Flax Yarn Polymer Matrix Composites:
Effects of Yarn Structure and Prestressing

By

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Dedications

I would like to dedicate this work to my beloved parents (Ghulam Muhammad, and Kalsoom Fatima) whose love and prayers encouraged me to pursue my higher studies and to my wife Humaira for endless love and support.
Abstract

This thesis is a study of the effects of flax yarn structural parameters and prestressing applied during the curing process on the mechanical properties of parallel-laid yarn composites with unsaturated polyester matrix. Three types of flax yarn structures were used as reinforcement in the unidirectional composites: singles, two-ply and wrap spun yarns.

The mechanical properties of the singles yarn reinforced composites were influenced by the amount of twist in the constituent flax yarn and the level of prestressing applied to the preform during resin curing. Composites prepared from low twist yarn (i.e. 3.8 Twist Multiplier or TM) showed higher mechanical properties than those prepared from medium and high twist yarns (i.e. 4.8 and 6 TM, respectively). The optimum level of prestressing (of up to 3% strain) varied according to the level of twist in the constituent yarn.

The study related to plied yarn based composites showed that with the change of ply/singles yarn twist ratio, the overall fibre orientation in relationship with the plied yarn axis changes. The ply/singles twist ratio can be tuned to achieve optimum alignment with the plied yarn axis, which in turn maximise the mechanical performance of the corresponding composites. The plied yarn based composites performed the best when the ply/singles twist ratio was between 30% to 35%, which matched well with theoretical prediction. Contrary to the singles yarn, prestressing of the plied yarns (up to 3.4% strain) had a detrimental effect on the mechanical properties of the resulting composites.
The study on wrap spun yarn based composites has shown that the filament wrapping density (wrap twist) and the presence or absence of false twist during spinning have significant effects on the mechanical properties of the corresponding composites. The composites prepared from wrap yarns spun with no false twist (NFT yarns) showed better mechanical properties than those prepared from yarns spun with false twist because of lower tortuosity and better fibre alignment. Prestressing further improved the mechanical properties of the resulting composites.

The research demonstrated that the mechanical properties of the composites prepared from the wrap spun yarns composites possessed the highest mechanical properties (~20 GPa and ~250 MPa flexural modulus and strength respectively), which is comparable to glass fibre reinforced polyester composites when the lower density of the flax fibre is taken into consideration. From a manufacturing point of view, the production cost of coarse count wrap spun yarns is lower than ring spun yarns because high productivity can be achieved and therefore this type of yarn structure have high potential for the manufacture of ecologically sustainable bio-composites for many structural applications.
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1. Introduction

The study presented in this thesis focusses on the structure-property relationship of flax yarn reinforced composites. This chapter provides a brief introduction to the research field, the general aims and scopes of this research work, and a summary of the contents of the thesis.

1.1 Introduction to Composite Materials

Plant fibres have been used as reinforcements in composites for thousands of years. The Egyptians used straw to strengthen mud bricks for wall construction [1], whilst the Mayan civilizations used plant fibres to strengthen bricks and pottery [2]. Over the past 50 years, however, synthetic fibres especially glass fibres and carbon fibres, have been the dominant reinforcement in the composites market [3].

Synthetic fibres originate from non-renewable (mostly petroleum based) resources, which are increasingly limited in supply. These fibres have disadvantages such as difficulties in recycling [4], higher greenhouse emissions and negative environmental impacts. Because of these drawbacks, composites based on natural fibres have attracted the attention of many researchers in recent years. There are an increasing number of review articles [5-13], and books [1, 14-18] on biofibre based composites in recent years.

Bast fibres are fibres obtained from the outer cell layers of the stems of plants [19]. These fibres, such as flax, hemp, sisal and kenaf, have many technical, economic and ecological benefits compared to synthetic fibres (Table 1-1). For
example, they are widely available and environmentally friendly, low cost and possess low specific gravity (1.38~1.5) and high specific properties [20-22]. As a result, these fibres are being used as substitutes to synthetic fibres for composites applications [5, 23-25].

Plant fibre reinforced composites (PFCs) have found applications in non-structural automotive composite parts, such as door components and instrument panels [14, 26-28]. They are also extensively used in application areas such as construction and infrastructure [14, 28-31] (beams, roof panels and bridges), sports and leisure (canoes, bicycle frames, tennis rackets) [14, 28, 30], furniture and consumer articles like chairs, tables, packaging [14, 26, 28, 30, 32, 33], pipes and tanks (water drainage, transportation) [11, 14, 28, 31, 33] and small scale wind energy generation (rotor blade material) [34, 35].

Flax is the most widely utilized bast fibres. It has low density, high specific mechanical properties comparable to glass fibres. It is also reported that flax fibre composites has vibration absorbing [36] and ultraviolet rays blocking properties. It is more suitable for composites designed for structural applications than other bast fibres. Due to these characteristics, flax fibres have been selected for use in the present study.
Table 1-1 Comparison between plant and synthetic fibres [37].

<table>
<thead>
<tr>
<th>Properties</th>
<th>Plant fibres</th>
<th>E-glass fibre</th>
<th>Carbon fibre</th>
</tr>
</thead>
<tbody>
<tr>
<td><strong>Economical</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Annual global production [tonnes]</td>
<td>31,000,000</td>
<td>4,000,000</td>
<td>55,000</td>
</tr>
<tr>
<td>Distribution for composites [tonnes]</td>
<td>Moderate (40,000)</td>
<td>Wide (600,000)</td>
<td>Moderate (35,000)</td>
</tr>
<tr>
<td>Cost [$/kg]</td>
<td>Low (~0.8–2.5)</td>
<td>Low (2)</td>
<td>High (19.0)</td>
</tr>
<tr>
<td><strong>Technical</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Density [g cm$^{-3}$]</td>
<td>Low (~1.40)</td>
<td>High (2.55)</td>
<td>Low (1.75)</td>
</tr>
<tr>
<td>Tensile strength [GPa]</td>
<td>Low (~0.4–1.5)</td>
<td>Moderate (2.0–3.5)</td>
<td>High (4.0–5.5)</td>
</tr>
<tr>
<td>Failure strain [%]</td>
<td>Low (~1.4–3.2)</td>
<td>Low (2.5)</td>
<td>Low (1.5)</td>
</tr>
<tr>
<td>Specific stiffness [GPa/g cm$^{-3}$]</td>
<td>Moderate (~20–60)</td>
<td>Low (29)</td>
<td>Very high (134)</td>
</tr>
<tr>
<td>Specific strength [GPa/g cm$^{-3}$]</td>
<td>Moderate (1.1)</td>
<td>Moderate (1.3)</td>
<td>High (2.7)</td>
</tr>
<tr>
<td><strong>Ecological</strong></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Energy consumption [MJ/kg of fibre]</td>
<td>Low (4–15)</td>
<td>High (30–50)</td>
<td>Very high (130)</td>
</tr>
<tr>
<td>CO$_2$ emissions [kg/kg of fibre]</td>
<td>None</td>
<td>Moderate</td>
<td>Very high</td>
</tr>
<tr>
<td>Renewable source</td>
<td>Yes</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Recyclable</td>
<td>Yes</td>
<td>Partly</td>
<td>No</td>
</tr>
<tr>
<td>Biodegradable</td>
<td>Yes</td>
<td>No</td>
<td>No</td>
</tr>
<tr>
<td>Abrasive to machinery</td>
<td>No</td>
<td>Yes</td>
<td>Yes</td>
</tr>
<tr>
<td>Toxic</td>
<td>No</td>
<td>Yes</td>
<td>Yes</td>
</tr>
</tbody>
</table>
Presently, both thermosets and thermoplastics are used in plant fibre composites [38]. However, there is a decreasing trend for the use of thermoset matrix materials especially in the automotive industry, increasing applications in the use of thermoplastic materials. This is due to the requirements of faster and cleaner processing with no toxic by-products, easy recycling and low cost of materials [26, 38, 39]. However, thermosets are high performance matrix materials because of the formation of large, cross-linked three-dimensional molecular structures after curing. They form better interfaces with hydrophilic plant fibres and can be processed at relatively low temperature when they are at their low viscous state, as compared to thermoplastics. Therefore a thermoset matrix was chosen to be used in the present study.

In composites industry, plant fibres are mostly used as random discontinuous form (non-woven mats) [12]. However, the resulting composites are not suitable for structural applications due to the low mechanical properties caused by discontinuity, short length (< 3-30 mm) [12, 14, 40] and random orientation of the natural fibres.

To be able to use plant fibres in structural applications, the reinforcing fibres must be aligned because they have high length and orientation efficiency factors, as dictated by the Krenchel fibre orientation efficiency equation [41]. Unlike manmade (or synthetic) fibres, which normally consist of continuous strands, natural fibres often consist of staple fibres having a short discrete length. In order to produce an aligned yarn structure, natural fibres can be spun into yarns using conventional textile spinning methods.
The conventional yarn production method for staple plant fibres is the twisting method and the resulting yarn has fibres aligned at a specific inclination angle to the yarn axis. The inclination of the twisted fibres results in poor alignment, which is detrimental to the resulting twisted yarn reinforced composites [42]. To improve the fibre alignment, a plying technique is employed in which two strands of yarn are put together and twisted in a direction opposite to that of the original single yarns. This results in improvement of fibres orientation to the yarn axis without any negative effect on the yarn strength [43].

Ideally, yarn in structural composites should be twistless so that all fibres are perfectly aligned to the yarn axis. Zhang L et al. [44] have demonstrated such a twistless yarn structure produced on a hollow spindle wrap spinning machine. In practice, fibres in the twistless yarn are not perfectly aligned because the yarn body follows a somewhat tortuous path due to the tension and torque balance between the flax fibre body and the fine wrapping filament [45]. Weaving can cause further fibre misalignment, due to the presence of yarn crimps in the resulting fabrics. Miao M et al. proposed a modified nonwoven process to produce highly aligned natural fibre tapes (or mats) to avoid the expensive spinning and weaving process for the twistless yarn approach [46]. By comparing the mechanical performance of composites from the highly aligned nonwoven tape to that from the twistless yarn, they hypothesized that the residual fibre stress from the spinning and weaving processes might have a beneficial effect on the mechanical performance of the final composites.

Residual stresses in a composite are generated during processing and in-service due to the processing stress, the anisotropic thermal expansion and the
elastic properties of their constituents. During composite processing at elevated temperatures, residual stresses can develop due to (1) cure-induced shrinkage of polymer matrix resin during the polymerization reaction, which forms cross-linked networks [47, 48] and (2) result in volume changes due to the mismatch of the thermal expansion of the constituent fibre and matrix materials. The presence of residual stresses may result in dimensional instability of composite parts and can even cause cracking within the matrix if they are too high. Of various techniques to minimise the process-induced residual stresses, fibre pre-stressing has been demonstrated successfully theoretically and experimentally with glass and carbon fibre composites during the last decade [49-51].

Prestressing techniques have traditionally been used in civil engineering to enhance the mechanical properties of precast reinforced concrete using tensioned tendons (usually steel bars). When the pretension applied to the tendons is released after the concrete is cured, it is transferred to the concrete as compression. Mechanical properties of the polymer-matrix composites have also been shown to improve [52-54] using prestressing applied during composite fabrication.

The first objective of this research is to investigate how the mechanical properties of unidirectional (UD) plant fibre composites (PFCs) are affected by different yarn structures and parameters for a particular yarn structure. The second objective is to study the changes in different properties of the PFCs by applying pre-stressing to the natural fibre spun yarns during the resin curing process. The natural fibre spun yarns have a complex twisted structure and are viscoelastic. The yarn structure and stress conditions undergo changes while a tensile load is applied and maintained over a period of time during curing.
1.2 Project Aims

The work aimed to identify ways to enhance the mechanical properties of the plant fibre based composites for use in structural applications. Flax fibre was chosen as a reinforcement for this study since it has been used widely in natural fibre based composites in various application fields.

Because plant fibres have a short length, they were spun together using various textile spinning technologies to produce yarns with varying structural parameters for use in composites fabrication. Three different yarn structures, namely single yarns, plied yarns and wrap spun yarns were used to produce unidirectional composites with an unsaturated polyester resin. The effect of pre-stressing during fabrication on the properties of the corresponding composites was studied. The properties of fibres, yarns and composites were inter-correlated to establish structure-property relationships.

1.3 Thesis Outline

The work presented in this thesis is composed of seven chapters. Chapter 2 reviews the literature in the field of natural fibre composites, bio-composites and pre-stressing in composites. Details of the materials and experimental methods used to prepare and characterise the materials used in this study are described in Chapter 3. The effect of yarn structures, namely singles, plied and wrap spun, on the properties of the resulting composites are presented in Chapter 4, 5 and 6, respectively. The effect of pre-stressing will be systematically studied throughout all chapters. Finally, Chapter 7 summarises the findings of this study, providing conclusions to the work and outlining paths for future developments in the area.
2 Literature Review

The aim of this chapter is to provide a broad understanding of the plant fibre structures and their composites, focussing on different yarn structures and methods to enhance mechanical properties of the resulting composites. It starts with a general introduction of composite materials and their classification followed by structure-property relationships in plant fibre composites. Different yarn structures used for composites manufacturing are discussed in the second part of the chapter. The last part is related to the concept of prestressing, including theories and experimental work on prestressed composites.

2.1 Composites and Their Structure

A composite material is a macroscopic combination of two or more distinct materials, having an identifiable interface between them [55]. Modern composite materials are usually designed to obtain a particular balance of properties for a given range of applications. The two constituent components of the composite are matrix and reinforcement. The matrix component binds together and provides continuous form to an array of a stronger and stiffer reinforcement constituent. The resulting composite material has a balance of structural properties that are superior to either constituent material alone.

2.1.1 Classification of Composites

Composite materials are classified at two different levels (Figure 2-1). The first type of classification is with respect to the matrix constituent. The major classes include ceramic-matrix composites (CMCs), metal-matrix composites
(MMCs) and organic-matrix composites (OMCs). The OMCs include two further types which are polymer-matrix composites (PMCs) and carbon-matrix composites (also called carbon-carbon composites) [56].

Figure 2-1 Classification of composite materials.

The second type of classification is based on the reinforcement form. The reinforcement can be particles, whiskers, continuous fibres and woven fibres (the braided and knitted fibre reinforcement included in this category) as shown in Figure 2-2.

Figure 2-2 Common forms of reinforcements.
2.1.2 Polymer-Matrix Composites

These are the composites in which the matrix is a polymeric material. The matrix polymer can be of two types namely thermoset and thermoplastic polymers [57]. There has been a growing trend of using thermoplastic matrix particularly in automobile industry [26, 39] due to a number of benefits. The thermoplastic composites have unlimited shelf life, fewer health risks to chemicals during processing, easier to recycle and are less expensive for high volume processing. But they do have some disadvantages as compared to thermoset-matrix composites like high processing temperature, high viscosities of the involved materials, and limited processing methods. On the other hand, thermoset composites are high performance materials and form a better interface with the hydrophobic plant fibres, need low processing temperatures, and can be used in a variety of processes for composites manufacture such as filament winding and pultrusion.

2.1.3 Plant Fibres as Reinforcement in Composite Materials

It has been stated in chapter 1 that plant fibres (lignocellulosic fibres) have many technical, economic and ecological benefits as compared to synthetic fibres especially E-glass (Table 1-1). Table 2-1 shows the comparison of mechanical properties of different plant fibres with those of glass fibre. In recent years there is increased awareness to replace glass fibre composites (GFRPs) with plant fibre composites (PFCs) which dominate today’s FRP markets [39]. The market for PFCs in commercial applications has been increasing annually over the past 15 years but these are mostly aimed to be used for non-structural automotive components mainly as a replacement to wood fibre composites [38, 58]. The plant fibres have drawbacks such as inferior and naturally variable mechanical properties, high
moisture absorption (resulting in poor mechanical properties) and weak fibre/matrix interface (which hinders load transfer from the fibres to the composites). Due to these drawbacks the plant fibres cannot be used for structural applications [5].

With a particular plant fibre, the composites prepared from short random fibres have inferior mechanical properties than those prepared from aligned long fibre reinforced (yarns, fabrics etc.) composites [46]. If the plant fibres are to be used for structural load bearing applications, the short plant fibres, technically known as staple fibres must be processed to continuous yarn structures so as to fully utilize the mechanical performance of the plant fibres. Yarn structures normally used for composites are singles yarn, plied yarns (normally 2 ply) and wrap spun yarns.
Table 2-1 Comparison of mechanical properties of different plant fibres and E-glass. Sources include those quoted and [11, 23].

<table>
<thead>
<tr>
<th>Fibre</th>
<th>Density (g/cm³)</th>
<th>Tensile modulus (GPa)</th>
<th>Specific tensile modulus (GPa/g/cm³)</th>
<th>Tensile strength (MPa)</th>
<th>Specific tensile modulus (MPa/g/cm³)</th>
<th>Failure strain (%)</th>
<th>Source</th>
</tr>
</thead>
<tbody>
<tr>
<td>Bast</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Flax</td>
<td>1.45-1.55</td>
<td>28-100</td>
<td>19-65</td>
<td>343-1035</td>
<td>237-668</td>
<td>2.7-3.2</td>
<td>[59]</td>
</tr>
<tr>
<td>Hemp</td>
<td>1.45-1.55</td>
<td>32-60</td>
<td>22-39</td>
<td>310-900</td>
<td>214-581</td>
<td>1.3-2.1</td>
<td>[8]</td>
</tr>
<tr>
<td>Jute</td>
<td>1.35-1.45</td>
<td>25-55</td>
<td>19-38</td>
<td>393-773</td>
<td>291-533</td>
<td>1.4-3.1</td>
<td>[59]</td>
</tr>
<tr>
<td>Leaf</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Pineapple</td>
<td>1.44-1.56</td>
<td>6-42</td>
<td>4-27</td>
<td>170-727</td>
<td>118-466</td>
<td>0.8-1.6</td>
<td>[60, 61]</td>
</tr>
<tr>
<td>Banana</td>
<td>1.30-1.35</td>
<td>8-32</td>
<td>6-24</td>
<td>503-790</td>
<td>387-585</td>
<td>3.0-10</td>
<td>[60]</td>
</tr>
<tr>
<td>Other</td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>Bamboo</td>
<td>0.60-1.10</td>
<td>11-30</td>
<td>18-27</td>
<td>140-230</td>
<td>210-233</td>
<td>1.3</td>
<td>[12]</td>
</tr>
<tr>
<td>E-glass</td>
<td>2.55</td>
<td>78.5</td>
<td>31</td>
<td>1956</td>
<td>767</td>
<td>2.5</td>
<td>[62]</td>
</tr>
</tbody>
</table>
2.1.4 Structure Property Relationship in Composites

Composites are heterogeneous materials in which reinforcing fibres are embedded in a continuous matrix. The fibres provide strength and stiffness to the composites while the matrix component transmits the externally applied loads to the reinforcing fibres via shear stress at the fibre/matrix interface, hence protecting the fibres from external damage. The mechanical properties of the composites are dependent on the intrinsic properties of the constituents (fibre and matrix) and their volumetric composition in the composites. For composites with unidirectional fibre orientation and continuous fibres, these properties are modelled by the rule of mixtures (ROM) Equation 2-1 [63].

\[ E_c = V_f E_f + V_m E_m \]

where \( E \) is the stiffness, \( V \) is the volume fraction, and subscripts \( c, f \) and \( m \) indicate composite, fibres and matrix respectively. This model has been re-adapted for composites comprising discontinuous fibres with non-unidirectional orientation, and is called the combined ROM model [64-66], Equation 2-2.

\[ E_c = \eta_f \eta_l V_f E_f + V_m E_m \]

where \( \eta_f \) is the fibre orientation efficiency factor which incorporates the effect of fibre packing arrangement and orientation in the composite, and \( \eta_l \) is the fibre length efficiency factor.

The ROM and combined ROM models are based on certain assumptions which include: (i) all fibres have identical geometry and properties, (ii) the fibres are distributed homogenously and uniformly in the matrix, (iii) there are iso-strain conditions within the composite, (iv) the fibre and matrix have elastic deformation
and (v) no porosity content $v_p$ in the composites. Although many of these assumptions are not true for FRCs in general, the ROM model has been widely applied for the prediction of properties of synthetic fibre composites.

The effect of porosity on the stiffness of plant fibre composites has been demonstrated by including the factor $(1-V_p)^2$ in the rule of mixtures model in modified ROM [67-72].

$$E_C = (\eta_f \eta_l E_f, V_f + V_m E_m)(1-V_p)^2$$

Equation 2-3

In Equation 2-3, the length efficiency factor for modulus $\eta_l$ can be estimated using Cox shear lag model [73] using Equation 2-4.

$$\eta_l = 1 - \frac{\tanh(\beta L)}{2}, \beta L = 2L \sqrt{\frac{D}{G_m}} \left( \frac{G_m}{E_f \ln(\frac{k}{V_f})} \right), \eta_l \in [0,1]$$

Equation 2-4

where $L$ is the fibre length, $D$ is fibre diameter, $G_m$ is shear stiffness of the matrix and $k$ is a constant to address the difference between true noncircular and variable cross sectional area of the fibre and apparent cross sectional area calculated by the measurement of apparent fibre diameter [7, 70, 71]

The fibre orientation distribution factor $\eta_o$ in Equation 2-2 and Equation 2-3 can be estimated using Krenchel orientation distribution factor [41] which is given by Equation 2-5, where $a_n$ is the fraction of fibres with orientation angle $\theta_n$ with respect to loading axis. The reinforcement orientation distribution factor ranges from 0 (fibres aligned transverse to loading direction) to 1 (all fibres aligned parallel to the loading direction).
\[ \eta_o = \sum a_n \cos^4 \theta_n \] \hspace{1.5cm} \text{Equation 2-5}

From Equation 2-2, Equation 2-3 and Equation 2-5 it can be seen that fibre orientation (angle) in the composites along the loading direction is a determining factor (while keeping all other factors unchanged) to enhance the mechanical properties of the PFCs. The fibre orientation and arrangement is different in different yarns and the structural parameters like twist can be different too, even within the same yarn. Therefore the mechanical properties of the resulting composites can be modified by changing the yarn structure as well as structural parameters for a certain type of yarn used for preparation of composites.

### 2.1.5 Fibre Orientation in Composites

The fibre orientation has a significant effect on the mechanical properties of the composites because of the anisotropic nature of the fibres. The fibre anisotropy results from natural structure of the fibres (in cellulose based fibres) [74] or from large aspect ratio along fibre axis as compared to cross-sectioned aspect ratio [75].

The fibre orientation distribution in the composites is dictated by the manufacturing technique. In a three dimensional random fibre orientation, the fibre orientation factor has a value of \(1/5 = 0.20\) [40].

In injection moulded PFCs, fibres are distributed in 3D random order, but they normally show a preferred orientation as found in study by Serrano et al. [76]. Garkhail et al. [40] and Bos et al. [77] found \(\eta_o\) to be 0.375 and 0.21 in their studies related to short flax fibre reinforced PP composites using injection moulding process. Serrano et al. [76] and Vallejos et al. [78] calculated the \(\eta_o\) to be 0.346 and 0.286 respectively for old-newspaper (ONP) fibre reinforced PP and hemp-PP
injection moulded compounded composites. Conventional nonwoven plant fibre composites have two dimensional random fibre orientation but they may show a preferred orientation. Bos et al. [77] have found values of 0.39–0.41 for nonwoven flax fibre reinforced composites. The orientation distribution factor for composites prepared from multiaxial fabrics depends on the ply orientation and therefore have a range of $\eta_o$ values. The orientation distribution factor has a value of 0.5 and 0.25 for composites prepared from balanced biaxial reinforcements in a [0,90] and [±45] stacking sequences, respectively [79]. To enhance the orientation distribution factor $\eta_o$ close to unity (maximum orientation possible), unidirectional fibre structures are required.

2.1.6 Fibre and Fibrous Structures

Fibres have been defined by the Textile Institute [19] as units of matter characterized by flexibility, fineness and a high ratio of length to thickness. If the fibre is to be used for textile purposes, then it should have sufficiently high temperature stability and a certain minimum strength and moderate extensibility. To apply the fibres to useful applications, they must be converted to an assembly which can easily be handled and have sufficient strength to endure various types of stresses during the manufacturing process. Fibres are of two different types with respect to their length, i.e. staple fibres (having short discrete length) and continuous fibres. All natural fibres including plant fibres are staple fibres with the exception of silk which is continuous filament fibre. Manmade fibres (regenerated and synthetics) are normally extruded into continuous lengths and are considered as continuous filament fibres, which later on are cut into short discrete length to use as staple fibre for textile processing.
2.1.7 Yarn Structure, Types and Production

Lignocellulosic fibres have been used as reinforcement in aligned composites due to low density and high specific properties [42, 80, 81]. However, due to short length of technical plant fibres, the reinforcement requires to be in the form of staple fibre yarns to be used for the preparation of aligned unidirectional plant fibre composites [44, 80-86].

Yarns are normally of two types. *Staple-spun* yarn and *continuous filament* (CF) yarn. Staple spun yarn is defined by the Textile Institute as “a linear assembly of many fibres in the cross-section and along the length, held together usually by the insertion of *twist*, to form a continuous strand, small in diameter but of any specified length. It is used for interlacing in processes such as knitting, weaving and sewing” [19]. The fibre can be either natural (like wool, cotton, flax, jute etc.) or manmade or synthetic fibres cut in varying short lengths (like polyester staple, nylon staple, cellulose acetate staple fibre etc.) at the manufacturing plant for blending with natural fibres like polyester/cotton blended fabric, wool/polyester blended fabric.

Staple yarn is normally produced using ring spinning process. In this process a parallel strand of fibres called *sliver* is passed through a series of rollers rotating at increasing surface speed, so that the strand is *drafted* to the required linear density. Twist is imparted to the fibres in the last stage of the spinning process to impart inter-fibre friction to enable the yarn to be used for further processing like fabric formation. The level of twist imparted to the yarn affects the stress transfer between the fibres within the yarn and this influences both (i) the yarn strength and (ii) fracture mechanism of the yarn (Figure 2-3). With the increase of twist, the fibre to fibre friction generated by the coaxial fibre helices increases up to a certain level,
shown in left side of the curve in Figure 2-3. In this region the yarn failure occurs due to fibre slippage. After achieving a maximum level of strength, further twist imparted to the fibres results in increased fibre obliquity (fibre inclination angle) to the yarn axis and high fibre-to-fibre friction. The yarn failure in this region occurs due to fibre breakage [43]. In the textile industry, twist is described by three parameters i.e. (i) twist direction (S or Z), (ii) twist level, $T$ (twist/m or twist/inch) and (iii) twist multiplier, which are related by the equation $TM = T \sqrt{tex}$ which is further explained in section 2.1.7.1.

Figure 2-3 The effect of twist on yarn strength and failure mechanism [87].

Folded yarn (also called plied yarn prepared by twisting two or more single yarns) and wrap spun yarn are also composed of short staple fibres but have different geometry than that of staple spun yarn [19].

*Filament yarn* is composed of unbroken length of continuous filaments which include natural (silk) filaments and filaments extruded from synthetic polymers like polyester, nylon, polypropylene, Kevlar etc. These filaments are slightly twisted to form a yarn structure suitable for processing at later stages.
Magnified photographs of twisted filament yarn and staple-spun yarn are shown in Figure 2-4.

Figure 2-4 Scanning electron micrograph of a polyester continuous filament yarn (above) and a ring spun yarn (below). Longitudinal and cross section images are positioned on the left and right, respectively [88].

2.1.7.1 Twist

Twist is the helical arrangement of fibres constituting the yarn. The helical arrangement (twist) of fibers imparts frictional contact between the fibres and imparts strength to the yarn. If the fibres are too loosely packed so that they can move around in the interstitial space, the yarn appears bulkier and has large diameter than the fibres which are closely packed. The helical arrangement of fibres in the yarns is represented by the simple helix model which is shown in Figure 2-5.
The twist is denoted by the inclination angle $\alpha$ of the surface fibre. The inclination angle is given by the Equation 2-6:

$$\tan \alpha = \frac{2\pi R}{h} \quad \text{Equation 2-6}$$

where $R$ is the radius of surface fibre in the yarn; $\alpha$ is the yarn twist angle; $h$ is the length of one turn of twist.

The twist level (or degree of twist) in yarn is the number of turns of twist per unit length. In Imperial units, it is described by twist per inch (tpi) while in metric system it is described as twist per metre (tpm). The twist angle $\alpha$ has important effects on yarn properties. The twist angle $\alpha$ is related to the twist level shown in Equation 2-7:

$$\tan \alpha = \pi D t \quad \text{Equation 2-7}$$
Twist factor, or twist multiplier, is a parameter used as a means of specifying the level of twist in a yarn which is independent of the yarn linear density and is a measure of hardness or softness of the yarn. It describes the helical angle of fibres inside the yarn. If two yarns are having equal twist factor and have different linear density, then the helical angle \( \alpha \) of the fibres inside the yarn will be the same. The relationship between twist factor, twist and linear density of yarn is given by Equation 2-8.

\[
TF = \text{turns} / \text{meter} \cdot \sqrt{\text{tex}}
\]

Equation 2-8

where \( TF \) is the twist factor; and tex is the linear density of the yarn in g/km.

The amount of twist in the yarn has a direct effect on the strength of the yarn. With the increase of twist multiplier \( TM \), the yarn strength increases to the maximum and the corresponding twist is called the optimum twist \( (t_{op}) \) or twist multiplier \( (TM_{op}) \). This part of the curve is associated with the interfibre frictional resistance to fibre slippage and is called the \textit{coherence region}. Beyond the optimal twist, the strength decreases with twist resulting from a reducing contribution of fibre modulus to yarn modulus as the helix angle becomes more oblique. This latter part of the curve is called \textit{obliquity region}.
2.1.7.2 Fibre Migration and Helix Model of Yarn Structure

The simple helix model depicted in Figure 2-4 is used to describe a yarn structure in which twist is used to bind fibres that are assembled with their lengths straight and parallel before spinning. After being spun into a yarn, the fibres lie along the helix of twist. In such a yarn, the fibres follow a path which varies in length according to the distance from the axis. To achieve this, each fibre would have to be delivered for twisting at a rate proportional to the position where a fibre occupies in the yarn, but this is not practicable.

Practically a self-locking structure is achieved by fibre lengths passing from outer to the inner regions of a yarn, throughout the fibre length, as they are twisted to lie along the helix model. In this way the fibres become interlaced to give cohesion to the spun yarn. This action is called as fibre migration and is defined as:

![Figure 2-6 Effect of twist on yarn strength [88].](image)
“Fibre migration is the cyclic change in the distance of elements of a fibre or filament (along its length) from the axis of a yarn, which occurs during production of the yarn [89].” Figure 2-7 shows cyclic path migration of the fibres moving from cylindrical layer to another.

![Figure 2-7 Tracer fibre showing migration path in conventional ring-spun yarn structure [88].](image)

Fibre migration is mainly caused by the existence of varying fibre tensions at the point where the strand is twisted (Figure 2-8). Fibres at high tension try to relieve their tensions by migrating to the core. Relatively slack fibres are displaced outwards. As fibres are fed to the twisting zone during manufacture, they occupy varying positions along the front roller nip. Fibres at the outer fringes of the strand tend to form the outer ring of the yarn and have to follow the longer helices in Figure 2-4 and therefore become highly tensioned. As a result, these fibres migrate to the core. Vice versa, fibres in the middle of the strand tend to follow the shorter helices to form the yarn core and gradually become slack. The slack fibres are pushed out from the core by the high tensioned fibres. This variation of fibre tension in the yarn has a negative effect on the mechanical properties of the final composite, especially initial modulus. The slack fibres will not contribute effectively to the composite at the initial loading stage.
2.1.8 Plied Yarn and its Properties

Plied yarn is prepared by bringing two or more single yarn strands side by side and twisting them together. The process is known as doubling, plying or twisting. Plied yarns are mostly produced from ring spun yarns because of their specific structure and suitability to plying. Plied yarns produced from ring spun yarns by twisting in the opposite direction to that of original single yarn are less twist lively (i.e., less residual torque), more even and less hairy and can have higher tenacity, breaking extension and abrasion resistance than the component singles yarns [90]. A schematic diagram of the manufacture of plied yarn is shown in Figure 2-9.

Figure 2-9 Down twisting process [88].
2.1.9 Wrap Spun Yarns

Wrap spinning (also called hollow-spindle wrap spinning) is a process in which drafted ribbon of parallel fibres that constitute the bulk of the spun yarn is wrapped by a continuous filament or fine count yarn so as to impart coherence and strength to the resulting yarn [88]. A schematic diagram for preparing wrap spun yarn by hollow spindle technique is shown in Figure 2-10. The hollow-spindle wrap spinning system consists of a roller drafting unit, a hollow spindle with a mounted pirn / bobbin of wrap filament, a pair of delivery rollers and a package winding unit. The spindle has a false-twister unit integrated on the bottom.

![Figure 2-10 Hollow-spindle wrap spinning system [88].](image)

The drafted fibre ribbon emerging from the drafting system passes down the centre of the hollow spindle and is threaded up to be false-twisted by the twisting device. The twist which propagates to the nip of the front drafting rollers, prevents any uncontrolled drafting of the length of the twisted ribbon within the hollow spindle. The filament is also made to pass down the hollow spindle and go around the pin twister. However, since the pirn rotates itself with the spindle, the filament
is not false-twisted. The basic reason for using a false twist is that it provides a certain temporary strength to the yarn core before it is wrapped by the filament.

Wrap spun yarns have some benefits as compared to ring spun yarns like high productivity, high yarn tenacity and uniformity, less hairiness, greater covering ability and possibility of using coarser fibres for finer yarns. A comparison of the structure of conventional twisted yarn, plied yarn and wrap spun yarn is given in Figure 2-11.

This can theoretically give the maximum orientation of natural fibres in the composites (Krenchel orientation factor = 1). In practice, this theoretical maximum cannot be reached because of uneven fibre tension at spinning and yarn tortuosity caused by the pressure from the wrapping filament [45].

![Figure 2-11 Yarn structure](image)

Figure 2-11 Yarn structure (a) single yarn (b) two-ply yarn (c) wrap spun yarn with twist less fibres in the core held by a thin continuous filament [13].
2.2 Prestressing Techniques

The prestressing in composites is taken from the principle of prestressed concrete. The prestressing is the deliberate introduction of stresses during manufacture. In the case of concrete, a prestress force is applied to create permanent internal stresses in a structure to improve its performance. The applied prestress force induces an axial compression which counteracts all, or part of, the tensile stress set up in the member by applied load [91].

There are two categories of fibre prestressing in composites manufacture, (1) prestressing to the fibres is carried out in a separate stage and the prestressed fibres are processed into composites in an unstressed condition, and (2) prestressing is applied and maintained (partially) in the fibre during the curing process of the polymer resin. In the first category, prestressing breaks weak fibres prior to reinforcement and reduces the dynamic effect of weak fibre failure on its adjacent fibres in the final composite [92, 93].

In the second category, in which prestress is applied to the fibres during curing, the applied strain in the composite induced by fibre pretension reduces the amount of thermal residual stresses resulting from the curing process [49]. The matrix compressive stresses also impede crack propagation and reduce composite strain resulting from external tensile and bending loads in typically glass fibre/epoxy composites [52, 54]. For viscoelastic fibres (for example nylon fibres) that are prestressed and immediately made into a polymer matrix composite, the fibres recover from creep as the matrix solidifies and any residual fibre stress left in the final composite is transferred to compression stress in the polymer matrix [94, 95].
Prestressed composites are prepared by applying a tensile preload to the fibre laminate before curing. The uncured polymeric matrix possesses negligible stiffness, so all the applied pre-load is carried by the fibres. During the curing process, the polymeric matrix gels and solidifies. When the composite is cooled down after the cure, the applied preload is removed, and the resulting elastic contraction of fibres relieves the tensile residual stresses in the matrix by inducing compressive stresses which result from curing cycle. The process of fibre prestressing is shown in Figure 2-12.

Figure 2-12 Schematic explanation of fibre prestressing principle. (1) Prestressing the fibres in uncured matrix. (2) Matrix curing and formation of fibre/matrix bonding and development of tensile residual stress in matrix (3) release of fibre pre-stress at ambient temperature which induces compressive stress to the matrix [96].
2.2.1 Developments on Fibre Prestressing in Composites

Effect of fibre prestress on residual stresses and study of failure in continuous fibre composites have been mathematically predicted using micro- and macro-mechanics theory. Here is a summary of prestress theories reported in the literature.

Tuttle [49] has proposed a simple micro-mechanics theory based on the rule of mixtures to predict the effect of fibre prestress on the matrix residual stresses in composites. He made three assumptions in his theory; (1) perfect bonding between fibre and matrix (2) both fibre and matrix are isotropic linear elastic materials (3) fibre and matrix properties are independent of temperature. He predicted the final residual strain in the matrix (\(\varepsilon_m\)) given as:

\[
\varepsilon_m = - \frac{\sigma_f^0 V_f}{E_{ii}} + \Delta T \alpha_{ii}
\]

Equation 2-9

where \(\sigma_f^0\) is the applied prestress, \(V_f\) is the fibre volume fraction, \(E_{ii}\) is the Young’s modulus of composite in fibre direction, \(\alpha_{ii}\) is the thermal expansion coefficient of composite in the fibre direction and \(\Delta T\) is the temperature change.

Tuttle predicted that in a non-prestressed \((\sigma_f^0 = 0 \text{ MPa})\) carbon/epoxy composite with \(V_f = 60\%\), the matrix is subjected to a tensile residual stress of about 24 MPa in the fibre direction which is 60\% of the ultimate tensile strength of epoxy resin. By applying a prestress of 1.4 GPa, he showed that the tensile residual stresses of matrix could be reduced to 0.98 MPa which is 2.5\% of the epoxy ultimate strength.
Hadi and Ashton [50] worked for further development of Tuttle’s model to predict the tensile strength of E-glass reinforced epoxy composites. The tensile strength of a unidirectional composite ($\sigma_c$) is given by the rule-of-mixtures as:

$$\sigma_c = \sigma_f V_f + \sigma_m V_m$$  \hspace{1cm} \text{Equation 2-10}$$

where $\sigma_f$ and $\sigma_m$ are the tensile strengths of fibre and matrix respectively and $V_m$ is the volume fraction of matrix. Hadi and Ashton predicted that the tensile strength of a prestressed unidirectional composite ($\sigma'_c$) is given by:

$$\sigma'_c = \sigma_f V_f + \sigma_m V_m + V_f \sigma'_f$$  \hspace{1cm} \text{Equation 2-11}$$

Results from above model show that tensile strength of UD composites increases with an increase in the fibre prestress. However, Motahhari and Cameron [51, 52] have shown that all prestress applied before matrix curing is not released to the matrix and some part of it remains in the fibre as tensile residual stress.

A theoretical analysis of enhancement of strength in previously prestressed fibres has been done by Manders and Chou [92]. They found that when the peak dynamic stress concentration factor associated with a fibre fracture is greater than the static stress concentration, it is possible to increase the strength of aligned fibre composite by pre-stressing the reinforcing fibre. The analysis shows that optimum level of pre-stress is a function of composite size and is higher for smaller composites. Too high prestress level weakens the composite. Regarding the variability of fibre strength, they found that fibres having less variability in strength were more susceptible to stress concentration and had great potential for strengthening by pre-stressing.
Chi and Chou [93] studied the effect of fibre prestressing on the mean strength of composites and dispersion of composites strength. They compared different properties of non-prestressed and prestressed loose carbon fibre composites with epoxy resin. They found that composite strength for low failure probability is low. They identified the optimum range for prestressing as well which resulted in about 25% of strength increase for 99.9% survivability. Bending of loose strands through a pair of circular bars at room temperature also proved an effective means of prestressing. The prestressing apparatus used is shown in Figure 2-14.

![Figure 2-13 A schematic view of fibre prestressing by bending [93].](image)

Zhao and Cameron [53] have studied the effect of level of prestressing on the reinforcing fibres on mechanical properties of polypropylene matrix glass fibre composites. They found that tensile, flexural and interlaminar shear strength increase with pre-stress up to a certain prestress level and then these properties show a slight decrease before they reach a relatively stable state and there exists an optimum pre-stress level to obtain best properties. They also concluded that the prestress levels for optimum properties are different for different types of loads applied to the composites. The pre-stress level for tensile properties to gain
maximum values was found higher than that required by flexural properties. Figure 2-14 shows a diagram of the apparatus used for the application of prestress to the fibre for composite manufacturing. The results of tensile and flexural strength and modulus as a function of prestressing are shown in Figure 2-15.

![Figure 2-14 Design for the fibre stretching frame (units in inches) [53].](image)

Motahhari and Cameron [51] analysed the micro-residual stresses in the glass fibre-epoxy prestressed composites. They evaluated the residual strains in the pretensioned fibres subsequent to the curing process. They found that residual stresses in the fibre prestressed composites are a linear function of applied fibre prestress during curing and the ratio of the residual strain to the total fibres’ strain is a constant value and it did not depend on fibres’ pre-tension. The results also showed that fibre prestressing has two opposite effects on composite strength, it can improve the strength by preventing crack opening mechanism in the matrix and it can decrease the strength by increasing the debonding taking place at the interface due to increased residual shear stresses. The apparatus used for prestress application to the fibres is given in Figure 2-16.
Figure 2-15 Tensile strength and modulus & flexural strength and modulus as a function of prestress [53].

Figure 2-16 A schematic diagram of production method. Fibres are under tension while resin is being cured [51].
In another paper the above authors [52] evaluated the impact strength of glass fibre-epoxy prestressed composites. They showed that the impact strength of prestressed composites increased up to 33% as compared to non-prestressed composites. The increase of impact strength was due to formation of a large new area created during splitting of the prestressed samples which was not shown by the unstressed samples. Results are given in Figure 2-17.

![Figure 2-17 Impact test results as a function of fibre prestressing [52].](image)

In a similar study by the same researchers [54] the flexural properties of glass-epoxy composites were shown to improve by the application of prestress during curing process. They concluded that the fibre prestressing level, at which the highest mechanical properties are obtained, is a function of curing temperature. Results of flexural tests are shown in Figure 2-18.

The improvement in flexural modulus was due to the fact that fibres are under tension in fibre prestressed composites [51]. The tensile residual forces in the fibres produce a component which is normal to the direction of fibres in the composites. This force component acts against the bending force during the bending test. As a result, more bending force is required to produce a certain...
amount of deflection as compared to unstressed specimen as shown in Figure 2-19. This leads to a higher flexural modulus [54].

Figure 2-18 Flexural modulus and flexural strength vs. prestressing level of E glass-epoxy composites [54].

Figure 2-19 Vertical components of residual force in the fibre \( F_p \sin \theta \) works against the bending force, resulting in increase of flexural modulus [54].

Fancey [97] described a method to produce compressive stresses within a composite material, for improving resistance to crack propagation. They stretched the fibres under a load and released that load before moulding the fibres into a matrix material. After matrix solidification, continuing viscoelastic recovery of the strained fibres imparted compressive stresses to the surrounding material. Nylon 6.6 fibre-polyester resin composites were shown to have absorbed 25% more impact energy than non-stressed samples. In a subsequent study, Fancey [94] evaluated the probable service life of these composites through accelerated ageing.
(by exposure to heat). Long term creep recovery data of Nylon 6, 6 and impact test data showed that prestressed polymeric matrix composites (PPMC) may be expected to have a service life of the order of several years.

In another study, Pang and Fancey [98] extended previous work on the viscoelastic recovery of Nylon 6,6 fibre-polyester resin composites [94, 97] by applying principles of time-temperature superposition. Subjected to 342 MPa creep stress for 24 h, viscoelastic strain recovery characteristics of Nylon 6.6 were found to be the equivalent of $9 \times 10^5$ hours (100 years) at 20°C. Composite samples did not show any significant deterioration in impact performance after being exposed to the similarly accelerated aging conditions.

In a similar study by the same authors [95], tensile properties of Nylon 6.6 fibre-epoxy resin composites were studied at different levels of fibre volume fraction $V_f$. They showed that at about 35~40% value of $V_f$, the tensile strength, modulus and toughness achieved maximum values in the viscoelastically induced prestressed composites. An increase of 15%, 30% and 40% was observed in the tensile strength, modulus and toughness values.

Ziaxia et al. [99] studied the tensile properties of pre-stretched weft knitted glass fibre (GF) fabric reinforced polypropylene (PP) composites. Tensile stiffness and tensile strength increased with the increase of pre-stretch level up to a certain point beyond which both decreased slightly because of the breakage of more glass fibres. The failure strain of the composites decreased with an increase in the pre-stretch ratio because of decrease of stretchability of knitted composites. The fabrication process and the results of tensile tests are shown in Figure 2-20 and Figure 2-21 respectively.
Figure 2-20 Fabrication process of pre-stretched composites. (a) Step 1: stretching and (b) Step 2: hot pressing [99].

Figure 2-21 (a) Tensile strength and (b) Tensile stiffness of GF/PP composites with different stretch ratio [99].

Schlichting et al [100] evaluated the flexural properties of glass fibre polymer matrix composites made from two resin systems (Quixfil and Adoro for dental implant applications), and the influence of unidirectional glass fibre reinforcements, with and without pre-tensioning. The results showed that prestressing increased the flexural modulus of Adoro but not Quixfil. Prestressing also significantly increased the flexural strength of beams in both Adoro and Quixfil groups from approximately 443 to 569 MPa and from 425 to 568 MPa, respectively. The prestressing device used is shown in Figure 2-22.
Jorge et al. [101] presented a method for applying prestress on the fibres during curing. A tow of fibres was passed around pins along a flat panel and weights were hung at the ends of the fibres to impart pre-stress to them. The pins were lubricated to enable the prestress to be applied evenly across the panel. The fibres were impregnated and cured under a glass plate at room temperature for 24 hours followed by post cure at 80 °C for 3 hours. The authors claimed an overall increase in tensile strength and modulus although there were no distinctive trends because of small number of data points at each level of prestress. The apparatus used is shown in Figure 2-23.

Figure 2-22 (A) Prestressing device. (B) Capstan stainless steel grip (30 mm diameter). (C) Initial tension of 215 N applied to fibre bundles by placing weights [100].

Figure 2-23 Setup used by Jorge et al. for the application of prestress to the fibres [101].
2.3 Summary

This chapter firstly presents a review of plant fibre based preform composites with emphasis on their structure-property relationship. It is evident from the available micro-mechanic models available in the literature that the mechanical properties of composites are strongly affected by a number of factors including fibre alignment (expressed by the Krenchel Orientation factor, Equation 2-5), volume fraction of constituent and their inherent mechanical properties.

In order to fully utilize the reinforcement potential of plant fibres in load bearing applications, technical plant fibres naturally available in discontinuous form need to be assembled into a continuous yarn structure that can be used for manufacturing structured composites. There are different ways to assemble staple fibres into yarns, each having their own set of physical and mechanical properties.

Public domain literature on the effect of prestress on static mechanical properties of composites has been reviewed. By applying an optimum fibre prestress, composites tensile strength and flexural strength may be improved. The fibre alignment in composites can be improved by prestressing which results in enhancement of mechanical properties of the composites.

Despite all these studies related to manmade fibres especially glass fibres and carbon fibres, there is no study available related to the effect of prestressing on the mechanical properties of the plant fibre based composites. Plant fibre based yarns have complex structure and prestressing can change the structure to fully utilize the environmentally friendly plant fibres for structural composites applications. This study focusses on the structure property relationship of plant fibre composites.
prepared from different yarn structures like singles yarn, wrap spun yarns and plied yarns.
CHAPTER THREE

3 Materials and Methods

3.1 Materials

3.1.1 Flax Fibre and Yarns

The flax fibres were supplied in sliver form by a linen spinning mill in China. The fibres were grown, retted and scutched in Europe and processed into sliver form for use in linen yarn production by the linen mill. The mean linear density of the sliver was 14 ktex (g/m). The slivers were spun into yarns of varying twist levels on a lab scale Caipo SRL ring spinning machine and a hollow spindle wrap spinning machine.

For experiments on single yarn based composites (Chapter 4), yarns were spun at three different levels of twist, namely 3.8 TM (twist multiplier), 4.8 TM and 6.0 TM having S-twist direction, with an average linear density of 374 ± 6, 385 ± 5 and 405 ± 7 tex (gm / km) and a nominal twist of 185, 235 and 285 twist/m respectively. TM = \( T/N_e^{1/2} \), where \( T \) is the number of twist turns per inch of yarn and \( N_e \) is the yarn count according to the English cotton system (which is equal to the number of hanks each of 840 yarns weighing one pound). The delivery speed on the ring spinning machine was kept between 10 ~ 14 m/min.

For two-plied yarn based experiments (Chapter 5), a fresh batch of singles yarns with nominal twist levels of 3.8 TM, 4.8 TM and 6.0 TM and S-twist direction, were spun with an average linear density of 186 ± 7 tex, 192 ± 5 tex and
195 ± 4 tex, respectively. These singles yarns were then twisted together using a Calvani Italy Ring twisting machine (shown in Figure 3-2) in the twist direction opposite to that of the singles yarns i.e. Z-twist. The spindle speed on the ring twister was fixed at 6000 rpm. Plied yarns were prepared at different levels of ply twist/singles yarn twist ratio for all 3 singles yarn twist levels. There were 8 levels of ply twist/singles yarn twist ratio, namely 0.2, 0.25, 0.30, 0.35, 0.40, 0.50, 0.7 and 1.0. The singles and plied yarn specifications are given in Table 3-1.

Table 3-1 Singles and plied yarn specifications for plied yarn experiments.

<table>
<thead>
<tr>
<th>Twist multiplier (TM)</th>
<th>Singles yarn</th>
<th>Plied yarn</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>3.82 ± 0.07</td>
<td>4.78 ± 0.05</td>
</tr>
<tr>
<td>Nominal twist (t/m)</td>
<td>254 ± 10</td>
<td>350 ± 5</td>
</tr>
<tr>
<td>Linear density (tex)</td>
<td>186 ± 7</td>
<td>192 ± 5</td>
</tr>
</tbody>
</table>

<table>
<thead>
<tr>
<th>Plied/singles yarn twist ratio</th>
<th>Nominal plied yarn twist / m (tpm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.20</td>
<td>-</td>
</tr>
<tr>
<td>0.25</td>
<td>64</td>
</tr>
<tr>
<td>0.30</td>
<td>76</td>
</tr>
<tr>
<td>0.35</td>
<td>89</td>
</tr>
<tr>
<td>0.40</td>
<td>102</td>
</tr>
<tr>
<td>0.50</td>
<td>127</td>
</tr>
<tr>
<td>0.70</td>
<td>178</td>
</tr>
<tr>
<td>1.0</td>
<td>254</td>
</tr>
</tbody>
</table>

In wrap spun yarns based experiments (Chapter 6), two different types of filaments were used as wrapper for the parallel flax fibres. One was a textured polyester multifilament, designated as 33.9/13 tex (33.9 tex containing 13 strands),
which was used as a wrapper for the parallel flax fibres. A custom-made wrapping machine was used for wrap twisting of the yarns, as shown in Figure 3-1.

In this setup, fibrous assembly delivered from the front delivery roller of the drafting unit was fed to the hollow spindle of the wrap spinner. Upon exit from the hollow spindle, the wrap filament that was already wound on the bobbin was wrapped on the parallel fibres strand. The wrap spun yarn was then wound on a bobbin for further processing.

Figure 3-1 Yarn strand being fed from the ring spinning machine to wrap twister.

The spindle speed of the twisting element on the wrap spinning machine was set at 1400 rev/m for 90, 115 and 140 wraps/m yarns, and 2800 rev/m for 180, 230 and 280 wraps/m yarns. The wrap twist of the yarn was changed by changing the delivery from the drafting unit while the wrap spindle rotation speed was kept constant.
Wrap spun yarns were prepared in two different types, with false twist (FT) and without false twist (NFT). Six different levels of wrap twist, 90, 115, 140, 180, 230 and 280 wraps/m, were selected for the experiments in each set of the FT and NFT yarns. The average linear density of the wrap twist yarns was 337 tex in NFT yarns and 375 tex in FT yarns.

![Calvani ring twisting machine](image)

Figure 3-2 Calvani ring twisting machine.

### 3.1.2 Resin

Unsaturated low styrene emission (LSE) polyester resin ESCON F62-333 and MEKP Interox NR20 hardener were supplied by FGI Australia. The F62-333 LSE resin had density of 1.10 g/cm³, tensile and flexural strength of 60 MPa and 100 MPa and flexural modulus of 4000 MPa, respectively [102].

### 3.2 Composite Manufacturing

The flax yarns were first prepared into hanks (100 rounds of 1 m-long loops) using a hand driven laboratory wrapping machine. The hanks were dried in a
convection oven at 100°C for five hours before being used for composite fabrication.

An open-ended, two-part leaky mould technique [103] was used to produce “unidirectional composites” from the yarn hanks because of ease in the preparation of samples and accuracy with the applied strain to the yarns. Figure 3-3 shows the open-ended two part mould which was used to prepare a single composite bar of controlled thickness. The leaky mould was placed inside a steel channel frame fitted with a fixed pin and a movable pin. The yarn hank was loaded under prestress between the two pins. Before the application of prestress, the part of the yarn hank in the range of the leaky mould was impregnated with the resin system using a brush (fibre and resin in approximate 50/50 ratio by weight), with the rest of the hank kept dry. The movable pin was then moved by the screw mechanism to apply a predetermined level of strain to the yarn hank (it was accurate to ±0.001). The applied strain was based on the load-elongation curve of the yarn with maximum applied strain kept below the minimum value of the breaking strain of the yarn in each case. The leaky mould was closed and the device was moved to a hydraulic press. A constant pressure (45 kN) was applied on top of the leaky mould while the resin was cured for 24 hours at room temperature. The applied tension was then removed by releasing the screw and the composite specimen was taken out from the mould. The specimen was post cured at 80°C in a convection oven for two hours. The fabrication procedure is illustrated in Figure 3-3. The samples were approximately 3 mm thick. One bar for flexural test and three bars for tensile test per prestress level were prepared and the test pieces were cut out of the bars using a bench saw.
3.3 Characterization

3.3.1 Fibre Testing

3.3.1.1 Fibre Tensile Testing

All fibre samples were conditioned for 24 hours in a controlled laboratory environment, 20°C ± 2°C and 65 ± 2 % relative humidity according to ISO 139:2005, before testing. All mechanical testing of samples was carried out in the controlled environment at 20°C ± 2°C and 65 ± 2 % relative humidity.

Tensile testing of the flax fibres was carried out on a Favimat™ single fibre tensile testing machine using a load cell of 1200 cN capacity. The Favimat machine tests both tensile properties and the linear density of the fibre specimen, from which the fibre tenacity is calculated. The fibre density measurement is based on the
vibroscope principle [104]. The gauge length was 10 mm and the cross head speed was set at 1 mm/min. A pretension of 0.5 cN/tex was applied during the test. A total of 95 fibres were tested.

### 3.3.1.2 Fibre Density

The flax fibre density was measured using a calibrated Pycnometer Ultrapy 1200e manufactured by Quantachrom. Helium was used as a displacement gas for measurement of true density of the flax fibres $\rho_f$ [105, 106]. The pycnometer works by measuring the amount of displaced gas [Figure 3-4]. The pressure is measured when (i) filling the specimen chamber (with volume $V_s$) $P_1$ and then (ii) evacuating it into a second empty expansion chamber (with volume $V_e$) $P_2$, which allows for the calculation of the sample solid phase volume $V_s$ (Equation 3-1). The final density reading is the mean reading $\rho_f$ from three samples, where each density reading is an average of nine systematic readings from nine purges and runs. A purge pressure of $P_1 = 28.0$ psig, equilibrium rate of $0.05$ psig/min and specimen chamber temperature of 240 °C was used. The mass of fibre samples was measured using a digital balance having a sensitivity of 0.1 mg. The measured density measurements were precise to three decimal points.

![Figure 3-4 Schematic diagram of a gas Pycnometer.](image)

Figure 3-4 Schematic diagram of a gas Pycnometer.
\[ V_s = V_e - \frac{P_2}{P_1 - P_2} V_e \]  

Equation 3-1

### 3.3.1.3 Fibre Stress Relaxation Testing

Stress relaxation of flax fibres was carried out on an Instron 5567 using a 10 N load cell. The gauge length was kept at 100 mm. The fibre specimen was first extended to a predetermined strain of 0.020. The applied strain was accurate within ±10% value. The cross head of the tensile testing machine was then stopped and held in position. The decay of stress in the fibre was recorded over a period of an hour.

### 3.3.2 Yarn Testing

#### 3.3.2.1 Yarn Tensile Testing

All the yarn samples were conditioned for 24 hours at controlled laboratory environment, as described in section 3.3.1.1 before testing. Mechanical testing of samples was carried out in a controlled environment, using an Instron 5567 testing machine using a 100 N load cell, in accordance with ASTM D 2256 [107]. The machine worked on the Constant Rate of Extension (CRE) principle. The tests were carried out at a cross head speed of 200 mm/min for an effective gauge length of 500 mm with pretension of 0.5 cN/tex. Each test result was a mean of 30 specimens taken from one yarn sample. The breaking force and breaking elongation of yarn were normalised by using linear density of the yarn for each sample to produce specific stress (cN/tex) versus extension (mm) curves. Tensile modulus of the yarn was calculated from the slope of the initial part of the load-extension curve for each yarn specimen.
3.3.2.2 Stress Relaxation Testing of Yarn

Stress relaxation tests on flax yarns were carried out using the Instron 5567 testing machine with a 100 N load cell. The yarn specimen with gauge length of 500 mm was first extended to a predetermined strain of 0.024. The cross head of the tensile testing machine was then stopped and held in position. The decay of stress in the yarn was recorded over a period of one hour. One data point per second was recorded for the duration of the test.

3.3.2.3 Yarn Geometry Tests

The effect of tension load on the geometry (diameter and twist) of the dry yarn was studied using a Canon DSLR camera fitted with macro lens. The yarn sample, with a length of 200 mm, was mounted on the Instron tensile testing machine. Images of the yarn were taken at predetermined levels of strain. The images were used to measure yarn diameter and angle of surface fibre inclination over a range of applied strain levels using ImageJ software. 10 images were taken along the length of the yarn and 5 readings each of yarn diameter and fibre inclination angle were taken resulting in a total of 50 readings for each yarn sample.

3.3.3 Composite Testing

3.3.3.1 Composites Flexural Testing

Flexural properties of the composites were determined by the three-point bending test performed on an Instron 5567 machine using a 30 kN load cell in accordance with ASTM D 790-03 [108]. The specimen size was 70 mm (length) × 10 mm (width) × 3 mm (thickness), while the cross head speed was 1.6 mm/min.
Six specimens were tested for each composite sample. The flexural strength ($\sigma_f$) and flexural modulus were calculated using the formulae given below [108]:

$$\sigma_f = \frac{3PL}{2bd^2} \quad \text{Equation 3-2}$$

$$E_b = \frac{mL^3}{4bd^3} \quad \text{Equation 3-3}$$

where

$E_b$ = modulus of elasticity in bending (MPa)

$$\sigma_f = \text{flexural stress (MPa)}; \quad L = \text{span length (mm)};$$

$$b = \text{width of specimen (mm)}; \quad d = \text{depth of the specimen (mm)};$$

$m = \text{slope of the tangent to the initial straight-line portion of the load-deflection curve (N/mm) of deflection}.$

3.3.3.2 Composites Tensile Testing

Tensile testing of the composite samples was performed by using Instron 5567 machine with a 30 kN load cell in accordance with ASTM D 3039/D 3039M-00 [109]. The specimen size was 250 mm (length) $\times$ 15 mm (width) $\times$ 3 mm (thickness). Six specimens were tested for each composite sample.

3.3.3.3 Composites Fibre Weight Fractions and Volume Fractions

The fibre weight fraction ($W_f$) of each specimen was calculated by taking the ratio of the weight of fibres in the composite specimen to the total weight of the specimen. The fibre weight in a unit length of the specimen is known from the total weight of the yarn hank and the length of the hank (500 mm). To calculate the fibre
weight per unit length for prestressed composites, the strain applied to the yarn was added to the initial hank length (i.e. 500 mm + elongation due to applied strain).

The fibre volume fraction \((V_f)\), matrix volume fraction \((V_m)\) and void content \((V_p)\) were then calculated from \(W_f\), \(\rho_c\) and the density of the fibres \((\rho_f)\) and matrix \((\rho_m)\) using below formulae [81].

\[
V_f = \frac{\rho_c W_f}{\rho_f} \quad \text{Equation 3-4}
\]

\[
V_m = \frac{\rho_c}{\rho_m} (1 - W_f) \quad \text{Equation 3-5}
\]

\[
V_p = 1 - (V_f + V_m) \quad \text{Equation 3-6}
\]

### 3.3.3.4 Microscopy Analysis

Five samples, with dimensions 10 mm (width) × 5 mm (length) × 3 mm (thickness), were cut from each composite specimen. These samples were fixed in epoxy resin for grinding and polishing using a Struers RotoPol-21 polishing machine. The polished surfaces were imaged using a DP 70 camera system fitted on an Olympus BX50 optical microscope.

Yarn diameter in the final composites was measured from the microscopic images using ImageJ software. A minimum of five readings were taken from the cross section of an image, while a minimum of five images were taken for each sample to get the average value from the 25 reading.

### 3.3.3.5 Dynamic Mechanical Thermal Analysis (DMTA)

Dynamic mechanical thermal analysis of the composite samples was carried out using a TA DMA Q800 instrument in a dual-cantilever bending flexural loading
mode at frequencies of 1 Hz, 10 Hz and 50 Hz. The testing was carried out between 
-20°C and 125°C at a heating rate of 3°C/min and strain of 0.01%. The storage 
modulus (E') and loss tangent (tan δ) of the composite specimens were measured 
as a function of temperature (from -20 °C to 125 °C). The specimen sizes were 35 
mm (length) × 10 mm (width) × 3 mm (thickness). The glass transition temperature 
was defined as the maximum of the loss tangent (tan δ).
4 Study of Singles Yarns Based Unidirectional Composites

4.1 Introduction

In this Chapter a range of flax fibre singles yarns having various levels of twist will be produced by conventional ring spinning method. These yarns were then utilised to fabricate polyester composites using an in-house designed mould capable of introducing a prestress during the curing stage of the resin. The first part of this Chapter will study the effect of yarn twist on the physical properties (composite density, void content and fibre volume fraction), thermo-mechanical (DMA) and mechanical (flexural and tensile) properties of the resulting composites.

In the second part, the effect of prestress on the properties of the composites will be investigated. The changes of yarn structure due to prestressing will also be determined using optical microscopy and the results will be correlated with the mechanical properties to establish the structure-property relationship.

4.2 Flax Fibre Characteristics

A number of fibre tests were carried out to determine the properties of the flax fibres used for this work. These properties include microscopic structure (morphology), density and tensile properties.

4.2.1 Morphology

Optical reflection microscopy analysis of the fibres was carried out to observe the fibre surface and to measure their diameters. The fibre linear density was measured using the Favimat™ machine. The average fibre linear density was
153.1±131.4 dtex based on measurement of 95 randomly selected fibres. Based on the flax fibre density of 1.51 g/cm³ [20], the fibre diameter was then calculated from the fibre linear density. The average fibre diameter was 88 μm with a standard deviation of 35 μm from the 95 fibres tested. Figure 4-1 shows an image of a flax fibre from the sliver used to prepare the yarns in this study.

![Figure 4-1 Optical microscope image of a flax fibre with defects.](image)

Fibre defects can be seen as bands perpendicular to the length of the fibre. These defects are generally considered to be local misalignments of cellulose microfibrils in the cell wall and are known by different names in the literature, like nodes, kink bands, slip planes, misaligned zones or micro-compressions [110].

It has been shown in a recent study that the amount of defects in flax fibres increases due to the processing of the fibres [111]. The increasing number of processing steps were correlated to the decrease in strength of the fibres in another study related to flax and hemp fibres [112]. Therefore performance of plant fibre reinforced composites is dependent on the amount of defects in the fibres generated during the growth and processing of fibres because these defects act as stress raisers in the polymer matrix in the composites giving rise to fibre failure [113] and debonding at the fibre-matrix interface [114].
4.2.2 Mechanical Properties and Density of Flax Fibre

The tensile strength, breaking elongation and linear density of the flax fibres was measured using Favimat™ machine. The tenacity (specific tensile strength) and specific modulus were calculated by taking into consideration the linear density of the fibres measure by Favimat™. Figure 4-2 shows the tenacity distribution of the tested flax fibres. The average tenacity, breaking elongation and specific modulus were 46.0 cN/tex, 3.1% and 20.4 N/tex, respectively. The fibre density measured by the Pycnometer was found out to be $1.512 \pm 0.006 \text{ g/cm}^3$, which is close to the average of most lingo-cellulosic fibres. Using this fibre density, these values can be converted to engineering values of breaking stress 690.3 MPa and elastic modulus 30.6 GPa respectively. These values are at the low end of the flax fibre properties reported in literature [115]. Taking the measured linear density of the flax fibre, the specific tensile strength and specific tensile modulus come to be 456 MPa/gcm$^{-3}$ and 20.2 GPa/gcm$^{-3}$ respectively.

![Figure 4-2 Tenacity (specific tensile strength) distribution of flax fibre.](image)
4.3 Study of Yarn Properties

4.3.1 Characterization of Twisted Flax Yarns

A typical micrograph of ringspun flax yarns is given in Figure 4-3. The flax fibres in the yarn are twisted with a left-handed angle to the yarn axis, indicating an S-twisted yarn.

The yarns were spun at two nominal linear densities, 385 tex and 190 tex, each with three levels of twist multiplier (TM), 3.8 TM, 4.8 TM and 6.0 TM. The yarns spun at nominal linear density of 190 tex (actual linear density was 186 ± 7 tex, 192 ± 5 tex and 195 ± 4 tex for 3.8 TM, 4.8 TM and 6.0 TM yarns respectively) were used to prepare two-ply yarns used for experiments mentioned in chapter 5 (related to the study of plied yarn based composites), while this chapter covers the study of the singles yarns based composites prepared from the 385 tex singles yarns. The tensile test curves of the flax yarns at three different levels of twist are presented in Figure 4-4. The Instron force-displacement tensile data was converted to specific tensile strength referred to as Tenacity in the textile industry. This was achieved by taking into account the linear density of the yarns. It can be seen that the yarns have much lower tenacity but higher breaking strain than the constituent fibres. Figure 4-5 (A) shows the tenacity of 385 tex singles yarns at different twist factors, while Figure 4-5 (B) shows the tenacity of 190 tex yarns.

Figure 4-5 compare flax yarn tenacity and breaking strain at three levels of twist. The tenacity increased when the twist was increased from 3.8 TM to 4.8 TM and then decreased when the twist was further increased to 6.0 TM. This is because with the increase of twist, the cohesion between the fibres increases, as illustrated in Figure 2-3.
Figure 4-3 Optical microscope image of a twisted (3.8 TM) flax yarn.

Figure 4-4 Tensile test curves of flax fibre and corresponding processed yarns at different twist levels.
Figure 4-5 Tenacity of flax yarns at different twist levels. (A) 385 tex yarns, (B) 190 tex yarns.

Figure 4-6 Tenacity distribution of 385 tex flax fibre spun yarns.

The decrease in yarn strength at the high twist level 6.0 TM can be attributed to increase of fibre obliquity (i.e. helical angle) in the yarns [88]. It can be seen in the Table 4-1 that the fibre inclination angle of the high twist yarn (6.0 TM) is higher than that of low twist (3.8 TM) yarn. Also there was a decline in specific tensile strength (tenacity) observed after an initial improvement from low to medium twist yarn. On the other hand, as shown in Figure 4-5, the breaking
extension increased with the increase of yarn twist throughout the twist range because of the long helix path of the fibres and the large fibre strain before the break of highly twisted yarns.

The distribution of tenacity of the yarns has been shown in Figure 4-6. The yarn strengths are sufficiently high for textile processes and handling in composite fabrication. The physical and tensile properties of twisted yarns are given in Table 4-1.

Table 4-1 Summary of flax yarns characteristics.

<table>
<thead>
<tr>
<th>Yarn twist multiplier</th>
<th>3.8</th>
<th>4.8</th>
<th>6.0</th>
</tr>
</thead>
<tbody>
<tr>
<td>Linear density (tex)</td>
<td>374.2 (9.58)a</td>
<td>385.2 (6.61)</td>
<td>405.8 (7.37)</td>
</tr>
<tr>
<td>Twist angle (°)</td>
<td>21 (1.35)</td>
<td>27.4 (1.24)</td>
<td>32.7 (2.76)</td>
</tr>
<tr>
<td>Diameter (mm)</td>
<td>0.63 (0.05)</td>
<td>0.60 (0.07)</td>
<td>0.58 (0.04)</td>
</tr>
<tr>
<td>Tensile strength (cN/tex)</td>
<td>13.99 (3.1)</td>
<td>16.78 (2.4)</td>
<td>14.08 (3.73)</td>
</tr>
<tr>
<td>Tensile modulus (N/tex)</td>
<td>6.20 (1.21)</td>
<td>6.29 (0.76)</td>
<td>3.59 (0.99)</td>
</tr>
<tr>
<td>% Elongation at break</td>
<td>3.7 (0.48)</td>
<td>5.0 (0.60)</td>
<td>6.6 (1.12)</td>
</tr>
</tbody>
</table>

a Average (Std Dev.)

4.3.2 Yarn Stress Relaxation

Viscoelastic material, when subjected to a stress load below the yield strength will slowly creep (i.e. extend) as time goes on. Upon removal of the stress load, a partial instantaneous recovery will be observed but full recovery of the material will require a certain amount of time. This phenomena is often investigated using stress relaxation study.

The flax yarn structure is held together by fibre-to-fibre friction. When a constant load is applied to the yarn, the fibres tend to slide relative to each other slightly by overcoming the fibre-to-fibre friction, which is an internal friction to the yarn and displays as a gradual decrease of the yarn tension (i.e. the yarn is
viscoelastic). Figure 4-7(a) shows the initial part (first 60 seconds) of the low twist (3.8 TM) yarn tension decay curves at two prestressing levels, 0.01 and 0.024 applied strains. The complete stress-time curves (one hour relaxation) at the two prestressing levels are replotted in Figure 4-7(b), in which the instantaneous yarn tension is normalized by the maximum yarn tension corresponding to the applied prestressing strain.

As can be seen, the stress drop is related to the prestressing strain applied. At low prestressing level (applied strain 0.01 in Figure 4-7), the initial yarn stress was 2.52 cN/tex and the end stress after 1 h of relaxation was 1.06 cN/tex for low twist 3.8 TM yarn, the yarn stress dropped by 58%. At high prestressing level (applied strain 0.024 in Figure 4-7), the yarn stress dropped by 42% after 1 h, significantly less than the stress drop at the low strain level. Similar results have been reported by other studies [116, 117]. This is due to the fact that the fibres in the flax yarn are held together by fibre-to-fibre friction. On the application of load to the yarn, the straight fibres are able to slip relative to the neighbouring fibres so that the slack fibres can straighten. This results in a small permanent increase of yarn length and equalization of fibre tension, i.e. slack fibres in the yarn buildup their tension while high tension fibres reduce their tension because the external load is now shared by more fibres. The permanent increase in yarn length causes part of the stress drop as shown by the results in Table 4-2 and Figure 4-7. At a low prestressing level, fibres straightening and slippage dominates, which is irrecoverable yarn yarn extension. On the opposite side, at high prestressing level, after the initial fibre straightening and slippage, the fibres themselves start to elongate elastically and this part of yarn elongation can recover if yarn tension is released [118].
The yarn stress relaxation test results at four applied strain levels summarized in Table 4-2 confirm that the stress drop decreased consistently as prestressing level increased. The yarn stress was converted to MPa values by taking into account the flax fibre density = 1.5 g/cm³ and using the formula: Yarn stress (MPa) = Yarn specific stress (cN/tex) x Density (1.5 g/cm³) x 10.
Table 4-2 Yarn stress relaxation test results of yarns at three twist levels.

<table>
<thead>
<tr>
<th>Yarn TM</th>
<th>Applied strain</th>
<th>Initial specific stress (cN/tex)</th>
<th>Initial stress (MPa)</th>
<th>End specific stress after 1hr (cN/tex)</th>
<th>End stress after 1hr (MPa)</th>
<th>Stress drop</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.8 TM</td>
<td>0.01</td>
<td>2.52 (0.13)</td>
<td>37.8 (1.95)</td>
<td>1.06 (0.02)</td>
<td>15.9 (0.3)</td>
<td>58% (2%)</td>
</tr>
<tr>
<td></td>
<td>0.014</td>
<td>3.20 (0.71)</td>
<td>48 (10.65)</td>
<td>1.56 (0.40)</td>
<td>23.4 (6.0)</td>
<td>52% (2%)</td>
</tr>
<tr>
<td></td>
<td>0.024</td>
<td>7.90 (0.54)</td>
<td>118.5 (8.1)</td>
<td>4.58 (0.32)</td>
<td>68.7 (4.8)</td>
<td>42% (1%)</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>11.62 (0.90)</td>
<td>174.3 (13.5)</td>
<td>7.27 (0.65)</td>
<td>109.1 (9.8)</td>
<td>37% (1%)</td>
</tr>
<tr>
<td>4.8 TM</td>
<td>0.024</td>
<td>5.35 (1.38)</td>
<td>80.25 (20.7)</td>
<td>2.80 (0.79)</td>
<td>42.0 (11.9)</td>
<td>48% (2%)</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>11.23 (1.61)</td>
<td>164.5 (24.2)</td>
<td>7.20 (1.07)</td>
<td>108.0 (16.1)</td>
<td>36% (0.4%)</td>
</tr>
<tr>
<td>6.0 TM</td>
<td>0.024</td>
<td>4.43 (0.8)</td>
<td>66.45 (12.3)</td>
<td>2.25 (0.49)</td>
<td>33.8 (7.4)</td>
<td>50% (2%)</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>6.57 (0.9)</td>
<td>98.55 (13.5)</td>
<td>3.86 (0.53)</td>
<td>57.9 (7.9)</td>
<td>41% (0.1%)</td>
</tr>
</tbody>
</table>

* Average (Std. Dev)

Figure 4-7 Stress relaxation curve of low twist (3.8 TM) flax yarn at two prestressing levels, 0.01 and 0.024 applied strain for (a) First 60 seconds, (b) total time of 60 minutes.
4.3.3 Diameter and Twist Angle of Dry Yarn under Strain

The yarn diameter and angle of fibre inclination on the yarn surface of the low twist (3.8 TM) and high twist (6.0 TM) yarns were measured while the yarn was being elongated in the tensile testing machine. At the beginning of the test, marks were applied at 5 points uniformly separated along the length of the yarn between the two jaws and images were taken using digital SLR camera. Images were taken again at the predetermined prestress (strain) levels at the same points and variation in the diameter was calculated using the image analysis of the photographs using ImageJ software. As shown in Figure 4-8, the yarn diameter of both low and high twist yarns contracted significantly but the contraction in the low twist yarn was higher than that in the high twist yarn. The reason is that fibres in high twist yarn are packed much closer to each other than in low twist yarn and on the application of tensile load, the capacity for fibres to pack further (and hence diameter decrease) is much lower than for fibres in the low twist yarn. This is shown by the lower value of Poisson’s ratio to be discussed below.

The low twist yarn had an extremely high Poisson’s ratio of 6.6 (at 0.03 applied strain there was ~ 20% reduction in yarn diameter) while the high twist yarn had lower Poisson’s ratio of 3.9. These values are 10-15 times higher than the Poisson’s ratio of common solid materials, such as steel (~0.3) and rubber (~0.5) and fibres like wool (0.4-0.6) [119] and nylon (0.37-0.42) [120]. This extremely high Poisson’s ratio of the twisted yarn is not a reflection of the Poisson’s ratio of the constituent flax fibres. The contraction of yarn diameter is mainly a result of increased fibre packing density in the yarn, i.e., the disappearance of voids between the fibres in the yarn. Higher twisted yarns will have much lower porosity and therefore much smaller Poisson’s ratio [121].

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Under external load, the yarn becomes thinner and longer, in other words, the length of the fibre helix in the yarn \((h=1/T, \text{ is the yarn length containing one complete turn of twist})\) become larger and the helix diameter \((d)\) becomes smaller at the same time. Consequently, the fibre angle of inclination \(\theta\) decreases very quickly following the relationship \(\tan \theta = \pi d/h\) [122]. The experimentally determined angle of inclination \(\theta\) at all twist levels as a function of the pre-stress are plotted in Figure 4-9. \(\theta\) decreased steadily with the increasing strain, and at the highest strain of 0.03 strain, the \(\theta\) in low twist (3.8 TM) yarn decreased by about 28.6% from 21° to 15° while for high twist (6.0 TM) yarn it decreased by 18.7% from 33.2° to 27°.

Figure 4-8 Changes in dry yarn diameters as a function of applied strain in low (3.8 TM) and high (6.0 TM) twist yarns.
Figure 4-9 Changes in yarn inclination angle ($\theta$) as a function of applied strain in low (3.8TM), medium (4.8 TM) and high (6.0 TM) twist yarns.

4.4 Composites Properties

4.4.1 Density and Fibre Volume Fraction

The density and fibre volume fraction of the composites manufactured from yarns having different twist levels and at different levels of prestress are shown in Figure 4-10.

In Figure 4-10 we can see that the density of low and medium twist (3.8 TM and 4.8 TM) yarn based composites only slightly increased with increasing prestress. The fibre volume fraction slightly decreased with increasing prestress during the composite fabrication process. The reason is that with the application of prestress force, the fibres come closer to each other resulting in decreased yarn radius and cross sectional area (as previously discussed, see Figure 4-8). This leads to the reduction of fibre volume per unit volume of the composite i.e. reduction of fibre volume fraction $V_f$ and increase of composite density because fibres are occupied in less space. In high twisted yarn (6.0 TM), the fibres are tightly packed...
and therefore the chances for the fibres to squeeze further under the applied
prestress decrease.

The mean fibre volume fraction for the low twist (3.8 TM), medium twist
(4.8 TM) and high twist (6.0 TM) based composites were all found to be similar,
0.466 ± 0.016; 0.471 ± 0.014 and 0.486 ± 0.029 respectively. The density of these
composites was found also to be very comparable (within 5%) 1.244 ± 0.022, 1.248
± 0.005 and 1.252 ± 0.029 for 3.8 TM, 4.8 TM and 6.0 TM based composites,
respectively.
Figure 4-10 Effect of prestress on the density and fibre volume fraction of composites prepared from yarns of three different twist levels. (A) 3.8 TM, (B) 4.8 TM and (C) 6.0 TM.
4.4.1.1 Fibre Packing in Composites

During composite fabrication, the voids between the flax fibres are either infiltrated by resin \( (V_m) \) or remain as voids \( (V_p) \). Because the resin was applied to the yarn hank before the prestressing, it can relatively easily infiltrate into the centre of the yarns through connecting voids between the fibres. This level of infiltration into the centre of the yarn depends on the packing density of the fibres, which varies with twist of the yarn. It will be easy for the resin to infiltrate between the fibres for a low twist yarn as compared to a high twist yarn. Some of the resin initially infiltrated into the yarn may be squeezed out of the yarn when the prestress is applied.

Figure 4-11 shows the micrographs of the composites cross sections prepared from three different levels of twist without applying prestress. Figure 4-12 shows the images of composites cured under the applied prestress. It can be seen in Figure 4-11 that the cross section in the low twist yarn and medium twist yarn based composites had noncircular cross section while the high twist yarn (6.0 TM) based composites had round cross section in response to the lateral pressure applied during the compression moulding process. The high twist yarn maintained its circular cross section due to high fibre packing density in the high twist yarn.
Figure 4-11 Microscopic images of the composites prepared without prestress. 
(a,b) 3.8 TM; (c,d) 4.8 TM; (e,f) 6.0 TM. Scale bar is 200 μm.

Figure 4-12 shows that the yarns have circular cross section for all levels of twist under the application of prestress as opposed to noncircular cross sections in low and medium twist yarn based composites without prestress. This shows that the prestress has increased the fibre packing density in the yarn and the yarn’s resistance to compression. It is also evident that the distribution of fibres within the compact yarn is uneven and there are noticeable resin-rich regions in the composite.
cross section, while it can also be observed that yarn bundles in the composites are distributed uniformly within the matrix.

Figure 4-12 Microscopic images of the composites prepared with a prestress (a,b) 3.8 TM at 0.03 strain; (c,d) 4.8 TM at 0.03 strain; (e,f) 6.0 TM at 0.038 strain levels.
The void content of the singles yarn based composites was calculated using Equation 3-6 [81] and are given in Figure 4-13. It can be seen that the void content of the composites decreased with the increase of applied prestress. The decrease is due to the improvement in fibre alignment and better resin impregnation under the application of prestress. The void contents for these composites were all in the range of 3 ~ 6.5%, which is higher than that of glass fibre based composites (1~3%). Normally void content of <1% is required for aerospace applications, but void contents of up to 5% are acceptable for less demanding areas like automotive and marine applications [123, 124]. Although there is an economic benefit of producing parts with higher void content due to ease of manufacturing, increased porosity will impart adverse effects on mechanical properties, especially over a period of time due to a potential increase in moisture absorption [125].

![Figure 4-13 Void content of singles yarn based composites at different levels of applied prestress, prepared from singles yarn at three different twist levels. (A) 3.8 TM, (B) 4.8 TM, (C) 6.0 TM.](image)

The cross sectional area and diameter of the yarns in the final composites is plotted as a function of the applied strain in Figure 4-14. The average yarn cross section area decreased by 30%, 14% and 5.3% at applied strain of 0.03, 0.03 and 0.038 for 3.8 TM, 4.8 TM and 6.0 TM based composites respectively. This
reduction in yarn cross sectional area translates to 16%, 7.2% and 3% reduction in the yarn diameter for the 3.8 TM, 4.8 TM and 6.0 TM yarn based composites respectively. The difference of mean diameter between the dry yarn and the yarn in the final composites may be attributed to the presence of the resin between the fibres and the pressure applied to the composite during compression moulding. In low twist yarn it may be easier for fibres to get closer to each other and slide relative to each other, resulting in higher reduction of yarn diameter. On the other hand, in high twist yarns, the fibres are already very close to each other and application of prestress does not result in so much reduction in yarn diameter.

![Figure 4-14 Yarn cross section area and diameter change due to prestressing in composites.](image)

**4.4.2 Mechanical Properties**

The main objective of fibre prestressing is to improve the mechanical properties of the composites. In this study, flexural properties and tensile properties of composites have been tested to determine the effect of prestressing during manufacturing on the mechanical properties of the resulting composites.
4.4.2.1 Flexural Properties

Three point bending tests were carried out on the composites samples prepared from yarns having three different levels of twist i.e. 3.8 TM, 4.8 TM and 6.0 TM. The flexural strength and flexural modulus results of the composites are given in Figure 4-15 ~ Figure 4-17. The maximum applied force during each flexural test was used to calculate the flexural strength of the samples using Equation 3-2.

Figure 4-15 Flexural strength (A) and Flexural modulus (B) vs. Prestressing for 3.8 TM yarn based composites.

In Figure 4-15, it can be seen that the flexural strength and flexural modulus of the composites increased with the increase of applied strain during the composites fabrication. The flexural strength increased by 29% from 133.6 MPa to 172.07 MPa and flexural modulus by 37% from 10.94 GPa to 15.0 GPa with the application of 3% (0.03) strain for low twist 3.8 TM yarn based composites. The increase of flexural strength for medium twist 4.8 TM yarn based composites (Figure 4-16) was 12% from 117.90 MPa to 132.5 MPa and flexural modulus by 18% from 11.02 GPa to 13.0 GPa at an applied strain as 3% (0.03). For high twist 6.0 TM yarn based composites (Figure 4-17), the increase in flexural strength was
found to be 9% from 114.71 MPa to 125.14 MPa and flexural modulus by 48%
from 7.66 GPa to 11.29 GPa when a prestressing strain of 0.026 was applied.
Further increase of prestressing caused the flexural stress to decrease.

Figure 4-16 Flexural strength (A) and Flexural modulus (B) vs. Prestressing for
4.8 TM yarn based composites.

Figure 4-17 Flexural strength (A) and Flexural modulus (B) vs. Prestressing for
6.0 TM yarn based composites.

The statistical analysis \textit{t-test} results performed on the flexural test results
are given in Table 4-3. As can be seen in Table 4-3, the flexural strength and
flexural modulus of the low twist (3.8 TM) yarn based composites were
significantly improved by the prestressing. In medium twist (4.8 TM) yarn based
composites, the flexural modulus was found to be significantly improved by
prestressing, but the improvement in flexural strength was not statistically
significant. The improvement in flexural strength and flexural modulus in the high
twist (6.0 TM) yarn based composites was also found to be statistically significant until a prestressing level of 0.026 strain, beyond which both properties declined.

Table 4-3 Probability of prestressing having no effect on flexural properties. *p*-values less than 0.05 have been highlighted.

<table>
<thead>
<tr>
<th>Yarn TM</th>
<th>Applied strain</th>
<th>Flexural strength</th>
<th>Flexural modulus</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0.01</td>
<td>0.02</td>
</tr>
<tr>
<td>3.8 TM</td>
<td>0.01</td>
<td>-</td>
<td>0.153</td>
</tr>
<tr>
<td></td>
<td>0.02</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>0.026</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>4.8 TM</td>
<td>0.01</td>
<td>0.302</td>
<td>0.110</td>
</tr>
<tr>
<td></td>
<td>0.02</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>0.026</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>6.0 TM</td>
<td>0.01</td>
<td>0.047</td>
<td>0.025</td>
</tr>
<tr>
<td></td>
<td>0.02</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>0.026</td>
<td>-</td>
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<td>0.000</td>
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<tr>
<td></td>
<td>0.02</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>0.026</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

Similar improvements in mechanical properties have been reported by other studies related to the effect of prestressing on glass fibre and carbon fibre reinforced...
composites [54, 101, 126]. Jorge et al. [101] reported that the tensile strength of glass fibre reinforced polyester composites increased with the increase of applied prestress and stabilised after reaching a certain level. Motahhari reported that the flexural strength of glass fibre reinforced epoxy composites showed a slight decline after reaching the maximum. In the present study the flexural strength has been found to increase in low twist (3.8 TM) and medium twist (4.8 TM) yarn based composites, while for high twist (6.0 TM) based composites, the strength increased up to a strain level of 0.026 and then declined.

Table 4-4 Changes caused by prestressing in composites prepared from singles yarns at three twist levels.

<table>
<thead>
<tr>
<th>Yarn Twist</th>
<th>3.8 TM</th>
<th>4.8 TM</th>
<th>6.0 TM</th>
</tr>
</thead>
<tbody>
<tr>
<td>Total improvement in flexural strength</td>
<td>29%</td>
<td>12.4%</td>
<td>9.1%</td>
</tr>
<tr>
<td>Initial fibre angle (°)</td>
<td>21</td>
<td>27</td>
<td>30.2</td>
</tr>
<tr>
<td>Final fibre angle (°)</td>
<td>15</td>
<td>20.3</td>
<td>27</td>
</tr>
<tr>
<td>Krenchel orientation factor change (%)</td>
<td>7%</td>
<td>10.8%</td>
<td>12.9%</td>
</tr>
<tr>
<td>Unprestressed composites fibre volume fraction ($V_f$)</td>
<td>0.482</td>
<td>0.474</td>
<td>0.515</td>
</tr>
<tr>
<td>Prestressed composites fibre volume fraction ($V_f$)</td>
<td>0.451</td>
<td>0.445</td>
<td>0.492</td>
</tr>
<tr>
<td>Change of fibre volume fraction (%)</td>
<td>-6.4%</td>
<td>-6.1%</td>
<td>-4.5%</td>
</tr>
<tr>
<td>Dry yarn residual stress after 1 hr (MPa)</td>
<td>68.7</td>
<td>42</td>
<td>33.7</td>
</tr>
</tbody>
</table>

When comparing the flexural test results with those of the constituent yarn results presented in Figure 4-5, it is seen that the flexural properties of the composites declined with increasing twist of the constituent yarns, while the yarn strength increased by increase of twist up to a level and then declined. This is due
to the fact that in composites, the mechanical properties are dictated by the fibre alignment with the yarn axis. With the increase of yarn twist, the fibre inclination increases resulting in decrease of Krenchel fibre orientation factor in the rule of mixtures equation (Equation 5-2). But in yarns the strength initially increases due to increase of friction between the fibres reaching a maximum level, beyond which the strength decreased due to increasing obliquity of the yarns.
4.4.2.2 Tensile Properties

The tensile test results of composites prepared at different levels of prestress are given in Figure 4-18. As it can be seen from Figure 4-18, the tensile strength and tensile modulus increased with the increase of prestress in low twist 3.8 TM yarn based composites. For both the medium twist and high twist yarn based composites, the tensile strength and modulus increased up to their maximum at 2% strain and then declined.

![Figure 4-18 Tensile test results of composites prepared from (A) 3.8 TM yarn, (B) 4.8 TM yarn & (C) 6.0 TM yarn based composites.](image)

The results seem to suggest that each yarn has its own optimum level of prestress. The low twist yarn had a higher optimum prestressing level than the medium and high twist yarns. These trends have similarity to that observed from the flexural test results of these composites in the previous section. Statistical analysis (t-test) was performed on the tensile test results of the prestressed
composite to determine if the improvement in tensile properties caused by prestressing was statistically significant. The t-test results are shown in Table 4-5.

Table 4-5 Tensile test results t-test analysis. *p*-value showing probability of prestressing having no effect.

<table>
<thead>
<tr>
<th>Yarn twist level</th>
<th>Applied strain</th>
<th>Tensile strength</th>
<th>Tensile modulus</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td>0.02</td>
<td>0.03</td>
</tr>
<tr>
<td>3.8 TM</td>
<td>0</td>
<td>0.013</td>
<td>0.025</td>
</tr>
<tr>
<td></td>
<td>0.02</td>
<td>-</td>
<td>0.365</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>4.8 TM</td>
<td>0</td>
<td>0.025</td>
<td>0.347</td>
</tr>
<tr>
<td></td>
<td>0.02</td>
<td>0.04</td>
<td>-</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>-</td>
<td>-</td>
</tr>
<tr>
<td>6.0 TM</td>
<td>0</td>
<td>0.0037</td>
<td>0.0089</td>
</tr>
<tr>
<td></td>
<td>0.02</td>
<td>-</td>
<td>0.1374</td>
</tr>
<tr>
<td></td>
<td>0.03</td>
<td>-</td>
<td>-</td>
</tr>
</tbody>
</table>

The tensile strength and tensile modulus results of prestressed composites are statistically significant (*p* < 0.05) between unstressed composites at all levels except for 4.8TM yarn composite sample at the prestressing level of 0.03 strain. The improvements in tensile properties observed here are in agreement with results observed by Jorge et al. [101] and Zhao and Cameron [53] on studies related to glass fibre reinforced polyester composites.

### 4.4.3 Dynamic Mechanical Properties of Composites

The dynamic mechanical analysis (DMA) technique can be used to evaluate the performance of composites over a wide range of temperatures. During the test, the response of composites is measured as it is deformed under sinusoidal or other periodic stresses. From the DMA we can get a better view about its quality related to fibre/matrix adhesion, damping behaviour and elasticity [127-132]. Table 4-6 summarises the results of the DMA test. Figure 4-19 and Figure 4-20 are the plot
of storage modulus and loss tangent tan (δ) versus temperature for the composites prepared from the three different levels of twist, i.e., 3.8 TM, 4.8 TM and 6.0 TM. The storage modulus of composites decreased with the increase of temperature. This reduction in modulus is associated with the softening of matrix at higher temperature. When the fibre orientation deteriorates with the increase of yarn twist, there was a decrease in the storage modulus.

Table 4-6 DMA test results of composites from different twist levels.

<table>
<thead>
<tr>
<th>Yarn twist level (TM)</th>
<th>Fibre volume fraction (Vf)</th>
<th>Max. storage modulus (MPa)</th>
<th>Tg (°C) from tan δmax</th>
<th>Peak tan δ</th>
</tr>
</thead>
<tbody>
<tr>
<td>3.8</td>
<td>0.435</td>
<td>11891.5</td>
<td>93.7</td>
<td>0.207</td>
</tr>
<tr>
<td>4.8</td>
<td>0.474</td>
<td>10859.5</td>
<td>90.5</td>
<td>0.195</td>
</tr>
<tr>
<td>6.0</td>
<td>0.494</td>
<td>10244.9</td>
<td>89.3</td>
<td>0.190</td>
</tr>
</tbody>
</table>

Figure 4-19 Effect of twist on the storage modulus of composites.
Figure 4-20 Effect of twist on the loss tangent ($\tan(\delta)$) of composites.

The damping parameter $\tan(\delta)$ is the ratio of the loss modulus to the storage modulus. It is a dimensionless property which is related to the ability of a material to absorb vibrational energy. The damping nature of the composite depends on factors like nature of the matrix, interface or delamination and damping due to energy dissipation in the matrix crack and broken fibres area [131]. Figure 4-20 shows that the value of $\tan(\delta)$ decreased with the increase of yarn twist because poorly aligned fibres are less effective in passing on vibration and more effective in absorbing energy. It could be due to less frictional effect in tightly bound (high twisted) yarns.

The effect of prestress on the dynamic mechanical properties of the composites, DMA was performed on the composites prepared from low twist (3.8 TM) yarn at 3 different levels of prestress. The Figure 4-21 shows the storage modulus of the composites at different prestress levels while Table 4-7 shows the results of DMA test.
Table 4-7 DMA test results of composites prepared at different levels of prestress.

<table>
<thead>
<tr>
<th>Applied strain</th>
<th>Fibre volume fraction (Vf)</th>
<th>Max. storage modulus (MPa)</th>
<th>T_g (°C) from tan δ_max</th>
<th>Peak tan δ</th>
</tr>
</thead>
<tbody>
<tr>
<td>0</td>
<td>0.438</td>
<td>10745.5</td>
<td>101.2</td>
<td>0.167</td>
</tr>
<tr>
<td>0.02</td>
<td>0.433</td>
<td>10608.5</td>
<td>103.8</td>
<td>0.181</td>
</tr>
<tr>
<td>0.03</td>
<td>0.418</td>
<td>10177.4</td>
<td>104.2</td>
<td>0.186</td>
</tr>
</tbody>
</table>

Figure 4-21 Effect of prestress on the storage modulus of composites.

It can be observed that both the storage modulus and the fibre volume fraction (Vf) of the composites decreased with increasing levels of prestress as shown in Table 4-7. With the increase of fibre content, the storage modulus also increases because of inherent stiffness imparted by the fibres resulting in efficient stress transfer. Similar results have been found in other studies [133-135].

The value of tan δ and glass transition temperature T_g increased with the increase of prestress level (coincident with decrease of fibre volume fraction). The tan δ peak became sharper with the increase of prestress level. The improvement
in the value of $\tan \delta$ may be due to the fact the fibre alignment was improved by the application of prestress to the fibres, which resulted in an improvement in the transfer of vibrational energy and reduction in energy absorption.

Figure 4-22 Effect of prestress on the loss tangent ($\tan \delta$) of composites prepared from low twist (3.8 TM) yarns.

4.5 Conclusion

The effect of prestressing during the manufacturing of composites containing unidirectional oriented flax yarn at three levels of twist has been investigated in this chapter. The structure-property relationships were established by correlating the yarn structural parameters such as fibre packing density, angle of inclination in the yarn and fibre alignment to the mechanical properties and thermo-mechanical properties of the resulting composites. It is found that the tension applied during prestressing caused an increase in fibre packing density and a reduction in the angle of inclination in the yarn, resulting in improved fibre
alignment and a decrease in fibre volume fraction in the final composite. The prestressing also results in residual fibre stress in the final composite, which is transferred into compression in the matrix, leading to further improvement of the mechanical performance of the final composite.

The yarn prestressing has resulted in an enhancement of flexural strength and modulus of the low twist 3.8 TM yarn based composites by 29% and 37% respectively. For medium twist 4.8 TM yarn based composites, the respective improvements were 12% and 18% respectively, and 9% and 47% in high twist yarn 6.0 TM based composites respectively.

The flexural properties of the composites showed improvement with the application of prestress prepared from yarns at all three levels of twist. The level at which maximum strength and modulus are exhibited depended on the level of yarn twist and the level of applied prestress during the curing stage.

The low twist yarn based composites showed better mechanical properties than that of the medium twist and high twist yarn based composites. The mechanical properties were shown to decline with the increase of yarn twist. From manufacturing point of view, low twist yarns can be spun at lower costs than medium or high twist yarns, because the throughput of the yarn spinning machine decreases with increasing twist.
5 Study of plied yarn based composites

5.1 Introduction

In this chapter, a range of two-ply yarns are produced by folding two flax singles yarns. The characteristics of plied yarn and its benefits above singles yarn will be discussed. This will be followed by a study of the mechanical properties of plied yarns produced from different single yarns at different levels of ply twist. A study of the structure-property relationship in flax yarn reinforced composites prepared without and with prestressing will be presented. The effects of yarn twist and applied prestress on the properties of the resulting composites will be analysed.

5.2 Plied Yarn Structure and Properties

The fibres in a singles yarn are normally inclined to the yarn axis. The extent of inclination is dictated by the amount of twist imparted to the yarn during spinning. The yarn twist-strength relationship has been discussed in the previous chapter. The fibre inclination angle relative to the direction of applied load should be as low as possible to maximise the mechanical properties of the fibre reinforced composites. The low twisted yarn (to keep the fibre angle to a minimum) does not have sufficient strength to withstand the processing tension. The fibre inclination angle in a twisted yarn can be reduced without compromising the yarn strength by making a two-ply yarn structure. The two-ply yarn is prepared by bringing together two singles yarns in parallel and twisting them together in a direction opposite to that of the constituent singles yarns. The ply twisting technique is widely used in
the textile industry to improve yarn strength and abrasion resistance, to reduce twist liveliness and to achieve certain visual effects [43].

Because the fibre inclination angle in singles yarns varies from zero at yarn centre to a maximum at yarn surface, the ply twist imparted in the opposite direction cannot turn all fibres to the direction parallel to the ply-yarn axis. By changing the level of ply-twist, fibres at one radial position in the singles yarn can be turned to the direction parallel to the two-ply yarn axis while fibres at other radial positions are at varying degrees of angle to the two-ply yarn axis. This results in a reduced average fibre inclination angle and thus an improvement in fibre alignment along the two-ply yarn axis.

5.2.1 Plied Yarn Tensile Properties

Plied yarns were prepared from singles yarn having three different nominal twist levels namely 3.8 TM, 4.8 TM and 6.0 TM with the average linear density of 186 ± 7 tex, 192 ± 5 tex and 195 ± 4 tex respectively. The tensile test results of these constituent singles yarns have been discussed in section 4.3.1. Each type of singles yarn was then plied at varying twist levels with another identical yarn to form a two-ply yarn with average linear density of 370 ± 6 tex, 378 ± 7 tex and 387 ± 8 tex respectively for 3.8 TM, 4.8 TM and 6.0 TM yarns respectively. Figure 5-1 shows the tensile tests results of the plied yarns produced from these three singles yarns. The tenacity of the plied yarns prepared from 3.8 TM singles yarns increased with the level of ply twist. The strength of plied yarns from the 4.8 TM singles yarn increased with ply twist up till the ply/singles twist ratio reached about 70% and then started to decline. The plied yarns prepared from the high twist 6.0 TM singles yarn reached maximum strength at a smaller ply/singles twist level of 35%.
medium ply twist levels, it is seen that the tenacity of plied yarns increased with increase of singles yarn twist (tenacity of 4.8 TM yarns is higher than that of 3.8 TM yarns) and then declined for 6.0 TM yarns. Similar trend was found for singles yarns tensile test results discussed in Figure 4-5. The increase of tenacity from 3.8 TM to 4.8 TM plied yarns is attributed to the increase of friction between the fibres, while the decline of tenacity for 6.0 TM based plied yarns is due to obliquity of the fibres at high twist.

The breaking elongation of the plied yarns is shown in Figure 5-2. The percentage elongation at break of the yarns increased with the increase of singles yarn twist, as well as with the increase of ply twist (for any particular level of singles yarn twist). The increase of breaking elongation with increase of ply twist is due to the fact that with increase of twist, the frictional force between the fibres increases and it takes more time to take the fibres apart, which is shown by higher elongation at break.
Figure 5-1 Tensile test results of plied yarns at different ply twist levels, prepared from singles yarns having three different levels of twist (3.8, 4.8 and 6 TM).

Figure 5-2 Breaking elongation (%) of plied yarns at different ply/singles twist ratios.
5.3 Composites Properties

5.3.1 Density and Fibre Volume Fraction

The composites were prepared in three different sets of plied yarns and each set of the plied yarns were prepared from the 3.8 TM, 4.8 TM and 6.0 TM singles yarns. The ply/singles twist ratio was changed in each set. The density and fibre volume fraction of composites prepared from these plied yarns are presented in Figure 5-3.

The change of ply twist did not have a significant effect on the density and fibre fraction of the prepared composites in each set of singles yarn twist. In fact, the density of the composites prepared at different ply twist levels was all within 3% from the average, i.e., 1.235 ± 0.031, 1.241 ± 0.025 and 1.25 ± 0.026 for 3.8 TM, 4.8 TM and 6.0 TM singles yarn based composites, respectively. Similarly the fibre volume fraction of the as-prepared composites was within 3% of each other 0.455 ± 0.011, 0.458 ± 0.014 and 0.457 ± 0.014 for 3.8 TM, 4.8 TM and 6.0 TM singles yarn based composites, respectively.

Figure 5-4 shows the effect of prestressing on the fibre volume fraction and density of the composites prepared from plied yarns at the 30% ply/single twist ratio for all the three singles yarns 3.8 TM, 4.8 TM and 6.0 TM.
Figure 5-3 (A) Density and (B) Fibre volume fraction of plied yarn based composites.
Figure 5-4 Fibre volume fraction ($V_f$) and density of the composites prepared at different levels of prestress using 30% ply / singles twist ratio.

With the increase of prestress (applied strain), both the fibre volume content and the density of the resulting composites decreased. This is because all the hanks used in the study had the same number of rounds of yarns and the same total fibre volume. The leaky mould used in the experiments produces a composite sample with fixed gap and width, which leads to fixed volume per unit length of the final composite sample. The prestress applied to the yarn hank elongates it due to fibre elongation and fibre slippage in the yarn, i.e. the fibres are spread over a longer length in the final composite sample, which means a reduced fibre volume per unit length of the final composite sample. A similar decrease of the density of the respective composites was observed due to reduced mass of the fibres as a result of prestressing. Similar results have been obtained for the composites prepared from the singles yarns discussed in the previous chapter and the wrap spun yarns to be discussed in the next chapter.
5.3.2 Fibre Packing in Composites

Optical microscope images of the composites cross sections were used to assess the microstructure of the composites. Figure 5-5 shows the cross sections of 3.8 TM singles yarn based composites at ply/singles twist ratios of 0%, 25%, 50% and 75%. The yarns are uniformly distributed in the composites cross section. The yarns in 0% ply twist composites (3.8 TM singles yarn laid parallel to each other without any plying twist), Figure 5-5(A) show nearly circular yarn cross sections. The cross section shape starts to deform with the increase of ply twist. This is because with the increase of the opposite direction ply twist, the twist in the singles yarn decreases and the singles yarn becomes easier to deform under the pressure of compression moulding, as shown in Figure 5-5 B-D.

Figure 5-5 Micrographic images of 3.8 TM singles yarn based plied yarn composites at (A) 0%, (B) 25%, (C) 50% and (D) 75%.
The void content of the composites prepared from plied yarns at different ply/singles twist ratios are given in Figure 5-6. The void content first decreased and then increased with the increasing ply twist with a minimum at ply/single twist ratio of about 25%. This coincides with the ply/single twist ratio of 28% for maximum fibre orientation factor [43]. The void content of the high twist singles yarn (6.0 TM) based composites was always higher than that of the corresponding low twist singles yarn (3.8 TM) based composites. This is due to the fact that with the increase of twist, the fibre packing density in the yarn is higher, resulting in hindrance in resin impregnation. The void content of the composites was in the range of 2.0% ~ 4.2% for 3.8 TM singles yarn based composites and 2.1 ~ 4.8% for the 6.0 TM singles yarn based composites, all of which are in the acceptable range for less demanding applications such as automotive and marine [123, 124, 136].

![Figure 5-6](image.png)

Figure 5-6 Void content of 3.8 TM, and 6.0 TM single yarn based composite at different ratios of ply/singles yarn twist.
5.3.3 Mechanical Properties

Three point bending test and tensile tests were carried out to assess the effect of ply twist and prestressing on the mechanical properties of the resulting composites. Firstly the effect of ply twist/singles yarn twist ratio on composites mechanical performance was assessed for the three singles yarns. Based on these results, the ply/singles twist ratio that produced the maximum flexural strength was determined. The optimum two-ply yarns were then used to study the effect of prestressing.

The results of flexural and tensile tests of the composites prepared from the three sets of plied yarns are presented in Figure 5-7 and Figure 5-8.

As can be seen from Figure 5-7 and Figure 5-8, the flexural properties and tensile properties increased with the increase of ply/singles twist ratio up to about 30% ~ 35% and then declined. This coincides with the theoretical optimum ply/singles twist ratio of 28% [43]. Besides, both flexural and tensile properties decreased with the increase of twist in the constituent singles yarns, which has also been predicted by Gu and Miao using the geometrical model of two-ply yarns proposed [43].

The effect of Krenchel fibre orientation factor given in Equation 5-1 [41] on composite mechanical properties can be assessed using the rule of mixtures equation (Equation 5-2) [137].

\[
\eta_v = \sum a_n \cos^4 \theta_n \quad \text{Equation 5-1}
\]

\[
E_v = \eta_v \eta_i V_f E_f + (1-V_f)E_m \quad \text{Equation 5-2}
\]
Figure 5-7 Flexural test results of plied yarn based composites at different levels of ply/singles twist ratios prepared from (A) 3.8 TM, (B) 4.8 TM and (C) 6.0 TM singles yarns.
Figure 5-8 Tensile test results of plied yarn based composites at different levels of ply twist prepared from (A) 3.8 TM, (B) 4.8 TM and (C) 6.0 TM singles yarns.
As can be seen in Figure 5-7, the flexural properties in all three sets of composites (prepared from 3.8 TM, 4.8 TM and 6.0 TM singles yarns) achieved a maximum value at a ply twist level of 30% ~ 35% ply twist level. Similarly the tensile properties achieved a maximum level at 30% ~ 35% ply twist level for the same set of composites. This indicates that the contribution of fibre orientation (described by Krenchel fibre orientation factor) achieved a maximum value at these 30% ~ 35% ply twist levels. These experimentally found values of ply-yarn twist / singles yarn twist are very close to the theoretical value of 28% (ply twist ratio of 0.28) predicted by Gu and Miao [43] analysing the differential geometry of fibre trajectory using an idealised twisted yarn model. The mechanical properties declined with the increased of twist in the constituent singles yarns.

5.3.4 Effect of Prestressing on Mechanical Properties

Based on the previous results, 30% is the level of ply/singles twist ratio to achieve maximum flexural and tensile properties. Figure 5-9 shows the results of flexural tests of the composites fabricated without prestressing and with two levels of prestressing.

It can be seen from Figure 5-9 that both the flexural strength and modulus slightly decreased with the application of prestress during the composite curing. Statistical t-test was performed on the flexural test results to determine if there was a significant difference in results due to prestressing. The results of the t-test performed on the flexural test results of prestressed composites are presented in Table 5-1. The decline in flexural strength and flexural modulus due to prestressing was statistically significant at most of the high prestressing levels. The decline in flexural properties may be attributed to the deterioration of fibre orientation due to
applied prestress and the slight decrease in the fibre volume fraction of the prepared composites. The fibre volume fraction decreased because all the hanks used in this study had the same number of rounds of yarn and all composite samples had same volume. The leaky mould produced all composite samples with fixed height and width, leading to a fixed volume per unit length of the final composite sample. With the application of prestress, the fibres elongate and slip over each other resulting in increased yarn length and corresponding decrease of fibre mass per unit length of the composite [118]. This in turn results in decrease of fibre volume fraction with the application of prestress.

The surface fibre inclination angle with the plied yarn axis at different levels of applied prestress was measured using digital SLR camera and the results are given in Figure 5-10. It is seen that the fibre inclination angle increased with the increase of applied prestress at all levels of twist. Figure 5-11 shows the fibre volume fraction of composites at different levels of prestress. It is seen that the fibre volume fraction decreased with the increase of applied prestress.

![Figure 5-9](image_url) (A) Flexural strength, (B) Flexural modulus of plied yarn based composites manufactured at different levels of applied strain.
The effect of prestressing on composites prepared from plied yarns is opposite to that on composites prepared from singles yarn (presented in section 4.4.2.1). The improvement in flexural properties in singles yarns has been attributed to the improvement in fibre alignment due to prestressing. Whereas in composites prepared from plied yarn, the prestressing resulted in deterioration of fibre alignment to the yarn axis.

Table 5-1 *t-test* results performed on flexural test results of plied yarn based prestressed composites.

*p-value = probability of prestressing having no effect.*

<table>
<thead>
<tr>
<th>Yarn TM</th>
<th>Applied strain</th>
<th>Flexural strength</th>
<th>Flexural Modulus</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>0</td>
<td>0.012</td>
<td>0.018</td>
</tr>
<tr>
<td>3.8</td>
<td>0</td>
<td>-</td>
<td>0.2493</td>
</tr>
<tr>
<td></td>
<td>0.012</td>
<td>-</td>
<td>0.4321</td>
</tr>
<tr>
<td>4.8</td>
<td>0</td>
<td>0.016</td>
<td>0.022</td>
</tr>
<tr>
<td></td>
<td>0</td>
<td>-</td>
<td>0.3051</td>
</tr>
<tr>
<td></td>
<td>0.016</td>
<td>-</td>
<td>0.0757</td>
</tr>
<tr>
<td>6.0</td>
<td>0</td>
<td>0.022</td>
<td>0.034</td>
</tr>
<tr>
<td></td>
<td>0</td>
<td>-</td>
<td>0.0474</td>
</tr>
<tr>
<td></td>
<td>0.022</td>
<td>-</td>
<td>0.0920</td>
</tr>
</tbody>
</table>
Figure 5-10 Fibre inclination angle in plied yarns at different levels of applied strain.

Figure 5-11 Fibre volume fraction of composites at different levels of prestress.
5.3.5 Summary

The effect of yarn structure and prestressing during the fabrication of composites containing unidirectional plied yarns prepared from singles yarns having a range of twist levels and the plied yarns having varying levels of ply yarn / singles yarn twist ratio has been investigated in this chapter. The structure-property relationships were determined by relating the plied yarn structure parameters (singles yarn twist and level of plied / singles twist ratio) imparted during yarn production to the mechanical properties of the resulting composites. We found that the level of ply twist in the yarn was an important factor to the mechanical properties of the resulting composites.

With the change of ply twist, the resulting fibre orientation in relation to the plied yarn axis changes and at a certain ply twist level, the overall fibre alignment to the plied yarn axis reaches its optimum. The composite showed maximum mechanical performance at the ply/singles twist level of 30% to 35%, which coincided with the theoretical prediction based on idealised yarn geometry. This optimum ply/singles twist level is smaller than the 50% ply/singles twist ratio normally adopted for textile applications.

Prestressing did not improve the mechanical performance of the two-ply yarn based composites. The prestressing force was found to deteriorate fibre orientation on the surface of the plied yarn.

It is also found that regardless of the singles yarn twist, the fibre orientation in the resulting plied yarn can be varied by changing the ply twist ration, to bring the fibres to an optimum overall alignment to the plied yarn axis to fully utilize the reinforcing efficiency of the two-ply yarn.
The manufacture of the plied yarn involves an additional twisting process, which adds cost and energy consumption. With a careful selection of singles yarn twist and the level of (ply) twist to be imparted to the plied yarn, a compromise can be made between the cost and mechanical performance of the resulting composites.
CHAPTER SIX

6 Study of Wrap Spun Yarns Based Composites

6.1 Introduction

This chapter covers the study related to unidirectional composites prepared from flax wrap spun yarns. It starts with the description of wrap spun yarn and the effect of different structural parameters on its mechanical properties. The structure-property relationship in flax yarn reinforced composites prepared from these wrap spun yarn will be presented by inter-correlating the wrap yarn structure to the physical and morphological properties of the composites. This is followed by the study related to effect of prestressing on the mechanical properties of the as-prepared composites.

6.2 Wrap Spun Yarn Structure and Properties

Wrap spun yarn (also known as hollow spindle wrap yarn, cover spun yarn or parallel yarn) has a complex structure which consists of a core and a wrapper. The core is formed by a twistless parallel staple fibre strand which is normally delivered from the front roller nip of the drafting system, while the wrapper is normally a multifilament or monofilament thread, as shown in Figure 6-1. The wrapping filament applies fibre-to-fibre pressure which is needed to create the friction between the fibres and to impart yarn strength [138]. Because of low fibre-to-fibre pressure generated by the wrapping filament as compared to that generated by the twist in the yarn, wrap spun yarns generally have lower
tensile strength and are less compact as compared to ring spun yarns at their optimal twist level.

![Figure 6-1 Structure of a wrap spun yarn.](image)

The structure of the hollow spindle wrap yarn is also affected by the linear density of the wrapping filament because the fibres in the staple core show some degree of disorientation relative to the yarn axis [139]. Most of the wrap spinning machines are equipped with false twist hooks fixed to the exit end of the spindle bore. The main purpose of using a false twister is to provide a certain temporary strength to the yarn core before it is wrapped by the filament wrapper.

The photographs in Figure 6-2 and Figure 6-3 were taken from pairs of wrap yarns produced under similar conditions except for the application of false twist during spinning. It can be seen from the two figures that with the increase of wrap twist, the difference between yarns produced with or without false twist (FT) becomes prominent. The FT yarns in Figure 6-2 are wavier (more tortuous) than the corresponding NFT yarns in Figure 6-3.
In order to understand the effect of the type of wrapper filament and the wrapping density (twist) on the tensile properties of the corresponding wrap yarns, two different types of filaments were used. Yarns were prepared at different levels of wrap twist with and without the application of false twist. The characteristics of the multifilament yarns are given in Table 6-1.
Table 6-1 Characteristics of polyester multifilament yarns used as wrapper.

<table>
<thead>
<tr>
<th>Description</th>
<th>Number of filaments</th>
<th>Linear density (dTex)</th>
<th>Initial Modulus (cN/tex)</th>
<th>Tenacity (cN/tex)</th>
<th>Elongation at Max. Load (%)</th>
<th>Breaking Elongation (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Flat Filament</td>
<td>24</td>
<td>78.4</td>
<td>360.3</td>
<td>24.59</td>
<td>16.8%</td>
<td>19.7%</td>
</tr>
<tr>
<td>Textured filament</td>
<td>13</td>
<td>33.9</td>
<td>510.9</td>
<td>36.58</td>
<td>23.8%</td>
<td>25%</td>
</tr>
</tbody>
</table>

The wrap yarn twist which is expressed as number of wraps (rounds) of the wrapping filament around the straight fibre core was measured on a hand-driven twist tester while the wrapping filament helix angle was measured from images taken using a Digital SLR camera. Figure 6-4 and Figure 6-5 show the actual wrap twist compared to nominal wrap twist and wrapping filament helix angle using flat and textured filaments, respectively. The wrap twist in the yarns spun with false twist was slightly higher than those without wrap twist and their difference became prominent as the nominal wrap twist (ratio of spindle speed to front roller surface speed) increased. A larger difference was seen when the wrapping filament helix angle was plotted against nominal twist.

Figure 6-4 (A) Actual wrap twist and (B) Wrapper filament helix angle at various levels of nominal wrap twist of wrap yarns spun using flat filament.
Figure 6-5 (A) Actual wrap twist and (B) Wrapper filament helix angle at various levels of nominal wrap twist of wrap yarns spun using textured filament.

6.2.1 Tensile Properties

Breaking load, tenacity and initial modulus of yarns prepared from flat and textured filaments are shown in Figure 6-6 and Figure 6-7. Figure 6-8 shows the typical tensile test curves of the tested yarns.

It can be seen that generally the strength and tenacity increased with the increase of wrap twist imparted to the yarns. In the flat filament based yarns, strengths (breaking load and tenacity) of FT yarns and NFT yarns were approximately the same below 140 wraps/m, above which the tensile strength of the FT yarns exceeded that of the corresponding NFT yarns. In case of textured filament yarns, the strength and tenacity of FT yarns was higher than that of NFT yarns and the difference between FT & NFT increased with the increase of twist. It can also be observed that the strength and tenacity of flat filament based yarns were higher than those of textured filament based yarns.

The increase of breaking load and tenacity with increase of wrap twist is caused by the fact that with the increase of wrap twist, the mutual support between the fibres increases which is similar to the role of twist in case of ring
spun yarns where the friction between fibres increased with the increase of twist [88]. The breaking load and tenacity of flat filament based yarns were higher than those of textured filament based yarns because of the fact that with the increase in linear density of the wrapping filament, the inter-fibre friction forces also increase due to development of higher radial forces and an increased filament-to-fibre surface contact. This may cause an increase in mutual support between the fibres resulting in an increase of yarn strength and tenacity. The flat filament used in this study had breaking load of 189.4 cN which is around 55% higher than that of the textured filament, which has a breaking load of 121.9 cN. Similarly the linear density of the flat filament was 78.4 dTex, which is more than double the linear density of the textured filament (33.9 dTex). The results in this study are similar to the results reported by H. M. Behery et al [139] and Rottmayr and Brosch [140].
Figure 6-6 Breaking Load (A, B), Tenacity (C, D) and Initial Modulus (E,F) of yarns prepared from flat and textured filaments at different levels of wrap twist, with and without false twist. Flat filaments: A, C, E; Textured filaments: B, D, F.
Figure 6-7 Effect of filament type and wrap twist on breaking elongation of FT & NFT yarns prepared with textured and flat filament as wrapper.

Figure 6-8 Typical tensile test curves for wrap spun yarns prepared from (A, B) Flat filament, (C, D) Textured filament, at different wrap twist levels.
The yarn modulus in flat filament NFT yarns increased with the increase of wrap twist up to a level and then became stable, while modulus of FT yarns declined after attaining a certain maximum level. Conversely the yarn modulus of the FT and NFT yarns prepared from textured filament increased with the increase of the wrap twist in the yarns. Yarn modulus reflects the fibre modulus (assuming the yarn is twistless) and the degree of inter-fibre movement (i.e. slippage) during tensile loading. When the lateral pressure in the core is sufficiently high so that no inter-fibre slippage can take place, further increase in wrapping density and tension have little effect on the yarn modulus. The decline of FT yarn modulus after achieving the maximum value is due to the increased tortuosity which increases with increase of wrap twist in FT yarns as explained in next section. The modulus of textured filament based yarns increased with the increase of wrap twist in the tested range (90 ~ 180 wrap/m) like that of the flat filament based wrap yarns.

Typical tensile curves are shown in Figure 6-8. It can be seen that with the increase of wrap twist of both FT and NFT yarns, the tenacity increased with gradual decrease of breaking elongation of the yarn. In both textured and flat filament based NFT yarns, the breaking elongation was much higher than that of the corresponding FT yarns at same wrap twist levels. At low wrap twist density in NFT yarns, the fibres slipped over each other by the application of tensile load while the filament was being stretched. This is shown by the fluctuation in the tensile curve. The yarn failed at the filament breakage under tensile load.
6.2.2 Wrap Yarn Tortuosity

During the wrap yarn spinning, false twist is sometimes inserted to the fibres emerging from the delivery roller of the spinning machine, to impart temporary strength to the fibrous strand. This false twist forms spiral shaped yarn structure or tortuous yarn. The false twist was found to play an important role in the yarn structure with the formation of spiral shape in the wrap spun yarns, affecting significantly the mechanical properties as observed in the tensile testing results presented in Figure 6-6 and Figure 6-7.

Figure 6-9 shows a typical cell of the wrap yarns shown in Figure 6-2 and Figure 6-3. The twistless staple core has a circular cross section (diameter $d$), and the wrapping filament and the core each form a helix (with helix radii $d_1/2$ and $d_2/2$, respectively).

Yarn tortuosity factor $r$ is defined as the ratio:

$$ r = \frac{d_2}{d}. $$
A straight cylindrical wrap yarn as proposed by Xie et al. [138] has zero tortuosity because \( d_2 = 0 \).

It can be calculated by taking into consideration the yarn linear density, twist factor and yarn volume density, and can be calculated by using the formula given in Equation 6-1 [45].

\[
r = 1 - 89.34\sqrt{\frac{\tan \theta}{\alpha_t}}
\]

Equation. 6-1.

where \( \alpha_t = T \sqrt{C} \) is the twist factor (tex\(^{1/2}\).turns/cm), \( C \) = yarn count in tex, and \( \rho \) = yarn volume density (g/cm\(^3\)) which can be calculated from yarn tex count and yarn diameter measured from digital images.

The effect of yarn wrap twist and false twist on the tortuosity of the yarns are shown in Figure 6-10. It can be seen in Figure 6-10 (A & B) that with the increase of wrap twist, the tortuosity in the FT yarns increased while decreased in the NFT yarns. In Figure 6-10 (A), the tortuosity reached 0.1 approaching perfect straight cylindrical yarn structure. Also it can be seen that the tortuosity of textured filament yarns was higher than the flat filament yarns. This trend can be explained by the yarn formation model presented by Miao et al. [45]. At low wrap twist of yarn, the buckling moment generated by the wrapping filament is the major cause of the tortuosity for both FT and NFT yarns. As the wrap twist increases, the wrapping pitch \( (l / T) \) becomes smaller so that bending span shortens and yarn core plays a smaller role to the wrapping filament buckling action. For the FT yarns, higher wrap twist results in increased core twist.
contraction during the initial wrapping stage, which in turn results in continued increase in yarn tortuosity with increasing wrap twist.

Figure 6-10 Effect of wrap twist on yarn tortuosity (A) Flat filament yarns, (B) Flat filament versus textured filament yarns.
6.3 Composites properties

6.3.1 Density and Fibre Volume Fraction

Composite samples were prepared from both false twist (FT) and no false twist (NFT) yarns at varying levels of wrap twist to study the effects of false twist and wrap twist on the composites properties. Figure 6-11 (A, C) shows the results of NFT yarns based composites, while Figure 6-11 (B, D) shows the results of FT yarns based composites.

Figure 6-11 Effect of wrap twist on density and void content of the composites prepared from (A, C) NFT yarns, (B, D) FT yarns.
In Figure 6-11 we can see that there was no clear trend in density variation with the increase of the constituent yarn twist. On the other hand, the void content of the composites demonstrate a tendency of increase with the increase of yarn wrap twist. This is because that with the increase of twist, the fibres are packed tighter inside the yarn, which results in decreased resin penetration inside the yarn. The void content of the FT yarns based composites was slightly higher than that of NFT yarn-based composites, which indicates that the resin impregnation in NFT yarn based composites is better than FT yarn based composites. The mean fibre volume fraction of the FT & NFT yarn based composites at different twist levels were all found to be similar, 0.455 ± 0.017 and 0.471 ± 0.021 respectively and are given in Figure 6-12.

![Figure 6-12 Fibre volume fraction of the composites prepared at different levels of wrap twist prepared from (A) NFT yarns, (B) FT yarns.](image)

Figure 6-13 shows the effect of prestressing on the fibre volume content and void content of the composites manufactured from both FT & NFT yarns. The fibre volume content ($V_f$) showed a slow trend of decrease with the applied prestress in both FT & NFT composites. The unstressed composites had an
average fibre volume fraction of 0.47 which decreased to 0.43 at 0.05 strain level. The decrease in fibre volume fraction in the composites is consistent with the level of strain applied to the yarns during compression moulding. This is because all the hanks used in the study had the same number of rounds of yarns and all composite samples had the same volume. The leaky mould used in the experiments produces a composite sample with fixed gap and width, which leads to fixed volume per unit length of the final composite sample. Prestress applied to the yarn hank elongates it due to fibre elongation and fibre slippage in the yarn, i.e. the fibres are spread over a longer length in the final composite sample, which means a reduced fibre volume per unit length of the final composite sample. Similar results were obtained for composites prepared from 140 t/m and 180 t/m yarns in both FT & NFT types which are presented in Figure 6-14.

Figure 6-13 Effect of prestress on (A) fibre volume content and (B) void content of the 90 t/m yarn based composites.
As shown in Figure 6-13 (B), the void content reduced dramatically with the applied prestress for both FT & NFT yarn based composites. This is due to the fact that during prestressing, the fibres slide over each other and become better aligned to the yarn axis as a result of tensioning. The improvement of fibre alignment is shown by the decline in tortuosity of the wrap yarns due to prestressing in Figure 6-15. Wrap yarn tortuosity due to prestressing was determined on the Instron tensile testing machine. The yarn specimen between the jaws was stretched to a predetermined strain level and the machine was stopped to take photographs of the yarn using a digital SLR camera. The digital
images were processed using ImageJ software to measure wrapping filament orientation and yarn diameter at the corresponding strain levels, from which the yarn tortuosity was calculated. Figure 6-16 shows representative images of 280 t/m FT yarns at different levels of prestress.

Figure 6-15 Wrap yarn tortuosity at different strain levels.

Figure 6-16 Images of 280 t/m FT yarns at different strain levels. (A) No strain, (B) 0.03 strain, (C) 0.05 strain.
Figure 6-15 shows that with the application of strain, the wrap yarn tortuosity for all the yarns decreased. All the FT yarns had higher tortuosity than the corresponding NFT yarns. The decline in the yarn tortuosity with the application of strain (prestress) results in improved alignment of the fibres, improved resin impregnation and decreased void content in the composites, as shown in Figure 6-11 to Figure 6-14. In Figure 6-16 it can be seen that with the application of prestress, the yarns become straighter which means that the tortuosity of the yarn decreased with the application of prestress.

### 6.3.2 Fibre Packing in Composites

Optical microscope observations of the composite cross sections were used to assess the microstructure of the composites. Figure 6-17 shows representative images of composites prepared at different levels of wrap twist in both FT and NFT yarns. It is evident that the flax yarns are well dispersed in the matrix and fibres are well impregnated with the matrix with few occurrences of voids. It can be seen that with the increase of wrap twist, yarn becomes more compact and more instances of resin rich regions occur in the cross sections.

Figure 6-18 shows representative images of composites prepared from 90 wraps/m FT & NFT yarns at different prestress levels. As can be seen in the images, in the absence of prestress, the FT & NFT yarns did not show any boundaries, indicating that loose yarn structure was pressed together. At applied prestress of 0.03 and 0.05, yarn boundaries became clear. The reason is that with the application of prestress, the fibres in the yarn are packed together and yarn assumes a near circular cross section, which is evident in Figure 6-18 (B, C, E and F).
When comparing these images with those of singles yarn based composites presented in chapter 4 (Figure 4-11 and Figure 4-12), it can be seen that the wrap spun yarn were not as circular in cross section as the ring spun singles yarns. Fibres in the wrap spun yarn based composites are distributed more evenly throughout the cross sectional area while in the singles yarn based composites, fibres in the yarns had concentrated distribution with noticeable resin rich regions within the boundary of the yarn.

It is also evident that the flax yarns are well dispersed in the matrix and fibres are well impregnated with the matrix with few occurrences of voids. Figure 6-17 shows the cross section images of composites at different levels of wrap twist in both FT & NFT yarns. It can be seen that with the increase of wrap twist, yarns become compact which is shown by more instances of resin rich regions in the cross sections.

The void content of the wrap spun yarn based composites, shown in Figure 6-17 was found out to be in the range of 1.5 ~ 4.0%, which is comparable to that of glass fibre based composites (1~3%) [80].
Figure 6-17 Microscopic images of composites prepared from different twist levels. (A) 90 wpm NFT, (B) 140 wpm NFT, (C) 180 wpm NFT, (D) 90 wpm FT, (E) 140 wpm FT and (F) 180 wpm FT.
6.3.3 Mechanical Properties of Composites

6.3.3.1 Effect of Wrap Twist Level and False Twist

Three point bending tests were used to measure the mechanical performance of composites so as to assess the effects of wrap twist and false twist of wrap spun yarns. The flexural test results are presented in Figure 6-19.
The flexural strength and flexural modulus of the composites prepared from NFT yarns (Figure 6-19 A) increased with the increase of wrap twist up to a certain level and then declined. The increase of wrap twist in NFT yarn is accompanied by the decrease of yarn tortuosity in the whole range of wrap twist levels. On the other hand, both flexural strength and flexural modulus of the composites prepared from FT yarns continuously declined with increasing wrap twist in the yarn (Figure 6-19 B). In this case, the tortuosity of yarns increased with increasing wrap twist.

These results can be explained on the basis of the structure of the wrap spun yarns. In FT yarns, the false twist imparted to the fibres forms the yarn in spiral shape which is described by the term “tortuosity”. The higher the tortuosity, the poorer will be the fibre alignment along the yarn axis. Due to the decline in fibre orientation factor, the overall mechanical performance of the composites declines, as evident in Figure 6-19.
6.3.3.2 Effect of Prestressing on Composite Mechanical Properties

Composites were prepared from four different wrap twist levels namely 90, 140, 180 and 280 wraps/m from FT yarns and NFT yarns. Yarns were prestressed to strain levels of 0.03 and 0.05 (3% and 5% of the original length) in all yarns during resin curing. The flexural and tensile properties of the prestressed composites were compared to those of the unstressed composites. Figure 6-20 shows the flexural test results while Figure 6-21 shows the tensile
test results of the composites prepared at different levels of prestress and twist levels.

The flexural properties of both FT and NFT yarn based composites were enhanced by different levels between 9% to 39.7%, except for the composite material prepared from the highest wrapping density FT yarn (280 wraps/m), where no improvement in flexural performance was observed.

Similarly, the improvements in tensile strength and modulus were higher for composites prepared from NFT yarns than for those prepared from FT yarns. The tensile properties were also found to be improved by varying extent for composites prepared from both FT and NFT yarns except for the 280 wraps/m FT yarn based composites, as shown in Figure 6-21. This might be attributed to the high lateral pressure exerted on the flax fibres by the wrapper filament and the high tortuosity of the yarns. The applied prestress force would not introduce fibre slippage and straightening and instead the yarn would undergo elastic elongation.

The fibre volume fraction of the prestressed composites prepared from wrap spun yarns were found to slightly decrease with increasing prestress; which was similar to that found in prestressed composites prepared from singles and plied yarns discussed in sections 4.4.1 and 5.3.1. It can be inferred that the observed improvement in mechanical properties was not affected by the decrease of fibre volume content of the prestressed composites.
Figure 6-20 Flexural test results of composites from four levels of wrap twist at different levels of applied prestress.
Figure 6-21 Tensile test results of composites from four different levels of wrap twist at different levels of applied prestress.

In summary, in the wrap spun yarns, the twistless parallel staple fibres are held together by the continuous-filament wrapper which exerts a radial pressure providing necessary frictional forces between the individual staple fibres. With the increase in wrapping density (wraps/m of the filament around the parallel
fibres), the inter-fibre friction forces also increase due to the development of higher lateral pressure exerted by the wrapper and greater filament-to-fibre surface contact. All of the fibres in the yarn are not at same tension level exerted by the wrapper filament, the fibres on the surface which are in contact with the wrapper filament are under higher tension than those located inward and in the centre of the yarn. When an external load is applied to the yarn, the fibres located in the middle or centre of the yarn can slip slightly relative to the fibres located on the surface and in contact with the filament so that these fibres can straighten, resulting in a small permanent increase of yarn length. As a results, the fibre tension tends to equalize, i.e., the fibres on the surface build up tension and the fibres in the middle or in the core reduce their tension because the external load is shared by more fibres. At low prestressing levels, fibre straightening and slippage dominate, causing irrecoverable yarn elongation. When higher prestressing tension is used, after the initial fibre straightening and slippage, the fibres themselves tend to elongate. The fibre straightening as a result of slippage and fibre elongation by the further application of prestressing force, depends on the levels of wrap twist and whether false twist is applied to the yarns or not. As can be seen in the results presented in this chapter, the flexural strength and flexural modulus of both FT and NFT yarn based composites improved by prestressing, except for the 280 wraps/m FT yarn based composites, where no improvement in flexural performance was observed. This can be attributed to very high later pressure exerted by the wrapper filament and high tortuosity of the yarns so that the applied prestress force would not introduce fibre slippage and straightening.
6.3.4 Dynamic Mechanical Properties

The dynamic mechanical analysis (DMA) was used to evaluate the performance of composites over a range of temperatures. Table 5-2 shows the results of DMA test performed on the composites prepared from different levels of wrap twist in both FT & NFT yarn types, while Table 5-3 shows the results of prestressed composites prepared from 280 wraps/m FT & NFT yarns.

In Table 6-2 and Figure 6-22 it can be seen that the storage modulus of the composites increased with the increase in wrap twist of the NFT yarn based composites, while that of FT yarn composites decreased with increasing wrap twist. It means that the addition of wrap twist in NFT yarns allows greater stress transfer at the interface and ultimately increased the storage modulus. Conversely the storage modulus of the FT yarn based composites decreased with increasing wrap twist. These results are consistent with the flexural static test results of these composites where flexural properties reported in section 6.3.3.1. Because the fibre content of the tested specimens was nearly the same, the change in storage modulus can be attributed solely to the effect of wrapping twist density on the mechanical properties.
Table 6-2 DMA test results of composites prepared from different levels of wrap twist in both FT & NFT yarns.

<table>
<thead>
<tr>
<th>Sr. #</th>
<th>Yarn Wraps / m</th>
<th>FT / NFT</th>
<th>Avg. $V_f$</th>
<th>Max. storage modulus (MPa)</th>
<th>$T_g$ from $E''_{\text{max}}$ (°C)</th>
<th>$T_g$ from Tan $\delta_{\text{max}}$ (°C)</th>
<th>Peak height of Tan $\delta$ curve</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>90</td>
<td>NFT</td>
<td>0.478</td>
<td>10652.9</td>
<td>96.1</td>
<td>106.3</td>
<td>0.222</td>
</tr>
<tr>
<td>2</td>
<td>140</td>
<td>FT</td>
<td>0.460</td>
<td>11397.8</td>
<td>94.5</td>
<td>103.8</td>
<td>0.213</td>
</tr>
<tr>
<td>3</td>
<td>180</td>
<td>FT</td>
<td>0.475</td>
<td>12254.3</td>
<td>93.4</td>
<td>102.9</td>
<td>0.233</td>
</tr>
<tr>
<td>4</td>
<td>280</td>
<td>FT</td>
<td>0.467</td>
<td>13005.9</td>
<td>88.1</td>
<td>102.0</td>
<td>0.173</td>
</tr>
<tr>
<td>5</td>
<td>90</td>
<td>FT</td>
<td>0.493</td>
<td>11378.8</td>
<td>89.4</td>
<td>102.4</td>
<td>0.196</td>
</tr>
<tr>
<td>6</td>
<td>140</td>
<td>FT</td>
<td>0.462</td>
<td>9711.9</td>
<td>95.6</td>
<td>104.2</td>
<td>0.212</td>
</tr>
<tr>
<td>7</td>
<td>180</td>
<td>FT</td>
<td>0.447</td>
<td>9384.3</td>
<td>84.7</td>
<td>103.2</td>
<td>0.207</td>
</tr>
<tr>
<td>8</td>
<td>280</td>
<td>FT</td>
<td>0.438</td>
<td>8643.2</td>
<td>89.3</td>
<td>102.4</td>
<td>0.219</td>
</tr>
</tbody>
</table>

![Graphs](image)

Figure 6-22 Storage modulus of composites at different wrap twist levels (A) NFT yarns, (B) FT yarns; and different prestress levels, (C) 280 wraps/m NFT yarns at different prestress levels, (D) 280 wraps/m FT yarns at different prestress levels.
Table 6-3 DMA test results of composites prepared from 280 wrap/m FT & NFT yarns at different levels of prestress.

<table>
<thead>
<tr>
<th>Sr. #</th>
<th>FT / NFT</th>
<th>Applied strain</th>
<th>Avg. $V_f$</th>
<th>Max. Storage Modulus (MPa)</th>
<th>$T_g$ from $E''_{max}$ ($^\circ$C)</th>
<th>$T_g$ from $\tan \delta_{max}$ ($^\circ$C)</th>
<th>Peak height of $\tan \delta$ curve</th>
</tr>
</thead>
<tbody>
<tr>
<td>1</td>
<td>NFT</td>
<td>0</td>
<td>0.467</td>
<td>13005.9</td>
<td>88.1</td>
<td>102.0</td>
<td>0.173</td>
</tr>
<tr>
<td>2</td>
<td>NFT</td>
<td>0.03</td>
<td>0.449</td>
<td>13258.5</td>
<td>97.1</td>
<td>106.3</td>
<td>0.205</td>
</tr>
<tr>
<td>3</td>
<td>NFT</td>
<td>0.05</td>
<td>0.432</td>
<td>12204.5</td>
<td>96.8</td>
<td>104.5</td>
<td>0.215</td>
</tr>
<tr>
<td>4</td>
<td>FT</td>
<td>0</td>
<td>0.438</td>
<td>8643.2</td>
<td>89.3</td>
<td>102.4</td>
<td>0.219</td>
</tr>
<tr>
<td>5</td>
<td>FT</td>
<td>0.03</td>
<td>0.425</td>
<td>8542.0</td>
<td>87.5</td>
<td>103.6</td>
<td>0.221</td>
</tr>
<tr>
<td>6</td>
<td>FT</td>
<td>0.05</td>
<td>0.418</td>
<td>8419.3</td>
<td>93.6</td>
<td>106.0</td>
<td>0.235</td>
</tr>
</tbody>
</table>

Figure 6-23 Loss modulus of composites at different wrap twist levels (A) NFT yarns, (B) FT yarns; and different prestress levels, (C) NFT yarns at different prestress levels, (D) FT yarns at different prestress levels.
In case of effect of prestressing on the storage modulus, it is shown that there was a slight increase in storage modulus of the composites prepared from NFT yarns and then the modulus declined, while in case of 280 wraps/m FT yarn based composites, a slight decline was observed in the storage modulus. Again these results are similar to the flexural test results.

The damping parameter \((\tan \delta)\) which is the ratio of loss modulus to the storage modulus shows the damping nature of the composites which depends on factors like nature of the matrix, interface or delamination and damping due to energy dissipation in the matrix crack and broken fibres area [131]. It is observed in Figure 6-24 that the damping factor increases with the increase in temperature, and reaches at a maximum level in transition region then decreases in the rubbery region. It is also observed that the peak values of the \(\tan \delta\) curves decreased with the increasing twist in NFT yarn based composites despite of no change in their fibre volume fraction. The results can be attributed to the fact that with the increase of wrap twist, more pressure is exerted by the wrap filament on the fibres. This results in reduced molecular mobility and hence decreasing trend in \(\tan \delta\) values. In the case of FT yarn based composites, the peak \(\tan \delta\) values increased with increasing wrap twist, which may indicate increased molecular mobility besides the fact that the storage modulus decreased with increasing twist. It is also shown in Table 6-2 that the fibre content of these samples decreased with increasing twist, which may also contribute towards the increase of \(\tan \delta\) peak values. As shown in Table 6-3, the \(\tan \delta\) peak of the prestressed composites in both NFT & FT yarns increased with the increase of prestress. This is due to the decrease of fibre content with increasing prestress, because the composites were prepared using a controlled gap between the mould parts.
(thickness), and the fibre content decreased with increasing prestress. If the process would have been done with a controlled prestressing force (achieving thinner samples with prestressing) the results obtained would have been opposite as $V_f$ would have gone up with prestress. An increase of peak tan δ values also indicates increased mobility of molecular chains with increase of prestress to both FT & NFT yarn based composites. It means that with the application of prestress, there was a relaxation in the fibres after being extended by the applied tensile force and decrease of lateral pressure exerted by the wrapping filament.

Figure 6-24 Damping factor (tan δ) of composites at different wrap twist levels (A) NFT yarns, (B) FT yarns; and different prestress levels, (C) NFT yarns at different prestress levels, (D) FT yarns at different prestress levels.
6.4 Conclusion

In this chapter, the effect of yarn structure and prestress applied during the manufacturing of composites has been studied. Flax fibre wrap spun yarns with varying levels of wrap twist were prepared with and without applying false twist. The structure property relationships were established by correlating the yarn structural parameters such as wrap twist density and presence or absence of false twist, and the manufactured yarn to the mechanical properties and thermo-mechanical properties of the resulting composites. We found that the wrap twist density and the false twist applied to the yarn during its manufacture significantly affected the curliness of the yarns technically known as tortuosity. The yarn tortuosity in turn had a prominent effect on the mechanical properties of the manufactured composites. The prestressing also results in residual fibre stress in the final composite, which is transferred into compression in the matrix, leading to further improvement in the mechanical performance of the resulting composites.

It was found that the false twist applied to the yarns during manufacture adversely affected the mechanical properties of the composites because of higher tortuosity of the constituent yarn, on the other hand increasing the wrap twist (without imparting false twist) resulted in enhancement of mechanical properties.

The yarn prestressing has resulted in the enhancement of flexural strength by 16.2%, 15.3% and 31.2% and flexural modulus by 26.7%, 18.2% and 35.1% respectively of the composites prepared from 90, 140 and 180 wraps/m NFT yarns. Similar improvements were found in the mechanical
properties of the composites prepared from FT yarns. The prestressing level applied to the yarn during manufacturing process dictates the level of improvement and there exists an optimum strain level like 0.05 found in the present study for NFT yarn based composites resulting in the maximum enhancement in the mechanical performance.
CHAPTER SEVEN

7 Conclusions

The work in this thesis has focused on studying the structure-property relationship between flax yarn structures and the mechanical performances of resulting unidirectional yarn composites. Three different types of yarn structures, namely singles yarns, wrap spun yarns and plied yarns, were used in the study. For each type of yarn, the effects of yarn structural parameters and level of pre-stressing during composite fabrication on the mechanical properties of the resulting composites were studied.

In singles yarns based composites, the composites mechanical properties were found to be dependent on the level of yarn twist as well as applied prestress. In the absence of prestress, the mechanical properties were found to decline with the increase of twist in the singles yarn. At a given twist level, prestressing improved the mechanical properties of the resulting composites up to a certain level before declining, but the degree of improvement of mechanical properties due to prestressing decreased with the increase of twist in the constituent singles yarn. Prestressing caused an increase in fibre packing density and a reduction in fibre inclination angle in the yarn, which resulted in improvement in fibre alignment in the final composites. The prestressing force resulted in a residual tensile stress in the fibres which was transferred into compression stress in the matrix, leading to a further improvement of the mechanical performance of the composites.

The low twist yarn based composites showed better mechanical properties that the medium twist and high twist yarn based composites. From manufacturing point of view, low twist yarns have lower process related costs than medium or high
twist yarns, because the throughput of the yarn spinning machine decreases with increasing twist, leading to higher production costs for higher twist yarns.

The study related to two-ply yarn based composites involved the use of plied yarns at different levels of ply/singles twist ratio, prepared from singles yarns with three twist levels. With the increase of ply twist, the mechanical properties of the composites improved up to a certain level and then declined. Maximum composites mechanical properties were achieved at a plied yarn to singles yarn twist ratio of 30% to 35% for all singles yarn twist levels. The optimum ply/singles twist ratio is lower than the 50% ratio normally applied to two-ply yarns for textile applications.

Prestressing on two-ply yarns did not improve the mechanical performance of the resulting composites, because the fibre orientation in the plied yarn had a detrimental effect on fibre orientation.

In the study related to wrap spun yarns, two different types of wrap yarns, with false twist (FT) and without false twist (NFT), were used at varying levels of wrap twist. The structure-property relationships were established by intercorrelating the wrap yarn structural parameters like wrap twist density and presence or absence of false twist during spinning with the mechanical properties and thermo-mechanical properties of the resulting composites. It is found that the wrap twist density and the false twist imparted to the yarn during yarn manufacturing significantly affect the curliness in the yarns, known as tortuosity. This yarn tortuosity in turn has a prominent effect on the mechanical properties of the manufactured composites.

It is found that the false twist imparted to the yarns during manufacture
adversely affects the mechanical properties of the composites because the tortuosity of the yarn increases with increasing wrap twist in FT yarns which results in fibre misalignment in the yarn. Increasing the wrap twist (without imparting false twist) results in improvement of mechanical properties. From a manufacturing point of view, however, a higher twist results in a slower production and rise in manufacturing cost.

Prestressing also resulted in a further improvement of the mechanical performance of the resulting composite. Improvements of flexural strength by 16.2%, 15.3% and 31.2% and flexural modulus by 26.7%, 18.2% and 35.1% respectively of the composites prepared from 90, 140 and 180 wraps/m NFT yarns were observed. Similar improvements were found in the mechanical properties of the composites prepared from FT yarns.

Through comparison of mechanical properties of the composites prepared from the three yarn structures, it is found that the wrap spun yarns composites possessed the highest mechanical properties. From a manufacturing point of view, the manufacturing cost of coarse count wrap spun yarns is lower because high productivity can be achieved. The production of plied yarns involves an additional twisting (folding) process which adds to the production costs of spinning finer singles yarns.
Suggestions for Future Work

Future research work can be conducted in the following directions:

1. Prestressing has been shown to improve the mechanical properties of composites prepared from singles and wrap spun yarns. Further work can be conducted to develop the methods and equipment for applying prestress to singles and wrap spun yarns during continuous filament (yarn) winding composites fabrication processes.

2. Research work can also be conducted to study the effect of prestressing on pultruded natural fibre yarn reinforced composites, as well as development of means of yarn tension control during pultrusion process.
Bibliography


