tough and strong fiber raw materials for feeding in the LFT process, process modification, followed by optimization of materials compositions including PP, jute, viscose and maleated polypropylene and also explore the ways to overcome the default limitation regarding the mechanical performance, limited fiber length, dispersion, poor interfacial adhesion and degradation at high temperature.
Preparation of continuous long jute fiber reinforced thermoplastic with regenerated cellulose (viscose fiber) as impact modifier using (LFT) process seems to be a promising technique, to enhance the energy absorption without much decline in the other mechanical properties. In order to optimize the thermoplastic composites, the mechanical properties (such as tensile strength and modulus, flexural strength and modulus, impact strength, work of adhesion) microscopical analysis on fracture mechanism are experimentally evaluated. To avoid the agglomeration of the cellulosic fiber and to achieve better interface a suitable surface modifier or coupling agent can be combined. Controlled shear during the processing tends to achieve higher fiber length which may result in superior mechanical properties along with toughness and energy absorption. To investigate the amount of energy required to break the impact modified long jute fiber reinforced composite various impact strength analytical techniques were performed. The failure mechanisms of post impact damage system were microscopically analyzed for crack initiation, crack opening at the fiber matrix interface, fiber breakage, debonding and pullout of fiber. The viscoelastic properties of the impact modified and the MAPP modified viscoelastic properties were characterized using dynamic mechanical analysis (DMA). From the developed impact modified composites the fiber content, fiber length after the extrusion process and fiber dispersion has been measured, and were they found to play an important role for fracture toughness and energy dissipation of the composites.

To improve the viscose fiber dispersion the alternate feeding technique was followed. In this process the viscose fibers were fed in a same inlet of PP thus produce higher shear to breaks the fiber bundles and achieves improved fiber dispersion. The resultant optimum design of impact modified long jute fiber reinforced composites was achieved through LFT-process could be
commercialized to the industry after prototype trials. A successful outcome of this project will lead to replacement of polypropylene-long glass fiber composites used in automobile application.
2 Materials and methods

2.1 Jute fibers

Jute is extracted from the bark of the white jute plant, Corchorus capsularis and to a lesser extent from tossa jute (C. olitorius); mostly grown in Southeast Asian countries. Jute is called as "golden fiber" because jute fibers are long, soft and shiny. The length of the fiber roving can be up to 4 m. It is one of nature's strongest vegetable fiber and ranks second only to cotton in terms of production quantity (Ahmaruzzaman et al., 2005). The quality of the fibers depends on various aspects, such as the wealth of the soil, technical processing, genetic manipulation, retting practices (Sen 2009), methods of degumming (Wang et al., 2008) i.e., mechanical, chemical and biological methods. Jute has multi-laminate structure; the cell wall is made up of a number of layers, namely middle lamella, primary wall, secondary wall and lumen shown in Figure 2.1 (Maschinenwesen et al., 2006). The secondary wall again made up of the three principal constituents, namely α-cellulose, hemicellulose, and lignin; with minor amounts of protein, extractives and inorganic (Sen et al., 2011), shown in Table 2.1 and the cellulose structure shown in Figure 2.2.

<table>
<thead>
<tr>
<th>Chemical composition</th>
<th>Cellulose (wt %)</th>
<th>Lignin (wt %)</th>
<th>Hemicelluloses (wt %)</th>
<th>Pectin (wt %)</th>
<th>Wax (wt %)</th>
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<th>Specific tensile strength (σ/ρ)</th>
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<td></td>
<td>1.3-1.45</td>
<td>25-200</td>
<td>393-773</td>
<td>0.31-0.38</td>
<td>13-26.5</td>
<td>1.16-1.5</td>
</tr>
</tbody>
</table>
The structure of the jute fibers is influenced by climatic conditions, age and the fermentation process. The chemical composition indicates the presence of various polar groups, leading to high moisture retention capacity. However, these fibers are covered with pectin and waxy substances, thus hindering the hydroxyl groups from reacting with polar matrices and forming mechanical interlocking adhesion with non-polar matrices (Mwaikambo et al., 2001). Thereby the natural fibers properties are based on the following factors:

- Source of the fiber
- Surface texture (flaws) present on the fiber
- Different external chemical treatment carried out onto the fiber
- Variation in the chemical composition
- Aspect ratio of the fiber

Figure 2.1: Cell wall structure of natural fiber (Villee et al., 1989)
Figure 2.2: Chemical structure of cellulose, (Pandey 1998)

Jute mainly and readily available in four different forms such as fiber, yarn, hessian cloth or as woven fabric, which are all used as the reinforcement in the polymeric composites. The single jute fiber is only 1-6 mm long and 5-30 µm wide (Mussig 2010). For most products, fiber bundles are used, which can be 200 µm or more in diameter and as long as the complete jute stalk. The physical appearance of the fibers and their properties; poised to become the fiber-composites for the future for various end-uses and applications. A few changes in technical nature of the fibers are made, for the development of yarns and fabric; yields variation in mechanical performance fibers thereby the composites (Mwaikambo et al., 2001). Jute yarn is spun from the fibers and produced in the form of single strand. The thickness and the weight of the yarn are maintained by their twists and counts. The strength of the raw fiber affects the yarn strength, its quality specifically relates to the tensile strength of the yarn (Rowell et al., 1998). The tensile strength of yarns can also be improved by maintaining the draft. Draft plays an important role on the quality of yarn, the excessive drafts cause worse quality yarns and low drafts causes the low strength yarn (Mahabubuzzaman et al., 2011). Jute yarn in single and multiple twists, ranging from 4 lbs to 120 lbs are wound in cylindrical, conical, spools or in
hanks as per requirements and packed in truss, pallet and bales. The aesthetic appearance of the hessian cloth is a form of jute cloth/fabric, the fibers yarn are weaved with warp and weft i.e. they are biaxial oriented. In simple words, it is a fine plain woven fabric of 5 to 12 ozs/yard. Here the fibers are transformed to fabric. The hessian cloth is generally nonwoven material. The jute fibers possess high specific strength and stiffness. Therefore, these are suitable to be used as reinforcement in a polymeric matrix. The considerable variation in the fiber properties usually exhibits based on their size, maturity and processing methods adapted for the extraction of the fibers. Properties such as density, electrical resistivity, ultimate tensile strength and initial modulus are related to the internal structure and chemical composition of fiber (Mohanty et al., 2001; Wang 2004). Jute fibers are of low cost and have low-abrasion, they are biodegradable, and recyclable nature (Kafi et al., 2006; Khan et al., 2009; Dash et al 1999). The presence of hollow and cellular nature in natural fibers enables to have reduced bulk density, excellent acoustic and thermal property (Venkateshwaran et al., 2012).

### 2.2 Regenerated cellulose

Regenerated cellulose fibers are produced by dissolution of wood pulp in a suitable solvent and subsequent spinning process was performed on viscose fiber for production facility (Hughes et al., 2006; Adusumali et al., 2006; Di et al., 2011). They appear in the form of fiber bundles with fine, smooth surface. They also have regular twisted staples. In regenerated celluloses, the unit cell structure is an allotropic modification of cellulose. The structure of cellulose derivatives could be represented by a continuous range of states of local molecular order rather than definite polymorphic forms of cellulose which depend on the conditions by which the fiber is made (Jangala 2007). The structure of regenerated cellulose (viscose fiber) is shown in Figure 2.3.
Regenerated cellulose fiber such as viscose, modal and Lyocell are the most important fibers from the point of environment aspect.

**Figure 2.3:** Chemical structure of regenerated cellulose (viscose fiber) (Nisshinbo Ind. INC, Patent, 1995)

In this project viscose fibers were chosen as an impact modifier for jute reinforced polypropylene. Viscose fibers are inexpensive, light-weight and have a silky appearance and feeling. Viscose fibers are fine, regular filaments or staple fibers, having many tiny grooves which influence their characteristics. Their characteristics and properties vary according to the method of processing. Generally it has lower tenacity and greater elongation when compared to other regenerated cellulose such as Lyocell, cordenka and modal. The water retention value (swelling index) of viscose is very much higher compared to other regenerated cellulose fibers (Kreze et al., 2001). The natural structures and properties have influenced the usage of regenerated cellulose fiber in the thermoplastic composites. Several types of regenerated cellulose fibers are available in market; all exhibits same chemical composition whereas the different production process produces differences in the fiber structure. The viscose cellulose
usually has 98% of long chain α-cellulose and the remaining caustic content (Lewin et al., 1998). The long chain molecules are necessary to achieve physico-mechanical properties such as strength and extensibility. The typical chain length of the viscose fiber ranges from 300 to 1700 units. In the properties point of view, the viscose fiber has higher elongation to break in comparison to other regenerated cellulose represented in Table 2.2. Hence their reinforcement helps in improving the impact strength of the thermoplastic composite by enhancing the failure strain (Lewin et al., 1998). In addition they have advantages such as light-weight, recyclability and they are biodegradable.

Table 2.2: Physical and mechanical properties of regenerated cellulose fibers

<table>
<thead>
<tr>
<th>Regenerated fibers</th>
<th>Density (g/cm³)</th>
<th>Tensile Strength (MPa)</th>
<th>Young’s Modulus (GPa)</th>
<th>Elongation at break (%)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cordenka</td>
<td>1.8</td>
<td>770-890</td>
<td>19.0-21.0</td>
<td>11.0-15.0</td>
<td>Ganster et al., 2013</td>
</tr>
<tr>
<td>Lyocell</td>
<td>1.3</td>
<td>790-1400</td>
<td>30.5-36.0</td>
<td>6.0-10.3</td>
<td>Carrillo et al., 2010</td>
</tr>
<tr>
<td>Modal</td>
<td>1.3</td>
<td>368-506</td>
<td>11.0-15.4</td>
<td>8.6-12.2</td>
<td>Adusumali et al., 2006</td>
</tr>
<tr>
<td>Viscosea</td>
<td>1.3</td>
<td>293-323</td>
<td>7.0-15.0</td>
<td>23.0-25.0</td>
<td>Ozcelik Kayseri et al., 2010</td>
</tr>
</tbody>
</table>

Despite of the aforementioned merits, the regenerated cellulose fibers have some limitations. It is highly hydrophilic in nature and has low tenacity. The strength and stiffness of viscose fiber is less for viscose fiber in comparison to other regenerated cellulose. They decompose at 177 to 204°C. It does not melt or stick at elevated temperatures. Rayon is attacked by bleaches at very high concentrations and by mildew under severe hot and moist conditions. Prolonged exposure to sunlight causes loss of strength because of degradation of cellulose chains (Jangala et al., 1999).
2.3 Matrix properties and its role in composites

The matrix plays a major role in the performance of composites. The main function of the matrix is to bond the fibers together and to transfer the load between them through shear stresses at interface. The thermoplastic composites add advantages over thermosets such as easy processability, low cost, design flexibility etc. For maximum benefit, relatively long fibers are with control fiber orientation needed. The matrix surrounding the fibers helps to keep in desired location and orientation. Further they also protect the fibers from environmental damages such as elevated temperature and humidity. However, the incorporation of strong fibers decreases the energy absorption of the thermoplastic composites.

This research covers mainly to enhance the toughness of the composites without comprising strength and stiffness; in order to add the advantages in new avenues including the replacement of long glass fiber reinforced thermoplastic composites which are manufactured through the Direct-Long fiber thermoplastic (D-LFT) techniques. The basic property of the polypropylene material used in this work is shown in Table 2.3 as reported by manufacture.

| Table 2.3: Mechanical properties of virgin PP, (values in the bracket are standard deviation) |
|----------------------------------|----------------------------------|----------------------------------|----------------------------------|----------------------------------|----------------------------------|----------------------------------|----------------------------------|
| Flexural strength (MPa) | Flexural modulus (MPa) | Tensile strength (MPa) | Tensile modulus (MPa) | Elongation to break (%) | Impact strength (J/m) | MFI (g/10min) |
| 33.2 (0.8) | 867 (22) | 27.6 (0.8) | 634 (49) | 9.8 (0.1) | 28.0 (0.8) | 35 |

2.3.1 Surface modification of matrix

In general the mechanical properties of the fiber-reinforced polymers depend on the properties of the fiber and matrix, volume fraction of fiber and the interfacial adhesion between the fiber and matrix. As the natural fibers are hydrophilic and polymeric matrix is hydrophobic the poor
adhesion between the fiber and the matrix may result, which in turn leads to poor mechanical properties (Zaman et al., 2011). The strong fiber/matrix adhesion hinders the development of the energy absorption mechanisms such as debonding and fiber pull-out whereas sufficient and flexible interphase between the fiber and matrix enhances the energy absorption of the composites (Silva et al., 2006). Mueller reported that, fine natural fibers with high elongation capability offers higher impact strength, further it was noted that; flexible adhesion between fiber and polymer resulted in fiber pull-outs, whereas good adhesion on contrary results in abrupt fiber fracture with a minor energy degradation (Mueller 2004).

As the interfacial properties play an important role in analyzing the mechanical properties of the fiber reinforced thermoplastic composites. This work used MAPP as coupling agent in natural fiber reinforced polypropylene, the reaction mechanism of MAPP with the surface of natural fiber is shown in Figure 2.4. Karmaker et al also reported that the addition of MAPP in PP/Jute composite improved the tensile strength, flexural strength and E-modulus (Karmaker et al., 1991 & 1997). The improved wetting of fibers with matrix enhanced the stress transfer from the matrix to the fiber. Oksman et al reported that the improvement of strength and toughness was achieved using 2% MAPP coupling agent and facilitated stress transfer from the matrix to the natural fibers (Oksman et al., 2009).
Figure 2.4: Reaction mechanism of MAPP with the surface of natural fiber (Karnani et al., 1997)

2.4 Composite processing

The process flowchart shown in Figure 2.7 demonstrates the steps involved for development of hybrid bio-composites using LFT extrusion technique

2.4.1 Preparation of fiber-roving

In-line compounding and moulding of long natural fiber reinforced thermoplastic requires continuous fibers. In this work long jute fibers were used as reinforcement and viscose fibers as an impact modifier. Mixture of jute and viscose roving’s were manually prepared before compounding. Prior to the preparation of continuous roving’s the jute fibers were washed with water and dried at 60°C for 48 hours until achieving a uniform weight at different intervals of time. Similarly the viscose fibers were dried at 60°C for at least 2 hours. Jute and viscose were also combined and slightly twisted to make mixed fiber-rovings. In the preparation, weight fractions of viscose fibers (impact modifier) were varied based on the composition of the composites. In all the composition the jute fiber content was kept constant at 30 wt% and the viscose fiber content were ranging from 5 to 15 wt%.
2.4.2 Manufacturing of composites

The direct long-fiber extrusion technique (D-LFT) is a new improved one-step processing method, and has a key challenge to be used with natural fibers as reinforcement. Oksman et al developed the LFT process using natural fiber-rovings (Oksman et al., 2003 & 2009). They incorporated the fiber-roving into the side extruder and the fiber content in the composites was estimated according to feeding speed and the weight of the roving per meter. Oever and Snijder observed the processing properties of jute fiber reinforced polypropylene produced by continuous extrusion compounding, in their study the continuous jute sliver was unwound and fed downstream to the extruder and a vacuum degassing unit was positioned between the compounding section and the die (Oever and Snijder 2008). De´ak et al. developed the long-basalt-fiber-reinforced thermoplastic composites using the similar LFT process, their used a special extrusion tool and driven the rovings to impregnate in the molten polymer (De´ak et al., 2010). Once the fibers are introduced, narrower kneading blocks will provide distributive mixing to open the fiber bundles with minimal damage (Schut 2003). According to Knights, one vent is typically enough to remove moisture from fibers in the LFT process (Knights 2003). The direct feeding of fiber-roving produces longer fiber lengths and yields higher structural performance than the pre-chopped fiber reinforced thermoplastics (Malnati 2007). Thus the LFT technique proved to be an efficient method for long natural fiber reinforced thermoplastic composites.

The compression moulding process of LFT allows fabricating larger parts such as automotive front end carriers, instrument panel carriers, door panel supports, under body shields and number of other applications (Schut 2004). The parts made in LFT are routinely around 3-4 kg although wall thickness is below 1mm (Bristol 2011). The compression molding followed by LFT, uses the extrudate of required size and is placed in the mould cavity and pressed under heat and
pressure. This method is expected to produce the components with higher fiber length along with its orientation in direction parallel to the flow. Mcleod et al. reported a comparative study on the fiber lengths obtained from in-line compounding of D-LFT direct of injection and compression moulding processes (Mcleod et al., 2010). The results indicated that the extrudate from the compression system contained considerable longer fibers, typically more than twice the length of the fibers when compared with the injection purged system.

This present work adapted the LFT process using a co-rotating fully intermeshing twin-screw extruder with self-wiping screws. The used screws were double-flighted with L/D ratio of 48:1 with parallel extrusion series. In the process, the fibers were continuously fed directly into the side feeder of the extruder. The fiber content in the final composite was estimated according to the screw speed and the weight of the roving per meter. The thermoplastic resin was fed into the twin-screw compounding equipment through gravimetric feeding. The long fibers in the form of roving were fed through opening located downstream section into a molten polymer. The mixing elements were used to disperse the fibers homogeneously in the polymer in order to produce a composite material with uniform structure and properties. In this work composites were manufactured using two different methods, the fiber were fed in two different positions named LFT-1 and LFT-2.

In the LFT-1 method, the fiber roving was incorporated into a side feeder of the extruder, which was near to the die head region, i.e. in between the two atmospheric vent channels as shown in Figure 2.5 As the natural fibers have low thermal stability, the lower shear with respect to residence time of was expected to achieve higher fiber length.
In LFT-2 method, the fibers were fed into the extruder on the same inlet of PP-matrix shown in Figure 2.6.
In this method, the incorporated fiber passes from feed section to die head along with the matrix. The higher shear with respect to longer residence time was expected to achieve good dispersion in the impact modified thermoplastic composites.

![Process flowchart for composite manufacturing](image)

**Figure 2.7:** Process flowchart for composite manufacturing.

### 2.4.3 Compression molding process

A conventional compression moulding press, Hindustan Hydraulics, India with a load capacity of 150 tons was used for sample preparation. The mold temperature of 170°C and the pressure
about 35 MPa were used to prepare a sheet of with a dimension of 250 x 125 x 3 mm. Before, the
sheet was removed, the mold was cooled down to ambient temperature. However, it process is
cost effective and provides higher impact strength with low shrinkage compared to unmodified
jute reinforced thermoplastic composites.

2.5 Characterization methods

The following characterization methods were used to study the hybrid bio-composites or the
composites produced using LFT process. The izod and falling weight impact were used to study
how the impact modifier and the manufacturing process influenced the toughness of the prepared
composites. Heat deflection temperature (HDT) was used to measure the stiffness changes under
elevated temperature. The dynamic mechanical analysis, scanning electron microscopy were used
to study the structure and properties of composites produced. The soxhlet extraction was used to
extract the fibers by using boiling xylene and optical microscopy was used to measure the fiber
length after the extrusion process. Uniaxial tension-tension fatigue testing was used to predict the
service life of the developed thermoplastic composites.

2.5.1 Interfacial strength measurement

The strength of the fiber/matrix interface can be accurately measured using pullout test; it
involves pulling a partially embedded single fiber out of a block of matrix material. It is
determined by applying pullout load; in the first stage of loading the induced shear stresses along
fiber do not exceed the bond strength between the fiber and the matrix i.e. both fiber and matrix
deforms elastically. As the applied load is increased the shear stress also increases until the
bonding between the fiber matrix breaks. The crack initiated may or may not progress
catastrophically depending on the friction between the fiber and matrix in the debonding zone.
The frictional shear stress beyond the bonding strength results from slipage due to the elastic elongation of the fiber in the deboned zone. The pull-out experiments may be performed using the universal tensile testing apparatus in accordance with EN ISO 53812 and ASTM D 1577. The pull-out load against displacement can be recorded using a computer controlled plotter. The apparent shear strength $\tau$ calculated from the maximum load $F_{\text{max}}$ and fiber embedded area in the matrix using the mathematical relation (Bismarck et al., 2005).

$$
\tau = \frac{F_{\text{max}}}{\pi dl}
$$

where $L$ is the embedded length and $d$ is the diameter of the fiber. The pullout theory is used to determine the fracture toughness of the composite and it depends on various factors such as Volume fraction ($V_f$), critical length ($l_c$), maximum strength ($\sigma_f$) of fiber and interfacial shear stress ($\tau_f$).

### 2.5.2 Mechanical testing

The tensile and flexural properties of the composites were studied as a function of impact modifier and coupling agent MAPP. Tensile strength: The tensile testing is done to measure the mechanical properties of the composites. It can also be used to characterize the toughness of a material. The area under the stress strain curve is directly proportional to the amount of energy a material can absorb, i.e. the toughness (Brydson 1999). The tensile strength of the composites may be analyzed according to ASTM D638 using the specimen size of (0.125” x 0.5” x 6.5”). The test indicates the material's behavior under loading tension. Tensile testing provides data such as, tensile yield strength, tensile strength at break (ultimate tensile strength), tensile modulus (Young's modulus), and elongation at yield and break. The tensile strength ($\sigma$) is given by equation below where $F$ is load, $b$ the width of the sample and $h$ is the thickness of the sample.

$$
\sigma = \frac{F}{A} \text{ or } \sigma = \frac{F}{bh}
$$
Strain or elongation is evaluated from equation below where $\Delta l$ is the extension, $l_0$: the initial gauge length.

$$E = \Delta l / l_0$$

(3)

The Young's modulus in tension ($E$) calculated from stress vs strain curve, where $E$ is the modulus of elasticity, $F$ is the force or load; $A_0$ is the original cross-sectional area through which the force is applied; $\Delta l$ Change in length of the composite and $l_0$ is the original length of the composite.

$$E = \sigma / \varepsilon = (F/A_0) / (\Delta l / l_0)$$

(4)

Flexural strength: The flexural testing of the composite will be performed according to ASTM D790 using the specimen size of (0.125" x 0.5" x 5.0"); the test indicates rigidity and stiffness of the composites. This test provides flexural modulus ($E_f$) and flexural strength ($\sigma_f$), calculated by using the following equations:

$$E_f = \frac{ml^3}{4bh^3}$$

(5)

$$\sigma_f = \frac{3Fl}{2bh^2}$$

(6)

where $F$ is load, $m$ initial slope of the load vs. deflection curve and $l$: specimen length between two support points, $b$ is the width and $t$ is the thickness of the test piece.

### 2.5.3 Facture toughness and fatigue testing

Fracture mechanics provides a quantitative description of the resistance of a material to fracture. The fracture toughness is a material property, which can be used to predict the behavior of components containing cracks or sharp notches. The fracture toughness properties are obtained by tests on specimens containing deliberately introduced cracks or notches and subjected to prescribed loading conditions.
This present study adapted, the critical crack tip opening displacement to measurement of fracture toughness of the impact modified thermoplastic composites. The fracture test is carried out in displacement control at a constant rate of increase of stress intensity while recording load and crack opening displacement data, until the specimen breaks. Fracture mechanics concepts have been developed to assess the safety of components containing cracks. Fatigue cracking is one of the primary damage mechanisms of structural components. Fatigue cracking results from cyclic stresses that are below the ultimate tensile stress, or even the yield stress of the material. The fatigue life of a component can be expressed as the number of loading cycles required to initiate a fatigue crack and to propagate the crack to critical size. Therefore, it can be said that failure occurs in three stages – crack initiation; slow, stable crack growth; and rapid fracture (Mallick et al., 1997). The fatigue behaviour of composites will be influenced by the toughness; as a result, compatibilized impact modified thermoplastic composites achieved good fatigue resistance when compared to the unmodified one. The equations used to evaluate the fracture toughness and fatigue resistance are described in Chapter 7.

2.5.4 Impact testing

Energy per unit thickness required to break a test specimen under sudden load. The impact was provided with built-in pendulum hammer. The energy lost by the pendulum is equated with the energy absorbed by the test specimen. The standard test for impact strength of a material includes charpy test, Izod test. The izod impact test was performed according to ASTM D 256 of size (0.125” x 0.5” x 2.5”), notched izod impact test to find the energy absorption behaviour of the reinforced composites. Notched impact strength ($I_{no}$) is calculated by the following equation,

$$I_{no} = \frac{w}{bh} \times 10^3$$  

where w is the impact energy, b the width of the sample and h is the thickness of the sample.
The test was conducted in M/s. Tinius Olsen, Plastic impact tester (Model IT 503), Germany. The impact was provided with built-in pendulum hammer. At least ten specimens were tested for each set of samples and the mean values were reported.

The falling weight impact is used to evaluate the toughness of the composite, the main advantage of using falling weight impact test is, and its ability to reproduce under which real life component would be subject to impact loading. This method uses the free fall of a known weight to supply the energy to break thermoplastic composites. The test specimen of 60 x 60 mm sheet is clamped securely and the impact striker which falls through a vertical guide tube that guide tube, which is perpendicular to the impact surface. The kinetic energy of the falling weight was adjusted by varying its drop height. The resistance forces of the composites were measured by a load cell. The parameters force, deformation, energy absorption and velocity were recorded using a computer controlled plotter. The energy released from the drop weight test is calculated from following equation:

\[ E = mgh - l \]  

where E is energy (J); m: mass of the impact striker (kg); g: gravity (m/s^2), h: height in (m), l-loss incurred by friction and other source.

### 2.5.5 Dynamic mechanical analysis

Dynamic mechanical analysis has been widely used for investigating viscoelastic behaviour of polymeric materials. It determines the relevant stiffness and damping characteristics. It records the mechanical response (i.e. load and displacement) of a material as a function of temperature (time) while cycled at a single or multiple applied frequencies. DMA is a versatile technique that can be used in many deformation modes (e.g. tension, compression). From the dynamic
mechanical analysis the molecular mobility within materials and commonly used for measuring the changes in mechanical properties (such as storage modulus \( E' \)) and loss modulus \( E'' \) brought about by chemical reactions. The complex modulus \( E^* \), storage modulus \( E' \), loss modulus \( E'' \) and loss factor \( \tan \delta \) can be calculated by using the equations, given below:

\[
|E^*| = \frac{\sigma_A}{\varepsilon_A} \tag{9}
\]

\[
|E^*| = \sqrt{[E'(\omega)]^2 + [E''(\omega)]^2} \tag{10}
\]

\[
E'(\omega) = |E^*|. \cos \delta \tag{11}
\]

\[
E''(\omega) = |E^*|. \sin \delta \tag{12}
\]

\[
\tan \delta = \frac{E''(\omega)}{E'(\omega)} \tag{13}
\]

Dynamic mechanical analysis was performed using TA Instruments DMA Q800, (Delaware, USA). The measurement was conducted in three-point bending mode at a frequency of 1 Hz under nitrogen atmosphere. The dynamic storage modulus, loss modulus and loss factor of the composites were determined as a function of temperature ranging from -25 to 100°C.

### 2.5.6 Heat deflection temperature

A rectangular specimen is subjected to a three point bending load while immersed in a heat transfer medium. The temperature is raised at uniform rates, between 0.2 to 2 °C/min. The temperature of the medium is measured when the test bar has deflected 0.25 mm. The deflection versus temperature is plotted; the plots were initially adjusted to a zero displacement due to the negative displacement caused by the initial deflection of the test coupon under load. The result of this test is called the deflection temperature under flexural load. The load is calculated as follows:

\[
P = \frac{(2 \cdot S \cdot b \cdot d^2)}{3 \cdot L} \tag{14}
\]
where, $P$ is load, $S$ is maximum stress in the specimen, $b$ width of specimen, $d$ the depth of specimen and $L$ is span between supports.

2.5.7 Microscopy

The scanning electron microscopy (SEM) was used to study the morphology and microstructure of fibers as well as fracture surface of the composites. The fractured surfaces of the composites were analyzed to predict the interaction between the fiber and matrix. It was also used to examine the fiber dispersion in the hybrid bio-composites. The SEM was carried out in a vacuum chamber; with high voltage applied across the tungsten filament to generate a beam of electrons and serves the cathode. The generated electrons accelerates downward towards the sample and finally the condensing lens, condenses the electron in a beam and the objective lens focus the beam to a fine point on the sample. Thus generates, a very high-resolution image of sample surface, revealing details about less than 1 nm in size. Samples were sputter coated with gold to avoid charging.

2.5.8 Fiber extraction and fiber size measurement

The tensile strength and impact energy of the composites increases significantly with increase in fiber length. Therefore the long fiber extrusion technique was adapted in the study. It is obvious that fiber breakage takes place during the rotation of the screw, but the average fiber lengths or the range of fiber lengths after the process will be greater than the short fiber composites (Mallick and Broutman 1997). In this study the fibers were extracted from the composites by dissolving the polypropylene with hot xylene using soxhlet extraction apparatus. The residual fibers separated manually using water and their lengths were automatically measured using optical microscope.
2.6 Challenges of hybrid composite development

Hybrid LFT composites were developed with the direct incorporation of roving’s on molten polymer into the extruder. In the extruder the fiber length will be decreased due to the high shear forces. Reinforcing high bulk density fibers in continuous form is a challenging task. The used fiber rovings were hand-made and had therefore varied weight/length; therefore another important challenge was to achieve the targeted fiber content. The main advantage of the LFT process is the reduction of handling and manufacturing steps compared to the production of the semi-finished products.

2.7 Potential applications of LFT composite

LFT have gained an increasing market share in the automotive industry since the last decade (Krause et al., 2003). The continuous growth of LFT composites in Europe for automotive application is recorded (Henning et al., 2005). The typical applications of LFT thermoplastic composites are presented in Figure 2.8. As the LFT composites have a higher market demand in larger parts of automotive components, it is necessary to have economical production. Cost effective LFT material compositions with higher performance were developed.
Figure 2.8: Automotive applications of long fiber reinforced PP (Chaudhari et al., 2008)

2.8 Problem statement

Jute fibers have higher strength and stiffness than sisal, banana (Oksman et al., 2009). They are often used as reinforcement in thermoplastics in the form of short fibers. This thermoplastic composite tends to have lower energy absorption due to the synergetic effect. The problem could be overcome by several ways such as incorporation of impact modifier, improving the fiber dispersion, orientation and by minimizing the fiber breakage. Thereby the use of continuous fibers in the manufacture of thermoplastic composite has been adapted in the present work.

Generally jute fibers are available in long form, they are connected together to form “roving”. The regenerated cellulose are readily available in continuous roving form has higher degree of elongation behaviour which could be used as impact modifier. The mix of both jute and the
viscose fiber in the continuous roving form in the preparation of thermoplastic composites is a challenging task. The direct feeding of continuous fiber into the extrusion barrel, (LFT 1) process used for the preparation of the long fiber reinforced thermoplastic composites. This technique adapted with the aim of improving the fiber dispersion, orientation and to minimize the fiber breakage. However as both the fibers are hydrophilic in nature and as a result fiber disperses poorly in the non-polar thermoplastic because of strong hydrogen bonds between fibers. Further the natural fibers tend to degrade at lower temperatures i.e. above 200°C, the lower thermal stability of the fibers limits their matrix selection. The polypropylene has lower impact strength especially at lower temperature and they receive much attention in the natural fiber reinforced thermoplastic composites. The primary objective of the work is to use continuous viscose fibers as impact modifier on long jute fiber reinforced composite using LFT extrusion process.
3 Hypotheses and research objectives

3.1 Hypotheses

The research hypothesis that the combination of tough, strong fibers and relatively long fibers prepared with direct LFT extrusion technique will enhance energy absorption of the composites.

3.2 Objectives

The project aims to overcome the fundamental challenge; the preparation of long continues jute fiber reinforced thermoplastic composites using direct LFT technique. The objective of this research is as follows:

To investigate the energy absorption behaviour of natural fiber reinforced hybrid composites

1. To examine dispersion of jute and viscose fibers after extrusion process
2. To evaluate the interfacial adhesion between the PP matrix and the fibers
3. To study the effect of energy absorption behaviour of viscose fiber in thermoplastic composites
4. Examine the fracture mechanism from electron microscopy images
5. To predict fracture toughness of hybrid composite using micro-mechanical model.

3.3 Scope of the research work

The primary goal of the project was achieved through the incorporation of impact modifier. The experimental design is based on the research approach is shown in Figure 3.1. The long viscose (tough) fiber was opted in the study, which might function as an impact modifier to enhance the impact resistance of the composites. The long jute fiber will act as reinforcement that can
increase the strength and stiffness of the composite. Thereby the toughening of composite can be done without compromising the strength and stiffness of the thermoplastic composite.

**Figure 3.1:** Schematic representation of research approach

The direct LFT extrusion technique was adapted using twin-screw extruder where the fiber roving was fed into side-feeder of the extruder in which the extruder speed determines the rate at which fibers are pulled into the extruder. This production method also led to well dispersed and distributed fibers with longer fiber lengths, which are necessary for the improvement of all mechanical properties. The effect of work energy absorption of long jute fiber reinforced composites modified with viscose fiber was investigated based on various parameters as described in the chart given above.
3.4 Thesis outline

The finding of the above proposed objectives are presented in Chapter 4 to 7.

**Chapter 1** offers the scientific background for the toughening of long fiber reinforced thermoplastic (LFT) composites.

**Chapter 2** presents a compressive literature on materials, availability, properties, surface modification and processing of hybrid bio-composites, challenges involved in the development of hybrid long fiber reinforced thermoplastic (LFT) composites and problem statements.

**Chapter 3** describes the hypotheses, objectives, scope of the research and outline of the thesis.

**Chapter 4** describes the development of long jute fiber reinforced thermoplastic composites with long and tough regenerated cellulose as an impact modifier using direct LFT extrusion technique. Different weight concentrations of impact modifier were used and formulated the thermoplastic composite. The volume fraction of impact modifier was optimized to produce high-energy absorption composite without sacrificing much in the other mechanical properties. The effect of various weight concentration of impact modifier on mechanical properties of PP long jute fiber reinforced composites was investigated. The interfacial adhesion between the fibers and matrix are investigated by single fiber pullout test. These findings are also supported by Heat deflection/distortion temperature (HDT) by showing the dimensional stability of the composites. Further the natures of the fiber breakage in the composites were investigated using scanning electron microscopy (SEM). The results of this chapter are published in Journal of Applied Polymer Science (Ranganathan et al., 2015).

**Chapter 5** describes the effect of energy absorption of modified polypropylene long jute fiber reinforced thermoplastic composites using viscose fiber and MAPP. The amount of energy required to break the thermoplastic composites were investigated by various impact test. In this
chapter, the conventional Charpy impact method and instrumental falling weight impact test are performed. The dynamic mechanical analysis (DMA) is used to characterize the viscoelastic properties of the thermoplastic composites. The failure mechanisms of the composites are analyzed on the impacted samples using scanning electron microscopy (SEM). In addition the fiber dispersion of the composites was studied from the optical microscopy images of microtomed cross sections. The outcomes of the study are published in Journal of Applied Polymer Science (Ranganathan et al., 2016).

Chapter 6 describes the microstructure of fiber bundles, the longitudinal and cross sections and the effect of processing on fiber content, fiber length and the fracture toughness of the impact modified composites. In this study, the stress intensity factor under compact tension is analyzed for all the composites modified using impact modifier and MAPP. The target content of the fiber varied for all the compositions, which in turn affect the length of the fiber due to the variation in the shear during processing. The effect of fiber content and length of both fibers determines the toughness of the composites. The findings of this chapter are published in Composite Part-A (Ranganathan et al., 2015).

Chapter 7 illustrates the comparison of mechanical properties, toughness and fiber dispersion of viscose fiber reinforced thermoplastic composites developed using direct-LFT extrusion technique with different feeding types. The two different methods of feeding were carried to produce uniform fiber dispersion in the composites. In the first method i.e., (LFT-1), viscose fibers roving were incorporated into a side feeder of the extruder which is near to the die head region, whereas in the second method (LFT-2) the fiber were fed into the extruder on the same inlet of PP-matrix, but in both cases the fibers are fed directly into the polymer melt and the fiber weight fractions were controlled by the screw speed based on the target content, the weight of
the fiber roving per meter and the length per screw revolution and time. The effect of processing on the mechanical properties, toughness and the morphology of the thermoplastic composites is investigated. The mechanical properties and the toughness of the composites are experimentally measured using universal testing machine and the fiber dispersion was investigated by optical microscope on the microtomed cross-sections. The outcomes of the studies are accepted in Polymers for Advanced Technologies (Ranganathan et al., 2015).

Chapter 8 includes conclusion of work, study limitations and recommendations for future work.
4 Regenerated cellulose fibers as impact modifier in long jute fiber reinforced polypropylene composites: Effect on mechanical properties, morphology and fiber breakage

4.1 Introduction

The primary research on D-LFT composites using hybrid fibers is to achieve better mechanical performance of the thermoplastic composites together with improved energy absorption. The incorporation of fibers in the form of continuous roving using twin screw extruder will have a trend to retain higher fiber length and volume fraction (Grauer et al., 2012). LFT have excellent mechanical properties and stiffness-to-weight ratio, which is of great interest to the automotive industry (Häuptli et al., 2003).

The weakness of natural fiber reinforced thermoplastic composites is poor resistance to impact strength due to the lack of plastic deformation mechanism. As the PP has low impact strength at low temperatures and the addition of fibers causes the reduction of matrix deformation, which leads to further lowering of fracture toughness (Oksman et al., 1998). An intrinsic cause of low fracture toughness of the composites is inherent brittleness of the fibers. Therefore, improving the toughness with better interfacial adhesion would lead to better performance of the composites. Various potential toughening approaches have been attempted to solve this problem for natural fiber reinforced thermoplastic composites. One efficient way of improving the impact energy-absorbing ability of composite materials is to add tough materials to the host composites, such as high strain-to-failure fibers (Muhi et al., 2009). Adekunle et al 2011 successfully used Lyocell fibers (regenerated cellulose fibers) as an impact modifier in jute fiber reinforced
thermoset composite, and reported that the Lyocell fibers increases the impact resistance of the composites with longer pullout lengths (Adekunle et al., 2011). Graupner et al. reported, that the thermoplastic composites prepared with a mixture of bast fibers such as hemp, kenaf and cotton are suitable for various car components (Graupner et al., 2009). The combination of hemp, kenaf with cotton produced positive tensile characteristics of natural fibers with good impact properties of cotton.

In fiber-reinforced composites, fibers bridging the cracks in the matrix can provide resistance to crack propagation before these fibers break or are pulled out (Soon et al., 2000). However, the extent of improvement toughness and energy absorption of the composites depends on factors such as: fiber content, aspect ratio, orientation and interfacial adhesion between fiber and matrix.

The hypothesis of this study is that the combination of tough and strong fibers in the preparation of composites using direct LFT extrusion technique offers high fiber content, controlled aspect ratio and orientation. Thereby enhances energy absorption of the composites due to toughest fiber by providing longer fiber pullouts. Further the addition of maleated coupling agent increases the compatibility and improves the interfacial adhesion between the fibers and the matrix.

In this work, long and tough viscose fibers were used as an impact modifier in jute reinforced PP composites. The composites were manufactured through LFT process and the composites mechanical properties including impact strength and the energy absorption were investigated. In addition the microstructure and the extent of fiber breakage during the melt mixing process were studied using electron microscopy.
4.2 Experimental

4.2.1 Materials

Homopolymer polypropylene (PP) Propel 1350YG, extrusion grade, with MFI of 35g/10 min (230°C, 2.16 kg) was purchased from Indian oil corporation ltd., India used as matrix. A maleic anhydride grafted polypropylene, Epolene E-43, Sigma Aldrich, USA was used as a coupling agent. The jute fibers were procured from Chandra Prakash & Co. Pvt. Ltd., Jaipur, India. The fibers were used in the form of long fiber roving. The viscose fibers were supplied from Cheran Spinning Mills, Erode, India. They are derivative of wood pulp said to be regenerated cellulose processed by spinning method. The density of the fibers was 1.3 g/cm³, and used in the form of continuous fibers.

<table>
<thead>
<tr>
<th>Materials</th>
<th>Tensile strength (MPa)</th>
<th>E-modulus (GPa)</th>
<th>Elongation at break (%)</th>
<th>Density (g/cm³)</th>
<th>Fiber length (mm)</th>
<th>Fiber roving length (m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Jute</td>
<td>393-773ᵃ</td>
<td>13-26.5ᵃ</td>
<td>1.2-1.5ᵃ</td>
<td>1.3ᶜ</td>
<td>2-5ᵃ</td>
<td>1-2</td>
</tr>
<tr>
<td>Viscose</td>
<td>340ᵃ</td>
<td>12ᵃ</td>
<td>11ᵃ</td>
<td>1.3</td>
<td>38ᵃ</td>
<td>Continuous</td>
</tr>
</tbody>
</table>

ᵃ Data from literature [Oksman et al., 2009];ᵇ Data from the manufacturer;ᶜ Data from literature [Lewin et al., 1998]

Table 4.1 shows the properties of used fibers. Jute fibers are cheap raw material, having very good mechanical properties, the tensile strength and moduli of the fibers were found to be 393-773 MPa and 13-26.5 GPa with apparent density of 1.2 g/cm³ was used as reinforcement. They are available abundantly in India of various forms such as fiber roving, hessian cloth, yarn etc. The present work has opted in the form of long-fiber roving having 1-2 m length with a fiber diameter ranging from 20-25 µm. The appearance of the fiber “roving” is shown in Figure 4.1.
The viscose fibers have tensile strength and modulus of 340 MPa and 12 GPa respectively. They have high elongation at break, approximately 12%, which act as impact modifier. Similar to jute fibers the viscose fibers were also used in the form of long roving and were slightly twisted with the jute roving to make a mix of these fibers as shown in Figure 4.1.

![Figure 4.1: Handmade Jute fiber roving, viscose fiber in continuous form and a mixture of jute and viscose fibers](image)

### 4.3 Processing of composite material

#### 4.3.1 Compounding

Prior to compounding, the jute fibers were washed and dried at 60°C for 48 hours until achieving a uniform weight at different intervals of time. Similarly the viscose fibers were dried at 60°C for at least 2 hours. The composite materials were manufactured using direct LFT processing method, using a high performance co-rotating twin-screw extruder (ZE-25 model Berstorff Maschinenbau GmbH, D-3000 Hannover, Germany). The continuous roving was incorporated into a side feeder of the extruder, and fed directly into the polymer melt. The composites were prepared with various viscose fiber content ranging from 5 to 15 wt% and the jute fiber content was kept constant at 30 wt%. The fiber weight fractions was controlled by the screw speed and it was calculated using targeted fiber content, the weight of the fiber roving per meter, the length
per screw revolution and time. The compound was extruded in the form of profile and cut to the required lengths. The throughput was 5 kg/hr and the compositions of different materials and the processing setting along with the temperature profile are shown in Figure 4.2 and Table 4.2. The compositions of the prepared using various concentration of impact modifier on PP-J materials is shown in Table 4.3.

**Figure 4.2:** The schematic representation of the LFT extrusion process with processing parameters

**Table 4.2:** Processing parameters for the materials preparation using a twin screw extruder

<table>
<thead>
<tr>
<th>Parameters</th>
<th>Settings</th>
</tr>
</thead>
<tbody>
<tr>
<td>Screw speed (rpm)</td>
<td>100</td>
</tr>
<tr>
<td>Screw diameter (mm)</td>
<td>25</td>
</tr>
<tr>
<td>Screw L/D ratio</td>
<td>48:1</td>
</tr>
<tr>
<td>Temperature profile (°C)</td>
<td></td>
</tr>
<tr>
<td>Zone 1</td>
<td>150</td>
</tr>
<tr>
<td>Zone 2</td>
<td>155</td>
</tr>
<tr>
<td>Zone 3</td>
<td>160</td>
</tr>
<tr>
<td>Zone 4</td>
<td>165</td>
</tr>
<tr>
<td>Zone 5</td>
<td>170</td>
</tr>
<tr>
<td>Zone 6</td>
<td>175</td>
</tr>
<tr>
<td>Zone 7</td>
<td>180</td>
</tr>
<tr>
<td>Zone 8</td>
<td>185</td>
</tr>
<tr>
<td>Zone 9</td>
<td>190</td>
</tr>
</tbody>
</table>
4.3.2 Compression molding

Prior to compression molding, the cut profiles were dried at 60°C for 2 hours. Sheet size of 250 x 125 mm and 3 mm in thickness were prepared using a conventional compression molding press, Hindustan Hydraulics, India with a load capacity of 150 tons. The mold temperature was 170°C and the pressure was about 35 MPa. The samples for mechanical testing were cut from the sheets according to ASTM standards.

<table>
<thead>
<tr>
<th>Materials</th>
<th>PP (wt.%)</th>
<th>Jute fiber (wt.%)</th>
<th>Viscose fiber (wt.%)</th>
<th>MAPP (wt.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP</td>
<td>100</td>
<td>0</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>PP-J</td>
<td>70</td>
<td>30</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>PP-J-M</td>
<td>68</td>
<td>30</td>
<td>0</td>
<td>2</td>
</tr>
<tr>
<td>PP-V</td>
<td>70</td>
<td>0</td>
<td>30</td>
<td>0</td>
</tr>
<tr>
<td>PP-V-M</td>
<td>68</td>
<td>0</td>
<td>30</td>
<td>2</td>
</tr>
<tr>
<td>PP-J-V5</td>
<td>65</td>
<td>30</td>
<td>5</td>
<td>0</td>
</tr>
<tr>
<td>PP-J-V10</td>
<td>60</td>
<td>30</td>
<td>10</td>
<td>0</td>
</tr>
<tr>
<td>PP-J-V15</td>
<td>55</td>
<td>30</td>
<td>15</td>
<td>0</td>
</tr>
<tr>
<td>PP-J-V10-M</td>
<td>58</td>
<td>30</td>
<td>10</td>
<td>2</td>
</tr>
</tbody>
</table>

4.4 Testing and characterization

4.4.1 Mechanical testing

The tensile properties of the composites were performed according to ASTM D 638 using conventional tensile tester, Instron 3382, UK with a crosshead speed of 3 mm/min. The tensile strength, tensile modulus and elongation at break are calculated from the tensile test data. The flexural testing was performed according to ASTM D 790, with a support span of 16 times the
sample thickness and strain rate of 5 mm/min using the same equipment. The flexural strength is determined using $\sigma_s = (3P_{\text{max}}L)/(bh^2)$, where $P_{\text{max}}$ is the maximum load at failure (N), $L$ is the span (mm), $b$ and $h$ is the width and thickness of the specimen (mm), respectively. Flexural modulus was calculated from $E_f = (mL^3)/(4bh^3)$, where $m$ is the initial slope of the load deflection curve. Impact testing was performed according to ASTM D 256 on notched Izod specimens using M/s. Tinius Olsen, Model IT 503, Germany. The impact was provided with built-in pendulum hammer. The At least ten specimens were tested for each set of samples and the mean values were reported. Toughness (energy absorption) of the composites was calculated from the tensile results. The tensile toughness is defined here as the area under the stress- strain curve up to the complete rupture of the sample.

4.4.2 Single fiber pullout test

Single fiber pullout test was used to characterize the fiber-matrix interface. The bonding characteristics single fibers were investigated using pullout test. The samples were prepared by embedding a controlled length of the fiber in the PP matrix. The test was carried out using Instron 3382, Universal testing machine (UTM) tester, UK by pulling out the fiber from the resin block. The load and its extension were measured from the test in a force displacement. At least twenty five samples were tested on each composition.

4.4.3 Softening temperature

Heat deflection temperature (HDT) was measured by Ceast HDT apparatus, Italy. The testing was performed according to ASTM D 648 standard. The composites samples were tested at a rise in temperature of 2°C/min with a loading pressure of 0.45 MPa (66 psi). Five specimens were tested for each set of samples and the mean values were reported.
4.4.4 Electron microscopy

Fractured surfaces of the impact test samples of the composites with jute and viscose fibers were analyzed using scanning electron microscopy (SEM) with a Tesan Vega3 SBU, Czech Republic. An acceleration voltage of 5 kV were used and the sample surfaces were sputter coated with gold prior to SEM observation to avoid charging.

4.4.5 Fiber length measurements

The fibers were extracted by dissolving the composites with boiling xylene. The fibers were manually separated without overlapping using water and observed using optical microscope. Length of the fibers was calculated automatically from the images through software. The length given for each fiber is average of at least 100 measurements.

4.5 Results and discussion

4.5.1 Tensile properties

The mechanical properties of PP-J composites modified with various concentrations of VF and compatibilized with MAPP are shown in Table 4.4 and Figure 4.3. The PP-J composite is used as reference for the comparison. The addition of jute fibers did not increase the tensile strength of the composites, which is an indication of poor adhesion between the jute fiber and the PP matrix (Oksman et al., 2003). This was also seen because an addition of MAPP on the PP-J composite exhibited a higher tensile strength than the uncompatibilized material. Thereby the stress is not transferred from the matrix to the stronger fibers (Doan et al., 2006). The addition of viscose fibers showed a positive effect on the Izod impact strength but slightly decreased the tensile strength and modulus. The impact strength improved 73%, 170% and 147% with an addition of
5, 10 and 15 wt% of the impact modifier respectively while the tensile strength decreased with 7%, 12% and 14% as compared to PP-J composites. The decrease in the tensile strength is attributed to the lower mechanical properties of viscose fiber (Hes et al., 2010; Erdumlu et al., 2008). The lower mechanical properties of viscose fiber had a negative effect on the tensile properties of impact modified jute fiber reinforced PP system when compared to PP-J composites.

![Graph showing tensile properties](image)

**Figure 4.3:** Tensile properties of jute fiber reinforced polypropylene composites modified with viscose fibers as impact modifier and MAPP.

Generally, the addition of jute fibers increases the modulus of the composites. The modulus of the composites was progressively increased from 0.6 GPa of neat PP to 2.7 GPa for the composites with 30 wt% jute fibers, this is due to the high stiffness of these fibers. As expected the stiffness was reduced of all composites with impact modifiers which can be explained due to the low modulus of viscose fiber, whereas the addition of 2wt% of MAPP improved the tensile modulus of the composites. The PP-J-V10-M composites did not show much reduction in strength and stiffness compared to PP-J-M.
Table 4.4: Mechanical properties of PP composites containing 30 wt.% jute fibers modified with viscose fibers and MAPP (values in the bracket are standard deviation)

<table>
<thead>
<tr>
<th>Material code</th>
<th>Flexural strength (MPa)</th>
<th>Flexural modulus (MPa)</th>
<th>Tensile strength (MPa)</th>
<th>Tensile modulus (MPa)</th>
<th>Elongation to break (%)</th>
<th>Impact strength (J/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP</td>
<td>33.2(0.8)</td>
<td>867(22)</td>
<td>27.6(0.8)</td>
<td>634(49)</td>
<td>9.8(0.1)</td>
<td>28.0(0.8)</td>
</tr>
<tr>
<td>PP-J</td>
<td>47.1(7.1)</td>
<td>5269(482)</td>
<td>29.1(1.1)</td>
<td>2700(103)</td>
<td>3.3(1.0)</td>
<td>24.4(3.1)</td>
</tr>
<tr>
<td>PP-J-M</td>
<td>63.4(4.2)</td>
<td>6329(388)</td>
<td>32.3(1.5)</td>
<td>2793(184)</td>
<td>2.4(0.4)</td>
<td>22.1(3.4)</td>
</tr>
<tr>
<td>PP-V</td>
<td>38.8(2.1)</td>
<td>2996(71)</td>
<td>18.3(0.6)</td>
<td>1426(93)</td>
<td>8.9(2.3)</td>
<td>84.3(2.7)</td>
</tr>
<tr>
<td>PP-V-M</td>
<td>49.6(7.1)</td>
<td>3446(116)</td>
<td>19.1(0.6)</td>
<td>1452(68)</td>
<td>7.4(0.6)</td>
<td>79.6(4.0)</td>
</tr>
<tr>
<td>PP-J-V5</td>
<td>46.3(5.5)</td>
<td>4969(389)</td>
<td>26.4(1.2)</td>
<td>2446(171)</td>
<td>4.0(0.7)</td>
<td>42.1(2.3)</td>
</tr>
<tr>
<td>PP-J-V10</td>
<td>43.3(5.9)</td>
<td>4819(332)</td>
<td>25.1(1.1)</td>
<td>2408(53)</td>
<td>4.4(0.4)</td>
<td>65.8(3.4)</td>
</tr>
<tr>
<td>PP-J-V15</td>
<td>42.9(6.5)</td>
<td>4260(393)</td>
<td>24.5(0.8)</td>
<td>2399(191)</td>
<td>4.9(0.4)</td>
<td>60.1(3.2)</td>
</tr>
<tr>
<td>PP-J-V10-M</td>
<td>59.2(8.1)</td>
<td>5908(218)</td>
<td>30.4(1.0)</td>
<td>2688(95)</td>
<td>5.0(0.1)</td>
<td>67.4(2.7)</td>
</tr>
</tbody>
</table>

Figure 4.4 shows the elongation at break of PP-J composites modified with various concentrations of the impact modifier. The addition of viscose fibers on PP-J composites increased the strain to failure of the composites. Fu et al showed enhancement of strain to failure with the use of short glass fiber (SGF) in PP/short carbon fiber (SCF) composites, they reported the increase of fracture toughness due to the positive hybrid (synergistic) effect (Fu et al., 2000). This present study observed the increase in the concentration of viscose fiber has continuously increased the percentage of elongation values of the composites, which indicates the decrease in the stiffness. Graupner et al study observed, the incorporation of Lyocell fiber onto the PLA-hemp composites has increased elongation from 1.2 to 1.9%. They found the regenerated cellulose fiber have linear elastic behavior in the first section of tensile test and had higher elongation in the rest of the single fiber test (Graupner et al., 2009). In this work the addition of MAPP on the hybrid composites has accompanied the improvement in the ductility of the
composites. Almost 50% of improvement in the elongation values was observed with PP-J-V10-M when compared with PP-J composites.

![Graph showing elongation values for different composites.](image)

**Figure 4.4:** Elongation at break of jute fiber reinforced polypropylene composites modified with viscose fibers as impact modifier and MAPP

### 4.5.2 Flexural properties

Figure 4.5 shows the flexural strength of PP-J composites compared with the composites of different concentration of impact modifier and MAPP. The study observed that the addition of 5 wt% impact modifier does not show much variation in the flexural stress, whereas the increase in the concentration of impact modifier to 10 and 15 wt% resulted in 8.5 and 10.5% reduction in the flexural strength of the composites in comparison to PP-J. However, the MAPP compatibilizer had a positive effect on the impact modified composites. The PP-J-V10-M composite has resulted with 28% higher flexural strength than PP-J composites but 6% lower than that of
PP-J-M composites. The improved strength indicates the better stress transfer from matrix to fiber.

Table 4.4 summarizes the flexural modulus of the jute reinforced PP composites modified with various concentrations of the impact modifier and compatibilizer. The results are also shown in Figure 4.5. The composite with highest flexural modulus was achieved with the combination of compatibilizer and PP-J composites, because of the higher fiber stiffness and strong interface. The addition of impact modifier has significantly improved the flexible interphase between the fiber and matrix (Yazdani et al., 2006). The study observed that the, optimized interfacial adhesion was achieved with PP-J-V10-M component. The flexible interface attained has promoted the higher pullout of the fibers rather than the fiber breakage, which resulted with the efficient transfer of stress from matrix to fibers. Thereby the composites found to have improved the energy absorption without much sacrifice in the modulus.

![Graph showing flexural properties of jute fiber reinforced polypropylene composites modified with viscose fibers as impact modifier and MAPP](image)

**Figure 4.5:** Flexural properties of jute fiber reinforced polypropylene composites modified with viscose fibers as impact modifier and MAPP
4.5.3 Impact strength

Figure 4.6 represents the notched izod impact strength in turn the energy absorption of each compositions. The jute composites showed low impact strength of 24 J/m, which attributes to the presence of strong fiber reinforcement and poor fiber-matrix interface. The toughness of the composites is usually affected by the interfacial parameters and the mode of failure that are observed between the matrix and the fiber i.e. fiber fracture, debonding and pullout. The improvement in impact strength values were observed in all composites modified with various concentration of impact modifier, i.e. the incorporation of higher strain to failure fiber has improved the impact strength values. The higher strain rate determines the amount of energy absorbed by a material during fracture (Bledzki et al., 2007). The study depicts the addition of 5wt% impact modifier had shown the improvement of 73%, the increase in the volume content of impact modifier to 10 wt% has again raised the impact strength from 42 to 66 J/m. The higher improvement of impact strength may be explained due to the longer fiber pullout length of viscose fiber. Benevolenski et al reported that hybrid composites where flax fibers were partly replaced by Lyocell fibers improved the impact strength of the composites (Benevolenski et al., 2000). The microscopy study confirmed longer fiber pullout lengths in hybrid composites compared than the flax fiber composites; they speculated that the high ductility of Lyocell fibers is responsible for the longer pullout lengths. Hence, this study clearly indicated that the addition of viscose fiber changes the fracture behavior from brittle to ductile.

The crack propagation is the predominant toughening mechanism in the notched impact test (Wong et al., 2010). The improvement of impact strength characterizes higher stress transfer from matrix to fiber and able to absorb energy effectively. The better explanation on positive effect on energy absorption in this case is also expected due to the fiber length and its orientation.
(Norman et al., 2003). The longer the fiber will have effective energy dissipation; the tough fiber had a trend to maintain their fiber length and orientation after processing was the possible reason for improved toughness of the composite. The compatibilized system had a significant impact on the impact strength of the composite. The addition of 2 wt% MAPP on PP-J and PP-V system has shown a negative effect, this may be due to the strong interface between the fiber and matrix, but the compatibilized PP-J-V-10 had positive effect.

![Figure 4.6: Izod impact strength of notched samples of jute fiber reinforced polypropylene composites modified with viscose fibers as impact modifier and MAPP](image)

The reason for the drastic improvement in the impact strength is due to the weak (flexible) interface, which dissipates more energy through stress transfer from matrix to fiber whereas in case of uncompatibilized composites the higher dissipation of energy occurs for fiber pullouts and debonding due to their very weak interface. Hristov et al reported that the total fracture energy of unmodified PP/wood fiber composite slightly decreases compared to the virgin PP,
while addition of the MAPP leads to about 20% increase of the absorbed total energy (Hristov et al., 2005). Maleated polypropylene (MAPP) ensures better adhesion between the matrix and fibers leading to increased impact strength.

### 4.5.4 Toughness of impact modified composites

The toughness values shown in the Table 4.5 are retrieved from the tensile stress-strain curves. Typical load - displacement curves of PP-J, PP-J-V10 and PP-J-V10-M are illustrated in Figure 4.7. The figure clearly shows the unmodified PP-J composite exhibit brittle fracture, characterized by a sharp drop immediately after reaching the maximum tensile strength, the plastic deformation of the unmodified composite seems to be much lower than the other composites.

<table>
<thead>
<tr>
<th>Material Code</th>
<th>Without MAPP</th>
<th>With MAPP</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Interfacial strength (MPa)</td>
<td>Toughness (J/m³)</td>
</tr>
<tr>
<td>PP-J</td>
<td>9.5(0.3)</td>
<td>415</td>
</tr>
<tr>
<td>PP-V</td>
<td>1.9(0.2)</td>
<td>1637</td>
</tr>
</tbody>
</table>

The low value of strain to failure has contributed to limited toughness of the composite (Bledzki et al., 2007), which is probably due to the higher pullout of the fiber due to poor interfacial adhesion of fiber and matrix and higher stiffness of jute fibers. The system also exhibited with lower impact energy when tested in vertical direction along with a notch. The viscose fiber have a good strain to failure characteristic, the regenerated cellulose also have good fiber matrix adhesion than the natural fiber (Kim et al., 2008). Thus as expected the addition of viscose fiber
to small wt% of about 10% have shown higher strain to failure without much decrease in the mechanical performance of the composites, thereby contributing to higher energy absorption.

![Toughness of PP-J, PP-J-V10, PP-J-V10-M composites](image)

**Figure 4.7:** Toughness of PP-J, PP-J-V10, PP-J-V10-M composites

Novak and DeCrescente, stated in their report that the ability of the fibers within the composite to absorb large amounts of strain energy is a principal factor governing the amount of impact energy composite material can absorb (Novak and DeCrescente 1971). Present study, observes that viscose fibers have greater ability to absorb the strain energy of the composites. The energy absorption behavior of the composites is not only affected by the properties of fiber and matrix but also with several parameters such as interfacial strength, fiber length after the extrusion and the failure mechanism of the composite such as matrix failure, fiber debonding and fiber pullout. The effect of the compatibilizer on the composite was also examined and shown in Figure 4.7. The addition of 2 wt% of MAPP not only improved ductility, toughness but also the overall
mechanical performance of the composites. It is believed due to the better interface, which allowed greater debonding and pullout of the fibers.

4.5.5 Interfacial strength through single fiber pullout test

The interfacial shear strength of the composites was determined from fiber pullout test. The load-extension characteristics of jute viscose fiber bundle are shown in Figure 4.8 (a) and (b). Single fiber reinforced PP composites were prepared by placing single filament of fiber between two films of PP, 5 mm of fiber is embedded with matrix. Figure 4.8 (a), shows curves of IFSS and work of adhesion between single jute fiber in PP and PP/MAPP matrix. The adhesion bond strength between the fiber and the matrix was characterized by the values of the apparent interfacial shear strength ($\tau_{\text{app}}$) (Doan et al., 2006). The interfacial shear strength between the jute fiber and the PP matrix was measured to 9.5 MPa. It is seen that the jute fibers debonded more easily from matrix and pulled out from the PP matrix. The SEM micrograph study had also shown a partial adhesion of fibers with matrix; whereas the interfacial shear strength of single jute fiber embedded PP/MAPP is much higher than that of unmodified PP matrix, shown an improvement of 24%. This reveals and improved interaction between the fiber and the matrix and consecutively load transfer ability in the interface. The curve resembles a very strongly bonded interphase, i.e. the interface fails immediately after fiber extraction, as reported by (De´sarmont et al., 1991). This study also observed enhancement of load along with the modulus reveals the improved work of adhesion between the jute fiber and PP matrix, this clearly indicates the chemical interaction. Figure 4.8 (b), shows curves of IFSS between the viscose fiber bundle reinforced PP composite and MAPP modified PPV fiber bundle reinforced composites. The trend observed with viscose fiber pullout was a linear-elastic region in the first section followed by higher elongation at break. The curve clearly indicates sufficient interphase
between fiber and matrix; once the interphase has failed, the fiber were extracted in a controlled way and friction was measured until the fiber was completely pulled-out (Gonzaleza et al., 1999). The higher fineness and the specific bonding area might be the reason behind the lower IFSS of the viscose fiber. But the SEM micrographs of viscose modified composites observed to have less and fiber pullouts than the PP-J composites. So, highest izod impact strength values were also found in PP-V composite with almost 85 J/m.

**Figure 4.8:** Typical single fiber pullout test load-extension curves for jute and viscose fiber bundle reinforced PP and MAPP modified composites

### 4.5.6 Thermal analysis

The heat deflection/distortion temperature of all the composites as a function of fiber content is represented in Figure 4.9 and Table 4.5. The incorporation of jute on neat resin has shifted the HDT value from 90°C to 140°C, with the loss in the impact strength about 4%. The compatibilized PP-J composite had shown the higher HDT values due to the higher stiffness of the jute fibers. The HDT values were observed to decrease slightly with the addition of impact modifier. The tabulation clearly indicates the moduli/stiffness of the composites is reduced with
the addition of low moduli fiber. But when the concentration of impact modifier is increased to 10 wt% much reduction of HDT values was not observed.

![Figure 4.9: Heat deflection temperature of jute fiber reinforced polypropylene composites modified with viscose fiber as impact modifier and MAPP]

The incorporation of 2 wt% of MAPP on PPJ-V10 has gained the HDT of the composites are increased with the stiffness of the composites, indicates the volume fraction of reinforcement and impact modifier shows better dimensional stability of the composites. However further increase of impact modifier, shows fall in the HDT values along with impact strength. This is certainly due to the improper distribution and the dispersion of fibers and the insufficient amount of resin to properly wet the fiber are strongly decreased the properties of the composites.
4.5.7 Microscopy analysis

Scanning electron microscope images of impact fracture surface of PP-J and PP-J composites modified various concentrations of viscose fibers and MAPP are shown in Figure 4.10. The SEM images of all the composites observed having perfect fiber dispersion without local concentration. In Figure 4.10 (a), it can be clearly seen that the fracture surface are rough and more traces of jute fiber pullout holes in the matrix indicating lack of interfacial adhesion between the fibers and the polypropylene. This leads to easy fiber pullout during the impact (Mohanty et al., 2006). The Figures 4.10 (b) and (c) fracture surface image evident the traces of both fiber pullouts are quite low with addition of viscose fiber. Some fiber debonding lines are also observed on the fracture surface image, which indicates the fiber matrix adhesion is comparatively higher in case of 5 and 10wt% of viscose fiber. Johnson et al studies reported on PP/wood/Lyocell fiber reinforced hybrid composite, they found that the regenerated cellulose has less-pullouts during failure (Johnson et al., 2008). Kim et al study also reported a better adhesion between the PP matrix and rayon surface was observed than PP and pineapple fibers (Kim et al., 2008). The study of Adekunle et al observed that, good fiber–matrix adhesion, as it was very difficult to see the fiber pull-outs with Lyocell reinforced composites (Adekunle et al., 2011). They believed that the fibers were well-embedded in the matrix due to their microstructure. The Figure 4.10 (d) seems to have higher fiber loading and observed to have insufficient space for pullout paths. This reveals poor fiber wetting of the fiber occurs due to insufficient matrix material and fiber travelling gap, resulting in lowering impact strength. Figure 4.10 (e), fracture surface observed to have excellent fiber matrix adhesion. In image both the fibers jute and impact modifier are visible, they are covered with the polymeric matrix demonstrating the effectiveness of the coupling agent. The micrograph also indicates no voids around the fibers.
surface and found to have clear fiber debonding and pullout paths of both natural fiber and the viscose fiber. As reported by (Oksman et al., 2009) the improvement in almost all mechanical properties with pronounced deformation in presence of MAPP is evident, for the good interfacial adhesion between the hydrophilic fibers and hygroscopic matrix.

![Image of SEM micrographs](image)

**Figure 4.10:** SEM micrograph of fractured specimen shows jute fiber–polypropylene composites with viscose fibers as impact modifier and MAPP has compatibilizer

### 4.5.8 Fiber content and length measurements after extrusion

The fibers were extracted from the composites by dissolving PP in hot xylene using Soxhlet extraction apparatus. The fiber images are taken with M/s. OPUS vision measuring machine with a 0.75 X – 4.5 X observing magnification. A sufficient number of extracted fibers are
characterized; the length and the area of the fibers are calculated automatically from the images through software. At most care has been taken to avoid the duplicates.

Figure 4.11: Fiber length measurements (a) jute fibers, (b) impact modifier- viscose fibers

Figure 4.11 (a) and (b) shows the fiber length measurements of jute and viscose fibers respectively. The typical average fiber length values of jute and viscose fiber lengths are 1-1.2 mm and 6.8-7.3 mm respectively. The mechanical performance of the composites and the microscopic analysis of fiber length clearly demonstrate, that the viscose fiber acts as an impact modifier by bridging the cracks in the matrix and provides resistances to crack propagation and crack opening. Thereby fracture toughness of the composites is enhanced by larger deformation before complete pullout of fibers. Ganster et al reported the fiber length distribution determines the mechanical performance of the composite; their study observed unnotched charpy strength at room temperature were roughly doubled and notched impact strength increased almost five times from rayon reinforced PA 6.10 composites (Ganster et al 2008). They believed the drastic improvement energy absorption of the composite was due to their higher average fiber length in the final composites.
4.6 Conclusion

This study confirms that the impact strength of PP-J composites can be increased with viscose fibers as impact modifier. An increased viscose fiber concentration did slightly decrease the composites strength and stiffness as expected. The maximum improvement in the impact strength, toughness and elongation to break was found with 10 wt% addition of viscose fibers. This is an effect of the low strength and modulus with high elongation of the used viscose fibers. The addition of a MAPP had negative effect on impact strength and elongation at break and positive effect on tensile and flexural properties for PP-J and PP-V. But it was found that the addition of MAPP had a positive effect on all mechanical performance including impact strength when it was used in the hybrid composite of PP-J-V. This is believed due to sufficient and flexible interface achieved with the composite, which dissipates more energy through stress transfer from matrix to fiber, allowing the fiber debonding with higher energy absorption with longer pullout lengths.

The IFSS of MAPP modified single jute and viscose fiber composites have improved to 11.6 and 3.0 MPa from 9.6 and 1.9 MPa respectively, due the better interaction between the fiber and the matrix.

The electron microscopy images of fracture surfaces of the composites showed that PP-J composites had poor fiber matrix adhesion compared to those of impact modified composites. This is due to the ingredients that are presents in the fibers. The viscose fibers are chemically regenerated cellulose connected by hydrogen bonding, which can be wetted out with PP molecules, whereas the volatiles present in the jute fibers such as lignin and other extractible may restrict the adhesion with the PP.
The fiber length measurements after the extrusion process showed that the length of jute fibers were reduced to an average length of 1.0-1.2 mm while the viscose fiber length was around 6.8-7.3 mm. The higher fiber length after the extrusion indicates less sensitive fibers for the shear forces and higher toughness. The addition of viscose fibers influenced the impact strength due to the increased energy dissipation along the length of fiber. Thus, revealing the long fiber carry significantly higher fraction of load compared to short fibers.
5 Impact toughness, viscoelastic behavior and morphology of polypropylene-jute-viscose hybrid composites

5.1 Introduction

LFT composites have been gaining broad attention, particularly in automotive applications (Leonard et al., 2009). Oksman et al developed the high performance natural fiber reinforced thermoplastic composites using LFT extrusion technique (Oksman et al., 2009). Ganster et al have also adopted pultrusion technique using a conventional co-rotating twin-screw extruder for the preparation of thermoplastic composites (Ganster et al., 2008). The development of high-performance composite materials has mainly focused on achieving high modulus and strength, but from the automotive application perspective, the higher strength is not sufficient; the material’s ability to absorb energy and resist impact loading should also be the important criterion (Broutman et al., 1972). Oksman et al study reported that, the long jute fiber reinforced PP composites offered higher stiffness compared to sisal, banana and flax reinforced PP composites (Oksman et al., 2009). Sarkhel and his co-workers 2008 also reported that jute fiber composites possess high stiffness and strength to weight ratios but have very low energy absorption capability (Sarkhel and his co-workers 2008).

Extensive research concerning the energy absorption of natural fiber composites are being carried out by many researchers. Oksman and Clemons, investigated the effects of elastomer-compatibilizer combination on the morphology and mechanical properties of PP wood flour (WF) composites (Oksman and Clemons, 1998). They found that the combination of elastomeric impact modifier and MAPP as compatibilizer resulted in increased toughness. In this thesis Chapter 4 showed that addition of 10 wt% of viscose fibers with 2 wt% of MAPP in PP-J30
composites achieved optimized mechanical properties including toughness. The improvement of composites energy absorption or toughness have been achieved using matrix modification (Park et al., 1996), addition of impact modifier (Hristov et al., 2005), hybridizing with tougher fibers (Mallick et al., 1977), optimizing the interface between the fibers and matrix using coupling agent (Teh et al., 2005; Sain et al., 2005; John et al., 2008), optimizing the fiber content (Kim et al., 2008) and controlling the fiber length and fiber orientation (Baillif et al., 2009).

Regenerated cellulose is a cellulose derivative and is usually based on wood (Adekunle et al., 2011). Regenerated cellulose fibers are suitable for structural composites, due to their quality and performance. Table 5.1 demonstrates the mechanical properties of different regenerated cellulose fibers (Ganster et al., 2013; Ataollahi et al., 2011; Carrillo et al., 2010; Ozcelik Kayseri et al., 2010; Adusumali et al., 2006). It is seen that viscose fibers have lower strength and stiffness but higher elongation to break compared, for example, with Lyocell fibers. Therefore, it is expected that hybridization of PP-jute composites with higher extendable viscose fibers can increase the composites’ strain to failure and enhance the energy absorption of the composites.

The energy absorption or the toughness of the composites has been evaluated using various techniques such as un-notched and notched Izod and Charpy as well as instrumental falling weight (IFW) impact tests. Only a few studies about IFW testing of natural fiber reinforced composites can be found (Oksman et al., 1997). It has been reported that the jute fiber reinforced thermoplastics have inadequate impact strength (Santulli et al., 2000). The evaluation of falling weight impact properties is usually based on the hysteresis cycle of load vs. deflection. For a fiber reinforced plastic material, it is likely that the impact behavior is time dependent i.e., dependent on the velocity of the hammer when striking the specimen (Broutman et al., 1972). As
the results of tests are shown as 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 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 (Dhakal et al., 2007; Panthapulakkal et al., 2007). The high value of elongation at failure is expected to lead to improved ductile behavior, resulting in a good value of specific energy absorption compared with the unmodified jute reinforced polypropylene composites.

**Table 5.1:** Physical and mechanical properties of regenerated cellulose fibers

<table>
<thead>
<tr>
<th>Fibers</th>
<th>Density (g/cm³)</th>
<th>Tensile strength (MPa)</th>
<th>Young’s Modulus (GPa)</th>
<th>Elongation at break (%)</th>
<th>References</th>
</tr>
</thead>
<tbody>
<tr>
<td>Cordenka</td>
<td>1.8</td>
<td>770-890</td>
<td>19.0-21.0</td>
<td>11.0-15.0</td>
<td>Ganster et al., 2013</td>
</tr>
<tr>
<td>Lyocell</td>
<td>1.3</td>
<td>790-1400</td>
<td>30.5-36.0</td>
<td>6.0-10.3</td>
<td>Carrillo et al., 2010</td>
</tr>
<tr>
<td>Modal</td>
<td>1.3</td>
<td>368-506</td>
<td>11.0-15.4</td>
<td>8.6-12.2</td>
<td>Adusumali et al., 2006</td>
</tr>
<tr>
<td>Viscosea</td>
<td>1.3</td>
<td>293-323</td>
<td>7.0-15.0</td>
<td>23.0-25.0</td>
<td>Ozcelik Kayseri et al, 2010</td>
</tr>
</tbody>
</table>

a For comparison of viscose fiber properties with other regenerated cellulose fibers.

In the present work an attempt has been made to investigate the amount of energy required to break jute fiber thermoplastic composites and how this is affected by a modification with impact modifier (viscose fibers) and MAPP-compatibilizer. The modified and unmodified jute-PP composites’ viscoelastic properties were also characterized using dynamic mechanical analysis (DMA). In addition the post-impact damage and failure mechanism of fractured specimens were assessed and the fiber dispersion of the jute and viscose fibers in the PP matrix were studied using scanning electron microscopy (SEM).
5.2 Experimental

5.2.1 Materials

Matrix. The polypropylene (PP) used in this study is homopolymer Propel 1350YG, extrusion grade from Indian Oil Corporation Ltd., Chennai, India with an MFI of 35 g/10 min (230ºC, 2.16 kg).

Reinforcement. The jute fibers used in this study were in the form of continuous roving which were connected together manually according to our earlier study. The fibers were procured from M/s. Chandra Prakash & Co. Pvt. Ltd. (Jaipur, India). The diameters of the single fibers range from 20-25 µm and the density of the jute fibers were 1.45g/cm³.

Impact Modifier. The viscose fibers were procured from M/s. Cheran Spinning Mills (Erode, India). The viscose fibers used as impact modifier were also used in the continuous roving form. The appearance of the fiber was soft and silky. The fiber length and density are 38 mm and 1.3 g/cm³, respectively.

Coupling agent. A polypropylene grafted maleic anhydride, Epolene E-43, M/s. Sigma Aldrich, USA, with an average molecular weight (Mw) ~ 9,100, was used as coupling agent.

5.2.2 Processing of composite material

LFT composites manufactured using a high-performance co-rotating twin-screw extruder (ZE-25 model Berstorff Maschinenban GmbH, D-3000 (Hannover, Germany) reported elsewhere (Ranganathan et al., 2015). The continuous fiber roving was incorporated into a side feeder of the extruder, which feeds it directly into the polymer melt. The composites were prepared by varying the impact modifier concentration according to our earlier study and, the jute fiber content was kept constant at 30 wt% (Ranganathan et al., 2015). Material formulations and denotations used for comparison are shown in Table 5.2. The extrudates were compression
molded to rectangular sheets 250 x 125 mm and thickness of 3 mm using a conventional compression mold from M/s. 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 5.2: Material formulations and denotations

<table>
<thead>
<tr>
<th>Materials coding</th>
<th>PP (wt.%</th>
<th>Jute fiber (wt.%</th>
<th>Viscose fiber (wt.%</th>
<th>MAPP (wt.%</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-J30</td>
<td>70</td>
<td>30</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>PP-J30-M2</td>
<td>68</td>
<td>30</td>
<td>0</td>
<td>2</td>
</tr>
<tr>
<td>PP-J30-V10</td>
<td>60</td>
<td>30</td>
<td>10</td>
<td>0</td>
</tr>
<tr>
<td>PP-J30-V10-M2</td>
<td>58</td>
<td>30</td>
<td>10</td>
<td>2</td>
</tr>
</tbody>
</table>

5.2.3 Testing and characterization

The Charpy impact test was performed as per ASTM D 256 (notched) using 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 geometry of 75 x 10 x 3 mm with a notch depth of 2.54 mm and notch angle of 45°. The impact energies were measured for at least 10 test samples of every composition and average values are reported.

The low-velocity falling weight impact test was performed according to ASTM D 7136 on CEAST 9350 Fractovis plus, IFW apparatus (Pianezza, Italy). The instrument is equipped with a hemispherical impactor of 20 mm diameter fitted at a height of 458.9 mm and the total impact mass is 3.6 kg with a velocity of 3 m/s which gives the incident energy of 16.3 J. Test specimens measuring 60 x 60 x 3 mm were attached to the supporting ring. The impact striker fell to the
specimen, producing 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 using Ceast software. At least five specimens were tested on each category and the average values were reported.

Dynamic mechanical analysis was performed using TA Instruments DMA Q800, (Delaware, USA). The measurement was conducted in three-point bending mode at a frequency of 1 Hz under nitrogen atmosphere. The dynamic storage modulus, loss modulus and loss factor of the composites were determined as a function of temperature ranging from -25°C to 100°C.

The morphology of the composites was studied using scanning electron microscopy (SEM). The fractured cross-sections of the IFW specimens were sputter coated with gold and studied in a SEM at an acceleration voltage of 20 kV. The fiber dispersion was studied of microtomed cross section of the composite samples.

Dynamic mechanical analysis was performed using TA Instruments DMA Q800, (Delaware, USA). The measurement was conducted in three-point bending mode at a frequency of 1 Hz under nitrogen atmosphere. The dynamic storage modulus, loss modulus and loss factor of the composites were determined as a function of temperature ranging from -25°C to 100°C.

The thermal stability of the composites was measured using thermogravimetric analysis, TGA; Perkin Elmer, Pyris, (USA). The samples were heated from 0°C to 600°C at the rate of 20°C/min under nitrogen flow (50mL/min).
FTIR spectra of PP/jute composites modified using impact modifier and MAPP were recorded using Agilent Cary 630 FTIR Spectrometer, (USA). The spectrometer was used in the transmission mode with a resolution of 4 cm\(^{-1}\) in the range 500 to 4000 cm\(^{-1}\).

### 5.3 Results and discussion

#### 5.3.1 Impact toughness

Notched Charpy impact and IFW properties of PP-J30 and modified composites are seen in Table 5.3. The impact strength was improved with the addition of viscose fibers to the PP-J30 composite. The improvement in the impact strength may be due to several factors: properties of impact modifier, matrix, fiber length, adhesion between the matrix and fiber, etc. The results shown in Table 5.3 show that the addition of 10 wt% of impact modifier to PP-J30 has improved the impact strength from 3.2 to 7.5 kJ/m\(^2\), which is 134%, indicating ability of the viscose fibers to dissipate the energy along the length of fiber. The major mechanism to increase the energy absorption of the composites, accounted on this study is the pullout behaviour of fibers. We showed in our earlier investigation that debonding and fiber pullout, resulted in greater energy absorption capability on the jute fiber reinforced impact modified composite compare to the unmodified one. The addition of MAPP on the impact modified composites formed a weak (flexible) interface, which has increased the fibers pullout length and thus showing higher energy dissipation.

A typical load-time and force-deformation curves from the IFW impact test are presented in Figure 5.1 and the initial peak, maximum load and the time are presented in Table 5.3. The initial peak value of impact force represents the starting point of the damage, maximum load reveals the higher load bearing capability and contact duration is the time taken for damage initiation and
propagation. The peak load values for PP-J30 and PP-J30-M2 composites are 1175 and 1407 N, respectively. The PP-J30 composite exhibits lower peak load compared PP-J30-M2, indicating stress transfer from matrix to 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 5.1(a). This indicates the damage propagation extended beyond the peak load, which attributes to absorb energy until complete perforation of the specimen. This was also clearly noted from the Figure 5.3, that the impact modified composites did not have complete perforation on the back surface, whereas the unmodified composites have complete damage.

It is also noted that the contact duration of PP-J30-V10 composite was 4.2 ms higher compared to PP-J30 composites; the time elapsed for damage initiation to penetration on impact modified composite indicated the increase of energy absorption. The addition of MAPP on impact modified composites further increased the contact duration to 14.3ms indicates the difficulty in the complete perforation of composites shown in Figure 5.1(b).

The amount of energy absorbed is equal to the area of the impact hysteresis cycle force vs. deflection (Ghelli et al., 2012). When a material is subjected to any kind of impact loading, it absorbs energy by deflection/deformation. The deflection is usually accompanied by damage initiation and/or propagation in the form of fiber breakage, matrix cracking and fiber/matrix debonding. It can be noted from Figure 5.1(c) that the energy required to initiate the crack in the impact modified composites is lower than the unmodified PP-J30 composites. The unmodified composite exhibits brittle fracture with little propagation energy, the samples have fragmented after being subjected to the incident energy. Figure 5.1(c) clearly shows a sudden drop in the
force without much deformation. Thus, the total energy absorption of the composites was reduced due to their failure behavior. However, changes in the damage mechanism of the composites were observed with the introduction of viscose fiber to the jute fiber reinforced thermoplastic composites. A drastic increase in the deflection with a slight decline in damage initiation load was observed with the impact modified composites. This clearly indicates that specimens modified with impact modifier require higher penetrative energy, thereby enhancing total energy absorption of the composite.

![Graphs showing load vs time and force vs deformation curves of PP-J30 composites modified with viscose fibers and MAPP](image)

**Figure 5.1:** Typical load vs time and force vs deformation curves of PP-J30 composites modified with viscose fibers and MAPP
The improved damage mechanism with higher plastic deformation may be explained by the higher elongation behavior of viscose fibers. Besides the two different composites, shown in Figure 5.1(d) PP-J30-V10 modified with MAPP exhibited the higher energy absorption, this reveals that MAPP compatibilizer together with viscose fiber has formed the weak (flexible) interfacial adhesion.

Figure 5.2, shows the variation in energy absorption and velocity with respect to time of composites modified with viscose fibers and MAPP. It can be seen that there is an increase in the total energy absorption when viscose fibers are present. The energy used for the impact test was 16.2 J, whereas PP-J30-V10 samples absorb total energy of 12.9 J, as shown in Figure 5.2(a).

When the composites are modified with MAPP a noticeable change in energy absorption compared to unmodified composites is seen shown in Figure 5.2(b). The chapter 4 of this thesis has also shown that the mechanical properties and the impact strength increase in all modified composites.

**Table 5.3:** Charpy and IFW impact results of PP-J30 composites modified with viscose fibers and MAPP (values in the bracket are standard deviation)

<table>
<thead>
<tr>
<th>Material codes</th>
<th>Charpy impact strength (kJ/m²)</th>
<th>Charpy Impact energy (J)</th>
<th>Load (N)</th>
<th>Deformation (mm)</th>
<th>Time (ms)</th>
<th>Velocity (m/s)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-J30</td>
<td>3.2 (0.2)</td>
<td>8.0 (0.7)</td>
<td>1175.6</td>
<td>18.6</td>
<td>9.7</td>
<td>1.7</td>
</tr>
<tr>
<td>PP-J30-V10</td>
<td>7.5 (0.2)</td>
<td>12.9 (1.1)</td>
<td>1065.8</td>
<td>23.4</td>
<td>13.4</td>
<td>0.9</td>
</tr>
<tr>
<td>PP-J30-M2</td>
<td>3.1 (0.1)</td>
<td>6.8 (1.4)</td>
<td>1407.2</td>
<td>17.5</td>
<td>10.1</td>
<td>1.8</td>
</tr>
<tr>
<td>PP-J30-V10-M2</td>
<td>7.8 (0.2)</td>
<td>15.4 (0.6)</td>
<td>1187.0</td>
<td>24.8</td>
<td>14.3</td>
<td>0.1</td>
</tr>
</tbody>
</table>
In general, the area under the velocity-time curve represents the displacement. The impact velocity affects the energy dissipation of the composites during impact (Padaki et al., 2008). The full penetration of impactor on PP-J30 samples was observed depicted in Figure 5.3, and the residual velocity was 1.7 m/s. The possible reason may be more fiber breakage or the poor adhesion between the matrix and fiber.

**Figure 5.2:** Typical energy absorption and velocity vs time curves of PP-J30 composites modified with viscose fibers and MAPP

Figure 5.2(c) shows that the residual velocity of PP-J30-V10 was found to be 0.9 m/s; this shows that the addition of impact modifier content has driven the residual velocity to minimum value. Dhakal et al noticed the zero residual velocity for 4 and 5 layered non-woven hemp fiber reinforced polyester composites (Dhakal et al., 2007). They believed the effect was due to the higher impact energy dissipation.
Figure 5.3: Fragmentation characteristics of IFW test on PP-J30 composites modified with viscose fibers and MAPP
5.3.2 Microstructure

Figure 5.4 shows the influence of impact modifier and coupling agent on the microstructure of PP-J30 composites. Figure 5.4(a) shows more fiber pullouts leaving void in the matrix phase, due to worse bonding between the matrix and jute fibers (Mohanty et al., 2006; Johnson et al., 2008). This behavior led to composites with less plastic deformation after reaching the maximum load. The addition of impact modifier to the PPJ-30 composite’s microstructure Figure 5.4(b) shows less fiber pullouts compared to PP-J30; the fiber pullout paths are recognized, which proves that the viscose fiber has induced the plastic deformation after the maximum load, thereby contributing to the energy absorption of the composites by promoting debonding and pullout of fiber from matrix. Johnson et al reported about PP/wood/Lyocell fiber hybrid composite, they found that the Lyocell fiber resulted less pullouts during failure and favored higher elongation at break (Johnson et al., 2008). Similarly Kim et al, also noticed better fiber–matrix adhesion with rayon reinforced composites and they found improved toughness when compared to pineapple wood fiber reinforced thermoplastic composites (Kim et al., 2008).

The addition of the MAPP resulted in improved interfacial adhesion and the composite exhibited fiber breakage before the complete pullout of the fibers as shown in Figure 5.4(c). The effect was also observed in the load-deformation curve i.e., the higher load with less deformation was observed, which indicates that the strong interaction between the fiber and matrix led to the brittle failure. These findings are in agreement with results reported by (Goriparthi et al, 2012). They found a decrease in the impact strength along with energy absorption of compatibilized PLA/Jute composites due to the fiber breakage before debonding. However, the failure mechanism has been changed on the compatibilized impact modified PP-J30 composite demonstrated in Figure 5.4(d). This composite has recorded the maximum energy absorption this
is attributed to the formation of weak (flexible) interfacial adhesion between the fiber and matrix. This proves that jute fiber in the composites enhances the interaction with the matrix whereas the viscose fiber does not show much effect on interfacial adhesion with MAPP.

**Figure 5.4:** SEM images of fractured specimen from IFW testing of the composites modified with viscose fibers and MAPP

### 5.3.3 Viscoelastic properties

The storage modulus as a function of temperature for all the composites is represented in Figure 5.5 (a) and (b) and tabulated in Table 4. As reported by (Doan et al., 2006), a general trend of improvement in the thermo-mechanical properties with the addition of jute fibers in to the PP
matrix was observed, indicating improved stiffness of the composites. It is evident that the addition of a small amount of impact modifier causes a gradual softening of the PP-J30 composite. A possible explanation is that incorporation of viscose fibers has induced flexibility for the composite, due to the lower modulus values. This behavior was also confirmed by the static mechanical test. Adekunle et al noted a similar finding in flax fiber reinforced biobased resin using Lyocell as an impact modifier (Adekunle et al., 2011). The Lyocell hybridization was reported to have a slight reduction in the storage modulus compared to bio-based resin/ flax composite; the authors believed the effect is due to the Lyocell hybridization. Oksman and Clemons et al.\textsuperscript{22}, study showed that the addition of 10\% SEBS-g-MAH reduced the storage modulus; they believed that the flexible SEBS is the reason why the storage modulus is lowered; however, their results revealed improvement of the toughness of the composites (Oksman and Clemons et al., 1997). PP-J30-V10-M2 sample has shown higher storage modulus, and the same composite offered higher energy absorption during impact loading shown in Figure 5.5(b). The possible reason may be that, the jute fiber in the composites enhances its interaction with the matrix whereas the viscose fiber substantially has lesser effect with MAPP.
Figure 5.5: Storage modulus, loss modulus and tan delta as a function of temperature of PP-J30 composites modified with viscose fibers and MAPP

The loss modulus of composites modified with impact modifier and MAPP was compared with PP-J30, as shown in Figure 5.5(c) and (d), their values shown in Table 5.4. The PP-J30-V10-M2
composite shows higher value of loss of modulus compared to PP-J30 and impact modified composites. The higher loss peak clearly indicates the improved impact energy absorption of the composite (Gupta et al., 2009). The magnitude of loss modulus peak variation results in a severe decline in the storage modulus (Pothan et al., 2009). The broadening of the loss modulus peak was observed with the impact modified composites when compared to PP-J30. This indicates the increase in the energy absorption caused by the viscose fibers (Choudhury et al., 2007). The hybridization of viscose fiber with PP/jute composite has been found to affect the properties of the system. The incorporation of long viscose roving in the PP-J30 composite has lowered the loss modulus; this may be considerable, due to the property mismatch of the fibers. The addition of MAPP to the impact modified composite has improved the loss modulus corresponding to the viscous dissipation (Saha et al., 1999). The improvement in the modulus is due to reduced flexibility of the composite; these constraints on the segmental mobility were believed due to the enhanced interaction of jute fiber with matrix in presence of compatibilizer.

**Table 5.4:** Dynamic mechanical properties of PP-J30 composites modified with viscose fibers and MAPP

<table>
<thead>
<tr>
<th>Composition</th>
<th>Storage modulus (MPa)</th>
<th>Tan δ-peak (°C)</th>
<th>Loss modulus (MPa)</th>
<th>Tan δ peak Height</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-J30</td>
<td>1354</td>
<td>-3</td>
<td>88</td>
<td>0.065</td>
</tr>
<tr>
<td>PP-J30-V10</td>
<td>805</td>
<td>-1</td>
<td>53</td>
<td>0.066</td>
</tr>
<tr>
<td>PP-J30-M2</td>
<td>3606</td>
<td>-7</td>
<td>150</td>
<td>0.041</td>
</tr>
<tr>
<td>PP-J30-V10-M2</td>
<td>3133</td>
<td>-3</td>
<td>173</td>
<td>0.055</td>
</tr>
</tbody>
</table>

Figure 5.5(e) and (f) and Table 5.4 compare the tan δ value of the composites as a function of temperature. It is 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 indicates that once the deformation is induced in a material; the material will not recover its original shape (Gupta et al., 2009). This study shows that the peak amplitude has increased with the addition of impact modifier, indicating a greater degree of molecular mobility but the position of the peak has shifted to 3°C upward indication some kind of interaction between the viscose fiber and matrix. From the Figure 5.5(e), it is evident that the addition of MAPP to PP-J30 composites shifted the tan δ position about 4°C upward, which indicates enhanced interaction of the fibers with the matrix due to the addition of MAPP. (Oksman et al., 1997) noted similar findings; when wood flour and MAPP were used with the PP/EPDM system the tan δ peak shifted 4°C upward. But the addition of MAPP to the impact modified composite resulted with peak broadening along with the decrease in tan δ peak from 12°C to 11°C. This might be due to the formation of weak (flexible) interfacial adhesion between the fiber and matrix.

5.3.4 Interfacial properties

FTIR spectra of PP-jute composite modified with viscose fiber and MAPP is shown in Figure 5.6. The spectra of PP-J30 composite show a weak (flexible) intense peak in the range of 3200-3600 cm\(^{-1}\) attributes to the O-H stretching groups of jute fiber, CH\(_2\) symmetric and asymmetric peak appears at 2958 cm\(^{-1}\) and 2870 cm\(^{-1}\) due to alkyl chain of PP. The peak at 1455 cm\(^{-1}\) is attributed to the stretching of C–H bonds in the cellulosic structure. The peak at 1382 cm\(^{-1}\) is attributed to the O–H bending vibration and the strong band at 1040 cm\(^{-1}\) is attributed to the characteristic C–O–C stretching. In case of PP-J30-M2 composites, the intensity of the peak at 3200-3600 cm\(^{-1}\) corresponding to O–H frequencies appeared to be high when compared to PP-J30 composite. This is due to the hydrogen bond interaction between the ester group of the maleic anhydride and the hydroxyl group of the cellulose fiber, which enhances the interactions
with the matrix. The impact modified composites showed much broader peak, indicates the formation of interaction between the sulphite group of regenerate cellulose (viscose fiber) and O−H group of cellulose (Jute) fiber. The compatibilized impact modified jute reinforced composites shows reduced O−H intensity when compared to the PP-J30-V10 composites due to formation of weak hydrogen bonding, which deteriorates the interactions with the matrix (i.e. the viscose fiber substantially has lesser effect with MAPP).

Figure 5.6: FTIR spectra of PP-J30 composites modified with viscose fibers and MAPP

5.3.5 Thermal properties

The thermo-gravimetric curves of PP-J30, PP-J30-M2, PP-J30-V10 and PP-J30-V10-M2 are shown in Figure 5.7. The PP/Jute composites prepared at 30wt% fiber loading shows the initial
peak between 339°C to 386°C corresponds to the thermal degradation of cellulose. The second decomposition occurred between 378 °C to 414 °C, which is primarily attributed to aromatization and involving dehydration reaction (Mohanty et al., 2006).

![TGA curve](image)

**Figure 5.7:** TGA of PP-J30 composites modified with viscose fibers and MAPP

As seen from Figure 5.7 the impact modified composites exhibited slightly lower thermal stability than the PP-J30 composites. This may be attributed to the lower thermal stability of the viscose fiber. The modification of PP-J30 composites using 2wt% of MAPP has influenced on the thermal stability of the composites. This effect might be due to the stronger interaction between the fiber and matrix caused by the formation of the covalent bond at the interface (Doan et al., 2006). The addition of MAPP on impact modified composites showed improvement in the thermal stability of the composites, but found slightly lower than the PP-J30-M2 composites, due to presence of the weak (flexible) interfacial adhesion between the fiber and the matrix.
5.3.6 Fiber dispersion

Figure 5.8 shows SEM images of PP-J30 composites modified with viscose fiber and compatibilized with MAPP. The cross section of pure jute and hybrid composites are in respect to the fiber dispersion and orientation. In general, a good dispersion and distribution of fibers is essential to achieve high mechanical properties of composites (Oksman et al., 2009). The Figure 5.8(a) shows the jute fibers are well dispersed in the polymeric matrix without any aggregation. It is obvious that fibers with stiff structure and non-tangling behavior show better homogeneity in fiber distribution along the sample sections (El Sabbagh et al., 2013). Further, the jute fibers tend to oriented predominantly parallel to the flow direction. Figure 5.8(b) shows non-homogenous dispersion on impact modified composites, the viscose fibers are seen in bundles. The possible reason for the poor dispersion of viscose fiber in the composites could be the softness and toughness nature, since soft and tough fiber bundles are more difficult to separate during the processing. The effect was well agreed with Oksman et al study report, their result showed that the toughest fiber retained the fiber bundles in the composite (Oksman et al., 2009). Further several studies have demonstrated that it is difficult to disperse properly Cellulose fibers in polyolefin polymers because of strong hydrogen bonding between the fibers (Baillif et al., 2009; Dunnom et al., 1973; Klason et al., 1984; Saheb et al., 1999). Figure 5.8(b) also demonstrates that the viscose fiber bundles are aligned mostly in the same direction as that of the jute fibers. It is well known that the energy absorption is the function of plane orientation (Broutman et al., 1972). Thus the influence in the energy absorption with the incorporation of viscose fiber clearly indicates that the fiber bundles are oriented in the direction of the extrusion.
Figure 5.8: SEM micrographs on the cross-section of PP-J30 composites modified with viscose fibers and MAPP

However, the agglomeration of fiber bundles can be reduced with the addition of surface modifiers. Raj et al.\textsuperscript{42}, study detailed the importance of using surface modifiers to improve fiber dispersion in cellulose fiber/PP composites (Raj et al., 1989). The addition of MAPP on the impact modified and unmodified composite showed a slight improvement in the quality of dispersion in the polymeric matrix, evidenced from the Figure 5.8(c) & 6(d). Therefore the observation of this study predicts that, the further improvement in fiber dispersion without agglomeration in the composite may lead to achieve better impact properties along with other mechanical properties.
5.4 Conclusions

The present study is about the impact toughness, energy absorption, viscoelastic properties and fiber dispersion of modified long jute fiber reinforced thermoplastic composites. The prepared composites were studied using conventional Charpy impact and IFW impact test methods. The results showed that addition of viscose fibers improved the propagation energies, due to the higher energy dissipation. The Charpy and IFW tests showed 134% and 61% of improvement in the impact strength and energy absorption, respectively.

The microscopy analysis showed fiber pullout paths in impact modified composites which were not observed in the PP-J30 composites. This indicated that the toughness of the composites was influenced by viscose fiber, 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 fiber retained the fiber bundles in the composites.

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

Further, 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, thereby reduced the energy absorption of the composites.

The fractography micrographs of surface modified composites indicated the clear debonding and pullout of fiber due to the weak (flexible) interfacial adhesion developed between the fiber and
matrix. The SEM images have also shown slight improvement in the quality of dispersion on both the PP-J30-M2 and PP-J30-V10-M2 composite.

Similarly, the shift of the tan δ position about 4°C upward indicated the enhanced interaction of fibers with the matrix. But compatibilized impact modified composite showed enhanced toughness with the formation of weak (flexible) interfacial adhesion; in case of dynamic mechanical analysis the broadening peak along with the decrease in the peak position to 1°C was observed when compared to PP-J30-M2 composites. The FTIR spectra confirmed that the weak interfacial adhesion of PP-J30-V10-M2 composites whereas the TGA thermograms displayed a lower thermal stability when compared to PP-J30-M2 composites.

This study concludes that, improvement in impact performance was achieved with the incorporation of viscose fiber and by improving the efficiency of viscose fiber dispersion may further improve the impact properties along with other mechanical properties.
6 Structure property relation of hybrid biocomposites based on jute, viscose and polypropylene: Effect of fiber content and length on fracture toughness and fatigue properties

6.1 Introduction

In addition to improvement of the strength and stiffness, the use of natural fibre-reinforced thermoplastic composites can also increase the toughness. The measure of resistance to unstable crack growth is termed as fracture toughness ($K_{IC}$) (Dieter 1998). $K_{IC}$ represents the critical stress intensity factor, calculated on the basis of the maximum load; the fracture toughness tends to depend on the sample size. Generally, the composites should have high strength together with adequate toughness to resist rapid crack propagation (Nath et al., 2006). The crack propagation resistance or fracture toughness of fibre-reinforced composites depends on various factors, such as the toughness of the matrix, additives, the properties of the fibres, the dispersion and distribution of fibres and the orientation and length of the fibres after the manufacturing process. Further, the fracture toughness is greatly affected by applied stress, mode of fracture, matrix cracks, fibre debonding, fibre breakage, fibre frictional pullout and fibre bridging (Kim et al., 1992; Petersen et al., 2007). The incorporation of rigid reinforcements in the matrix reduces the ductility and impact toughness, which leads to differences in the crack propagation and the fracture behaviour (Arencon et al., 2009). The propagation of cracks depends on the debonding of the fibres at the fibre-matrix interface, fibre pullout and fibre and matrix failure (Meredith et al., 2012; Zarei et al., 2008; Agrawal et al., 2013). In fibre-reinforced composites, the applied stress is transmitted from the matrix to the fibre across the interface; hence, the interfacial
adhesion between the fibre and the matrix plays a major role in the crack propagation (Oksman et al., 1998; Bera et al., 2010; Adusumalli et al., 2010). When the composite is subjected to tensile load, the higher amount of stress will be borne by fibres, which causes the weak fibres to fail first. Further, application of the load on the composite causes failure of the intact fibres; this fibre fracture increases the stress concentration in the matrix, thus leading to matrix cracking (Mishnaevsky et al., 2009). In the case of a weak (flexible) fibre/matrix interface, the matrix crack defects along the fibre-matrix interface tends to have crack bridging along the fracture path and pullout of the fibres. As a result, the fibres provide higher resistance to crack growth by arresting and bridging the cracks. However, in the case of a strong fibre-matrix interface, the matrix crack propagates into fibres without leaving intact fibres to produce pullout, resulting in the lack of fracture toughening (Sakai et al., 1991). The fibre pullout process requires additional energy, and it increases with mean fibre length (Xia et al., 1994). Therefore, the current study adopted the direct long fibre thermoplastic (D-LFT) compounding process to maintain the length of the fibres during processing. The length of fibres is controlled by reduced shear, which also inclines the fibres to orient along the flow direction.

The main objective of the present study was to investigate the effect of jute and viscose fibres and the extent of their breakage on the fracture toughness and fatigue properties of PP composites manufactured via the D-LFT process. The effect of fibre length on mode I type fracture toughness of the composites was experimentally evaluated. The failure mechanism with the addition of viscose fibres in PP/Jute composites was identified and discussed.
6.2 Experimental

6.2.1 Materials and compounding

Homopolymer polypropylene (PP) Propel 1350YG, extrusion grade, with a melt flow index (MFI) of 35-g/10-min (230°C, 2.16 kg) was purchased from Indian Oil Corporation Ltd., India and was used as the matrix. Maleic anhydride grafted polypropylene, Epolene E-43, Sigma Aldrich, USA was used as the coupling agent. The jute fibres were procured from Chandra Prakash & Co. Pvt. Ltd., Jaipur, India. The fibres were in the form of long fibre bundles of 1-2-m length, and the fibres were connected manually to form continuous fibre rovings. In this study, the jute fibres were used in the form of long hand-made rovings, as shown in Figure 6.1.

The viscose fibres were supplied from Cheran Spinning Mills, Erode, India. Viscose is a derivative of wood pulp (also called as regenerated cellulose) processed via a spinning method. The fibres are staples or filaments of size ranging from 36-40 mm; the filament fibres are infinite in length, as shown in Figure 6.1 (b). The density of the used viscose fibres was 1.3 g/cm³.
A co-rotating twin-screw extruder with a side feeder ZE-25 model Berstorff Maschinenbau GmbH, D-3000 (Hannover, Germany) was used to perform the Direct-Long Fibre Thermoplastics (D-LFT) process. The extruder was equipped with a gravimetric feeder, two atmospheric vents and a vacuum vent, as described in our earlier study (Ranganathan et al., 2015). Prior to compounding, the jute fibres were washed and dried at 60°C for 48 hours until a uniform weight was achieved. Similarly, the viscose fibres were dried at 60°C for at least 2 hours. Hand-made roving of viscose and jute fibres was incorporated into side feeder of the extruder, which feeds it directly into the polymer melt. The fibre weight fractions was controlled by the screw speed, and it was calculated using targeted fibre content, the weight of the fibre roving per meter, the length per screw revolution and time. The compositions used for the comparison are shown in Table 6.1.

<table>
<thead>
<tr>
<th>Materials</th>
<th>PP (wt.%)</th>
<th>Jute fiber (wt.%)</th>
<th>Viscose fiber (wt.%)</th>
<th>MAPP (wt.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-J30</td>
<td>70</td>
<td>30</td>
<td>0</td>
<td>0</td>
</tr>
<tr>
<td>PP-J30-V5</td>
<td>65</td>
<td>30</td>
<td>5</td>
<td>0</td>
</tr>
<tr>
<td>PP-J30-V10</td>
<td>60</td>
<td>30</td>
<td>10</td>
<td>0</td>
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<td>0</td>
</tr>
<tr>
<td>PP-J30-V10-M2</td>
<td>58</td>
<td>30</td>
<td>10</td>
<td>2</td>
</tr>
</tbody>
</table>

**6.3 Testing and characterization**

**6.3.1 Scanning electron microscopy**

The morphology of the jute and viscose fibres was analyzed using scanning electron microscopy (SEM) with an instrument of Zeiss EVO-MA15, Germany. An acceleration voltage of 15 kV was
used, and the sample surfaces were sputter coated with gold prior to SEM observation to avoid charging.

### 6.3.2 Fiber content and length measurements

PP matrix was removed after boiling in xylene using a Soxhlet extraction apparatus to study the actual fibre content and determine how the extrusion process affected the fibre length. After extraction, the fibres were manually separated in water and then observed using an optical microscope M/s. OPUS with a magnification in the range of 0.75X – 4.5X. The actual fibre content ($A_{fc}$) was calculated based on the targeted rate, and the fibre length was measured automatically from the images using image analysis software. At least 200 fibres were measured of both fibres to calculate the average fibre length ($A_{fl}$).

### 6.3.3 Fracture toughness and fracture energy

The plane strain fracture toughness ($K_{IC}$) of the composites was measured from the single edge notch tensile (SENT) test. The SENT tests were performed using an Instron 3382 tensile testing machine in compact tension (CT) method using a cross-head speed of 1.5 mm/min. The CT specimens were cut from the compression moulded composite sheets. The dimensions of the specimens were determined according to ASTM D5045, the crack length, $a$, was selected such that $0.45 < a/w < 0.55$. Pre-cracks were initiated using a razor blade of 0.1-mm thickness and tapping gently with wooden hammer. The holes on the CT specimens were milled using a CNC machine. The fracture toughness $K_{IC}$ were calculated using the following equation (Leonard et al., 2009; Norman et al., 2003; Paluvai et al., 2015):

$$K_{IC} = \frac{P}{B\sqrt{w}} f \left(\frac{a}{w}\right)$$  ................................................................. (15)
for a CT specimen, \( f \left( \frac{a}{w} \right) \) is the geometry factor and is given by,

\[
f \left( \frac{a}{w} \right) = \frac{2 + a/w}{(1 - a/w)^{1.5}} \left[ 0.866 + 4.64 \left( \frac{a}{w} \right) - 13.32 \left( \frac{a}{w} \right)^2 + 14.72 \left( \frac{a}{w} \right)^3 - 5.6 \left( \frac{a}{w} \right)^4 \right] \ldots \ (16)
\]

\( K_{IC} \) is the fracture toughness of the specimen, \( P \) the load at failure, \( S \) the length of the span, \( a \) is the crack length, \( B \) and \( w \) are the thickness and width of the specimen respectively. The strain energy release rate or fracture energy, \( G_{IC} \) was calculated using the following equation

\[
G_{IC} = \frac{K_{IC}^2}{E} (1 - v^2) \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldots \ldot
6.4 Results and discussion

6.4.1 Scanning electron microscopy

Figure 6.2 shows the structure of the used fibres: 6.2(a) is the longitudinal view of jute fibre bundle, and 6.2(b) is the cross section. As shown in the image, jute fibre bundles have characteristic irregular shape, straight longitudinal appearance with non-circular cross-section. The elementary fibre size is approximately 20 µm. Figures 6.2 (c) and (d) show the longitudinal and cross section images of the viscose fibre bundle. The fibre bundle appears to be finer and smoother compared with the jute bundle. The twisted staples fibre appears to be smooth in the longitudinal direction and displays striation along the length. The fibre length ranges from 36-40 mm. The diameter of the fibre is 6.5 to 16.2 µm, and the fibres are tightly packed into a twisted bundle.
Figure 6.2: Microstructure of fiber bundle (a) longitudinal section of jute fiber bundle, (b) Cross section of jute fiber bundle, (c) longitudinal section of viscose fiber bundle and (d) cross section of viscose fiber bundle

6.4.2 Fiber content and fiber length after processing

The actual fibre content present in the composites along with the variation of fibre lengths of jute and viscose fibres are reported in Table 6.2. The results showed a slightly negative deviation of the actual fibre content ($A_{fc}$) for all compositions in comparison to the target fibre content. On average, the deviation was approximately 3.6%, due to the handmade roving, which does not have an exact weight/meter.
Figure 6.3, shows the jute and viscose fibre length distributions. The graph reveals that the average fibre length differs significantly ($A_{fl}$) among the different compositions. The result shows that increased fibre weight fraction of the viscose fibres increased the average fibre length of the composites, which may be due to increase in the weight of the roving per meter causing insufficient mixing, resulting in lower shear forces of the fibres. A similar observation was reported by (Gamon et al., 2013), the increase in the feeding rate with a slight increase in the screw speed was found to have higher mean aspect ratios. They believed that the screw speed appeared to be insufficient to break the fibres or to separate the bundles.

From Table 6.2, it is also observed that the viscose fibre length is longer compared with the jute fibre; (Oksman et al., 2009) reported a detailed study that indicated the tough fibres always remain in the form of fibre bundles and retain higher fibre length, in agreement with our observations. Similar results were reported by (Chakraborty et al., 1982) indicating that breakage of tough silk fibres was less compare to jute fibre

<table>
<thead>
<tr>
<th>Coding</th>
<th>$A_{fc}$ (%)</th>
<th>$A_{fl}$ (mm)</th>
<th>Failure load (N)</th>
<th>$(K_{IC})$ Mpa.m$^{1/2}$</th>
<th>$(G_{IC})$ kJ/m$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>$A_{fl}$</td>
<td>$(G_{IC})$</td>
<td>$Jute$</td>
<td>$Viscose$</td>
<td>$Jute$</td>
</tr>
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<td>PP-J30</td>
<td>29.6</td>
<td>0.9</td>
<td>--</td>
<td>166</td>
<td>3.9(0.2)</td>
</tr>
<tr>
<td>PP-J30- V5</td>
<td>33.4</td>
<td>1.2</td>
<td>6.6</td>
<td>234</td>
<td>5.8(0.1)</td>
</tr>
<tr>
<td>PP-J30-V10</td>
<td>38.1</td>
<td>1.2</td>
<td>7.2</td>
<td>384</td>
<td>9.1(0.2)</td>
</tr>
<tr>
<td>PP-J30-V15</td>
<td>42.5</td>
<td>1.3</td>
<td>7.8</td>
<td>338</td>
<td>7.9(0.3)</td>
</tr>
<tr>
<td>PP-J30-V10-M2</td>
<td>38.6</td>
<td>1.3</td>
<td>8.1</td>
<td>435</td>
<td>10.0(0.2)</td>
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</tbody>
</table>
Figure 6.3: Number average length of jute and viscose fibers extracted from extruded composite materials.

The addition of the MAPP on the modified composites resulted in a slight increase the average fibre length of the viscose fibres from 7.2 to 8.1 mm. This result might be due to the lubricating effect induced on the fibres and the PP matrix by the maleated polypropylene wax (MAPP); this lubrication reduced the shear forces between the individual fibres as well as between the fibres and the melt polypropylene. The results observed are in accordance with the study by Karmaker et al., who also noticed the similar increase in the fibre length on the compatibilized composite (Karmaker et al., 1996). They reported that the main reason for this behaviour is the reduction in
fibre attrition in the system with a coupling agent due to the lubricating effect on the lingo-
cellulosic filled PP composite.

6.4.3 Fracture toughness (KIC)

Table 6.2 shows the critical stress intensity or the fracture toughness ($K_{IC}$) and the critical strain energy release rate or the fracture energy ($G_{IC}$) values of the CT specimens. Figure 6.5, demonstrates the fracture behaviour of the CT composites. Figure 6.4 shows the following: 1) the jute based PP-composite exhibits a linear behaviour in the first section, followed by a minor displacement with a sudden load drop and 2) the MAPP modified jute based PP-composite exhibits a sudden drop in load immediately after the maximum load; this behaviour indicates the unstable crack growth. The fracture mechanism was observed to be different for all of the modified composites. All of the composites containing viscose fibres exhibited an initial load drop (corresponding to crack initiation) followed by an increment and a decrement, indicating that the presence of long length viscose fibre prevents the composites from catastrophic fracture or rapid propagation of cracks. Hence, the composites modified using viscose fibres have improved fracture toughness, as exhibited by stable crack growth, in contrast to the unmodified composites.

The fracture properties indicate both the trends in the fracture resistance behaviour as a function of impact modifier content and the values ($K_{IC}$) obtained from eqs. (1) and (2), where $P$ is the peak load in the corresponding composite and the geometrical properties $B$, $a$ and $w$ of the specimen and the ($G_{IC}$) values obtained by converting the fracture toughness values using eq. (3). The study shows that the addition of viscose fibres improved the fracture toughness and fracture energy of the PP-jute composites. The study indicates that the addition of 5 wt% viscose fibre
had shown improvement of 48% and 144% of $K_{IC}$ and $G_{IC}$, respectively. The increased volume fraction of viscose fibres to 10 wt% further raised the $K_{IC}$ and $G_{IC}$ values from 5.8 to 9.1 MPa m$^{1/2}$ and from 11.5 to 28.9 kJ/m$^2$, respectively. This result clearly indicates that the presence of viscose fibres prevented the rapid crack propagation and acts as a barrier for crack propagation. Many experimental studies proved that the long glass fibres bridges the stress across the crack, thereby providing increased work energy of the composites by hindering the propagation of crack, thus enhancing both the fracture toughness and the fracture energy energy (Fu et al., 2000; Yang et al., 2012).

![Figure 6.4: Typical CT specimens load-displacement curves of PP composites containing 30 wt.% jute fibers modified with viscose fibers and MAPP](image)

Further, the deviation in the crack direction was also observed in all composites modified with viscose fibres, as illustrated in Figure 6.5.
Table 6.3: Average fiber content, length and fracture behaviour of PP composites containing 30 wt.% jute fibers modified with viscose fibers and MAPP (values in the bracket are standard deviation)

<table>
<thead>
<tr>
<th>Composition</th>
<th>$A_c$ (%)</th>
<th>$A_l$ (mm)</th>
<th>Failure load (N)</th>
<th>($K_{IC}$) MPa.m$^{1/2}$</th>
<th>($G_{IC}$) kJ/m$^2$</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Jute</td>
<td>Viscose</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>PP-J30</td>
<td>29.6</td>
<td>0.9</td>
<td>--</td>
<td>166</td>
<td>3.9 (0.2)</td>
</tr>
<tr>
<td>PP-J30-V5</td>
<td>33.4</td>
<td>1.2</td>
<td>6.6</td>
<td>234</td>
<td>5.8 (0.1)</td>
</tr>
<tr>
<td>PP-J30-V10</td>
<td>38.1</td>
<td>1.2</td>
<td>7.2</td>
<td>364</td>
<td>9.1 (0.2)</td>
</tr>
<tr>
<td>PP-J30-V15</td>
<td>42.5</td>
<td>1.3</td>
<td>7.8</td>
<td>338</td>
<td>7.9 (0.3)</td>
</tr>
<tr>
<td>PP-J30-V10-M2</td>
<td>38.6</td>
<td>1.3</td>
<td>8.1</td>
<td>435</td>
<td>10.0 (0.2)</td>
</tr>
</tbody>
</table>

Thereby, the composite tends to have longer crack path along the interface, resulting in higher toughness. (Laranjeira et al., 2006), reported that the crack deviation on the weak (flexible) interface plays a role in the impact strength of the composites. In addition, note that the majority of the fibres are oriented perpendicular to the crack propagation direction, thereby retarding the crack growth (Clemons et al., 1999). The present study demonstrates the composites with higher fibre length achieves higher energy dissipation, thereby confirming that fracture toughness and fracture energy property not only depends on the mechanical properties of the fibre and matrix and the interface between fibre/matrix and dispersion but also depends on the aspect ratio of the fibre in the composite. This result can depend on the wetting of the fibres with the PP matrix, i.e., insufficient wetting of fibres in PP-J30-V15 composite led to the decreased $K_{IC}$ and $G_{IC}$ values. This result was corroborated by our earlier studies (Ranganathan et al., 2015).
The fibre length is expected to have important consequences on the mechanical properties of the composites (Baillif et al., 2009). Our earlier study showed that one reason for the higher energy absorption is the higher average fibre length in the final composites (Ranganathan et al., 2015). The greater fibre length in the composites is able to bridge the higher number of cracks in the matrix, thereby providing resistance to crack propagation and crack opening. Further, the addition of MAPP on the jute-viscous composite has shown better bridging of cracks by the formation of chemical bonds between the fibre and the matrix (Teh et al., 2005; Paunikallio et al., 2004). Thus, the composite showed larger displacement before complete pullout of fibre due to the formation of weak (flexible) interfacial adhesion. The observation was also confirmed with

**Figure 6.5:** CT specimens fracture behaviour of PP composites containing 30 wt.% jute fibers modified with viscose fibers and MAPP

The fibre length is expected to have important consequences on the mechanical properties of the composites (Baillif et al., 2009). Our earlier study showed that one reason for the higher energy absorption is the higher average fibre length in the final composites (Ranganathan et al., 2015). The greater fibre length in the composites is able to bridge the higher number of cracks in the matrix, thereby providing resistance to crack propagation and crack opening. Further, the addition of MAPP on the jute-viscous composite has shown better bridging of cracks by the formation of chemical bonds between the fibre and the matrix (Teh et al., 2005; Paunikallio et al., 2004). Thus, the composite showed larger displacement before complete pullout of fibre due to the formation of weak (flexible) interfacial adhesion. The observation was also confirmed with
the Karger-Kocsis and Friedrich report; their study demonstrated a slower crack growth rate for the composites with longer fibre lengths.

6.4.4 Fatigue properties

The results of constant-amplitude, tension-tension, cyclic fatigue tests for the composites are shown in Figure 6.6. It is clearly seen that the composites with viscose fibres show a greater fatigue life compared with the unmodified composites. The improvement in the stress level was observed over the entire range, indicating the enhancement in fatigue life of the modified composites. The experimental data of stress-life (S-N) curves of the PP based composites shown in Figure. 6 were fitted to Basquin’s law (Manjunatha et al., 2010; Shahzad et al., 2014):

$$\sigma_{max} = \sigma_f (N_f)^b$$

Where $\sigma_f$ the fatigue strength coefficient (FSC) and $b$ is is the fatigue strength exponent (FSE).

The values of FSC and FSE of the modified and unmodified composites are listed in Table 3. The results depicts that the addition of viscose fibres slightly increases the FSC. Figure 6 shows that the fatigue life of PP-J30-V10 is three-times higher than the unmodified jute composites due to improved fracture toughness and fatigue crack growth resistance because of the higher average viscose fibre length (Reifsnider, 2012). Goel et al. reported that fibre length also plays an important role in the fatigue behaviour of discontinuous fibre-reinforced composites; the long fibres always carry a significantly higher fraction of the load compared with short fibres (Goel et al., 2009). Evans et al. studied the fatigue crack growth in short carbon fibre-reinforced PEEK; their study report stated that improved crack growth resistance is attributed not only to the primary toughening mechanism (such as fibre bridging and pullout) but also to crack front bowing and deflection in the presence of fibres.
Figure 6.6: Fatigue life time (S-N) curve of PP composites containing 30 wt.% jute fibers modified with viscose fibers and MAPP

6.5 Conclusions

The present study investigated the effects of fracture toughness and fatigue properties based on the extent of jute and viscose fibre breakage during the extrusion process.

The fibre length measurements after the extrusion process showed that the lengths of the jute fibres were comparatively less than those of the viscose fibres. Further, the increase in the concentration of the viscose fibre has resulted in increased fibre length. It is believed that screw speed appeared to be insufficient to break the fibres or to separate the bundles.

The PP-J30 composites were found to have an unstable crack growth, revealing a lower fracture toughness and energy of 3.9 MPa.m$^{1/2}$ and 4.7 kJ/m$^2$, respectively, which increased to 9.1 MPa.m$^{1/2}$ and 28.9 kJ/m$^2$, respectively with the addition of 10 wt% viscose fibre due to stable crack growth. Further, the addition of 2 wt% MAPP to PP-J30-V10 composite was found to
increase the fracture toughness and energy to 10.0 MPa.m$^{1/2}$ and 31.2 kJ/m$^2$, respectively, due to inhibition of the crack propagation.

The fatigue life of the PP-J30-V10 composite was three-times higher than that of the PP-J30 due to improved fracture toughness and fatigue crack growth resistance because of the higher average viscose fibre length. Further, with the addition of 2 wt% MAPP, the FSE of impact modified jute-reinforced thermoplastic composite was found to increase from 40.2 to 41.0 MPa, thus demonstrating increased work energy by inhibiting the propagation of cracks.
7 Effect of LFT process on fiber dispersion and mechanical properties of viscose fiber reinforced thermoplastic composites

7.1 Introduction

The LFT process, is an extrusion technique in which continuous fiber rovings are directly fed into the extrusion unit which contains molten thermoplastic materials and then moulded in a single operation. LFT technique has been confirmed by several researchers (Krause et al., 2013; Henning et al., 2005) with significant improvements in stiffness, strength and toughness of composites over the short fiber reinforced thermoplastics (Zhuang et al., 2008). The shear during the process determines the fiber breakage, distribution and orientation. Additionally, the manufacturing process, residence time, fiber-fiber interactions and fiber-matrix interactions play a significant role in determining the dispersion of fiber (Chollakup et al., 2011). Our earlier studies have confirmed that the incorporation of regenerated cellulose fibers in the composite led to have clumping and agglomeration since the fiber bundles were soft and tough which were difficult to separate during the process. Further, many studies have demonstrated the reason behind the poor dispersion of fiber within the matrix due to the strong hydrogen bonds formed between them (Baillif et al., 2009; Dunnom et al., 1973; Klason et al., 1984; Saheb et al., 1999). Thus the clumping and agglomeration must be avoided to produce efficient composites (Sanadi et al., 1997). The efficiency of the LFT composite depends on the stress transferred from the matrix to the fibers, thereby contributing to higher strength, stiffness and toughness. Using long fiber rovings in LFT technique resulted in higher fiber length distribution, clumping and agglomeration which ultimately reduces the efficiency of the composites (Rowell et al., 1997).
The efficiency of the composites are improved by changing fiber feeding section, feed rate, process conditions and processing aids.

Czarnecki et al reported the extent of breakage was severe and rapid for glass fibers, and less extensive for kevlar fibers and least for cellulose fibers. Thus, the higher fiber lengths have the more tendencies to agglomerate (Czarnecki and White 1980). The use of dispersing and coupling agent could improve the dispersion of the cellulose fibers within the matrix. Raj et al indicates the importance of using surface modifiers to improve the fiber dispersion in the PP matrix (Raj and Kokta 1989). Woodhams et al has used stearic acid to study the dispersion of fiber in High density polyethylene (HDPE)/wood fibers. It concluded that the mechanical properties were improved along with improved fiber dispersion and improved wetting between the fiber and matrix (Woodhams et al., 1984). Malhotra et al also studied the addition of stearic acid during the compounding has greatly improved the fiber dispersion within the composites (Malhotra et al., 2012). The addition of silane agents and MAPP has been largely studied and resulted that the chemical treatments improved the fiber dispersion and also improves the adhesion with matrices (Baillif et al., 2009).

In this present work, the effort has been taken to improve the dispersion of regenerated cellulose fiber in order to obtain high performance LFT composite. The composites were manufactured through different methods of LFT technique. The fiber dispersion in the PP matrix has also been studied using scanning electron microscope (SEM). The comparison of mechanical properties including toughness has been investigated.
7.2 Experimental

7.2.1 Materials

Homopolymer polypropylene (PP) (extrusion grade with MFI of 35g/10 min) was purchased from Indian oil corporation ltd., India used as matrix. A polypropylene grafted maleic anhydride (Epolene E-43) was purchased from M/s. Sigma Aldrich, USA, with an average molecular weight (Mw) ~ 9,100, was used as coupling agent. Stearic acid, the dispersing agent was procured from Diucon Laboratory, Chennai. The viscose fibers (VF) were supplied from Cheran Spinning Mills, Erode, India. They are derivative of wood pulp said to be regenerated cellulose processed by spinning method. The properties of used fiber are reported in chapter 2 of the thesis. The linear density of the fiber was 1200 tex, and they were used in the form of continuous fiber bundles.

7.2.2 Processing of composite material

Prior to extrusion, the viscose fibers were dried completely at 60°C at least 2 hours until achieving a uniform weight at different intervals of time. The composite materials were manufactured using direct LFT processing method, using a high performance co-rotating twin-screw extruder (ZE-25 model Berstorff Maschinenbau GmbH, D-3000 Hannover, Germany) were used and reported elsewhere (Ranganathan et al., 2015). The composites were prepared using 10 wt% of the viscose fiber content and the two different methods of feeding were adapted to manufacture the composite. In the first method i.e., (LFT-1), viscose fibers roving were incorporated into a side feeder of the extruder which is near to the die head region as shown in Figure 7.1(a), whereas in the second method (LFT-2) the fiber were fed into the extruder on the same inlet of PP-matrix shown in Figure 7.1(b). In both cases the fibers were fed directly into
the polymer melt and the fiber weight fractions were controlled by the screw speed based on the target content, the weight of the fiber roving per meter and the length per screw revolution and time. The additives stearic acid and MAPP are directly mixed with PP granules before loading into the hopper, the reaction mechanism between regenerated cellulose fibers, SA, MAPP and PP are shown in Figure 7.2. Material formulations and denotations used for comparison are shown in Table 7.1. The compound was extruded in the form of continuous profile and cut to the length of 260-270mm. The extrudates were compression molded to rectangular sheets 250 x 125 mm and thickness of 3 mm using a conventional compression mold from M/s.Hindustan Hydraulics, India with a capacity of 150 tons. The process parameters in the compression moulding were set to a temperature of 170°C, pressure 35 MPa and time 15 min. The test samples were prepared as per ASTM standards from the compressed sheet using a contour cutter.

**Figure 7.1:** The schematic representation (a) LFT-1extrusion method, (b) LFT-2 extrusion method
Figure 7.2: Reaction mechanism between PP, viscose fiber, SA and MAPP.

Table 7.1: Material formulations and denotations

<table>
<thead>
<tr>
<th>Materials</th>
<th>PP (wt.%)</th>
<th>Viscose fiber (wt.%)</th>
<th>Stearic acid (wt.%)</th>
<th>MAPP (wt.%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LFTD-1</td>
<td>90.0</td>
<td>10</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>LFTD-2</td>
<td>90.0</td>
<td>10</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>LFTD-2+ SA</td>
<td>87.5</td>
<td>10</td>
<td>0.5</td>
<td>-</td>
</tr>
<tr>
<td>LFTD-2+ SA+M2</td>
<td>87.5</td>
<td>10</td>
<td>0.5</td>
<td>2.0</td>
</tr>
</tbody>
</table>
7.3 Testing and characterization

7.3.1 Microscopy analysis

The morphology of the composites was studied using scanning electron microscopy (SEM). The fractured cross-sections of the impact specimens were sputter coated with gold and studied in a SEM at an acceleration voltage of 20 kV. The fiber dispersion was studied on microtomed cross section of the composite samples.

7.3.2 Mechanical properties

*Tensile properties:* The tensile properties of the composites were performed according to ASTM D 638 using conventional tensile tester, Instron 3382, UK with a cross head speed of 3 mm/min. The tensile strength, tensile modulus and elongation at break are calculated from the tensile test data. At least five specimens were tested for each type of composite sheet to check the repeatability.

*Flexural properties:* The flexural testing was performed according to ASTM D 790 using Instron 3382, UK. The tests were carried out with a span-to-depth ratio of 16:1 at a crosshead and the strain rate of 5 mm/min. At least five specimens were tested and the results were averaged.

*Impact strength:* Impact testing was performed according to ASTM D 256 on notched Izod specimens using M/s. Tinius Olsen, Model IT 503, Germany. The edge-wise impact was provided with built-in pendulum hammer. Ten specimens were tested for each set of samples and the mean values were reported.

7.3.3 Fiber length measurements

The fibers were extracted by dissolving the composites with boiling xylene. The fibers were manually separated without overlapping using water and observed using optical microscope. Length of the fibers was calculated automatically from the images through software. The fiber
images were taken with M/s. OPUS Optical microscopy with a 0.75 X – 4.5 X observing magnification. The length given for each fiber is average of at least 100 measurements.

7.4 Results and discussion

7.4.1 Fiber dispersion

It is well known that the fiber dispersion plays an important role in deciding the properties of the thermoplastic composites. Scanning electron microscope (SEM) images of LFT-1, LFT-2, LFT-2+SA, LFT-2+SA+M2 composites are shown in Figure 7.3. Figure 7.3(a) shows the poor dispersion of viscose fibers with in the matrix, i.e. the fiber bundles agglomerations are clearly seen in the composites. This study has also shown a non-homogenous dispersion of viscose fiber in PP-Jute composites which was believed because of their soft and tough nature. The soft and tough fiber bundles were more difficult to separate during the processing. In addition, many authors reported that the natural cellulosic fibers in the composite lead to poor dispersion of the fibers due to strong inter-fiber hydrogen bonding which holds the fibers together (Saheb et al., 1999; Felix et al., 1991). Figure 7.3(b) represents the composite manufactured in LFT-2, it shows better fiber dispersion compared to the LFT-1, but still found to have some non-uniformity with reduction in the size of agglomerates. The improvements in the fiber dispersion obtained as a result of increased shear energy supplied to the composite material during processing (McCrum 1997).
Figure 7.3: SEM micrographs on the cross-section of LFT-1 and LFT-2 composites modified with stearic acid and MAPP

Figure 7.3(c) shows that the agglomerates are almost disappeared and the improvement in the fiber dispersion is mainly due to the incorporation of dispersing agent which indicated that the use of stearic acid hydrophobizes the fiber (Paul et al., 2010). Saheb et al studied that the effect of stearic acid on the fiber and it act as a highly dispersing agent which reduces the fiber to fiber interaction (Saheb et al., 1999). Many authors detailed the importance of using surface modifiers to improve fiber dispersion in cellulose fiber/PP composites (Raj et al., 1989). Figure 7.3(d) shows the dispersion of fiber in the polymeric matrix was improved with the addition of stearic and MAPP on the PP/VF composites manufactured in LFT-2 method. Therefore, uniform fiber
dispersion with the use of dispersing agent and compatibilizer on PP/VF composites achieved which may help to improve the mechanical properties.

7.4.2 Mechanical properties

The mechanical properties of PP/VF, SA modified PP/VF and SA+MAPP modified PP/VF composites manufactured using LFT-1 and LFT-2 were experimentally measured and compared, shown in Table 7.2.

Tensile properties: Figure 7.4 shows the comparison of tensile properties of PP/VF manufactured using LFT-1 and LFT-2 and modified with SA and MAPP. The tensile strength of SA and MAPP modified PP/VF composites manufactured using LFT-2 shows 20% higher than the LFT-1 composites. It was also found that the composite manufactured using LFT-2 with and without SA improves the tensile strength to 6% and 16% respectively, when compared to LFT-1 manufactured composites. This is attributed mainly due to the stress has been transferred from PP matrix to the viscose fiber. It may also be due to the fact that the viscose fiber bundles are prone to breakdown to smaller size by the high shear force and residence time has led to improved fiber dispersion compared to LFT-1 process. Further, the addition of dispersing agent has again might have improved the fiber dispersion within the matrix. This is corroborated by Woodhams et al reported that the use of stearic acid in high density polyethylene (HDPE)/wood fibers improved fiber dispersion and wetting between the fiber and matrix and thus resulted in significant improvements in mechanical properties (Woodhams et al., 1984). The addition of 2wt% MAPP on LFT-2-PP/VF-SA improves the tensile strength of the composites due to the stress transfer between the fiber and matrix. Similarly, Oksman et al has stated that MAPP compatibilizer has a positive effect on the tensile strength in natural fiber reinforced PP-composites (Oksman et al., 2009). The change in tensile modulus of the composites followed the
same pattern as that of the tensile strength. The variations in values of tensile modulus for the composites are presented in Table 7.2.

**Table 7.2:** Mechanical properties of PP/VF composites manufactured using LFT-1 & 2 and modified with stearic acid and MAPP (values in the bracket are standard deviation)

<table>
<thead>
<tr>
<th>Sample code</th>
<th>Flexural strength (MPa)</th>
<th>Flexural modulus (GPa)</th>
<th>Tensile strength (MPa)</th>
<th>Tensile modulus (GPa)</th>
<th>Elongation to break (%)</th>
<th>Impact strength (J/m)</th>
<th>Toughness (J/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>LFTD-1</td>
<td>33.7(0.5)</td>
<td>2.1(0.08)</td>
<td>24.5(0.8)</td>
<td>1.3(0.06)</td>
<td>4.5(0.5)</td>
<td>46.8(3.1)</td>
<td>1578</td>
</tr>
<tr>
<td>LFTD-2</td>
<td>39.3(1.2)</td>
<td>2.2(0.12)</td>
<td>26.0(0.8)</td>
<td>2.0(0.13)</td>
<td>5.2(0.6)</td>
<td>63.7(4.3)</td>
<td>1893</td>
</tr>
<tr>
<td>LFTD-2+SA</td>
<td>49.3(0.9)</td>
<td>2.8(0.08)</td>
<td>28.5(0.6)</td>
<td>2.0(0.09)</td>
<td>6.3(0.9)</td>
<td>79.4(4.7)</td>
<td>2965</td>
</tr>
<tr>
<td>LFTD2+SA+M2</td>
<td>50.7(1.4)</td>
<td>2.8(0.11)</td>
<td>29.4(1.1)</td>
<td>2.2(0.11)</td>
<td>6.5(0.7)</td>
<td>88.9(5.2)</td>
<td>3551</td>
</tr>
</tbody>
</table>

From Figure 7.4, it can be seen that the tensile modulus of composites are increased to 45% and 53% for LFT-2 and LFT-2+SA composites, respectively, compare to LFT-1. This attributed to the increase in modulus is mainly due to the improvement in the fiber dispersion. It is well agreed with the studies reported by Deshmukh et al mentioned that the stiffness and young’s modulus of the matrix depends on the extent of dispersion (Deshmukh et al., 2010). On the other hand, in case of LFT-2+ SA+M2, the modulus increment is more distinct to 65% upon the addition of 2wt% MAPP due to better fiber-matrix adhesion compared to unmodified composites which also enhances the stress transfer from matrix to fiber, the similar effect was also reported by (Oksman et al., 1998).
The elongation at break of PP/VF composites manufactured using LFT-1 and LFT-2 and modified with SA and MAPP are shown in Figure 7.5. In general, the viscose fiber exhibits higher elongation at break compared to other regenerated cellulose fibers as reported in our earlier study (Ranganathan et al., 2015). The composites manufactured using LFT-2 process
shows higher elongation than the LFT-1 composites and indicates the decreased stiffness which is due to the improved dispersion of fibers. The addition of SA on the PP/VF composites manufactured using LFT-2 has shown a slight improvement in elongation at break. Similar effect was reported by Nekkaa et al that the addition of dispersing agent has improved the elongation at break polypropylene/spartium junceum fiber composites (Nekkaa et al., 2012). Further, it is noted that the addition of 2wt% MAPP has accompanied the improvement in the ductility of the composites. Rana et al also found that the addition of coupling agent (MAPP) and elastomer on PP-jute composites has improved the elongation values along with higher resistance to impact strength (Rana et al., 2003).

**Toughness:** The toughness values shown in the Table 7.1 are retrieved from the tensile stress-strain curves. The Chapter 4 and 5 reported that the addition of viscose fiber has improved the energy absorption of thermoplastic composites by promoting the toughening mechanism such as debonding and pullout of fibers. This study predicted the variation in the tensile toughness values on the composites manufactured using different LFT methods. Table clearly shows that the LFT-2 composite exhibit higher ultimate tensile strength and plastic deformation than the LFT-1 method which has contributed to improved area under the curve. The viscose fiber have a good strain to failure characteristic (Ranganathan et al., 2014), thus the addition of small wt% (10wt %) of viscose fiber within the matrix have shown higher stain to failure, thereby contributing to higher energy absorption. Further, the addition of SA and MAPP has again improved the mechanical performance of the composites including toughness due to the improved fiber dispersion along with the formation of weak (flexible) interfacial adhesion between the fiber and matrix.
**Flexural properties:** Figure 7.6 shows the variation in the flexural strength of PP-VF composites manufactured using LFT-1 and LFT-2 and modified with SA and MAPP. As shown in Table 7.2, the composites manufactured by LFT-2 process shows higher flexural strength compared to LFT-1. This may be due to the increase in shear in LFT-2 process which separate the viscose fiber bundle into their individual fibers and increased the interface area between the fibrils and matrix. This factor could dramatically increase the flexural strength of the composites (Feng et al., 2007). Further, the addition of SA and combination of SA and MAPP to the PP/VF composite has raised the strength to 46% and 50%, respectively. This clearly demonstrates that the strength largely depends on both the fiber dispersion as well as the fiber –matrix interaction (Liu et al., 2013; Hargitai 2004; Oksman et al., 2002).

![Figure 7.6: Flexural properties of viscose fiber reinforced polypropylene composites manufactured using LFT-1 and LFT-2 method and modified with stearic acid and MAPP](image_url)

The change in the flexural modulus of the composites manufactured using LFT-1 and LFT-2 and modified with dispersing and coupling agent is shown in Table 7.2 and Figure 7.6. A slight increment of the moduli of the composites manufactured using LFT-2 was observed compared to
LFT-1. The composite with highest flexural modulus was achieved with the combination of SA and MAPP on PP/VF composites due to the very good fiber dispersion within the matrix and which increases the interfacial adhesion within the composites.

Impact properties: Figure 7.7 represents the notched izod impact strength of composites. The PP/VF composites manufactured using LFT-1 showed lower impact strength of 46.8 J/m which attributes to poor dispersion of fibers in the matrix. The properties of the thermoplastic composites are influenced by the processing parameters. Takase et al stated that the fiber length and dispersion needs to be optimum to obtain the enhanced mechanical properties (Takase and Shiraishi 1989). The increase the shear with respect to time of mixing proved to have better fiber dispersion along with the improvement in impact strength of 36% compared to LFT-1 process. Further, the addition of dispersing agent has influenced the dispersion of fibers within the matrix which in-turn enhanced the toughening mechanism of the composites. This effect was also observed by Feng et al that the addition of SA resulted in prevention of filler agglomeration and led to improved impact strength and elongation at rupture (Feng et al., 2007).

Further, the composites manufactured by LFT-2 with the addition of MAPP in combination with SA in the matrix has achieved a maximum improvement in notched impact strength of 90% which is believed due to the formation of weak (flexible) interfacial adhesion. The similar effect was observed by Oksman et al that the PP/wood fiber (WF) composites containing styrene-ethylene- butylene-styrene (SEBS) /maleic anhydride (MA) has shown the highest impact energies, and the addition of MAPP found to have little effect on the notched impact strength (Oksman et al., 1998). Paunikallio et al also reported that the viscose fiber–PP composites with MAPP had higher plastic deformation region compared to that the specimen prepared without MAPP (Paunikallio et al., 2003).
**Figure 7.7**: Izod impact strength of notched samples of viscose fiber reinforced polypropylene composites manufactured using LFT-1 and LFT-2 method and modified with stearic acid and MAPP.

### 7.4.3 Microscopy analysis

Images of impact fracture surfaces obtained by SEM reveal significant fiber pull-out shown in Figure 7.8. The Figure 7.8(a) and (b), shows some fiber debonding lines on the fracture surface image, which indicates the fiber-matrix adhesion. Some holes can be seen in the PP/VF composites, but fibers that were pullout are hard to detect. The study of Adekunle et al., observed that, good fiber–matrix adhesion, with Lyocell reinforced composites, as it was very difficult to see the fiber pull-outs. They believed that the fibers were well-embedded in the matrix due to their microstructure. The role of SA as dispersing agent was observed in viscose fiber/ PP composites shown in Figure 7.8(c). For the composites with MAPP, weak (flexible) interfacial adhesion between non-polar PP, stearated viscose fiber and viscose fiber-MAPP was expected. The Figure 7.8(d) also indicates no voids around the fibers surface and found to have clear fiber debonding and pullout paths of viscose fiber. The polymer composites with better fiber
dispersion and weak (flexible) interaction between with matrix improved the toughness, strength and stiffness of PP/VF composites.

**Figure 7.8:** SEM micrograph of fractured specimen from impact testing of LFT-1 and LFT-2 composites modified with SA and MAPP.

### 7.4.4 Fiber length measurements after process

The strength and toughness of the composites are also depends on the mean fiber length after processing (Karmaker et al., 1991). Figure 7.9(a) and (b) shows the viscose fiber length distribution of composite manufactured using LFT-1 and LFT-2. The typical average fiber length values of LFT-1 and LFT-2 are 6.9 and 4.4 mm, respectively. The reduction in the average fiber length was attributed to the increased shear energy produced from the residence time. Hietala et
al described that the shear forces reduces the particle size and separates the fiber bundle (Hietala et al., 2011). George et al found that the composites with higher fiber length had poor dispersion and fiber entanglements, whereas the composites with shorter fiber length observed to have good dispersion and are more easily aligned along the flow direction (George et al., 1995).

The composites manufactured by LFT-1 process in too short duration resulted with poor dispersion, while long residence favored improved fiber dispersion but reduction in the fiber length. Arino et al study reported that the fiber breakage reduces the reinforcement effect of the cellulose fiber (Arino et al., 2012). Nevertheless, it has also been reported that good dispersion of the fibers is an important parameter to improve the mechanical properties of the composite and which reduces the fiber length may result in better mechanical properties.

Figure 7.9: Fiber length measurements (a) LFT-1, (b) LFT-2

7.5 Conclusion

The objective of this study was to improve the dispersion of regenerated cellulose fiber and to obtain the high performance composites using LFT technique.
This study showed that composites manufactured using LFT-1 had poor fiber dispersion i.e. the fiber bundles retained in clusters, whereas the LFT-2 process offered homogenized fiber dispersion. This improvement in fiber dispersion was obtained as a result of the increased shear energy supplied to the composite material during processing. It was also observed that LFT-2 composites achieved higher mechanical performance including toughness compared to LFT-1 whereas the fiber lengths are found to be lesser than that of the LFT-1 composites. The highly dispersed composites along with weak (flexible) interfacial adhesion are the possible reason behind improved performance of the composites. Further, the addition of dispersing agent and surface modifier on the LFT-2 composites has again improved the fiber dispersion along with the mechanical performance of the composites.
8 Conclusions and recommendation

8.1 Conclusions

This dissertation describes a study on the improved impact behaviour of long jute fiber reinforced thermoplastic composites through hybridization using regenerated cellulose (Viscose fiber). This study gives an account for the manufacturing of long fiber hybrid thermoplastic composites and investigates the mechanical performance, impact properties and fracture toughness of the composites. Further it describes the factors influencing the impact performance and morphology of the developed composites. In addition the fiber breakage after extrusion and softening temperature for each composite were analyzed. Based on the observations and experimental results the following conclusions can be made about the work presented in this thesis.

- Hybrid bio-fiber based thermoplastic composite were manufactured using Direct-LFT) extrusion techniques. A mix of jute and viscose fiber rovings is a suitable way to fed into the extruder. The mechanical performance of the composites proved that the impact strength of PP-J composites can be increased with viscose fibers as impact modifier. The impact strength was considerably increased with the increase in the concentration of viscose fiber. The optimum impact strength, toughness and elongation to break were found with 10 wt% addition of viscose fibers, with a slight reduction in strength and modulus of the composites. The modification of matrix with 2wt% of MAPP had a positive effect on all mechanical performance including impact strength. The fiber length measurements after the extrusion showed that the viscose fiber had a length of 6.8-7.3 mm whereas the jute found to be 1.0-1.2 mm. The electron microscopy images of fracture surfaces showed that PP-J composites had poor fiber matrix adhesion compared to those
of impact modified composites. The improved impact strength on the modified 
composites has supported the hypothesis of energy dissipation along the length of the 
fiber.

- To obtain further understanding on the impact behaviour, the fragmentation 
characteristics on falling weight impact (IFW) were investigated. The developed impact 
modified composites showed higher propagation energies compared to unmodified jute 
reinforced thermoplastic composite. The crack bridging during the impact damage was 
influenced on the surface modified hybrid thermoplastic composite, due to the formation 
of weak (flexible) interfacial adhesion. The dynamic mechanical analysis results provided 
a characteristic of higher damping peaks for all impact modified composites. The fiber 
dispersion analysis using optical microscopy has proved insufficient fiber dispersion of 
viscose fiber on the developed composite when compared to jute fiber. 
Hence, the experimental finding concluded that the toughness of the composites was 
influenced by viscose fiber. It is also predicted that, energy dissipation along with other 
mechanical properties could be enhanced by improving the efficiency of viscose fiber 
dispersion on the composite.

- The D-LFT technology use the twin screw to chop the fibers, the length after the 
eextrusion, dispersion of fiber depend on the processing condition, mixing speed, shear, 
residence time and type of fiber. Our experimental study observed that the feeding of 
fibers in mix form using direct LFT extrusion technique have resulted in insufficient fiber 
dispersion, the viscose fiber retained in bundles due to its soft and tough nature. 
Therefore a comparative study on composites manufactured using LFT-1 and LFT-2 was 
carried out.
- In the LFT-1 viscose fibers roving were incorporated into a side feeder of the extruder which is near to the die head region, whereas in the second method LFT-2 the fiber were fed into the extruder on the same inlet of PP-matrix. This processing technique is aimed to improve the fiber dispersion. The morphology of the developed composites through LFT-2 proved the improvement in fiber dispersion. The composites achieved higher mechanical performance including toughness compared to LFT-1.

### 8.2 Study limitations and recommendations

- As the jute fiber, which was used as reinforcement has varying diameter all over its length; hence was very difficult to control the fiber content during the manufacturing of composites. The fiber was used in the form of roving without much twist and prepared manually, achieving the uniform consistency in thickness found to be impossible.

- The elevated temperature fracture toughness test was not performed due to the unavailability of environmental chambers (temperature controlled) fitted to universal testing machine (UTM).

- The automotive industry focuses in replacing non-sustainable materials with the environmental friendly materials with energy efficiency. To commercialize the bio-composite materials, a huge study and scientific approaches has to be adapted.

This study approached the direct LFT extrusion techniques for the development of composites, this extrusion process would require a significant improvement to disperse and distribute the fiber uniformly. The splitting of fiber bundle during processing is essential to achieve homogenous mixing. In the case of hybrid composites especially
when using regenerated cellulose more effort should be given during processing of composites in finding a viable way for the industries to obtain good dispersion of fibers in matrix.

- The further attempts could be made to study the fracture mechanisms at elevated temperatures and more detailed study is required to understand the fracture mechanism on various crack lengths induced on the samples, where the impact modifier acts as bridging element in the crack propagation.

### 8.3 Scientific and engineering contributions of the work

The energy absorption capability of composites materials is a consequence of the work of fracture that arises from several mechanisms such as interfacial adhesion between fiber and matrix, crack propagation in the matrix, pullout behavior of fiber, fiber length, fiber dispersion and its orientation.

- Our research work identified the various possible toughening mechanisms in LFT composite and then effectively combined these mechanisms to produce a tough composite.

- The impact modified thermoplastic composite absorbed higher energy through plastic deformation mechanism compare to the unmodified PP/Jute composite.

- The fracture mechanics of pullout theory was applied to study the processes involved in fiber-matrix interfacial debonding of long fiber reinforced thermoplastic composites.

- In addition to frictional coefficient, the surface modification induced weak (flexible) interfacial bonding between the fibers and matrix, which contributed higher toughness to the composites.
The energy dissipation occurs along the length of the fibers during the crack propagation of the composite. The crack bridging mechanisms contributed the crack growth resistance, i.e. the higher average fiber length of the fibers in the matrix restricted the crack propagation and resulted in impact toughening of LFT composites.

The linear elastic plastic fracture mechanics approach were used and determined the fracture toughness and critical energy release rate of impact modified thermoplastic composites.
References


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Oksman K, Skrifvars M, Selin JF. Natural fibers as reinforcement in Polylactic acid (PLA) composites. *Composites Science and Technology* 2003; 63(9):1317-1324.


Reifsnider K.L. Fatigue of Composite Materials, *Technology and engineering* 2012; Elsevier science publishing company INC.


Appendices

Appendix 1:

Optimization of fiber feeding with respect to screw speed

The screw speed calculation was carried out by considering the following factors

a. Weight of the roving per meter; b. the total weight of the fibers to be fed; c. length of
   the fiber/revolution; d. time.

Sample calculation

a. The total output rate is 5 Kg/hr

b. Total targeted fiber content 35 wt% - 1750 gms

c. The weight of the roving prepared - 11.5 gms

d. The length of the roving per revolution – 0.025m

e. Screw Speed = \(\frac{1750}{(0.025 \times 60 \times 11.5)}\) = 100 rpm

f. The screw speed has been kept constant for all composition, and the weight of the fiber
   roving per meter are varied according to the targeted rate of the fiber feeding.
Appendix 2:

Original image - Manufacturing of composites using LFT process

Appendix 3:

Original image of compressed sheet from extrudate
Appendix 4:

Optical microcopy images of microtome- PP-J30 composites modified with viscose fibers and MAPP.
### Appendix 5:

Table 1: Actual cost of thermoplastic, fibers, compatibilizer and other impact modifiers

<table>
<thead>
<tr>
<th>Materials</th>
<th>Cost (in US $)/Kg</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>Polypropylene</td>
<td>1.62</td>
<td>Indian oil corporation ltd.</td>
</tr>
<tr>
<td>Viscose fiber</td>
<td>1.52</td>
<td>Actuals procured from M/s. Cheran Spinning mill</td>
</tr>
<tr>
<td>Glass fiber - Direct Roving</td>
<td>1.61</td>
<td><a href="http://www.alibaba.com/showroom/fiberglass-roving-price.html">http://www.alibaba.com/showroom/fiberglass-roving-price.html</a></td>
</tr>
<tr>
<td>Polypropylene-graft-maleic anhydride average Mw ~9,100</td>
<td>122.30</td>
<td>M/S.Sigma Aldrich</td>
</tr>
<tr>
<td>Stearic acid</td>
<td>0.61</td>
<td>M/S.Adinath Petrochem</td>
</tr>
<tr>
<td>Styrene ethylene butadiene copolymer</td>
<td>3.11</td>
<td>(Taipol SEBS 6154)</td>
</tr>
<tr>
<td>Ethylene/propylene/diene terpolymer EPDM,</td>
<td>2.50</td>
<td>Shenzhen Comflex Industry Limited</td>
</tr>
</tbody>
</table>
## Appendix 6:

Table 2: General idea of cost comparison for PP-Glass, PP-Jute-Viscose, PP-PP-jute-EPDM, PP-jute-SEBS

<table>
<thead>
<tr>
<th>Material</th>
<th>PP-glass</th>
<th>PP-jute -viscose</th>
<th>PP-jute -EPDM</th>
<th>PP-jute-SEBS</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Weight</td>
<td>Cost</td>
<td>Weight</td>
<td>Cost</td>
</tr>
<tr>
<td>PP</td>
<td>580</td>
<td>0.94</td>
<td>580</td>
<td>0.94</td>
</tr>
<tr>
<td>Jute</td>
<td>--</td>
<td>--</td>
<td>300</td>
<td>0.20</td>
</tr>
<tr>
<td>Viscose</td>
<td>--</td>
<td>--</td>
<td>100</td>
<td>0.15</td>
</tr>
<tr>
<td>Stearic acid</td>
<td>--</td>
<td>--</td>
<td>5</td>
<td>--</td>
</tr>
<tr>
<td>Polypropylene-graft-maleic anhydride</td>
<td>20</td>
<td>2.44</td>
<td>20</td>
<td>2.44</td>
</tr>
<tr>
<td>Glass fiber</td>
<td>400</td>
<td>0.64</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Styrene ethylene butadiene copolymer</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td>Ethylene/propylene/diene terpolymer EPDM</td>
<td>--</td>
<td>--</td>
<td>--</td>
<td>--</td>
</tr>
<tr>
<td><strong>Total cost for 1 kg of composite</strong></td>
<td><strong>4.03</strong></td>
<td><strong>3.73</strong></td>
<td><strong>3.84</strong></td>
<td><strong>3.90</strong></td>
</tr>
</tbody>
</table>
## Appendix 7:

Table 3: Comparison of physico-mechanical properties of PP- Long glass fiber composite with various impact modified composites

<table>
<thead>
<tr>
<th>Composite</th>
<th>Izod notched impact strength (kJ/m²)</th>
<th>Izod notched impact strength (J/m)</th>
<th>Tensile strength (MPa)</th>
<th>Density (kg/m³)</th>
<th>Reference</th>
</tr>
</thead>
<tbody>
<tr>
<td>PP-Long glass fiber + MAPP</td>
<td>15.7</td>
<td>--</td>
<td>--</td>
<td>1183</td>
<td>Wollan E., 2015; Poucke et al., 2006</td>
</tr>
<tr>
<td>PP-Jute - Viscose+MAPP</td>
<td>8.0</td>
<td>67.4 ± 2.7</td>
<td>30.4 ± 1.0</td>
<td>1137</td>
<td>Ranganathan et al., 2014</td>
</tr>
<tr>
<td>PP/SEBS</td>
<td>8.0</td>
<td>--</td>
<td>20.0 ± 0.5</td>
<td>--</td>
<td>Lv L et al., 2015</td>
</tr>
<tr>
<td>PP/POE</td>
<td>4.5</td>
<td>--</td>
<td>22.0 ± 0.5</td>
<td>--</td>
<td>Lv L et al., 2015</td>
</tr>
<tr>
<td>PP-wood fiber- SEBS- MA +MAPP</td>
<td>--</td>
<td>54.1 ± 2.2</td>
<td>30.5 ± 0.5</td>
<td>--</td>
<td>Oksman et al., 1997</td>
</tr>
<tr>
<td>PP-wood fiber- EPDM- MA +MAPP</td>
<td>--</td>
<td>34.4 ± 1.4</td>
<td>27.7 ± 0.2</td>
<td>--</td>
<td>Oksman et al., 1997</td>
</tr>
</tbody>
</table>

Note:
SEBS: Styrene -ethylene –butadiene styrene
POE:Ethylene octene copolymer
Appendix 8:

Table 4: Comparison of weight savings for PP-glass fiber vs. PP-jute composites.

Consideration for calculation

1. The thickness of the PP-Glass fiber composite laminate is assumed as 10 mm
2. The weight reduction calculated for equal bending strength on both the composites

<table>
<thead>
<tr>
<th>Description</th>
<th>PP-Glass fiber</th>
<th>PP-Jute fiber</th>
</tr>
</thead>
<tbody>
<tr>
<td>Volume (cc)</td>
<td>10</td>
<td>X = 9.4</td>
</tr>
<tr>
<td>Density</td>
<td>1.2 (Wollan E., 2015)</td>
<td>1.1(This study)</td>
</tr>
<tr>
<td>Bending strength (MPa)</td>
<td>1</td>
<td>1</td>
</tr>
<tr>
<td>Thickness reduction (%)</td>
<td>--</td>
<td>6</td>
</tr>
<tr>
<td>Weight savings (%)</td>
<td>--</td>
<td>22%</td>
</tr>
</tbody>
</table>