Use of jute needle-punched nonwoven fabric as reinforcement in composite

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The effect of punch density, depth of needle penetration, fibre orientation and area density of jute needle-punched nonwoven fabric, for use as reinforcing material in jute-reinforced plastic (JRP) composite, has been studied and the composite properties, such as tensile strength and modulus, flexural strength and modulus, and impact strength optimized. Composites have been prepared using hand lay-up technique with polyester resin. The fabric made with optimized parameters has been chemically treated and the mechanical properties of its composite evaluated. It is observed that the cross-laid nonwoven produces better composite, considering its mechanical properties in the machine and cross directions, compared to parallel-laid nonwoven. Jute nonwoven with 10 mm depth of needle penetration, 250 punches/cm² punch density and 700 g/m² area density shows optimum mechanical properties of the composite. On comparing the properties of nonwoven composite with those of the composite made out of woven jute fabric, it is observed that the nonwoven fabric shows better properties than woven fabric. Chemical treatment further improves the properties. Bleaching of nonwoven fabric with H₂O₂ shows optimum properties among the different chemical treatments applied to the fabric.

Keywords: Composite, Jute-reinforced plastic, Mechanical properties, Needle-punched nonwoven fabric, Woven fabric

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1 Introduction

Composites are gaining popularity day-by-day as substitute of metal or wood due to their light weight and superior mechanical properties. Fibre-reinforced plastic (FRP) is a composite with two or more distinct physical phases, one of which is a fibrous phase. If the fibrous phase is jute, it is called jute-reinforced plastic (JRP). Jute is a strong, coarse and rigid fibre with very low extensibility, which makes it suitable to act as reinforcing material in the FRP composite. Jute is cheaper and process friendly, i.e. less wear and tear of tools. An extensive research on JRP has developed various products from composites using jute needle-punched nonwoven as reinforcement. In needle-punched nonwoven technology, barbed needles are continuously pushed into and through fibre web. Some fibres are hold by barbs and their orientation is altered as they transfer into the vertical plane of the resulting fabric. The reorientation and continuous presence of some fibres in both planes produce a coherent structure and affect the properties of nonwoven. Needling also creates web shrinkage along the fibre direction and stretch at right angles to the fibre direction. Moreover, needling changes compactness or packing of fibre assembly. All these events cause the structural changes in the fibre assembly. The extent of these changes depends upon the processing parameters, i.e. fibre orientation, needling density, depth of needle penetration and area density. Though the reinforcing material plays a significant role on the properties of fibre reinforced plastic, very little information is available for jute as reinforcement. In this work, a comparison of nonwoven fabric has also been made with the woven fabric as reinforcement and above-mentioned structural parameters are optimized with respect to the mechanical properties of composite.

Jute contains impurities on its surface. Hence, in jute-reinforced composites, jute fibre needs some chemical modification to improve its compatibility with polyester resin. Hence, several chemical treatments have been tried on jute needle-punched nonwoven fabric made with optimized structural parameters as stated above. An attempt has been made to further increase the mechanical properties of JRP composite by improving the compatibility.

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2 Materials and Methods

2.1 Materials

Tossa jute of grade TD3 was used for the preparation of needle-punched nonwoven fabrics. These nonwoven fabrics and commercially available jute hessian fabric were used for making JRP composites.

For chemical treatment of nonwoven fabrics, a commercial enzyme preparation ‘Biocellulase ZK’ was used. It contains 34.81, 96.30 and 136.0 units of cellulase, zylanase and pectinase per ml of the solution. Other chemicals used were hydrogen peroxide, trisodium phosphate, sodium hydroxide, sodium silicate, Ultravon JU and acetic acid. All the chemicals are of AR grade and used for the treatment of fabric.

Commercial grade general purpose polyester resin was used as matrix of the JRP. For composite preparation, methyl ethyl ketone peroxide, cobalt naphthanate and waxpol with polyvinyl alcohol were used as catalyst, accelerator and mould releasing agent respectively.

2.2 Methods

2.2.1 Preparation of Nonwoven Fabrics

The middle portion of jute reed (after removal of root and tip portion of reed) was processed through jute softener and breaker card after application of water only. The breaker card sliver was then fed to Dilo nonwoven plant. Cross-laid fabrics of different parameters were prepared as per the constructional details shown in Table 1, using 25 gauge RB (regular barb) foster needle. For parallel-laid fabric, a drum was used to prepare the web and then punched in needling loom.

2.2.2 Chemical Treatment of Nonwoven

Alkali Treatment—The nonwoven fabric was treated with 9% of sodium hydroxide solution for 2 h at room temperature. The fabric was washed thoroughly, neutralized and dried.

Scouring—Grey jute fabric was scoured with sodium hydroxide (1%) and Ultravon JU (2 ml/L) at boil for 60 min using 1:20 material-to-liquor ratio.

Hydrogen Peroxide Bleaching—Bleaching of grey, scoured and enzyme-treated fabrics was done at 90°C for 90 min, keeping the material-to-liquor ratio at 1:20 with hydrogen peroxide (1 vol), tri-sodium phosphate (5 g/L), sodium silicate (10 g/L), sodium hydroxide (1 g/L) and Ultravon JU (2 g/L). The pH of the bath was maintained at 10-11.

2.2.3 Preparation of JRP Composites

Composite sheet of 30 × 30 cm size was prepared by hand lay-up technique using a male-female type mould and tightening it by the screw. Two or required layers of each type of woven or needle-punched nonwoven were cut into desired dimension and then put on the mould after applying mould releasing chemical. Then polyester resin was applied on nonwoven, spreading it as evenly as possible along with 2% catalyst and 2% accelerator. The curing time was 30 min in summer under sunlight.

2.2.4 Evaluation of Properties

2.2.4.1 Nonwoven

The tensile properties of the nonwoven fabrics in machine direction were determined at 65% RH and 22-25°C on an Instron tensile testing machine. The test conditions were: test length, 10 cm; cross-head speed, 5 cm/min; and strip width, 2.5 cm. The fabric tenacity and elongation-at-break were determined using the following relationships:

\[
\text{Tenacity (cN/tex)} = \frac{\text{Breaking load (cN)}}{[\text{Specimen width (mm)} \times \text{Fabric area density (g/m²)}]}
\]

\[
\text{Extension-at-break (％)} = \frac{[\text{Elongation-at-break (cm)} \times 100]}{\text{Gauge length (cm)}}
\]

<table>
<thead>
<tr>
<th>Sample</th>
<th>Mass per unit area g/m²</th>
<th>Punch density punches/cm²</th>
<th>Depth of needle penetration mm</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>500</td>
<td>100</td>
<td>8</td>
</tr>
<tr>
<td>B</td>
<td>500</td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>C</td>
<td>500</td>
<td>100</td>
<td>12</td>
</tr>
<tr>
<td>D</td>
<td>500</td>
<td>150</td>
<td>10</td>
</tr>
<tr>
<td>E</td>
<td>500</td>
<td>200</td>
<td>10</td>
</tr>
<tr>
<td>F</td>
<td>500</td>
<td>250</td>
<td>10</td>
</tr>
<tr>
<td>G</td>
<td>500</td>
<td>300</td>
<td>10</td>
</tr>
<tr>
<td>H</td>
<td>300</td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>I</td>
<td>700</td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>J</td>
<td>900</td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>K</td>
<td>1100</td>
<td>100</td>
<td>10</td>
</tr>
<tr>
<td>W</td>
<td>250</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Samples A-K are nonwoven fabrics and sample W is woven fabric.

Enzyme Treatment—Scoured jute fabrics were treated with 4% (owf) enzyme at 55°C for 60 min and 120 min using 1:10 material-to-liquor ratio. The pH of the bath was maintained at 5 with acetic acid and sodium acetate. After the enzyme treatment, the temperature of the bath was raised to 90°C and this temperature was maintained for 15 min. Then the samples were washed and dried.
2.2.4.2 Composite

The tensile properties of composite were evaluated on an Instron tensile tester as per the ASTM standard (D683-86)\(^5\) using the 50 mm gauge length, 6 mm test width and 5 mm/min cross-head speed. The following relationships were used to calculate the tensile properties:

- Tensile strength (MPa) = Maximum load (N)/Initial cross-sectional area of the specimen (mm\(^2\))
- Tensile modulus (GPa) = [Stress (MPa)/Strain] \times 10\(^{-3}\)

Average of 20 tests was taken.

2.2.4.3 Evaluation of Flexural Properties and Impact Strength

A three-point loading system utilizing center loading on a simply supported beam has been applied to evaluate flexural properties on Instron material testing system following the ASTM standard (D790-81)\(^6\), using the sample dimension 80 × 10 mm, support span 64 mm and rate of cross-head speed 1.7 mm/min. The following relationships were used to evaluate flexural properties:

- Flexural strength (MPa) = \(3PL/(2bd^2)\)
- Flexural modulus (GPa) = \(L^3m/(4bd^3) \times 10^{-3}\)
- Maximum strain (%) = \(6Dd/L^2 \times 100\)

where \(P\) is the load at rupture (N); \(L\), the support span length (mm); \(b\), the width of the specimen at the centre of support span (mm); \(d\), the depth or thickness of the specimen at the centre of support span (mm); \(m\), the slope of the tangent drawn at the initial portion of load-deflection curve (Nmm\(^{-1}\)); and \(D\), the maximum deflection at the centre of the specimen (mm). Average of 20 tests was taken.

The impact strength was evaluated in IZOD-type cantilever beam impact tester following the ASTM standard (D 256-88).\(^7\) Average of 10 tests was taken.

3 Results and Discussion

3.1 Properties of Jute Nonwoven

Table 2 shows the physical properties of jute nonwovens in the machine direction. With the increase in depth of needle penetration (samples A, B & C), the bulk density and other properties improve initially and beyond 10 mm depth of penetration they decrease. As the depth of penetration increases for a given web, more barbs enter and fibre transfer in vertical direction is increased accordingly. The decrease in high depth of penetration is attributed to jute fibre breakage in severe action of barbs.\(^3,8\) With the increase in punch density (samples A, D, E, F & G), the extent of vertical rearrangement of fibre increases, resulting in more compact and entangled structure. But, for the fibres of low extensibility and rigidity like jute, there is an acute chance of fibre breakage due to the low fibre mobility and severe action of the needles. Table 2 shows that the bulk density increases with the increase in punch density, which suggests higher consolidation of fibrous web. Tenacity increases and breaking strain decreases up to 250 punches/cm\(^2\), and beyond that the trend is reversed. The similar trend is observed in case of work of rupture and bending modulus with the optimum value at 200 punches/cm\(^2\). Area density for samples H, A, I, J & K shows the increase in bulk density and bending modulus with the increase in fabric mass. The tensile properties increase with the increase in fabric mass up to 900 g/m\(^2\) and beyond that it decreases. The increase in the properties with the increase in g/m\(^2\) is mainly due to more entanglement and consolidation in structure.\(^8\) The decrease in tensile properties at high g/m\(^2\) is due to the breakage of jute fibre.\(^3\) Woven fabric (W) shows lower density due to the space between the yarns, higher strength, lower elongation and lower bending modulus compared to nonwoven fabric. From all these nonwovens, jute-reinforced plastic was made with polyester resin.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Bulk density g/cc</th>
<th>Tenacity cN/tex</th>
<th>Breaking strain</th>
<th>Bending modulus N/cm(^2)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>0.109</td>
<td>0.283</td>
<td>0.38</td>
<td>1.345</td>
</tr>
<tr>
<td>B</td>
<td>0.115</td>
<td>0.329</td>
<td>0.42</td>
<td>1.380</td>
</tr>
<tr>
<td>C</td>
<td>0.104</td>
<td>0.254</td>
<td>0.45</td>
<td>1.217</td>
</tr>
<tr>
<td>D</td>
<td>0.112</td>
<td>0.315</td>
<td>0.36</td>
<td>1.381</td>
</tr>
<tr>
<td>E</td>
<td>0.117</td>
<td>0.320</td>
<td>0.26</td>
<td>1.445</td>
</tr>
<tr>
<td>F</td>
<td>0.120</td>
<td>0.460</td>
<td>0.28</td>
<td>1.437</td>
</tr>
<tr>
<td>G</td>
<td>0.126</td>
<td>0.393</td>
<td>0.25</td>
<td>1.432</td>
</tr>
<tr>
<td>H</td>
<td>0.106</td>
<td>0.267</td>
<td>0.37</td>
<td>1.297</td>
</tr>
<tr>
<td>I</td>
<td>0.114</td>
<td>0.352</td>
<td>0.41</td>
<td>1.385</td>
</tr>
<tr>
<td>J</td>
<td>0.119</td>
<td>0.370</td>
<td>0.35</td>
<td>1.447</td>
</tr>
<tr>
<td>K</td>
<td>0.124</td>
<td>0.337</td>
<td>0.31</td>
<td>1.492</td>
</tr>
<tr>
<td>W</td>
<td>0.097</td>
<td>6.53</td>
<td>0.06</td>
<td>0.089</td>
</tr>
</tbody>
</table>

3.1.1 Comparison between Jute-Reinforced Plastics made from Woven Jute Fabric and Needle-Punched Nonwoven Jute Fabric

In woven fabric, two sets of compact and twisted threads are interlaced in the regular fashion, whereas in nonwoven fabric jute fibres are arranged in a particular order and then entangled by penetration of...
barbed needle. Air space in woven fabrics is higher than in nonwoven fabrics and their air space distribution is also different.

Figure 1a shows resin uptake of different jute reinforcing materials. The resin uptake indicates the percentage of resin absorbed (weight basis) by the reinforcing mat. The polyester resin is applied uniformly on the reinforcing material, so that it becomes fully wet. Then immediately it is squeezed under a fixed pressure. Higher resin uptake means better wettability as well as the ability to hold the resin inside the mat.

Figures 1b-d show the comparison between the composites made from woven and nonwoven jute fabrics. Both the composites contain about 1000 g/m² jute component, i.e. four folds of woven fabric and two folds of nonwoven fabric were used to make the composites. The resin uptake in nonwoven fabric is higher than that of woven fabric due to bulky and open structure of nonwoven fabric, facilitating better wetting of fibres compared to woven fabric. Composite from nonwoven shows higher tensile strength, flexural strength and impact resistance due to higher wettability and fibre-matrix interface. On the contrary, the yarn structure in the woven fabric is very much compact and it is very difficult to penetrate the resin inside the yarn structure. Hence, in spite of the higher mechanical properties of woven fabric compared to nonwoven fabric (Table 2), the mechanical properties of composite from woven fabric are inferior to that of nonwoven fabric. Compact structure of yarn prevents the wetting of fibres. Thus, the lower fibre-matrix interface is responsible for lower performance of woven fabric.

In the woven fabric, the difference between the composite properties in the warp and weft directions is mainly due to the yarn orientation in the composite caused by yarn crimp and weaker weft of the fabric. It is observed that the mechanical properties of composite from woven fabric in the warp direction are higher than in weft direction.

Due to better properties, needle-punched nonwoven structure is considered for the further studies. It also proves that the better wetting and higher fibre-resin interface play a significant role for better performance of composite.

3.1.2 Effect of Fibre Orientation

Parallel-laid and cross-laid nonwovens are very much common in use and easy to manufacture. These

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Fig. 1—Effect of fabric structure on composite properties [NW — nonwoven, Wp — warp, Wf — weft, Mc — machine direction, and Cr — cross direction]
fabrics are anisotropic in nature with respect to tensile strength. However, on latest needle-punched nonwoven machines, it is possible to have almost same tensile strength in both machine and cross directions. Figures 1a-d show the properties of JRP made from parallel-laid and cross-laid needle punched nonwovens. Machine direction (MD) of parallel-laid nonwoven shows the best mechanical properties in tensile, flexural and impact points of view. On the contrary, the cross direction (CD) of parallel-laid nonwoven shows the most inferior quality. Cross-laid improves mechanical properties in CD with slight deterioration in MD. In the parallel-laid nonwoven, majority of the fibres is oriented in the MD, whereas in the cross-laid nonwoven, majority of fibres is oriented in an angle (~20-30°) with the both sides of CD. These orientations of fibres play the main role in ascertaining the mechanical properties of nonwoven as well as composites. In the unidirectionally laid fibres, the strength in the direction of fibre laying depends on the fibre properties and at right angles to the fibres, the strength depends largely on the strength of bonding between the fibre and the resin. As most of the applications require strength, stiffness and toughness in both cross and machine directions, the cross-laid nonwoven has been chosen for this study.

3.1.3 Effect of Depth of Needle Penetration

According to Figs 2a-d, with the increase in depth of needle penetration, resin uptake increases gradually. Higher depth of needle penetration causes the fibre breakage as well as forms channel through the nonwovens. These channels, fibre reorientation and breakage allow the resin to penetrate inside the nonwoven very easily. Thus, the composite made from the nonwoven with 10 mm depth of needle penetration shows better properties. But with 12 mm depth of needle penetration, the composites show deterioration inspite of the increase in tensile strength. Strength increases due to higher resin content but other properties decrease due to severe jute fibre breakage. It is also reflected in the properties of nonwoven including decrease in bulk density in high depth of needle penetration. Hence, the fibres of nonwoven structure play a significant role in the properties of composite.

3.1.4 Effect of Needling Density

With the increase in needling density up to 250 punches/cm², resin uptake as well as mechanical properties improve (Figs 3a-d), in spite of the increase in packing of fibre assembly. Higher punches/cm² produces higher number of fibre pegs and holes, which may be responsible for better penetration of
resin. Higher fibre orientation and entanglement also improve nonwoven properties. These are responsible for better composite properties. Beyond 250 punches/cm², the packing of fibre mat becomes so high that it hinders to produce good fibre-matrix dispersion. This is also established by downward trend of resin uptake above 250 punches/cm². The nonwoven properties also decrease, may be due to high fibre breakage at high needling density. Hence, 200-250 punches/cm² may be considered as the optimum needling density.

3.1.5 Effect of Mass Per Unit Area
Figures 4a-d show that the mechanical properties in terms of tenacity, flexibility and impact strength of composite increase with the increase in mass/unit area of jute nonwoven reinforcement from 300 g/m² to 700 g/m². Beyond 700 g/m², these properties deteriorate. As the mass/unit area increases, the needling makes the fabric more compact, resulting in better peg formation and fibre reorientation/entanglement. It improves the wetting property and resin holding power as resin uptake increases, in spite of increase in packing of fibre assembly. But beyond 700 g/m², the packing is so high that it plays the predominant role and the resin uptake decreases, because resin cannot penetrate easily into the fibre assembly, which, in turn, affects the properties of composite. Moreover, at 1100 g/m², the nonwoven properties also deteriorate in spite of increase in bulk density, possibly due to excessive breakage of jute fibre. This may also reduce the mechanical properties of composite. Therefore, 700 g/m² is the optimum mass/unit area as far the mechanical properties of the composite are concerned, keeping all other parameters unchanged.

3.2 Surface Modification of Jute Nonwoven
Jute fibre needs some chemical modification to improve its compatibility with polyester resin. Several chemical treatments have been tried on jute, which basically remove the impurities in it and a small part of its chemical constituents, so that the fibre becomes flexible and more reactive with resin matrix. It is the cellulose ultimate, where the bonding must take place if an efficient composite is to be manufactured. Hence, the proper pretreatment of jute fibre may clean the fibre and break different linkages, releasing lignin, exposing cellulose ultimate and resulting in better and more stable bonds.
Modification of fibre has been done at nonwoven stage. Considering practical utility and ease of operation, following treatments have been identified. Each treatment has been optimized separately. The needle-punched nonwoven fabric with optimized physical parameters (700 g/m², 250 punches/cm² and 10 mm depth of needle penetration) has been considered as control fabric for chemical modification. The chemical treatments of fabric, such as hydrogen peroxide bleaching, alkali treatment and enzyme treatment, have been carried out.

3.2.1 Effect of Bleaching

To expose the reactive groups of jute fibre, jute nonwoven has been treated with oxidizing bleaching agent. Composites have been prepared from control and bleached nonwoven and their properties evaluated. Resin uptake increases considerably for the composite made from bleached nonwoven. The resin uptake is 150% in case of control, whereas it is about 300% in case of bleached nonwoven. From Table 3 it is clear that the increased resin uptake for bleached nonwoven results in increased tensile and flexural properties. Hence, the modification of jute fibre by bleaching method leads to jute fibre-reinforced plastic product with improved mechanical properties.

Table 3—Effect of surface modification of jute nonwoven on mechanical properties of JRP

<table>
<thead>
<tr>
<th>Sample</th>
<th>Tensile stress (MPa)</th>
<th>Tensile modulus (GPa)</th>
<th>Flexural stress (MPa)</th>
<th>Flexural modulus (GPa)</th>
<th>Impact strength (J/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Control</td>
<td>57.18</td>
<td>2.29</td>
<td>63.18</td>
<td>1.85</td>
<td>151.85</td>
</tr>
<tr>
<td>9% alkali treated</td>
<td>65.85</td>
<td>2.44</td>
<td>64.15</td>
<td>1.93</td>
<td>158.42</td>
</tr>
<tr>
<td>1% alkali + enzyme treated</td>
<td>60.14</td>
<td>2.49</td>
<td>77.02</td>
<td>1.96</td>
<td>169.05</td>
</tr>
<tr>
<td>Bleached</td>
<td>65.86</td>
<td>2.53</td>
<td>80.36</td>
<td>2.01</td>
<td>170.98</td>
</tr>
</tbody>
</table>

3.2.2 Alkali Treatment

Alkali treatment has been given to jute nonwoven mat, used to prepare the composite. Alkali (9%) treatment for 2h at room temperature gives the optimum effect.11 Tensile, flexural and impact strengths of the alkali-treated sample are found to be much higher than the control sample (Table 3). Removal of lignin-carbohydrate complex makes the structure loose, while xylan removal makes the fibre porous, thereby making a room for the large resin molecules to enter the matrix for crosslinking. Actually, alkali mediated pretreatment process helps
to increase the binding of resin with jute fibre. So, the pretreatment of jute fibre with alkali improves the jute fibre-reinforced plastic properties.

3.2.3 Enzyme Treatment

A mixed commercial enzyme (cellulase, xylanase and pectinase) preparation Bio-cellulase ZK (4% owm) was used for bio-treatment of 1% alkali-treated jute nonwoven mat. It is found that 1% alkali-treated jute nonwoven produces better composite with high tensile and flexural properties. The smooth fibres produced after enzyme treatment have high porosity (improved by 30%), make easy passage of resin to enter the fibre and perform crosslinking, resulting in better jute fibre-reinforced plastic products. The results of analysis are shown in Table 3. Improved crystallinity, softness, fineness and pliability of enzyme-treated fibre may be responsible for better flexural property of composite.

4 Conclusions

4.1 Jute needle-punched nonwoven produces better composites than jute woven fabric. Better wetting and higher fibre-resin interface play a significant role for better performance of composite.

4.2 In case of needle-punched nonwoven, fibre laying plays an important role in the composite properties. The cross-laid nonwoven produces better composite, considering its mechanical properties in the machine and cross directions compared to parallel-laid nonwoven.

4.3 Nonwoven with 10 mm depth of needle penetration, 250 punches/cm² punch density and 700 g/m² area density produces composite with better properties.

4.4 Modification of jute nonwoven by hydrogen peroxide bleaching process shows optimum properties of jute reinforced composites.

4.5 The removal of surface lignin exposes cellulose and helps to produce better fibre resin matrix interface. Alkali treatment removes surface lignin and improves porosity. Composites made from jute nonwoven modified by 9% alkali treatment are found to have improved tensile and flexural properties.

4.6 Enzyme treatment on 1% alkali treated jute nonwoven produces better composite with high tensile and flexural properties.

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