

## ABSTRACT

ICHHAPORIA, PRATIK KISHORE. Composite from Natural Fibers. (Under the direction of Dr. William Oxenham and Dr. Behnam Pourdeyhimi.)

Composites are widely used in our day to day life. Due to their low weight and ability to be tailored for specific end use they have gained a considerable ground in the high performance applications, such as aerospace and automobile industry. However, the use of polymers that can be recycled when used with carbon and other niche fibers renders the composite non-recyclable. This has become a major issue as the landfills are filling up at a faster pace along with the need for going green due to global warming.

To tackle these issues research in recent years has been focused on substituting olefin (polypropylene, polyethylene, etc) based composites with biodegradable ones. In order to achieve the goal of recyclable composites, natural fibers surfaced as the fiber of choice for reinforcing composites. The overhead for using natural fibers is their cleaning and processing needs. The current project uses hemp, a natural fiber, as a reinforcing media for producing a biodegradable polymer. The novel aspect is in terms of processing, in the elimination of carding, a cost and time intensive process. The web coming from the opening and cleaning line are needle-punched to provide integrity before being used as reinforcement.

After considering various materials for use in a matrix, a soy protein isolate and poly lactic acid(PLA) were selected, because the soy protein isolate used as matrix material can be dissolved in water. In order to form composites the water has to be completely evaporated from the hemp and soy protein isolate mixture. The cost for evaporating water became a hindering factor, along with poor composite properties such as porosity and brittleness. Thus poly lactic acid (PLA) was selected as material of choice for using as matrix. PLA in fiber

form was blended with hemp fibers during processing. The hemp-PLA webs produced were hot pressed in order to form composites. Various parameters studied to derive performance metrics were blend ratio of fibers and processing temperatures. The composites were analyzed for various mechanical properties like tensile and flexural properties.

Composites were fabricated from epoxy vinyl ester based resin reinforced with hemp fiber webs in order to compare their properties with the biodegradable composites formed from hemp-PLA.

The hemp-PLA composites though not as superior as the hemp-epoxy vinyl ester composite are made from renewable resources and can be used for non-structural applications and may provide superior thermal and acoustic insulation.

Composites from Natural Fibers

by  
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## DEDICATION

Dedicated to my dear Grandmother Ms. Dayagauri Ichhaporla, my aunt Ms. Indira Ichhaporla, my parents Mr. Kishore Ichhaporla and Mrs. Jaswanti Ichhaporla and my family for their unconditional love, boundless faith, unflinching support and blessings.

## BIOGRAPHY

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## *Chapter 1*

### INTRODUCTION AND LITERATURE REVIEW

Composites have become an integral part of our day-to-day life and can be found everywhere, e.g. rubber tire, spacecraft, asphalt, etc. Composites have been around for a long time with the classic example of bricks made from straw and mud. Another example is the Mongol bow which was constructed out of animal tendons, wood, and silk bonded together with an adhesive<sup>[38]</sup>. Nature also has its own composites in the form of wood, teeth, bones, muscle tissue, etc. In general the composite materials consist of a matrix reinforced with particles or fibers. Natural fibers were used for reinforcing the matrix until early in to the mid 20<sup>th</sup> century. However since 1950 there was an increased demand for stronger and stiffer, yet light weight, composites, in fields such as aerospace, transportation and construction. This led to the incorporation of high performance fibers for reinforcement. These newer composites have low specific gravity, superior strength and modulus when compared to the traditionally engineering materials like metals<sup>[08,09]</sup>. Due to their strength to weight ratio and comparable or better mechanical properties, composites are gaining grounds in industrial applications where traditionally metals were used. Table 1.1 lists some of the benefits of using a composite in lieu of metals.

#### *1.1 Fiber Reinforced Composites:*

High performance fiber reinforced composite materials are comprised of high strength and modulus fibers, embedded in, or bonded to, a matrix, with a distinct interface between them. In a composite, the fiber, as well as the matrix, retain their physical and chemical identities, but still provide a combination of properties that cannot be achieved with either

of the constituents alone. In general the fibers play the role of load bearer. The matrix, while keeping the fibers in the desired location and orientation, act as a load transfer agent and protects the fibers from external conditions such as chemicals, heat and moisture.

Table 1.1 Potential benefits of composites over metals<sup>[68]</sup>

- 
- Increased design flexibility
  - Better damage tolerance
    - Increased impact resistance
    - Increased fracture toughness
    - Greater scuff resistance
  - Better corrosion resistance
  - High specific strength and stiffness
  - Low coefficient of thermal expansion
  - Better fatigue resistance
  - Potentially lower component costs
    - Lower fabrication costs
    - Lower quality assurance costs
    - Lower scrappage rate
  - Integral construction of composite structures
    - Minimal plies
    - Fewer joints
    - Ply dimensions can correspond to principal load path
-

## 1.2 Composite Classification:

Composites can be classified based on the type of fibers or matrix used<sup>[09,38]</sup>.

Based on the **type of fibers**, composites can be further classified as:

- continuous fiber reinforced composite,
- short fiber reinforced composites.

Based on the **type of matrix**, composites can be broadly classified into three categories:

- metal matrix composites,
- ceramic matrix composites,
- polymer matrix composites.

Out of the three, polymer matrix composites are widely used and are the topic of discussion here<sup>[09]</sup>. Polymer matrix composites can be further sub-divided into two classes based on the type of matrix polymer i.e. thermoset or thermoplastic. A thermoplastic is a plastic that melts to a liquid when heated and freezes to a brittle, very glassy state when cooled sufficiently. Most thermoplastics are high molecular weight polymers whose chains associate through weak Van der Waals forces (polyethylene); stronger dipole-dipole interactions and hydrogen bonding (nylon); or even stacking of aromatic rings (polystyrene). Thermoplastic polymers differ from thermosetting polymers (Bakelite; vulcanized rubber) as they can, unlike thermosetting polymers, be remelted and remolded. Many thermoplastic materials are addition polymers; e.g., vinyl chain-growth polymers such as polyethylene and polypropylene.

The difference between thermoplastics and thermosetting plastics is that thermoplastics become soft, remoldable and weldable when heat is added. Thermosetting plastics however

cannot be welded or remolded when heated, simply burning instead. On the other hand, once a thermosetting is cured it tends to be stronger than a thermoplastic.

The advantages and disadvantages of each type of matrix polymer are delineated in Table 1.2<sup>[68]</sup>.

Composites that are reinforced with continuous fibers provide the best mechanical properties. These type of composites are usually reinforced with high performance fibers like Carbon, Kevlar, PEEK, etc. which are expensive. Due to high cost these composites find applications in niche areas where performance outweighs the cost.

Composites reinforced with short fibers are reinforced with staple fibers such as glass, cellulosic, or petroleum based fibers<sup>[13,38]</sup>. They are well suited for low strength and stiffness applications like particleboard, automotive applications, construction, etc. The short fiber reinforced composites are finding ever increasing applications in engineering and in consumer goods, like door panel inserts in cars, as they can offer a unique combination of properties and also because they are more economical than competing materials<sup>[13]</sup>. In addition to this, a short fiber reinforced composites can be processed in a manner similar to matrix. For thermoplastics, techniques such as injection molding can be adapted for mass production, even for the part with intricate contours. In contrast, continuous fiber reinforced composites cannot be readily adapted to mass production and are mostly designed for very specific products even though they have better mechanical properties. Another advantage of short fiber reinforced composites is that the fibers can be randomly distributed providing a more isotropic end product as compared to usually anisotropic laminas of continuous fiber reinforced composites.

Table 1.2 Advantages and disadvantages of thermosets and thermoplastics as composite

Property	matrix resins <sup>[68]</sup>	
	Thermosets	Thermoplastics
Formulations	complex	simple
Melt viscosity	very low	high
Fiber impregnation	easy	difficult
Prepeg tac	good	none
Prepeg drape	good	none to fair
Prepeg stability	poor	excellent
Processing cycle	long	short to long
Processing temperature	low to moderate	high
Processing pressure	low to moderate	high
Fabrication cost	high	low
Mechanical properties		
-54 to 93° C, hot/wet	fair to good	fair to good
Environmental durability	good	unknown
Solvent resistance	excellent	poor to good
Damage tolerance	poor to excellent	fair to good
Database	very large	small

### 1.3 Composite Mechanical Properties:

In the fiber reinforced composites, it has been shown that a significant improvement in properties of a resin/matrix can be achieved by adding suitable fibers and by controlling factors such as aspect ratio<sup>[08, 38]</sup>, fiber volume fraction, the dispersion and orientation of fibers, and the fiber-matrix adhesion<sup>[09, 13]</sup>. The effect of each of these factors on composite mechanical properties is described in detail, later in this chapter.

The properties of composites depend on the properties of its constituent components, their distribution and the interaction between them. The mechanical properties of the composite are usually considered the most important even though the composite may not be designed for load bearing application. At the very least, the composite product should be able to maintain its shape during the usage. Mechanical properties of continuous fiber reinforced composites can be predicted pretty accurately using the rule of mixtures (Equation 1.1) and the Halpin-Tsai equations<sup>[08, 09]</sup>. In contrast, for short fiber reinforced composites these properties are difficult to predict. This is due to factors such as: (1) fiber dispersion, (2) fiber orientation distribution, (3) fiber volume fraction and (4) the quality of interface between fiber and matrix that influence the composite properties. These factors, due to variation in fiber length and fiber length distribution in short fibers, along with inherent process variability, cannot be controlled precisely during manufacturing from part to part or from batch to batch<sup>[09, 13, 68]</sup>.

$$E_1 = (1-f) E_m + f E_f \dots \dots \dots \text{Equation 1.1}$$

Where,  $E_1$  = composite axial elastic modulus

$f$  = fiber volume fraction

$E_m$  = matrix elastic modulus

$E_f$  = fiber elastic modulus

The rule of mixture is a good approximation for axial stiffness. It indicates that the composite stiffness is a weighted mean between the moduli of the two components, depending only on the volume fraction of fibers. This expression assumes that the fibers are long and that there is equal strain on fibers and matrix. In the case of transverse stiffness, the rule of mixture does not give a good approximation and thus a more complex Halpin-Tsai model (Equation 1.2) needs to be employed<sup>[09]</sup>.

$$E_2 = \frac{E_m(1 + \xi \eta f)}{(1 - \eta f)} \dots\dots\dots \text{Equation 1.2}$$

$$\text{Here, } \eta = \frac{\left( \frac{E_f}{E_m} - 1 \right)}{\left( \frac{E_f}{E_m} + \xi \right)}$$

Where,  $E_2$ = Composite transverse elastic modulus

$E_m$ = Matrix elastic modulus

$E_f$ = Fiber elastic modulus

$f$  = fiber volume fraction

$\xi$ = adjustable parameter (usually 1)

Halpin-Tsai equation is not based on rigorous elasticity theory, but broadly takes into account the enhanced fiber loading, relative to equal stress assumption in the rule of mixtures. The above models gives a good prediction of the mechanical properties for continuous fibers, but are not that accurate for short fiber reinforced composites<sup>[09]</sup>. For aligned short fiber composites, the shear lag model proposed by Cox<sup>[13]</sup> and subsequently developed by Outwater, Rosen and Dow is employed. Later, a more complex, but more accurate, model to predict stiffness of aligned short fiber reinforced composites, with any cross-sectional shape was proposed by Eshelby<sup>[09,13]</sup>.

$$\sigma_1 = f \overline{\sigma_f} + (1 - f) \overline{\sigma_m} \dots\dots\dots \text{Equation 1.3}$$

$$\text{Where, } \overline{\sigma_f} = E_f \varepsilon_1 \left( 1 - \frac{\tanh(ns)}{ns} \right)$$

$$\overline{\sigma_m} \approx E_m \varepsilon_1$$

#### *1.4 Concept of Critical volume fraction:*

The equations for predicting the mechanical properties of both short and continuous fiber reinforced composites have fiber volume fraction as a common factor. This is because fiber volume fraction relates directly to composite strength<sup>[08,38]</sup>. This variation of axial strength in continuous fiber reinforced composite with fiber volume fraction is very well predicted by the rule of mixtures and can be depicted by Figure 1.1(a). As observed in the figure, tensile strength decrease drastically at low fiber volume fraction, which can be explained by a dilution of the matrix and the introduction of flaws in the matrix at the fiber ends, i.e. where the stress concentration occurs. At high volume fraction tensile strength improves, which can be explained by the of effects of fiber reinforcement outweighing the influence of matrix dilution. The tensile properties of composites improve with an increasing fiber volume fraction. The fiber volume fraction at which strength of composites stops declining and begins to improve is known as the critical fiber volume fraction. For the fibers to have reinforcing effect their proportion should be at least equal to the critical volume fraction. However, in order for the composite to perform well the fiber volume fraction must be above the volume fraction at which composite strength is more than that of the matrix strength. Fig 1.1 shows the strength of composite as a function of volume fraction. Once the critical fiber volume fraction is reached the composite failure might be governed by either failure due to matrix (shown by dotted line)( or by failure of fibers shown by solid line). The behavior of a composite during tensile loading is shown in Figure 1.2. As can be seen from the diagram, the composite undergoes three different phases under the load cycle before it breaks.

Phase I: Both fibers and matrix deform elastically

Phase II: Fibers continue to deform elastically, but matrix deforms plastically

Phase III: Occurs only if fibers deform plastically. Fibers fracture followed by fracture of the composite material

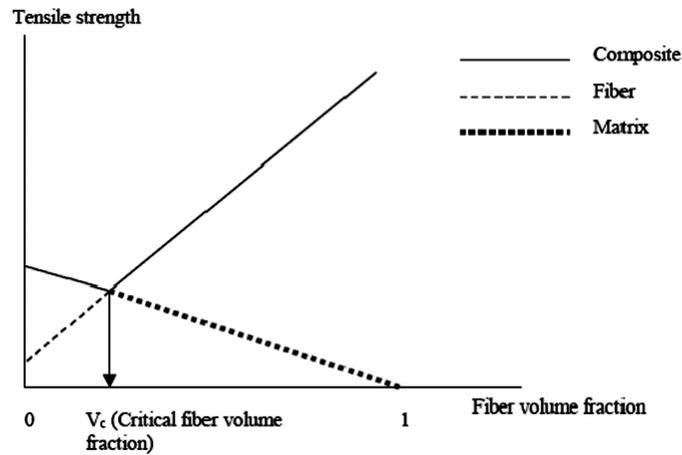


Figure 1.1 Composite tensile strength as a function of fiber volume fraction<sup>[09, 38]</sup>

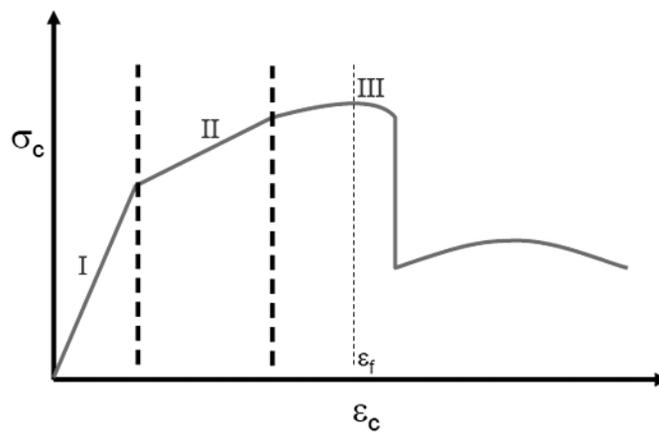


Figure 1.2 Composite stress-strain curve<sup>[09, 38]</sup>

*1.5 Critical fiber aspect ratio:*

Another important parameter governing mechanical properties, and hence the performance, especially for short fiber reinforced composites, is their fiber aspect ratio. Cox had done pioneering work by proposing a model, equation 1.4, relating critical fiber aspect ratio to shear strength based on Shear-lag analysis<sup>[09, 13]</sup>.

$$\frac{l_c}{d} = \frac{\sigma_f}{2\tau_y} \dots\dots\dots \text{Equation 1.4}$$

Where, d = fiber diameter,

$l_c$  = critical fiber length

$\sigma_f$  = ultimate fiber strength in tension

$\tau_y$  = interfacial shear stress

If we consider ultimate fiber strength in tension as constant, then Equation 1.4 shows an inverse relationship between critical aspect ratio and interfacial shear strength. It can thus be deduced that in order to have a lower critical fiber aspect ratio, a better interfacial shear strength is required<sup>[09, 13]</sup>. Interfacial shear strength can be varied by using coating/grafting on fiber surface, to improve the quality of the fiber/matrix interface. In composites, the load is transferred from the fiber to matrix by shear along the fiber/matrix interface. Figure 1.3<sup>[24]</sup> shows the variations in fiber stress and shear stress at the fiber/matrix interface along the fiber length. For maximum reinforcement, the fiber aspect ratio of any composite system should be above its critical value to ensure maximum stress transfer to the fiber before composite failure<sup>[56]</sup>. Fiber aspect ratio lower than the critical value results in insufficient stress transfer to fiber and thus the reinforcement is improper and in some cases, fiber just acts as fillers<sup>[09, 13, 38]</sup>.

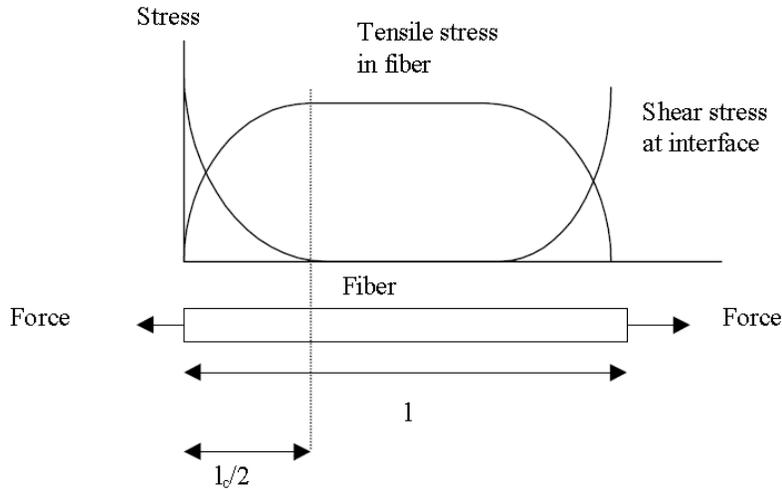


Figure 1.3 Tensile and shear stress variation along fiber length embedded in continuous matrix and subjected to tensile force in fiber direction<sup>[24]</sup>

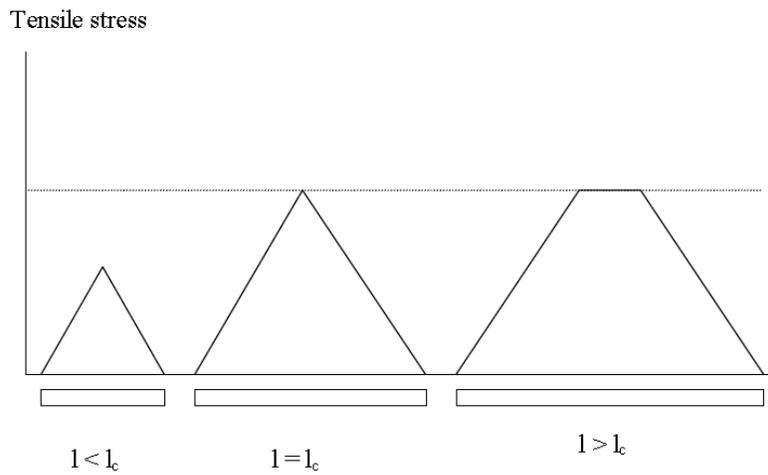


Figure 1.4 Effect of fiber length on fiber tensile strength<sup>[24]</sup>

In contrast if the fiber aspect ratio is too high, the fibers may get entangled during processing leading to poor mechanical properties, due to poor dispersion<sup>[13]</sup>. The fibers may be used as reinforcement in many different forms, i.e. short or long staple fibers, yarn, nonwoven web or fabric. Depending on the fiber form used for reinforcing the matrix, the fiber dispersion and hence the load transfer behavior from the matrix polymer to the fibers will be different. Effect of the fiber length on transferred fiber stress is shown in Figure 1.4<sup>[24]</sup>. The Problem with high performance fibers is that they tend to break during

processing. This changes the fiber length, and the fiber length distribution making predictions for the mechanical properties of composite a little complex<sup>[38, 72]</sup>. However, cellulosic fibers are flexible and are less prone to breakage during processing<sup>[27]</sup>. This ensures that the input fiber length distribution remains the same even after processing. Thus it is important to know fiber length/ fiber length distribution (in case of natural/short fibers) in order to determine the efficacy of reinforcement material.

### *1.6 Biodegradable Composites:*

With the development of new high performance fibers, composites began to compete with metals, and replace them, in myriad application. Fiber reinforced composites ventured into areas which were unthinkable few decades ago, making the products light weight, improving the performance of the product and in some cases improving the life time too. Better mechanical properties of these composites are due to excellent interfacial adhesion between fiber and matrix, in addition to good mechanical properties of fibers. However the good interfacial adhesion between fiber and matrix, which is beneficial in the product, is a significant disadvantage in the products “afterlife” since the fibers and matrix cannot be separated easily. This impairs the recycling of either or both. Also these fibers are not easily compostable; hence the composite cannot be used to recover energy. within today’s climate of growing environmental awareness and with depleting natural resources, people have resorted back to using natural fibers in lieu of polymeric fibers wherever possible<sup>[67, 66]</sup>. In recent years, more research is aimed towards achieving the goal of green composites or biodegradable composites<sup>[49]</sup>. Two main reasons for the interest in biodegradable materials are: (1) the growing problem of waste thereby resulting general shortage of landfill availability, and (2) the need for environmentally responsible use of resources

together with the carbon dioxide neutrality aspect<sup>[42]</sup>. Bio-composites or more specifically the “green composites,” consists of bio-fiber and bio-plastic from renewable resources and thus are expected to be biodegradable. Biodegradable polymers may be defined as those that undergo microbially induced chain scission, leading to mineralization, photo degradation, oxidation, and hydrolysis, which can alter a polymer during the degradation process. The degradation reactions that occurs when lignocellulosic materials (fibers) are exposed to nature are shown in Table 1.3; and Table 1.4 shows cell wall polymer component responsible for properties of lignocellulosic material in order of importance.

Definition of biodegradable plastic as provided by ISO<sup>[22]</sup> and ASTM<sup>[02]</sup> are given as follows. ISO definition of biodegradable plastic is: “A degradable plastic is one in which the degradation process results in lower molecular weight fragments produced by the action of naturally occurring microorganisms such as bacteria, fungi and algae”. ASTM defines biodegradable plastic as, “A degradable plastic in one in which the degradation process results from the action of naturally occurring organisms such as bacteria, fungi and algae”. Either way they provide a carbon dioxide neutral life cycle, and hence they don’t add to the gases causing green house effect. The cartoon in Figure 1.5 shows a general life cycle of compostable, biodegradable polymer.

Table 1.3 Degradation reactions that occur when lignocellulosic resources are exposed to nature<sup>[42, 55]</sup>.

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**Biological degradation**

Fungi, bacteria, insects, termites

Enzymatic reactions - Oxidation, hydrolysis, reduction

Chemical reactions - Oxidation, hydrolysis, reduction

Mechanical - Chewing

**Fire degradation**

Lighting, sun, man

Pyrolysis reactions - Dehydration, hydrolysis, oxidation

**Water degradation**

Rain, sea, ice, acid rain

Water interactions - Swelling, shrinking, freezing, cracking

**Weather degradation**

Ultraviolet radiation,- Oxidation, reduction, dehydration,

water, heat wind, hydrolysis

Chemical reactions

**Mechanical degradation**

Dust, wind, hail, snow, sand

Mechanical - Stress, cracks, fracture, abrasion

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Table 1.4 Cell wall polymer responsible for the properties of lignocellulosic in order of importance

---

**Biological degradation**

Hemicelluloses  
Accessible cellulose  
Non-crystalline cellulose

**Moisture Sorption**

Hemicelluloses  
Accessible cellulose  
Non-crystalline cellulose  
Lignin  
Crystalline cellulose

**Ultraviolet Degradation**

Lignin  
Hemicelluloses  
Accessible Celluloses  
Non-crystalline cellulose  
Crystalline cellulose

**Thermal Degradation**

Hemicelluloses  
Celluloses  
Lignin

**Strength**

Crystalline cellulose  
Matrix (Non-crystalline cellulose + Hemicelluloses + Lignin)  
Lignin

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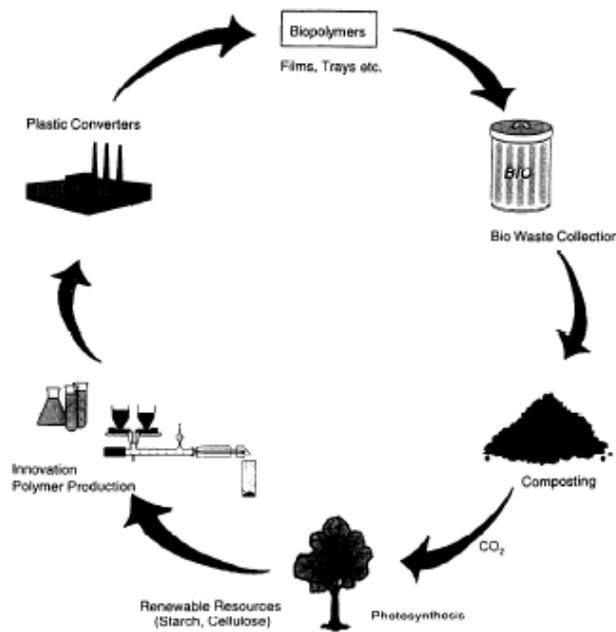


Figure 1.5 Life cycle of compostable, biodegradable polymers<sup>[34, 42]</sup>.

In order to replace the inorganic/olefinic fibers, natural fibers have to provide comparable if not better mechanical properties for reinforcing composites. The selection of suitable natural fibers for replacing high performance / olefinic fibers is determined by the required values of the stiffness. Further criteria governing the choice of a suitable reinforcing fiber are, for example, elongation at failure, thermal stability, adhesion of fibers and matrix, dynamic and long-term behavior, price and processing costs.

### 1.7 Natural Fibers:

Natural fibers in general can be classified based on their origin, and the plant-based fibers can be further categorized based on part of the plant they are recovered from. A general classification for natural fibers is provided in Figure 1.6.

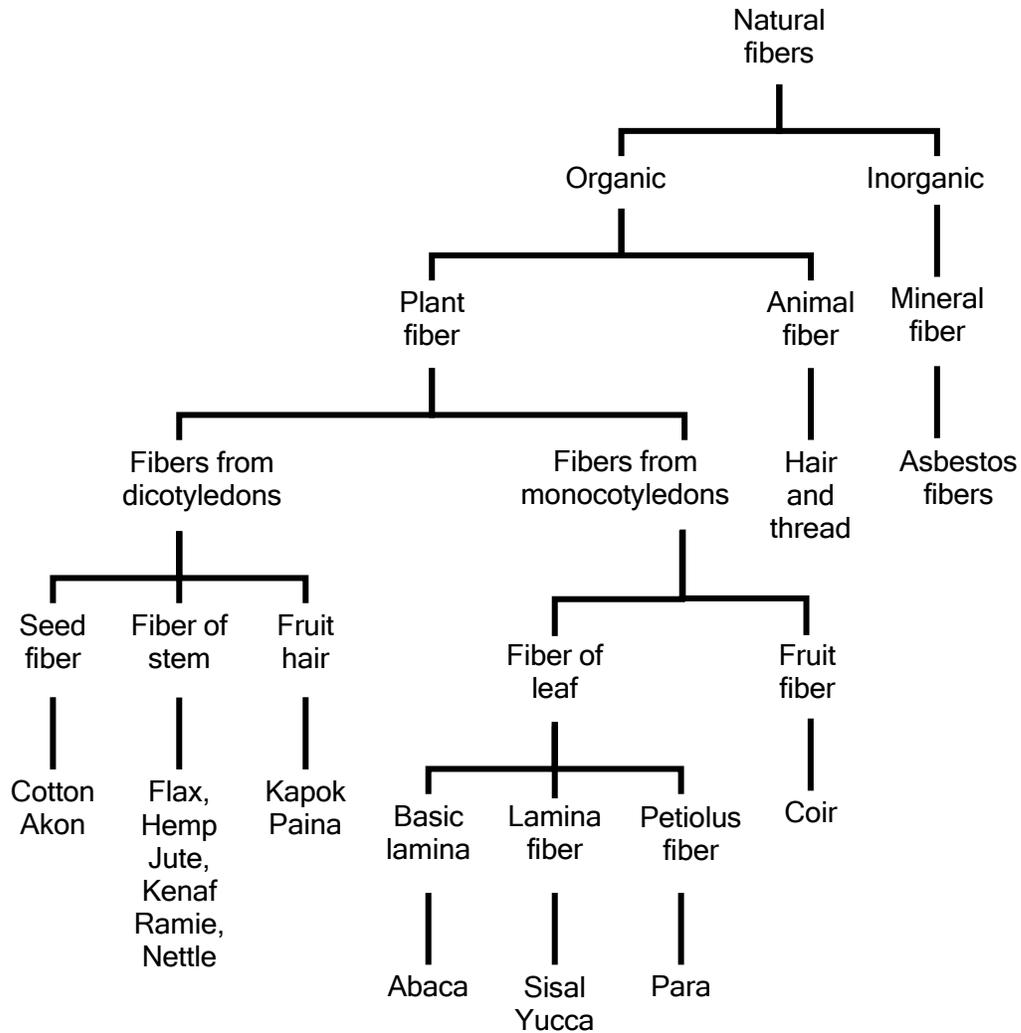


Figure 1.6 Classifications for natural fibers<sup>[43]</sup>.

### 1.8 Plant Fibers:

Plant fiber primarily consists of cellulose, hemicellulose and lignin. Chemical composition of some of them is provided in the Table 1.5<sup>[56]</sup>. The table also provides microfibrillar spiral angle for these fibers. Usually lower the spiral angle, i.e. as the inclination of microfibrill to fiber axis decreases, the higher the modulus and strength.

Bast fiber or skin fiber is the fiber collected from the Phloem (the "inner bark" or the skin) or bast surrounding the stem of a certain, mainly dicotyledonic, plants. Most of the technically important bast fibers are obtained from herbs cultivated in agriculture, as for instance flax, hemp or ramie. Since the valuable fibers are located in the phloem, they must often be separated from the xylem material ("woody core"), and sometimes also from epidermis. This process of fiber separation is known as retting. Bast fibers generally occur as a bundle of fibrils that are glued together by pectin and calcium ions. As bast fibers in general have low spiral angles they have high modulus and better tensile strength as shown in Figure 1.7<sup>[27]</sup>. Specific modulus and specific strength of some bast fibers like flax and hemp are almost comparable and in some cases better than the glass fibers. Some of the generic values for the fiber length and diameter distribution for natural fibers are provided in Table 1.6<sup>[43]</sup>, while Table 1.7<sup>[42]</sup> compares the strength and modulus of natural fibers with established high performance fibers with established high performance fibers.

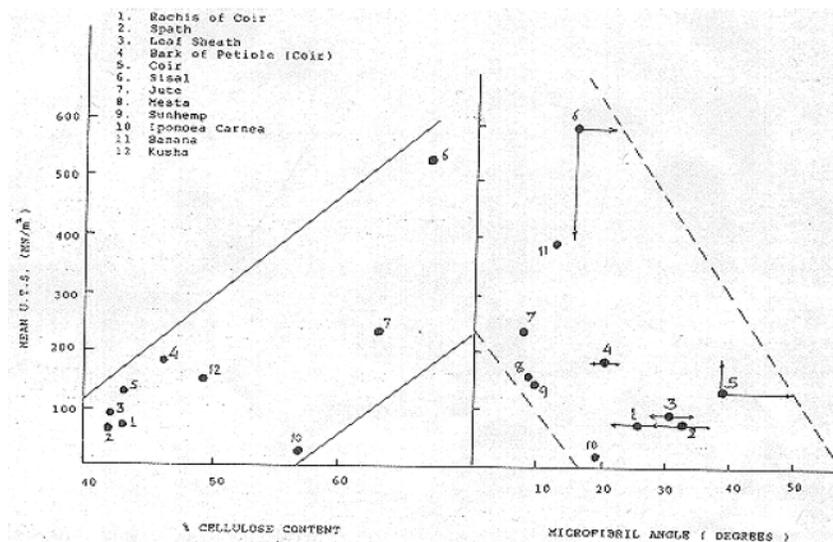


Figure 1.7 Relationship between cellulose content and microfibril angle to ultimate fiber tensile strength<sup>[27]</sup>.

Table 1.5 Chemical compositions of some common plant fibers<sup>[16, 28, 42, 56]</sup>.

Type of fiber	Cellulose	Lignin	Pentosan	Ash	Silica	Spiral angle(deg)
<b>Stalk Fiber</b>						
Straw						
Rice	28-48	12-16	23-28	15-20	9-14	-
Wheat	29-51	16-21	26-32	4.5-9	3-7	-
Barley	31-45	14-15	24-29	5-7	3-6	-
Oat	31-48	16-19	27-38	6-8	4-6.5	-
Rye	33-50	16-19	27-30	2-5	0.5-4	-
<b>Cane fiber</b>						
Bagasse	32-48	19-24	27-32	1.5-5	0.7-3.5	-
Bamboo	26-43	21-31	15-26	1.7-5	0.7	-
<b>Grass fiber</b>						
Esparto	33-38	17-19	27-32	6-8	-	-
Sabai	-	22	24	6	-	-
<b>Reed Fiber</b>						
Phragmites						-
Communis	44-46	22-24	20	3	2	-
<b>Bast fiber</b>						
Seed flax	43-47	21-23	24-26	5	-	8-10
Kenaf	44-57	15-19	22-23	2-5	-	8
Jute	45-63	21-26	18-21	0.5-2	-	8
Hemp	57-77	9-13	14-17	0.8	-	6
Ramie	87-91	-	5-8	-	-	8
<b>Core Fiber</b>						
Kenaf	37-49	15-21	18-24	2-4	-	-
Jute	41-48	21-24	18-22	0.8	-	-
<b>Leaf fiber</b>						
Abaca (Manila)	56-63	7-9	15-17	1-3	-	-
<b>Sisal</b>						
(agave)	43-62	7-9	21-24	0.6-1	-	10-25
<b>Seed Hull fiber</b>						
Cotton	85-96	0.7-1.6	1-3	0.8-2	-	-
<b>Wood fiber</b>						
Coniferous	40-45	26-34	7-14	<1	-	-
Deciduous	38-49	23-30	19-26	<1	-	-

Table 1.6 Dimensions of selected agricultural fibers<sup>[43]</sup>.

Fiber type	Fiber length, mm average(range)	Fiber fineness, mm average(range)
Flax (fiber bundle)	(250-1200)	(0.04-0.6)
Flax (single fibers)	33 (9-70)	0.019 (0.005-0.038)
Hemp (fiber bundles)	(1000-4000)	(0.5-5)
Hemp (single fibers)	25 (5-55)	0.025 (0.01-0.05)
Jute (fiber strands)	(1500-3600)	-
Jute (single fibers)	(2-5)	0.020 (0.010-0.025)
Cotton	10-60	
Kenaf	2-6	0.2
Ramie	60-260	0.040-0.080
Sisal	1-5	0.05-0.2

Table 1.7 Properties of natural fibers and synthetic fibers<sup>[42]</sup>.

Type of Fiber	Density g/cm <sup>3</sup>	Tensile Strength MPa	Young's Modulus GPa	Elongation at break %
Cotton	1.5-1.6	287-800	5.5-12.6	7.0-8.0
Jute	1.3-1.45	393-773	13-26.5	1.16-1.5
Flax	1.50	345-1100	27.6	2.7-3.2
Hemp	-	690	-	1.6
Ramie	1.50	400-938	61.4-128	1.2-3.8
Sisal	1.45	468-640	9.4-22.0	3-7
Pineapple	-	413-1627	34.5-82.51	1.6
Coir	1.15	131-175	4-6	15-40
E-glass	2.5	2000-3500	70	2.5
S-glass	2.5	4570	86	2.8
Aramid	1.4	3000-3150	63-67	3.3-3.7
Carbon	1.7	4000	230-240	1.4-1.8

In addition to having comparable mechanical properties, few other advantages of bast fibers over glass fibers are<sup>[43, 50, 58]</sup>:

- It provides better thermal and acoustic insulation properties, especially as an automotive interior or construction material part, due to presence of lumen/void in the fiber.
- They are easy to process on the traditional textile machinery for making reinforcement elements, like yarn, mat or woven fabric.
- They are not harsh, like glass fibers on the processing machinery like extruder, pelletizer, or injection molding machine.
- They are natural fibers, hence do not cause any allergies or lung diseases if breathed in or came in contact with.
- In automotive interior part, they function as better components as they do not show any sharp fractures.
- They are lightweight as compared to glass fiber reinforced composites, thereby providing better fuel efficiency or the weight saved, in interior and car trim panels, can be used in other car components to improve its performance.
- Provide economic incentive by being a green product, also they provide a marketing advantage.

Due to advantages outlined above along with comparable specific strength and modulus, bast fibers are rapidly replacing glass fibers as reinforcement in automotive interior components, like door trim panel, trunk and hood liners, etc.

Until few years back, research was aimed at using natural fibers for reinforcing olefin, polyurethane and few traditional matrix materials. Even with natural fiber reinforcement, it

is difficult to segregate the fibers at the end of product life cycle for degradation or composting. Also, presence of natural fibers in the plastic matrix renders them non-recyclable. Thus, the goal in recent years has been to develop a biocomposite using natural fiber and bio-plastic from renewable resources, which at the end of product life cycle can be completely biodegraded or composted for energy recovery.

Bioplastics were initially developed by packaging industry to solve the waste disposal and burial woes<sup>[55, 66]</sup>. As they were developed for packaging needs, their material properties did not meet matrix system requirements. Particular problems with these materials were associated with overly high breaking elongation and high processing viscosity, which proved to be disadvantageous. In recent year, many researchers have done breakthrough work to improve properties of these plastics and to develop new plastics for matrix materials. Bio-polymers can be broadly classified into: starch based, oil based, and lingo-phenol based materials. Table 1.8, provides information about the material class polymer belongs to, manufacturer and commercial name for some of the polymers.

Table 1.8 Classification of biopolymers on the basis of material class<sup>[42]</sup>.

Material class	Manufacturer	Product name
Cellulose acetate	Mazzucchelli	BIOCETA <sup>®</sup>
	Planet polymer	EnviroPlastic <sup>®</sup> -Z
Copolyester	BASF	Ecoflex
	Eastman	Easter Bio <sup>™</sup>
Polycaprolcatone	Birmingham polymers	Poly( $\epsilon$ -caprolactone)
	Planet polymer	EnviroPlastic <sup>®</sup> -C
	Solvay	CAPA <sup>®</sup>
	Union Carbide	TONE <sup>®</sup>
Poly(ester amide)	Bayer	BAK 1095
		BAK 2195
Poly(ethylene terephthalate)	DuPont	Biomax <sup>®</sup>

Table 1.8 (continued).

Material class	Manufacturer	Product name
Polyglycolide (PGA)	Alkermers	Medisorb <sup>®</sup>
	Birmingham polymers	Poly(glycolide)
	Boehringer Ingelheim	Resomer <sup>®</sup>
Polyhydroxyalkanoates (PHA)	PURAC	PURASORB <sup>®</sup> PG
	Metabolix	PHA
	Biomer	Biomer <sup>®</sup>
Poly(lactic acid) PLA	Monsato	Biopol <sup>®</sup>
	Alkermers	Medisorb <sup>®</sup>
	Birmingham polymers	Poly(L-lactide) & Poly(DL-
Poly(lactic acid) PLA	Boehringer Ingelheim	Resomer <sup>®</sup>
	Cargill Dow polymers	EcoPLA <sup>®</sup>
	Chronopol	Heplon <sup>™</sup>
Poly(lactic acid) PLA	Hygail	PLA
	Neste	Poly(L-Lactide)
	PURAC	PURASORB <sup>®</sup> PL/PD/PDL
Poly(vinyl alcohol) PVOH	Idroplast	Hydrolene <sup>®</sup>
	Novon	Aqua-NOVON <sup>®</sup>
	Planet Polymer	Aquadro <sup>®</sup>
Starch & starch blends	Texas Polymer	Vinex
	AVEBE	Paragon <sup>™</sup>
	BioPlastic(Michigan)	Envar <sup>™</sup>
Starch & starch blends	BIOTEC	Bioplast <sup>®</sup> , Bioflex <sup>®</sup> ,
	Buna Sow Leuna	Sconacell <sup>®</sup>
	Earth Shell	Starch-based composite
Starch & starch blends	Midwest Grain	Polytriticum <sup>™</sup> 2000
	Novamont	Mater-Bi <sup>™</sup>
	NOVON	Poly-NOVON <sup>®</sup>
Other Blends	Starch Tech	ST1, ST2, ST3
	Alkermers	Medisorb <sup>®</sup>
	BioPlastic(Colorado)	Biocomposite material
Other Blends	Birmingham polymers	Poly(DL-lactide-co-
	Boehringer	Poly(DL-lactide-co-
	Resomer <sup>®</sup>	Resomer <sup>®</sup>
Other Blends	Planet Polymer	EnviroPlastic <sup>®</sup> -U
	PURAC	PURASORB <sup>®</sup> PLG
	PURAC	PURASORB <sup>®</sup> PDLG

### *1.9 Biopolymer:*

A Brief review is provided below for each biopolymer along with more in-depth review about the soybean oil based biopolymer since this is the one of most interest to the present research. The first fully bio-synthetic/biodegradable polymer, poly-(3-hydroxy-butyrate) (PHB), although originally discovered in 1925, was developed on a semi-technical scale by ICI only in the 1980's by microbial fermentation of sugar. The development of this and related polyesters like the poly(alkanoates) (PHAs) is described by Hamond and Ligat<sup>[59]</sup>. Like PHA's, Poly(lactic acid) (PLA) also has an old history. It was synthesized in 1932 by Wallace H. Carothers<sup>[05]</sup>, but the molecular weight was very low and the polymer lacked attractive properties. It was further developed by DuPont and Ethicon. Further research was hindered till 1980's due to materials susceptibility to hydrolytic degradation. Later major breakthrough in process technology, coupled with decreased costs of biologically produced lactic acid, has led to commercial scale production of Polylactic acid (PLA) from lactic acid for non-medical applications. Recently Cargill-Dow LLC started up a commercial scale polylactide plant.

PLA is a versatile polymer made from renewable agricultural raw materials, which are fermented to lactic acid on degradation. Conversion of lactic acid to high molecular weight PLA can be achieved by two routes. One is using solvent free novel distillation process<sup>[18]</sup>, used by Cargill-Dow. Other is a solvent based process<sup>[15]</sup>, used by Mitsui Toatsu, in which a high molecular weight polymer is produced by direct condensation using azeotropic distillation to remove the water of condensation continuously. Figure 1.8 shows the steps involved in these two processes<sup>[35]</sup>. PLA polymers range from amorphous glassy polymers

with a glass transition of 60°C to semi crystalline/highly crystalline products with melting point ranging from 130°C to 180°C. The melting point depends on the level of crystallinity. Polylactic acid (PLA) is normally a linear chain molecule. In comparison to polyolefin's, they have poor melt elasticity, which might be due to lower degree of molecular chain entanglement. PLA are aliphatic polyesters, and the susceptibility to moisture is the primary force driving the degradation reaction. The rate of hydrolytic degradation is temperature and humidity dependent.

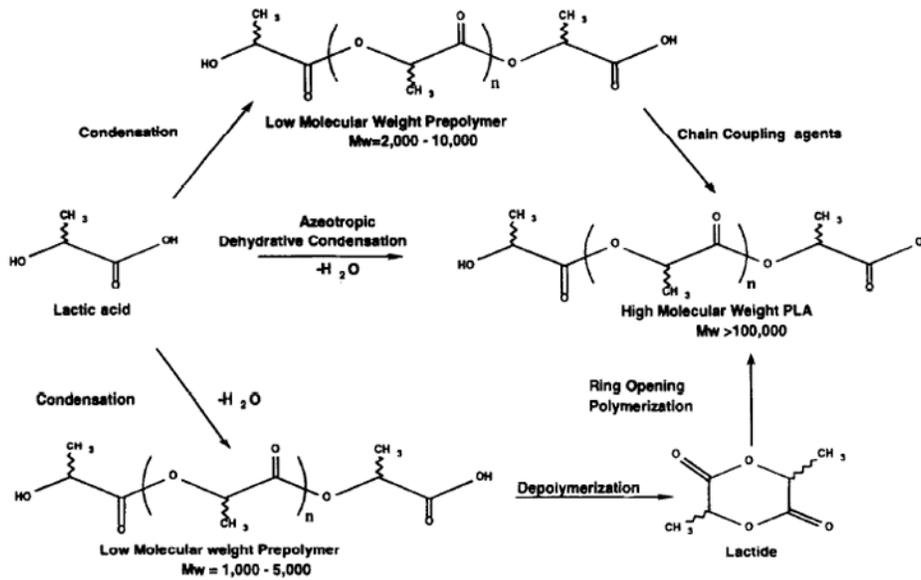


Figure 1.8 Manufacturing routes to Poly(lactic acid)<sup>[35]</sup>

Poly(lactic acid) (PLA) and its composites were primarily used for medical applications due to comparatively high cost and inadequate availability. But now with the beginning of commercial scale production and reduction in cost, PLA has been used as fiber and matrix for various composite applications. Oksman et. al. used PLA as matrix material for

reinforcing enzyme retted flax fibers in the form of roving and characterized the composites for mechanical properties<sup>[48, 49]</sup>. Later Plackett et. al. used L-PLA as matrix for reinforcing jute fiber nonwovens. They also characterized the mechanical properties and studied the composite degradation behavior using gel permeation chromatography<sup>[53]</sup>. This work was extended to Bamboo fibers by Lee et. al. They modified the surface of the bamboo fibers, using maleic anhydride both as coupling agent and a compatibilizer, and kneaded them at 180°C into sheets. Mechanical and water absorption behavior of the composites were observed along with its morphology<sup>[25]</sup>.



Figure 1.9 Ford's Model U car with numerous biobased materials technologies<sup>[39]</sup>

Thus a lot of work is being carried out as use of Polylactic acid (PLA) as fiber and/or matrix for biodegradable composite, but they have yet to reach the usage volume that they have as disposable, degradable packaging materials. One of the latest uses of the PLA that has been explored is as a diaper coverstock. One of the promising news for PLA use is that Ford recently unveiled a new Model U (Figure 1.9) concept vehicle that uses PLA polymer for its canvas roof and carpet mats. Similarly Toyota also plans to use PLA resin for its Prius hybrid car for interior parts which are reinforced with kenaf fibers<sup>[39]</sup>. Large-scale use of PLA by automotive giants should help to cross the barrier for PLA fiber and make it a

commercially viable polymer for resin matrix material.

Starch based polymers are also competing with olefins and other plastics for a place in resin matrix materials. Starch is inexpensive and is abundantly available from annual crops like corn and potatoes. Starch, like Polylactic acid (PLA) and soy based resins are also biodegradable in wide variety of environments and can produce totally degradable products for specific market demands.

Starch consists of two major components: amylose and amylopectin. Starch can be gelatinized by extrusion cooking technology. Gelatinized materials with different starch viscosity, water solubility, and water absorption can be prepared by altering the moisture content of the raw product and the temperature or the pressure in the extruder. The extrusion cooked starch can be solubilized without any formation of maltodextrins, and that the extent of solubilization can be controlled by extrusion temperature, moisture content of the starch before extrusion and the amylose/amylopectin ratio.

When the starch is heated above its glass transition and melting temperatures in presence of plasticizers, an exothermic transition takes place. This transformation leads to formation of thermoplastic starch that can be used in plastic sector. The thermoplastic starch though can be used in plastic sector, is very susceptible to humidity which limits its use for various applications. Tomka et. al<sup>[67]</sup> thus developed a thermoplastically processable starch (TPS) stable against moisture, which is a modified native starch obtained without water. Here instead of water, a plasticizer or additive is used<sup>[67]</sup>. Novamont has lot of intellectual properties for development of TPS and complexed starch. One of their products is Mater-Bi<sup>T.M.</sup> which is widely used in Europe as a packaging material and additive<sup>[07]</sup>.

Starch based plastics were developed initially for packaging industry to solve the waste disposal problem, however later research lead to development of starch based plastics suitable for using as resin matrix materials for biodegradable composites. Thermoplastics starch can be processed similar to olefins but has two primary disadvantages: (1) it is mostly water-soluble, and (2) it has poor mechanical properties. The water resistance has been improved by mixing it with certain synthetic polymers, adding cross-linking agents such as Calcium and Zirconium salts<sup>[64]</sup> and the problem of poor mechanical properties of the starch based plastics can be tackled by using synthetic polymers or natural fibers as reinforcement. The use of natural fibers as reinforcement to improve the mechanical properties will be the focus here. One of the initial studies for fiber reinforced eocomposite using starch plastic as resin matrix material was carried out by Curvelo et. al<sup>[10]</sup>. They used bleached pulp having fiber length of approximately 1 mm for reinforcing, a mixture of glycerin and starch matrix. The composite was prepared by mixing fiber and polymers in mixer and then hot pressing them at 160°C. The composites were then analyzed for mechanical properties, water absorption and morphology. Later, on similar biodegradable composites based on Polycaprolactone and starch matrix were studied by Cyras et. al<sup>[11]</sup> and Wu<sup>[73]</sup>. They not only determined the mechanical properties of the composite but also studied the effects of various parameters on it. However, an in depth studies of various parameters, like filler content, fiber length and nature, moisture content; affecting thermal and mechanical behavior of the reinforced starch plastic was carried out by Averous et. al.<sup>[03]</sup>. Foam composite using starch matrix reinforced with jute and flax fibers were prepared Soykeabkaew et. al<sup>[65]</sup>. They also studied the effect of fiber and moisture content on mechanical and thermal properties.

Thermal and biodegradation behavior of composites based on starch plastics has been carried out by Wool et. al.<sup>[71]</sup> and Alvarez et. al.<sup>[01]</sup>. Dynamics of starch degradation in polyethylene/starch composites was investigated by using aerobic biodegradation and two models for starch degradation were also considered in the studies. It was determined from the study that accessibility of starch is dependent on the starch concentration in the composite, the distribution of starch particles, the fractal microstructure, the degradation time, and the biological process by which microorganisms degrade the starch in the composite. Thus starch based biopolymers are slowly but steadily entering the composites market and making a place for themselves especially in the field of eco-composites.

Biodegradable Polyesters (PET), like Poly(hydroxyl butyrate) (PHB), its copolymers such as Poly(hydroxybutyrate-co-valerate) (PHBV), and Poly(butylene succinate) (PBS), produced by various microorganism are the recent biopolymers receiving attention in composites field for use as matrix material. Mechanical properties of these polymers are comparable to traditional thermoplastics such as polyethylene and polypropylene. Biocomposites were made from these polymers by reinforcing them with various natural fibers, (pineapple, bamboo, abaca, lyocell) and effect of various parameters were studied on thermal and mechanical behavior of the composites by various researchers<sup>[26, 36, 37, 61, 62, 63]</sup>.

For use in biodegradable composites some of the petrochemical based resins were also modified to render them biodegradable at the end of their life cycle. Pioneering work in this field was carried out by researchers at Bayer AG, Germany who came up with BAK 1095 and BAK 2095<sup>[17]</sup>. Both of these are polyesteramides that are synthesized by a special process, which renders them biodegradable. Since they have petrochemical base they can



Figure 1.10 Henry Ford, in 1941, using axe on Soy based car to show its strength.

still be processed and used similar to other olefins. Thus they have the advantage of being processable like olefin and yet are biodegradable.

Mishra et. al.<sup>[41, 54]</sup> has made composites using BAK and coir, sisal and pineapple leaf fibers and studied their mechanical behavior and biodegradability. The target applications for these composites are packaging, adhesives, coatings, films, bags, etc.

Soybean resins are based on triglycerides, which are the major component of plant and animal oils. Triglycerides are composed of three fatty acids joined at a glycerol juncture. Most triglycerides contain fatty acids that vary from 14-22 carbon atoms, with 0 to 3 double bonds per fatty acid. In 1941 Henry Ford was the first to use Soybean flour in car shown in Figure 1.10. In the car, the panels were compression molded from hemp and ramie fibers embedded in a matrix of urea formaldehyde with wood and soybean flours. Resins from soybean oil are sought as replacement for liquid molding resins, such as unsaturated polyester, vinyl esters and epoxy resins<sup>[04,23]</sup>.

Among various plant and animal oils, Soybean oil is the most popular oil, covering 22% of the total oil market. Hence it is of highest interest for applications in resins for composites. The structure of soybean oil varies from region to region as it depends on soil and weather condition<sup>[51]</sup>.

Unmodified triglycerides do not readily polymerize, the chemical functionality necessary to cause polymerization however can be added to the triglycerides. The main step to reach higher level molecular weight and crosslink density, as well as incorporate chemical functionalities known to impart stiffness in a polymer network, has been accomplished by

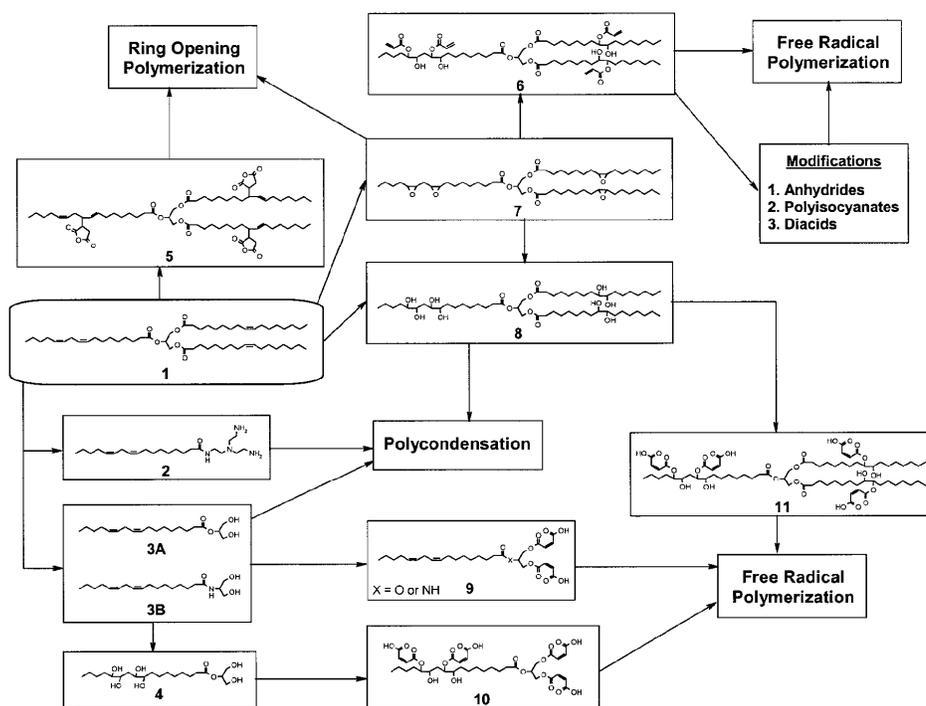


Figure 1.11 Chemical pathways leading to polymers from triglyceride molecules<sup>[23, 70, 72]</sup>.

Wool et. al., and is illustrated in Figure 1.11<sup>[23, 70, 72]</sup>. In structures 5, 6, 7, 8, and 11 (Figure 1.11) the double bonds of the triglycerides are used to functionalize the triglyceride with

polymerizable chemical groups. From the natural triglyceride, it is possible to attach maleates (5), or convert the unsaturation to epoxy (7) or hydroxyl functionalities (8). These transformations make triglyceride capable of reaction via ring opening or polycondensation polymerization. Reaction of the epoxy functional triglyceride with acrylic acid incorporates acrylates onto the triglycerides (6), whereas reaction of the hydroxylated triglyceride with

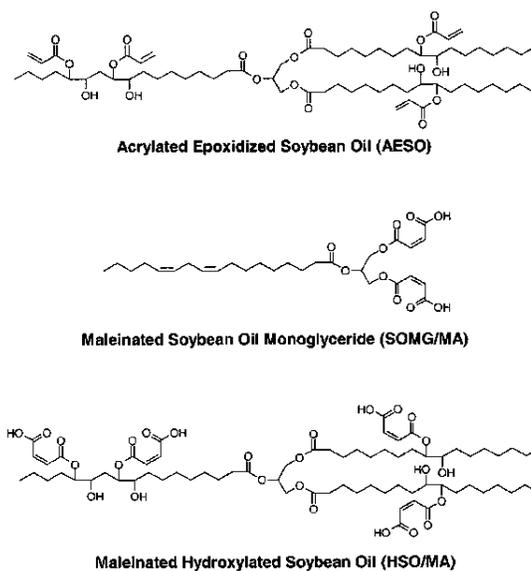


Figure 1.12 Triglyceride based monomers<sup>[23]</sup>.

maleic anhydride incorporates maleate half-esters and esters on to the triglyceride (11). These monomers can then be blended with a reactive diluent, and blended like conventional vinyl ester resins, and cured by free radical polymerization<sup>[23, 72]</sup>.

Various monomers that can be synthesized from a triglyceride are Acrylated epoxidized soybean oil (AESO), Maleinated soybean oil monoglyceride (SOMG/MA), and Maleinated hydroxylated soybean oil (HSO/MA). Structure of these monomers is shown in Figure 1.12.

The monomers of interest to us are AESO, they are synthesized by reaction between acrylic acid and epoxidized triglycerides, which are found in abundance in natural oils such as vernonia plant oil, or can be synthesized from unsaturated oils like, soybean oil or linseed oil by standard epoxidation reaction<sup>[14]</sup>. Epoxidized soybean oil is commercially available with functionality of 4.1 – 4.6 epoxy rings per triglyceride. Triglycerides can then be reacted via addition polymerization, with the addition of acrylates. One of the uses of AESO is as surface coating, and is commercially manufactured in forms such as Ebecryl 860<sup>[29]</sup>. AESO can also be blended with a reactive diluent, such as styrene, to improve its processability and control the polymer properties to reach a range acceptable for structural applications.

Wool et. al. were among the first to make composites from AESO polymers and flax and hemp fibers<sup>[23, 69, 72]</sup>. They also studied a biodegradable composite based on AESO polymer and keratin fibers. Here they carried out an in-depth examination of compatibility between fiber and resin, effect of comonomer concentration, vibration damping properties, mechanical properties, dielectric properties, thermal properties and water absorption properties of the composites<sup>[69]</sup>. Mohanty et. al. has also developed composites based on soy flour plastic. They have used grass<sup>[30]</sup> and pineapple leaf fibers<sup>[31]</sup> for reinforcing the matrix. The composites were prepared using extrusion followed by injection molding at a temperature of about 130°C. In the study, effect of fiber volume fraction, and compatibilizer on mechanical, thermal and water absorption was analyzed. They found that with increasing the fiber volume fraction the mechanical properties of composite improved, and it gave optimal properties at about 30%.

Netravali et. al.<sup>[32, 33, 45]</sup> is another group involved with research in soy based plastics. They have worked chiefly with ramie fibers, both short and long, and soy protein isolate. They also performed an extensive study of various factors affecting biodegradability and mechanical behavior of the composites. The processing temperature used by them was around 120°C to 130°C and they reached a volume fraction of about 30%. They characterized the interfacial surface characteristics between fiber and polymer using various techniques, and studied the effect of various additives on interfacial strength. Composites based on soy polymer resin has been used commercially by John Deere in their farm products as a cover for Round hay baler as shown in Figure 1.13<sup>[72]</sup>.



Figure 1.13 Round hay baler. The panel containing name John Deere was made from soy based resin<sup>[72]</sup>.

Research is continuously going on for modifying existing polymers to make them biodegradable or to synthesize new polymer from renewable resources having better mechanical properties that can fulfill the function of resin matrix for reinforcing fibers and

solve the waste disposal problem, making earth a greener and a better place.

The eco-composites are finding applications in various fields like transportation, construction<sup>[06]</sup>, automotive<sup>[21,40,46, 58,60]</sup>, agriculture<sup>[40,72]</sup>, furniture<sup>[46,50]</sup>, etc. due to their biodegradable nature along with improved mechanical properties. Pioneering work in biocomposites and their applications has been done at DLR institute in Germany. Researchers over there have developed various resins to fabricate composites suitable for replacing glass fiber and other high performance fiber reinforced composites. They even have been able to use many of their composites for commercial applications like in interior of train coaches, safety helmets, car interior, etc<sup>[06,20]</sup>.

One of the prime fields of application targeted by the biocomposite is automotive sector. The automotive sector has a huge growth potential, and if used in the interior of auto the composite does not need to have mechanical properties in the order of carbon, Kevlar or other high performance fiber reinforced composites. Also, when used inside most of the times it is not exposed to human eyes thus eliminating the requirements for aesthetic properties. Incorporation of biocomposite sometimes provides better thermal<sup>[74]</sup> and acoustical insulation<sup>[12, 47]</sup> properties as compared to traditional composites. Also, the weight is reduced leading to potential fuel saving. Moreover since the composites are biodegradable they can be easily degraded or composted to recover energy reducing polymeric waste.

The few drawbacks of using the biocomposites in automotive sectors are poor moisture stability, lack of fire retardancy, high cost, and poor impact properties. These problems have to be tackled in order for the biocomposites to compete with traditional composites and make an impact in auto sector.

## *Chapter 2*

### PROPOSED WORK

The goal of this project was to fabricate composites from natural fibers. Until now researchers have used fibers either in loose form, rope, carded web, or fabrics. In each case to achieve a uniform reinforcement media, fibers have to be opened properly. The opening process for natural fibers invariably involved carding, a time and cost intensive process. Novel aspect of this project was in terms of fiber processing, wherein the carding process was eliminated. The fibers after being opened and cleaned were taken, from Trutzschler<sup>®</sup> Scanfeed, to a needle-punching machine where the web was provided with strength and integrity. Elimination of the carding process not only saved time and money, but also increased the process speed as carding is usually a slower process in fiber processing. During needle punching process, the needles on a board enter perpendicular to the fiber web. While passing through the web the needle re-orient some of the fibers in Z-direction. These fibers lead to fiber entanglement providing for web strength. Thus for needle punching process to be successful fibers have to be flexible, otherwise while re-orientation via needles the fibers will break.

Among natural fibers, bast fibers have good mechanical properties and hence form an ideal candidate as the reinforcing media. Hemp, a bast fiber, has specific mechanical properties similar to the glass fibers and at the same time is flexible. Further hemp fibers can be easily processed on traditional opening and cleaning machines. Plenty of research is also being carried out in automotive market on olefin polymers reinforced with hemp fibers. The

above mentioned reasons, along with industry wide acceptance, led to selection of hemp fibers as reinforcement media in form of a nonwoven web.

To form matrix around the hemp fibers, soy-based resin was selected, as soy protein resin is abundantly available and has better future prospects. However, soy protein resin proved to be a very challenging material for processing in to a film as well as composites. Processing of soy resin was energy and cost intensive as water was used as a carrier medium for formation of films or composites from soy protein isolate powder. Further, the films and composites formed were brittle and stiff. To circumvent these problems of soy protein resin composites, other polymers were explored for serving as matrix.

Poly(lactic acid) (PLA), is a polymer from renewable resources. PLA is widely available and is a thermoplastic polymer. PLA could also be formed directly into a composite by hot pressing however, the composite would have been very brittle, and had high strain for very low stress (strain in the order of 40-50% percentage or more, for only few mega Pascal stress). Thus PLA was selected to be reinforced with hemp fibers. In order to take advantage of the novel fiber processing aspect of this project PLA in fiber form was selected. The PLA fibers were blended with the hemp fibers during opening process. The needle-punched web was then hot pressed at pre-selected temperatures. On hot pressing, the PLA fibers in nonwoven webs melted forming a matrix around the hemp fibers leading to the formation of hemp-PLA composite. The hemp-PLA fiber webs thus provide a thermoplastic web, that is portable and can be easily hot pressed to form a composite without the requirement of intricate and expensive machines. Further the composites can be formed into any contour or shape.

In order to determine the performance of hemp-PLA composites, they were compared with the epoxy-vinyl ester resin reinforced with the hemp fibers. Epoxy resin composites were chosen as they are widely used in the industry.

The parameters varied during fiber processing and composite fabrication were: fiber volume fraction, thermoplastic fiber %, web weight at Scanfeed, needling density and hot press temperature. The fibers and composites fabricated were characterized for mechanical properties.

### *Chapter 3*

#### EXPERIMENTAL WORK – SOY PROTEIN ISOLATE TRIALS

This chapter discusses the first experimental approach implemented towards fabricating composites from natural fibers that involved selection of soy protein resin for reinforcement of hemp fibers; and the procedure involved in casting Soy protein films and fabricating hemp fiber reinforced soy protein composites. To understand the role of soy protein as matrix, a plan was laid to prepare its resin, then cast it into films and finally to characterize its mechanical properties. Each of these steps are discussed in detail in subsequent paragraphs.

The first step was to prepare the resin. The Soy protein resin selected to serve as resin was PRO-FAM<sup>®</sup> 974 from Archer Daniel Mills (ADM). The procedure to prepare the resin involved dissolving Soy protein isolate (SPI) powder in water. The ratio of SPI powder to that of water was 1:9. In order to facilitate formation of uniform resin, the pH of the water was increased to 11, by adding 0.1N sodium hydroxide solution. Then the solution was stirred using magnetic stirrer at 90°C for 60 minutes.

To study the resin properties, an experiment was designed to cast films at different processing conditions and then to characterize the mechanical properties of those films. The most critical part of the experiment was to develop a robust method to cast films. The first step taken to cast a film was to prepare the resin using the method mentioned above. The prepared resin was poured in a shallow glass container. As the major content of the resin is water, the excess water has to be evaporated in order to achieve a uniform film. Thus, the glass container was placed in an oven for drying at selected temperature and time.

Once the films were almost dry, they were hot pressed at different temperature and pressure. The temperature range used for drying the films was from 60-90°C and time was varied from 15-30 hours. Two critical parameters, namely temperature and pressure were varied for hot pressing. The temperature range used for hot pressing was from 120 to 130°C and pressure was set at 0, 5000 and 10000 pound.

The film casting process proved to be very challenging. The obstacles encountered during the film casting process were; brittleness of film, air bubble formation, unevenness, and skin formation. Figure 3.1 below shows some of the above mentioned issues with the formed films.

Different techniques were tried to address these issues. For example, the glass container was lined with Teflon sheet and a vacuum was applied during oven drying of the resin to facilitate uniform drying of the film, potentially less bubble formation and ease of film removal. Additionally glycerol, a plasticizer, was used as an additive during resin preparation to overcome the brittleness of films. However, these approaches did not resolve the problems in film casting.

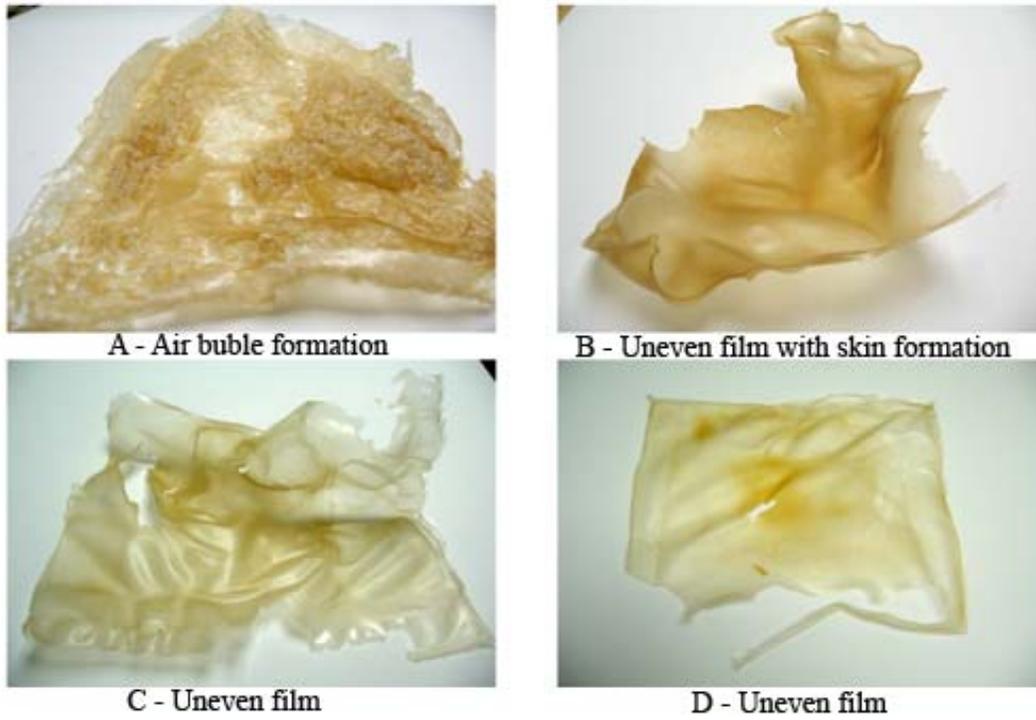


Figure 3.1 Soy Protein Films – 1

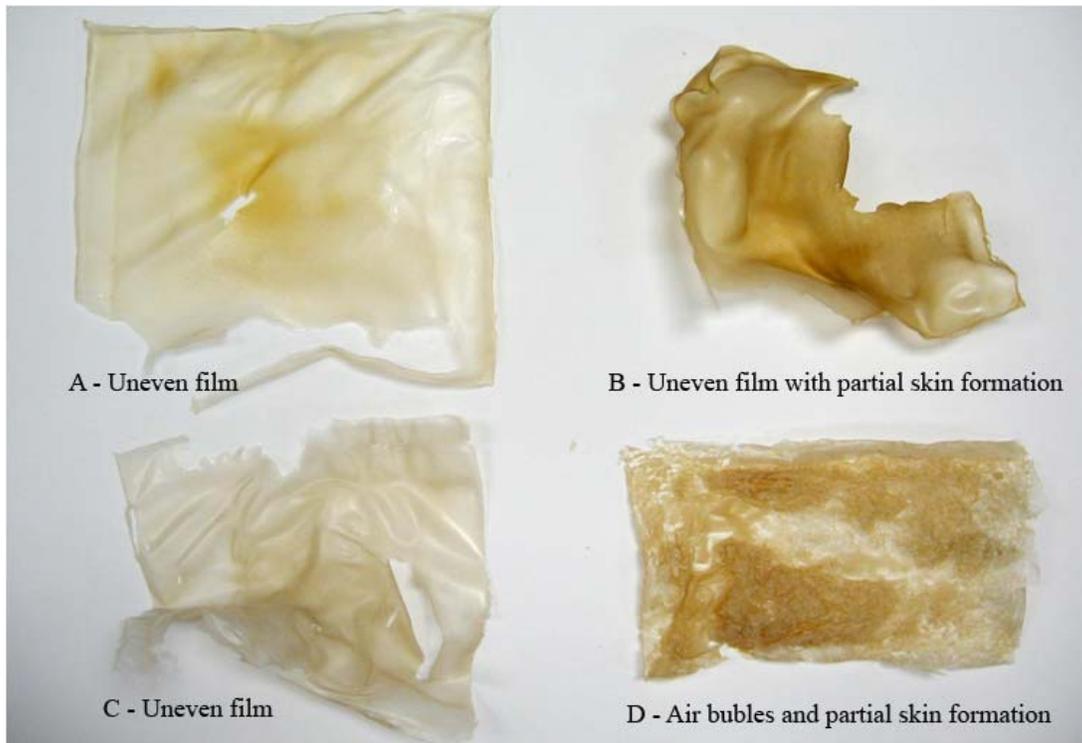


Figure 3.2 Soy protein films – 2

To circumvent problems faced during film formation, another experiment was designed to fabricate composites directly and characterize them. Based on prior literature work, some of the factors affecting composite properties were identified as fiber volume fraction, temperature for evaporation of water from resin, hot press temperature and time. These factors were varied during the composite formation process as mentioned below:

- Fiber volume fraction – 25, 30, 35, 40, 50 and 75%
- Temperature for evaporation of water from resin – 60, 70, 80 and 90°C
- Hot press temperature – 110, 120, 130 and 140°C
- Time for hot pressing – 3, 5 and 10 minutes.

Although it was possible to fabricate composites using certain processing conditions, the composites had issues similar to the cast films in terms of brittleness and porosity. It was observed that when the fiber volume fraction was low, the composites were porous and brittle. Further, when the hot press temperature was 130 or 140°C and time of pressing was 5 minutes or more the protein started to degrade. Figure 3.3 shows improper wetting and skin formation on hemp-SPI resin composites that were fabricated.



SPI - Hemp : Stir @ 90 C for 2 h  
Oven dry 60 C for 72 h; Hot press @ 120 C for 3 min



SPI - Hemp: Stir @ 90 C for 2 h  
Oven dry 75 C for 29 h; Hot press @ 120 C for 3 min



SPI -Hemp: Stir @ 90 C for 4 h  
Oven dry 90 C for 16 h; Hot press @ 120 C for 3 min



SPI - Hemp: Stir @ 90C for 2 h  
Oven dry 60 C for 24 h; Hot press @ 120 C for 6 min

Figure 3.3 Hemp reinforced soy protein composites – Top view



Figure 3.4 Hemp reinforced soy protein composites

It can be seen from Figure 3.4 that the composites formed were non-uniform, porous and brittle.

To achieve a composite with good mechanical properties, the fiber web has to be uniformly encapsulated by resin matrix. To study hemp fiber dispersion in SPI resin matrix, cross-sectional image of composite was taken by Scanning Electron Microscope (SEM). The SEM image (Figure 3.5) showed that the fiber web was not being impregnated by resin i.e. the resin formed an almost separate layer on top of the fiber web. This was subsequently confirmed by a high magnification SEM image (Figure 3.6). This lack of impregnation could be due to the high surface tension of the web, which would require a lower viscosity resin, or excessively high driving force for resin impregnation.

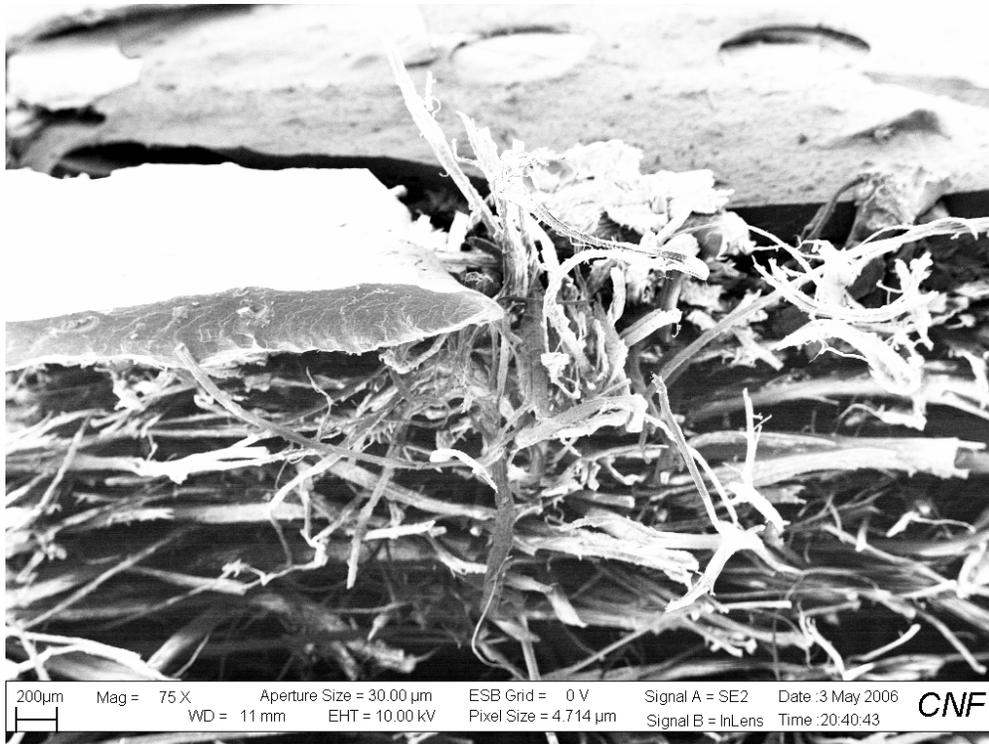


Figure 3.5 SEM of Cross-section of Hemp reinforced soy protein composites

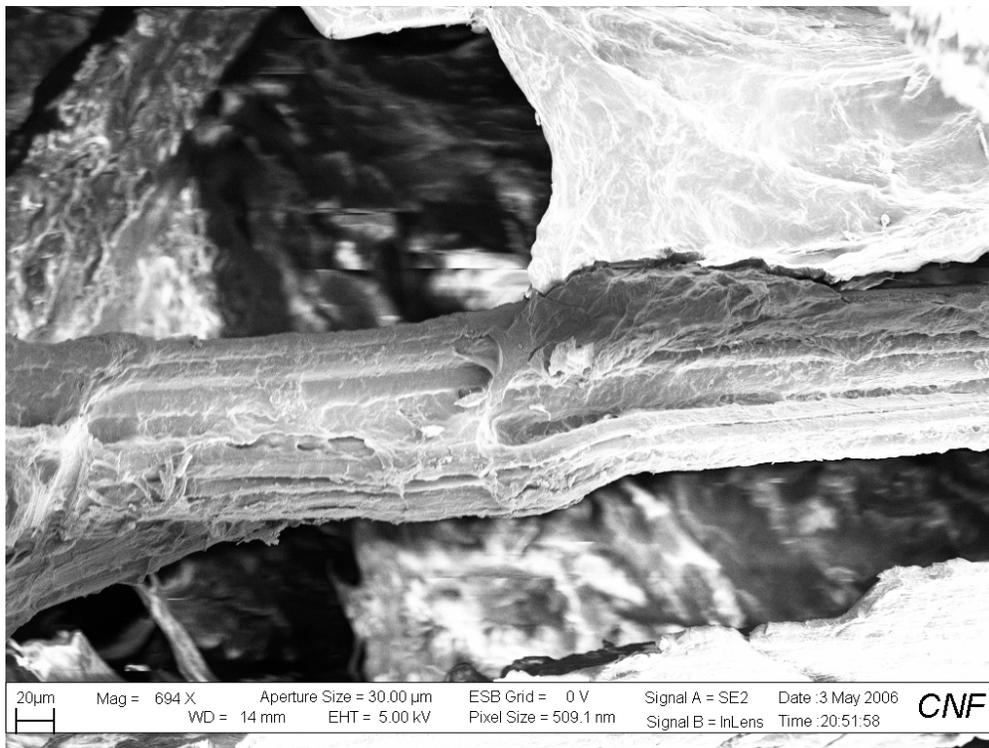


Figure 3.6 SEM – Close up of fiber and resin interface of composite

From the film casting and composite fabrication process trials carried out it is possible to offer some suggestions for future work in this area:

- When a resin system is comprised of 90% water, and the water has to be evaporated, especially from hydrophilic fibers like hemp, the process becomes very energy intensive. The drying process thus not only takes long time, which would inhibit a commercial processing system, but also becomes expensive. It would thus be essential to make resin system with less water content, however this may in turn limit the potential impregnation of the fibers..
- Hemp fiber web becomes compact after needle-punching thereby reducing web porosity. As the web becomes more compact it becomes difficult to impregnate it with resin, which results in the formation of separate layers of resin and fiber web with a weak interface at border. The process should thus be adapted so that hemp fiber webs when impregnated with resin are more receptive to resin, coupled with a resin having a viscosity low enough to encapsulate and form a matrix around the fibers.
- The surface of the hemp may also be potentially modified in order to achieve better adhesion between fiber and resin matrix.

Based on the discussion above it can be concluded that although the present investigation into the production of composites from hemp fibers/soy protein proved to be unsuccessful, the process was a learning experience and provides some guideline of potential pitfalls to avoid when considering the successful fabrication of films and composites.

## Chapter 4

### EXPERIMENTAL WORK

This chapter outlines the design of experiment used in the final trials for fiber processing, web consolidation and composite fabrication. The design for each process is described in detail in subsequent paragraphs.

#### *4.1 Plan for Fiber processing*

The fibers as they arrive at a textile processing facility are in a bale form. In a bale, the fibers are packed very tightly. Any further processing of the fibers thus requires opening of the fiber tufts into smaller tufts and ideally into individual fibers. If the fibers are derived from natural sources they also undergo the cleaning process while the opening process is performed.

Usually for making a nonwoven web, the fibers are opened and blended using opening machines. This is normally followed by carding or air-lay processing forming a uniform web. The web is then provided integrity and strength by bonding it using mechanical, chemical or thermal bonding methods.

Figure 4.1 shows the process outline for the fiber processing. As shown in the figure, the fibers received in the bale form were opened in the bale opener, first machine in the processing line. The fibers were further opened in the BXM opener. In case of forming the hemp-PLA webs; the fibers were also blended according to their weight fraction in the BXM opener. Further fiber opening was carried out in the finer FOL opener, followed by the Scanfeed machine. The Scanfeed machine as well as further opening the fiber tufts, also forms a uniform web that is normally fed to a carding machine. The carding machine

provides more intense opening and some blending and the web produced would typically then pass to the cross-lapper after which it is bonded. However, in the presently designed process flow, the carding process was eliminated and fiber batts were taken directly to a needle-punching machine (for bonding). It was believed that the additional uniformity achieved by the Scanfeed could potentially enable the elimination of the carding process for certain applications and this would result in a process shortening and significant cost benefits.

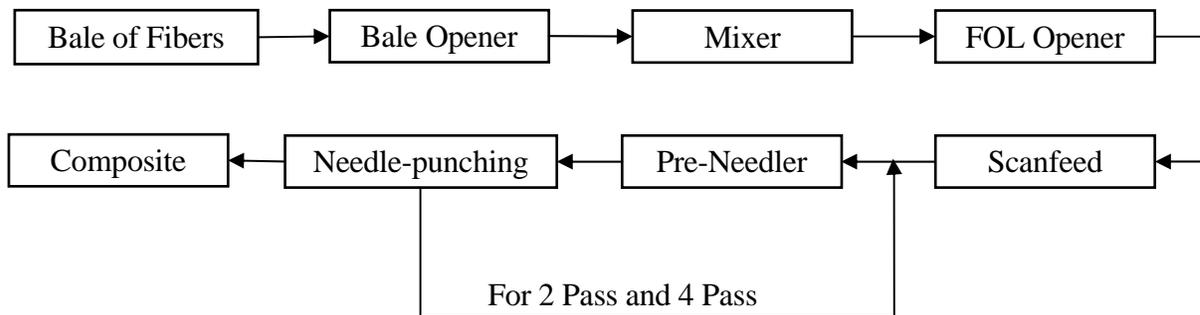


Figure 4.1 Process flow

#### 4.2 Experimental Design for web formation:

Fiber webs formed on the scanfeed machine were needle punched in order to provide the web with some strength and structural integrity, to be transported for further processing. A plan was thus formulated where the parameters were controlled on needle punching machine, and the resulting change in performance of the web, as well as the composites, were characterized. Two types of nonwoven webs were made. The first nonwoven web was made using only the hemp fibers. The web formation process had two variables, web weight and number of passes through the needle-punching machine (which essentially changed the needle punch density of the web). Since the Scanfeed machine provides a uniform web when operating above a basis weight of over 400 grams per square meter, the

plan was formulated to make webs with 500 and 1000 grams per square meter. Each web would then be processed through needle punching machine operating at 11.63 strokes per square inch for 1 pass, 2 passes and 4 passes. This would lead to formation of 6 different types of web: 3 webs having lower weight(500 g/m<sup>2</sup>) with 11.63, 23.26 and 46.52 strokes per square inch needle punch density; and 3 webs at higher weight(1000 g/m<sup>2</sup>) 11.63, 23.26 and 46.52 strokes per square inch needle punch density. The material passage through needle-punching machine, as shown in Figure 4.2, was as described below:

- For 1 pass one side was passed through needle-punching machine.
- For 2 pass web was needle-punched first on one side and then in the next passage the side was reversed and the back side was needle-punched.
- Similarly, for 4 pass, front and back side were needle-punched twice with reversal of side each time before passage through needle-punching machine.

The design of experiment for fiber processing is shown in figure 4.2 below

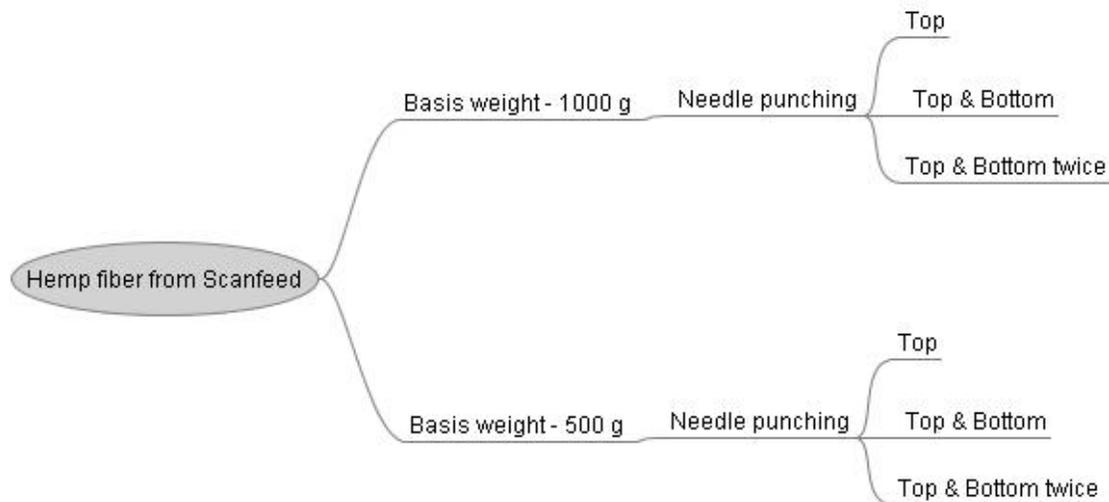


Figure 4.2 Design of experiment for hemp fiber web needle-punching

The second type of nonwoven web was made from the blend of hemp and polylactic acid (PLA) fibers. The fiber blending was performed at the BXM opener according to appropriate weight fraction of fibers. Three different blend ratio's were selected for web and hence composite formation: (1) 25% hemp – 75% PLA; (2) 50% hemp – 50% PLA; and (3) 75% hemp – 25% PLA. These proportions of the hemp fibers should provide a broad range of the fiber volume fraction and will provide information about the mechanical properties of reinforced composites.

#### *4.3 Experimental design for composite fabrication:*

The plan was formulated to convert the nonwoven fiber webs made from the hemp and hemp-PLA fibers in to composites, and perform a comparative study of the mechanical properties. It was intended that the nonwoven webs made out of the hemp fibers were to be infused with the resin. As epoxy resins are widely used in the industry they were selected to infuse the hemp fiber web to form composites that would serve as bench mark for hemp-PLA composites. The resin used for infusion of nonwoven webs was Derakane Momentum<sup>®</sup> by Ashland chemicals.

PLA in fiber form was selected to be used as matrix material due to the ease of processing and blending it with the hemp fibers. The three different blend ratios employed were: 25% hemp – 75% PLA, 50% hemp – 50% PLA, and 75% hemp – 25% PLA. Here PLA fibers upon melting would encapsulate hemp fibers forming a matrix. The melting point of PLA is in the range from 170°C -200°C. Thus, the processing temperature selected to fabricate the composite were 190°C, 200°C and 210°C. These temperatures were selected so that we had representative processing temperatures below the polymer melting point, close to

melting point and the last one above melting point of PLA polymer. Literature also suggested ideal processing temperature in vicinity of 200°C for PLA polymer.

The hemp-PLA webs at each selected blend ratio were heat pressed in between Teflon® sheets at the three selected temperatures for 3 minutes. This gave 9 different types of composites, i.e. 3 different blend ratios at each processing temperature or at each blend ratio composite formed at 3 different temperatures. Figure 4.3 below shows the experimental design for fabrication of the hemp-PLA composite.

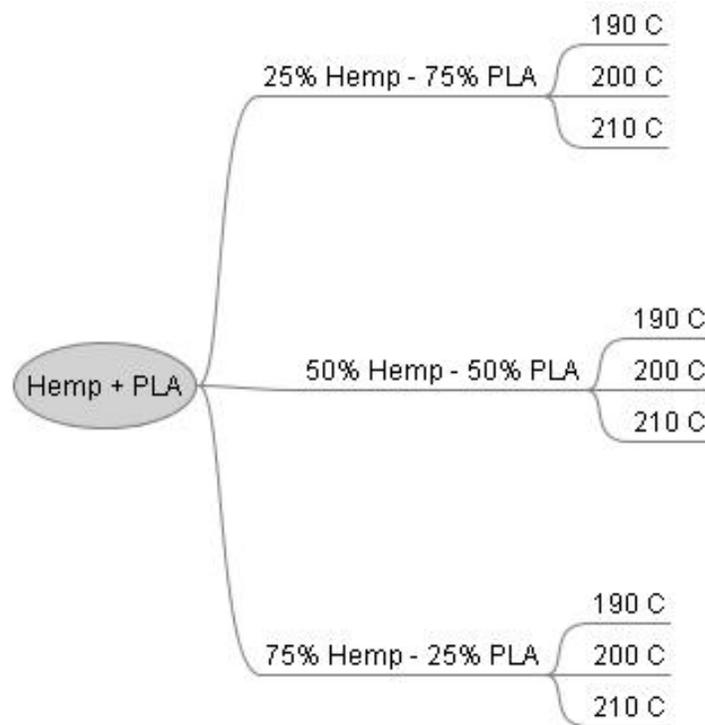


Figure 4.3 Design of Experiment for hemp-PLA composite fabrication

#### 4.4 Fiber Characterization:

The hemp fibers used for processing forms the building blocks of the composites and hence

were characterized for length distribution, fineness and tensile properties. The characterization of each property was performed for the hemp fibers at bale stage and sequentially at each processing stage until the Scanfeed in order to study the effect of the fiber processing. Length distribution analysis was performed using a Peyer machine. The fibers were then characterized for fineness by averaging the diameter measurements at 5 places along the length of the fiber using an optical microscope and an image analysis system. The same fibers were then tested for tensile properties on a constant rate of extension (CRE) type tester according to method ASTM D3822.

#### *4.5 Web characterization*

For each nonwoven web specimen thickness and weight were determined. The thickness measurements were performed at 3 places along the length of each specimen. Then the overall sample thickness was determined by averaging measurements for 10 specimens. Thickness was determined using a manual thickness tester having 1/1000 inch (0.0254 mm) precision as directed in test method ASTM D5729. Further, the specimen weight was determined using a weighing scale having precision of .001 g.

Although the target weight for the hemp webs were 500 and 1000 grams per square meter ( $\text{g/m}^2$ ) for 1 pass, 2 pass and 4 pass of web through needle-punching machine. Achieved web weights were:

- 1 Pass – 708 ( $\text{g/m}^2$ )
- 2 Pass – 634 ( $\text{g/m}^2$ )
- 4 Pass – 695 ( $\text{g/m}^2$ )
- 1 Pass – 1100 ( $\text{g/m}^2$ )
- 2 Pass – 1135 ( $\text{g/m}^2$ )
- 4 Pass – 1200 ( $\text{g/m}^2$ )

Each of the above webs used for reinforcing the epoxy vinyl ester resin, as well as all hemp-PLA webs were characterized for tensile properties using strip test (ASTM D5035),

for flexural properties using Cantilever test (ASTM D5732) and for air permeability (ASTM D737).

All the webs were also characterized for evenness using an adapted flat bed scanner together with proprietary software by Alasso industries, for analyzing the image of web for evenness.

#### *4.6 Composite characterization*

For each composite specimen the thickness and weight were determined. The thickness measurements were performed at 3 places along the length of each specimen. Overall sample thickness was determined by averaging measurements for 10 specimens. Thickness was determined using manual thickness tester having 1/1000 inch (0.0254 mm) precision as directed in test method ASTM D5729. Specimen weight was determined using a weighing scale having precision of .001 g.

Each hemp-PLA composite sample was characterized for evenness, using a technique similar to that used for the webs, utilizing a flatbed scanner and image analysis software. The composites were further characterized for tensile properties using test method ASTM D3039 and flexural properties using test method ASTM D790.

The hemp webs were infused using Vacuum assisted resin transfer molding (VARTM) in order to make uniform and compact composites. Vacuum pressure used for the infusion process was 30 mm on Hg scale. The composites were then characterized for tensile properties using test method ASTM D3039 and flexural properties using test method ASTM D790.

#### *4.7 Methods for nonwoven web characterization:*

The following tests were performed on both hemp and hemp-PLA nonwoven webs:

##### *4.7.1 Tensile test (ASTM D5035):*

For characterizing the tensile properties of the nonwoven webs, the strip test was used. Each specimen was 25 by 200 mm, with the long direction representing the test direction. The testing was performed on Q-Test, a CRE type tester using a 250 lb load cell. The gauge length for testing was 75 mm and the cross-head speed used was 300 mm/min. For each sample, 10 specimens were tested in each machine directions (MD) and cross-machine direction (CD). Data for load-elongation curve, elastic modulus, peak load and % strain data were acquired by the testing machine and provided at end of each test.

##### *4.7.2 Flexural properties (ASTM D5732):*

The stiffness of the nonwoven webs was measured by determining bending length as directed in test method ASTM D5732. Each specimen was 25 by 200 mm, with the long direction representing the test direction. The testing was performed on IDM tester, with the metal slide moving at rate of 120 mm/min. Bending length was measured for face and back of both ends providing four readings for each specimen.

After determining bending length, based on thickness and weight of specimen bending modulus and flexural rigidity were calculated.

##### *4.7.3 Air permeability (ASTM D737):*

To obtain an indication of the ease of wetting of webs by resin, air permeability of each sample was determined in accordance with method ASTM D737. 10 measurements were made for each sample and the air permeability was measured in terms of  $\text{ft}^3/\text{min}/\text{ft}^2$ .

#### *4.8 Methods for composites characterization:*

##### *4.8.1 Tensile test (ASTM D3039):*

The primary objective of this test was to evaluate the in-plane tensile properties of the hemp-PLA and hemp-epoxy vinyl ester resin composites. For each sample, eight to ten specimens were tested in each machine and cross-machine direction.

Each specimen was 25 by 250 mm, with long direction representing the test direction. In order to prevent slippage and premature failure at the grips, the hemp-epoxy vinyl ester resin specimen ends were bonded to tabs on the front and back sides using a fast curing epoxy. The tabs were made from the same material, i.e., hemp-epoxy vinyl ester composite and they were cut to 25 by 50 mm dimensions. After bonding tabs specimens were left in the laboratory for at least 24 hours to allow the epoxy to cure and for sample conditioning.

The specimens were tested on Sintech, a CRE type tester using 1000 lb load cell. Pneumatic grips, with grip pressure of 1600 psi, were used to prevent specimen from slipping.

The cross-head speed used was 2 mm/min, and gauge length was 150 mm. Load-elongation curve, breaking load, peak stress and % strain at peak stress were acquired in real time by machine and provided at the end of each test.

##### *4.8.2 Flexural properties (ASTM D790):*

Flexural tests were performed according to ASTM D790 to determine flexural strength and the elastic modulus in flexure of the composites. In the three point bending test the maximum axial fiber stress is located directly under the loading point and the midspan deflection is measured using the cross-head motion.

Each specimen was 25 by 127 mm, with long direction representing the test direction. The simply supported span was 76 mm. The specimens were tested on a CRE type tester with load cell of 500 N at rate of 24 mm/min.

Based on the measured deflection,  $D$ , the corresponding strain in the outer fibers,  $\epsilon_f$ , can be calculated as

$$\epsilon_f = \frac{6Dd}{L^2} \dots\dots\dots \text{Eq. 3.1}$$

Where:

$d$  = Depth of the beam, inch

$L$  = Support span, inch

Based on the measured load,  $P$ , the corresponding stress in the outer fiber,  $f_f$ , can be calculated as:

$$f_f = \frac{3PL}{2bd^2} \dots\dots\dots \text{Eq. 3.2}$$

For the hemp-PLA composites, an analysis was carried out to determine the uniformity of the material. The measurement was performed by scanning the samples under standardized condition and then analyzing the images using proprietary software from Allasso Industries. Measurements were done for the overall sample panel as well as for individual specimens utilized for tensile and flexural testing.

## Chapter 5

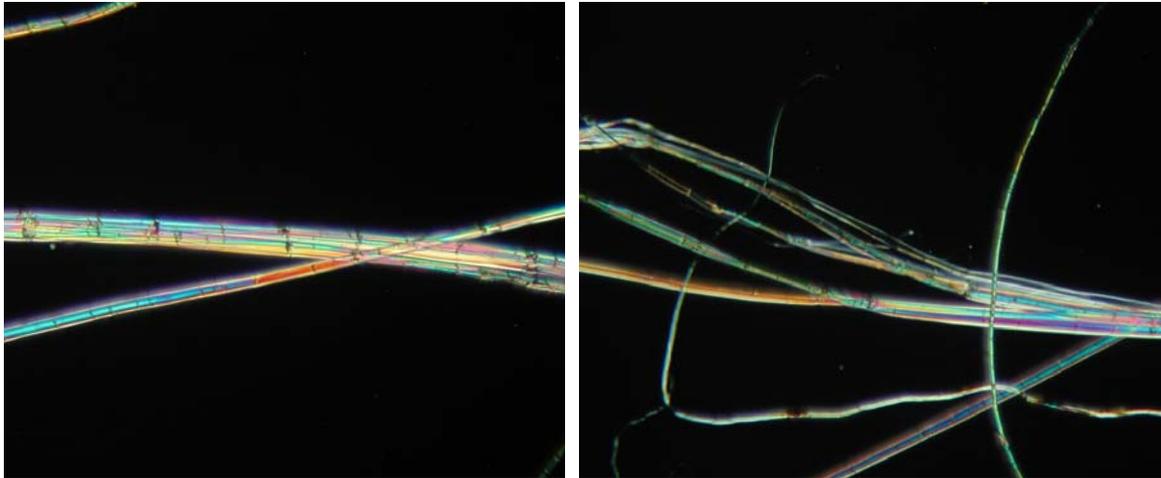
### RESULTS AND DISCUSSION

In this chapter the results of the experimental investigations for the fibers, the needle punched webs and the composites are tabulated, and analysis is provided for their behavior.

#### *5.1 Fiber testing results:*

The hemp fibers belong to the “bast fiber” category. The bast fibers are composed of a collection of fibrils “glued” together to form a fiber. As the fibers passes through various opening and blending machines, the glue loosens and the fiber separates into groups of smaller fibrils as shown in figure 5.1 below. This leads to formation of finer fibers. Hemp fibrils are much stronger than the collection of the fibrils forming the fiber (which is essentially a composite structure). Thus the opening process not only makes the fibers finer, by physically breaking the bonds holding the fibrils together, but also makes the measured “fiber strength” greater, since individual (or smaller agglomerates of) fibrils may now be measured. Since the fibrils are separated there is no significant change in length of fibers. These trends can be seen in graphs 5.2, 5.3 and 5.4.

Fiber length, fiber fineness and tensile properties at each sequential stage of the fiber processing are listed in Table 5.1. The change in those properties along the processing line is shown in Figure 5.2, 5.3 and 5.4.



(a) Hemp fiber

(b) Hemp fibers beginning to separate

Figure 5.1 – Hemp fiber – Microscopic view

Table 5.1 Fiber properties

Machine	# of fibers characterized	Tenacity(N/tex)		Fineness (tex)		Mean Length (mm)	
		Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
Unopened	631	0.58	0.75	10.15	8.88	23.4	3.3
Bale Opener	205	0.43	0.35	9.25	7.63	23.4	1.8
BXM	190	0.49	0.34	8.15	8.37	24.4	4.3
FOL	191	0.54	0.36	6.66	5.68	24.4	2.3
Scanfeed	192	0.54	0.38	6.24	7.44	24.9	1.3

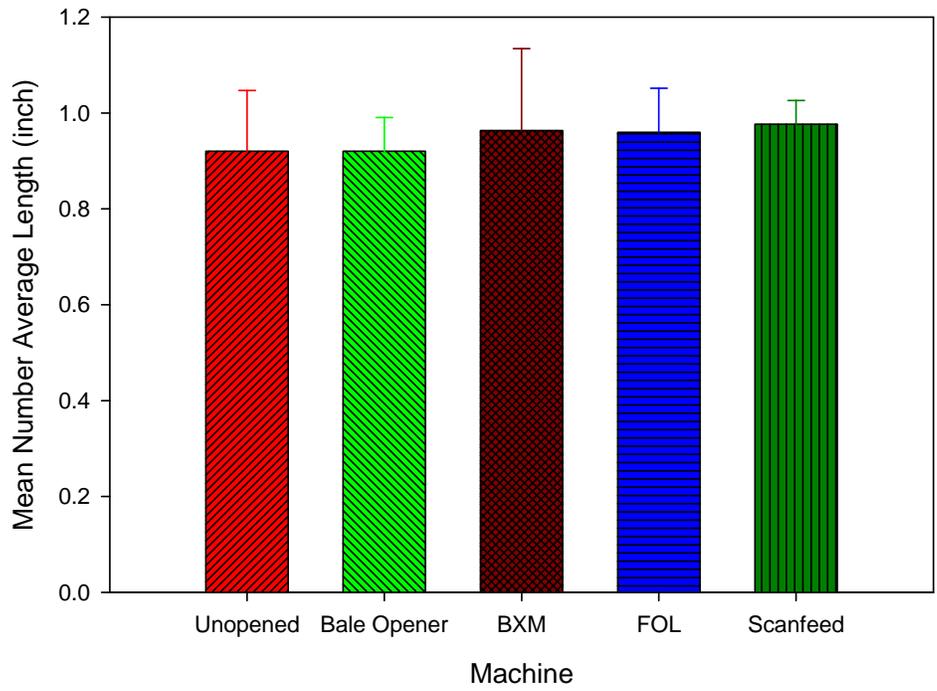


Figure 5.2 Fiber Length

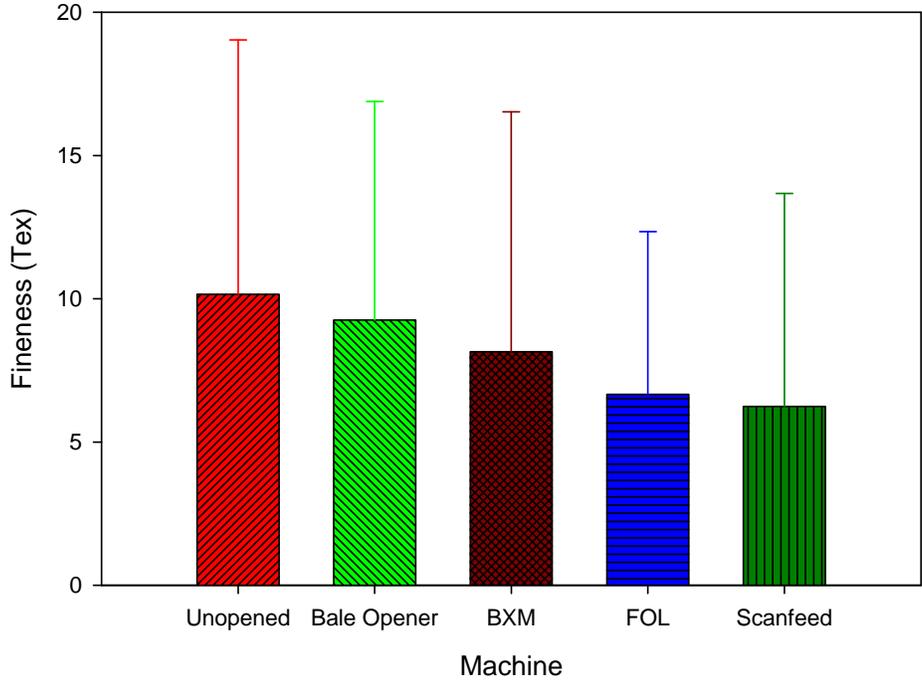


Figure 5.3 Fiber Fineness

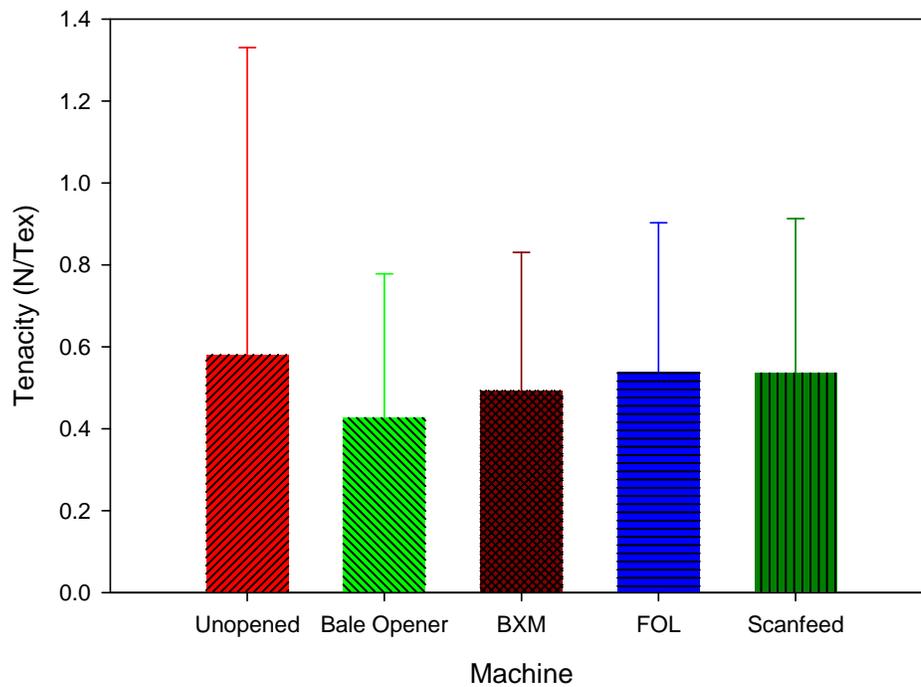


Figure 5.4 Fiber Tensile Strength

Hemp fibers from the bale, labeled as ‘unopened’ in above table and graphs, have high variability and are coarse. As the material passes through the opening machines, the fibers are opened. This in turn makes the fiber finer. Also, the process of opening the fibers leads to breaking of some of the weaker fibers and hence we see an increase in fiber strength (although not statistically significant) at each subsequent processing step.

Thus opening and cleaning of the fibers leads to finer fibers with less variability. Even though several hundred fibers were characterized, due to innate variability of the natural fibers there is a lot of variability as compared to the synthetic fibers.

### *5.2 Needle-punched nonwoven web characterization:*

The fibers at the end of the opening and cleaning process were converted into the web that was received from the scanfeed machine and then needle-punched. The needle punched web forms a reinforcing component for the hemp-epoxy vinyl ester composites; and it forms the composite for hemp- Polylactic acid fiber (PLA) webs. From the needle punched web characteristics inferences can be drawn regarding the composite properties. Thus the needle punched webs were characterized for their uniformity, mechanical properties and the air permeability.

5.2.1 Air permeability:

Table 5.2 Air permeability of needle punched webs

	Air permeability (ft <sup>3</sup> /min/ft <sup>2</sup> )										
	4Pass 1200 g/m <sup>2</sup>	4Pass 695 g/m <sup>2</sup>	2Pass 1135 g/m <sup>2</sup>	2Pass 634 g/m <sup>2</sup>	1Pass 1100 g/m <sup>2</sup>	1Pass 708 g/m <sup>2</sup>	25% Hemp 75% PLA-2P	50% Hemp 50% PLA-1P	75% Hemp 25% PLA-1P	50% Hemp 50% PLA-2P	75% Hemp 25% PLA-2P
1	30.20	81.30	32.40	108.00	32.20	76.20	28.60	30.40	35.76	40.48	44.69
2	41.50	83.35	32.00	107.00	41.36	61.50	31.20	29.60	33.50	42.34	47.45
3	34.04	82.53	36.58	133.40	37.54	87.68	31.40	28.60	39.36	43.17	52.14
4	37.38	86.92	34.40	122.90	36.58	89.20	31.80	29.20	36.74	43.17	53.38
5	35.42	122.90	32.40	130.10	30.60	74.40	29.00	29.00	37.38	44.00	53.38
6	34.40	90.65	32.96	107.00	36.42	105.00	30.20	26.17	34.74	45.38	54.62
7	41.36	91.90	35.93	128.30	50.21	76.20	30.60	26.40	30.40	45.38	55.24
8	33.14	103.00	36.10	107.00	31.00	88.06	32.20	27.28	30.40	46.07	57.65
9	32.20	124.70	29.80	111.00	31.00	71.16	31.00	24.79	33.68	46.07	58.75
10	34.22	79.40	29.60	113.00	36.90	98.56	31.60	24.10	31.00	47.45	59.30
Mean	35.39	94.67	33.22	116.77	36.38	82.80	30.76	27.55	34.30	44.35	53.66
Std Dev	3.70	16.80	2.49	10.73	6.02	13.25	1.18	2.14	3.09	2.10	4.70

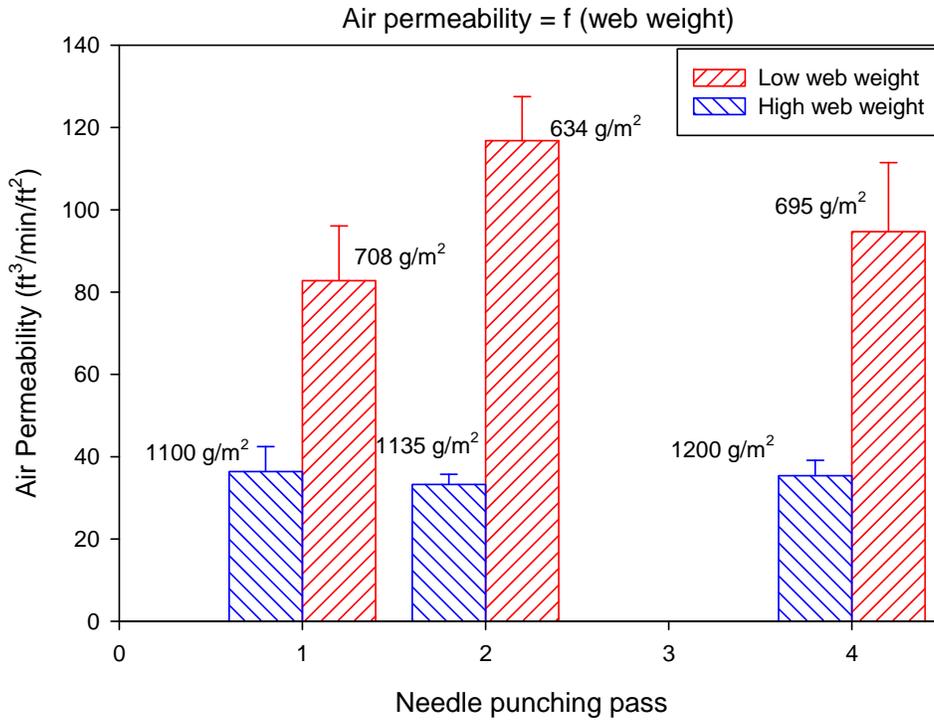


Figure 5.5 Air permeability of 100% Hemp fiber webs

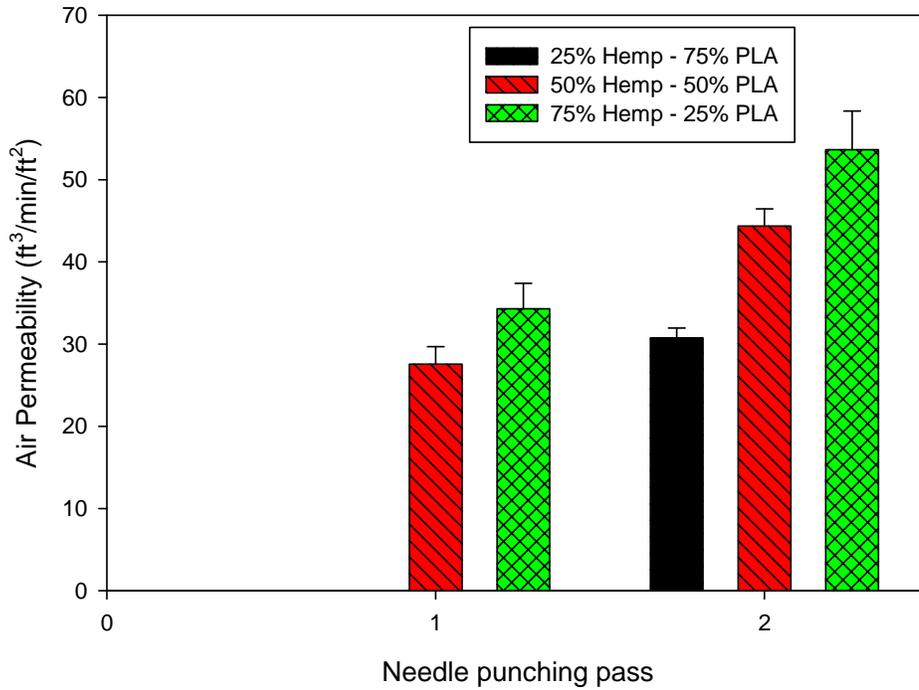


Figure 5.6 Air permeability of Hemp-PLA fiber webs

The air permeability of the needle-punched nonwoven web, provides the indication of resistance to wetting of the nonwoven web by resin, i.e. higher the air-permeability easier it will be to impregnate the web. The Air permeability of the needle-punched webs are listed in the table 5.2.

The air permeability of a web varies with its mass per unit area: as the web weight increases the fibers in web cross-section resisting air passage increases, leading to lower air permeability. The sample weight, and hence the air permeability of the samples, was divided into two groups for the hemp fiber webs. Due to significant weight difference between the two group of webs, the air permeability of those two group was significantly different as shown in Figure 5.5 above. During needle punching the needle passes in a plane perpendicular to the web re-orienting few fibers causing the fiber entanglement that provides web integrity and strength. This re-orientation and needle passage creates channels in the web especially when the needle punch density is high and webs are light weight. Needle punching also leads to web compaction and fiber breakage, that in low web weights with high punch density may result in holes in the webs. Thus for the webs within lower weight group, as the punch density i.e. needle punching passes increases the air permeability increases as shown in Figure 5.5.

In hemp- Polylactic acid (PLA) fiber webs, the fiber denier for the hemp fibers was in the range of 50 to 90 denier, where as the denier value of the Polylactic acid (PLA) fibers was approximately 1.5 denier. For the same web weight as the fiber fineness increases, the surface area resisting air flow increases, thereby reducing the air-permeability of the samples. For the hemp- Polylactic acid (PLA) fiber webs even though the web weight was between 500 to 700 g/m<sup>2</sup> the air permeability was lower and in range with the heavier

hemp webs. The lower values for the air permeability of the webs can be explained due to the use of finer Polylactic acid (PLA) fibers.

The air permeability of the hemp- Polylactic acid (PLA) fiber web is governed by both the number of pass through the needle punch machine as well as the blend ratio. As the proportion of the Polylactic acid fiber (PLA) fibers increases the surface area resisting the air flow increases, leading to lower air permeability. Thus the web with 75 % hemp and 25% PLA had the highest air permeability, whereas the web with 25% hemp and 75% PLA had the lowest air permeability.

Needle punching the web makes it more compact, and causes fiber breakage. Also, the passage of the needles through the web creates channels that might permit easy air passage. Thus increase in punch density i.e. number of passes through the needle punching machine leads to the higher values for air permeability of the web.

Table 5.3 Tensile properties of needle-punched nonwoven webs.

	Pass	Peak Stress (Mpa)				% Strain @ Peak load			
		MD		CD		MD		CD	
		Mean	$\sigma$	Mean	$\sigma$	Mean	$\sigma$	Mean	$\sigma$
25% Hemp-75% PLA	2	2.76	0.39	1.87	0.34	67.80	5.05	114.06	8.04
50% Hemp-50% PLA	1	1.90	0.16	1.71	0.21	75.46	6.51	88.20	6.10
75% Hemp25% PLA	1	1.37	0.13	1.21	0.19	47.55	7.24	71.98	7.85
50% Hemp-50% PLA	2	2.37	0.24	1.97	0.24	65.19	5.73	82.80	8.84
75% Hemp25% PLA	2	2.12	0.19	1.24	0.20	45.16	6.56	71.24	7.89
1Pass-708g	1	1.38	0.47	0.97	0.31	40.41	5.72	57.66	7.89
2Pass-634g	2	0.68	0.17	0.30	0.08	31.59	5.71	53.65	15.48
4Pass-695g	4	0.65	0.17	0.27	0.07	31.66	4.67	47.13	7.22
1Pass-1100g	1	0.91	0.28	0.82	0.46	25.33	7.19	47.46	11.23
2Pass-1135g	2	2.73	0.55	1.04	0.20	29.83	5.72	62.73	5.93
4Pass-1200g	4	1.24	0.24	0.50	0.16	21.66	4.79	56.80	7.84

5.2.2 Tensile properties:

In textile fiber processing, most of the machines that do the operation of opening and blending also partially orient the fibers in the machine direction. This partial orientation of the fibers leads to bias in the material properties rendering the machine direction stronger in terms of tensile properties as compared to the cross-machine direction as shown in the figures 5.7 to 5.10 below.

For hemp webs, as expected, with the increase in material weight the tensile strength characteristics of the needle-punched web improved. In lower web weights increase in number of passes through needle punch machine reduced the strength. This could be because increased needle punching would break the fibers more, deteriorating mechanical properties. Very high needle punch density even can lead to holes for lower web weight needle punched webs. For high web weight 2 passes through needle punch machine were found to be providing optimal properties.

For hemp- Polylactic acid fiber (PLA) webs, the blend ratio had the most significant effect. With increase in percentage of PLA fibers, the strength of the webs increased, even though the weight of the webs increased with decreasing PLA percentage. This can be attributed to higher number of PLA fibers that are finer as compared to hemp fibers, for same web weight, contributing for the mechanical properties. The graphs below shows peak stress as per test direction for each web type. Thus from all the results and graphs it can be deduced that the 1135 g/m<sup>2</sup> web in machine direction for 25% hemp – 75% PLA web was the strongest from peak tensile stress as well as energy to break criteria.

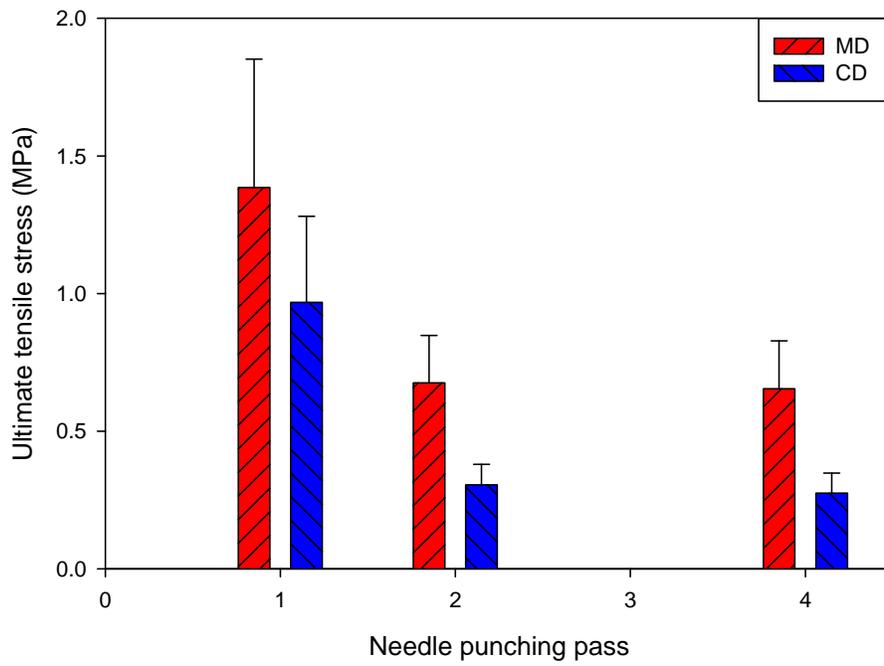


Figure 5.7 Ultimate tensile stress - 100% Hemp fiber webs  
Low web weight

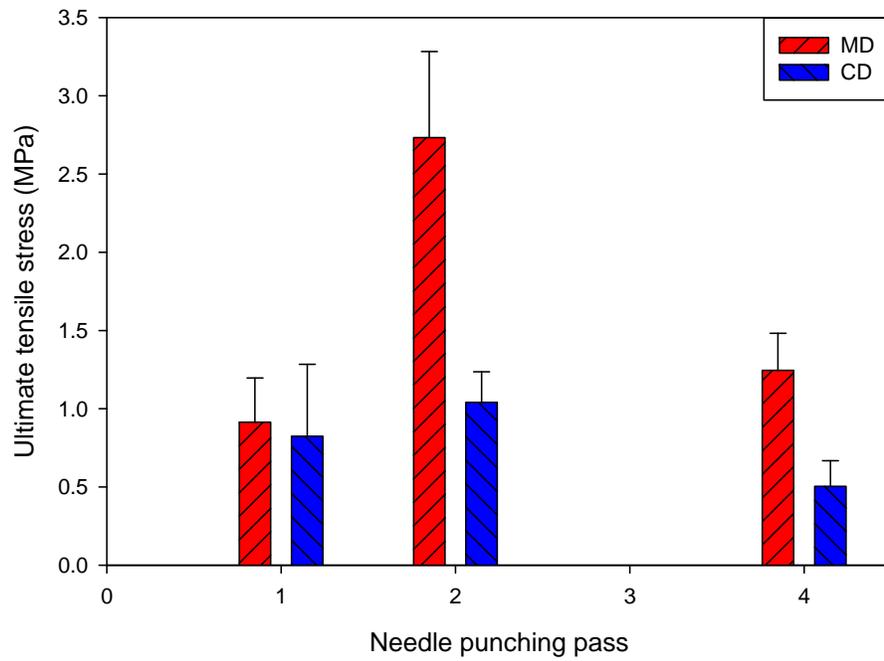


Figure 5.8 Ultimate tensile stress - 100% Hemp fiber webs  
High web weight

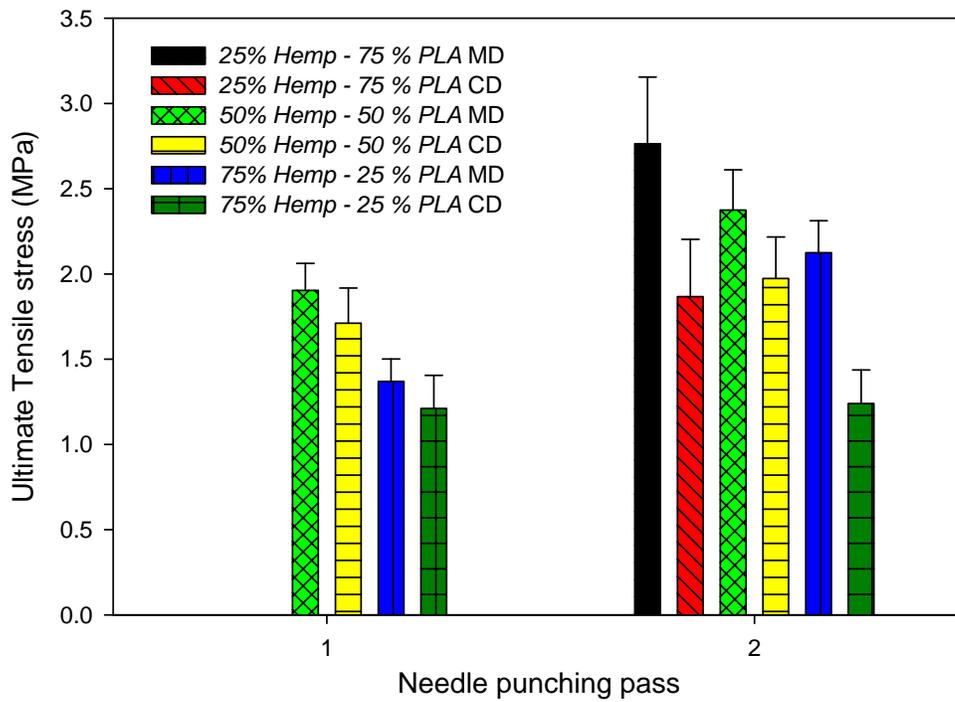


Figure 5.9 Ultimate tensile stress - Hemp-PLA fiber webs

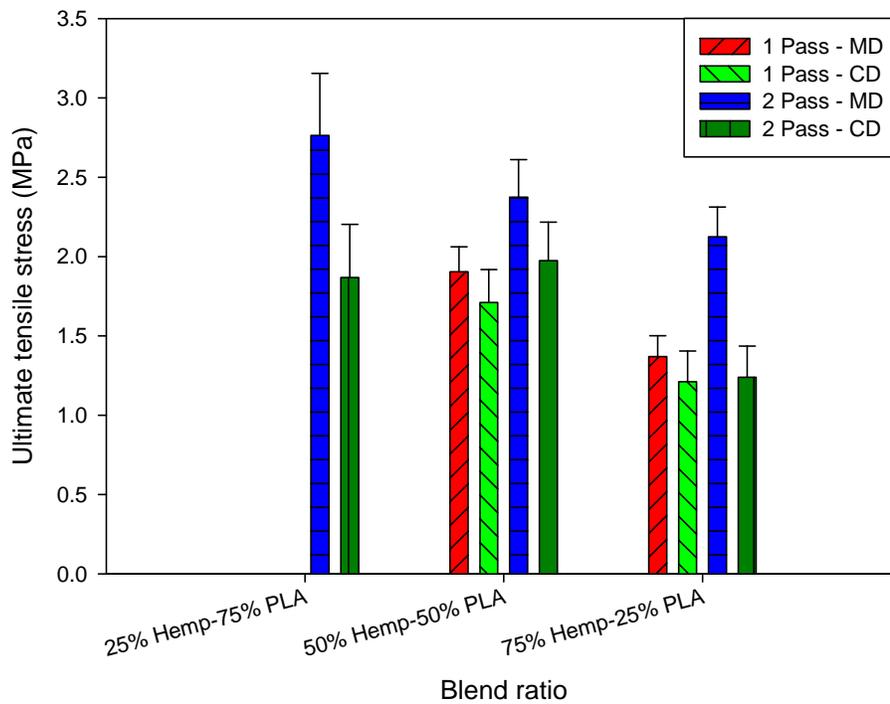


Figure 5.10 - Ultimate tensile stress Hemp-PLA fiber webs - Blend ratio.

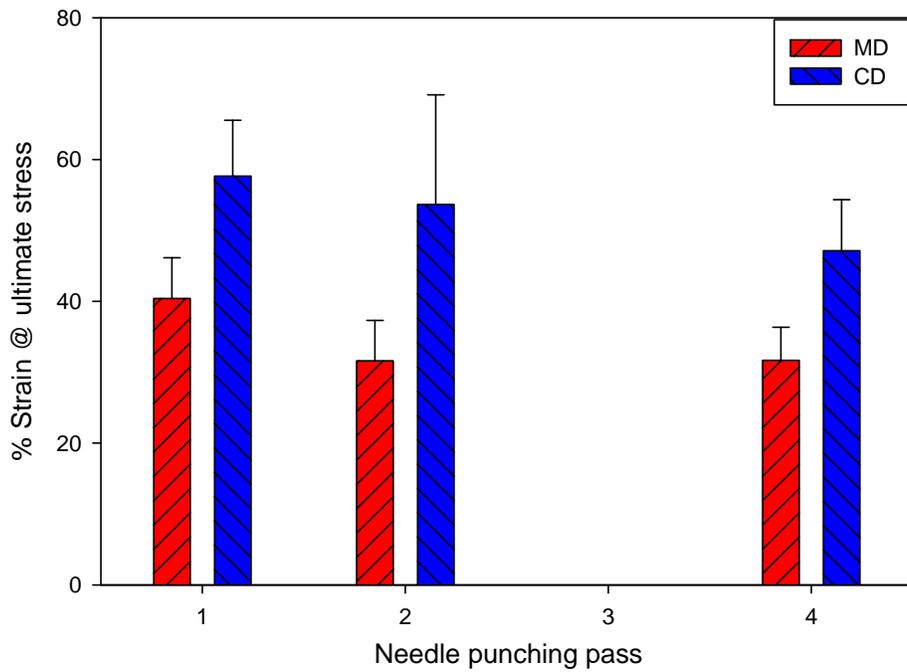


Figure 5.11 - % Ultimate tensile strain 100% Hemp fiber webs  
Low web weight

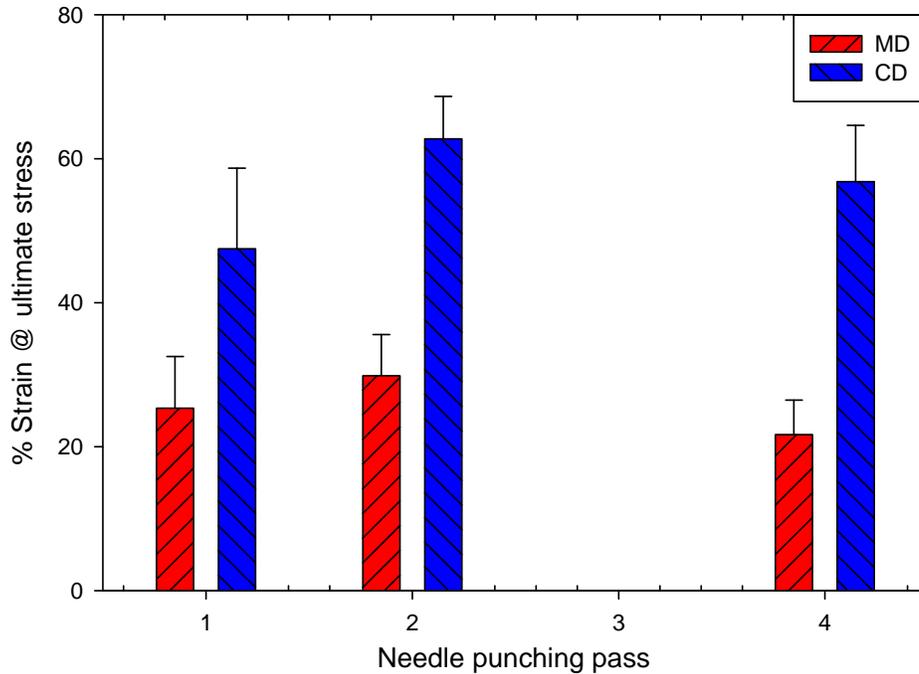


Figure 5.12 - % Ultimate tensile strain 100% Hemp fiber webs  
High web weight

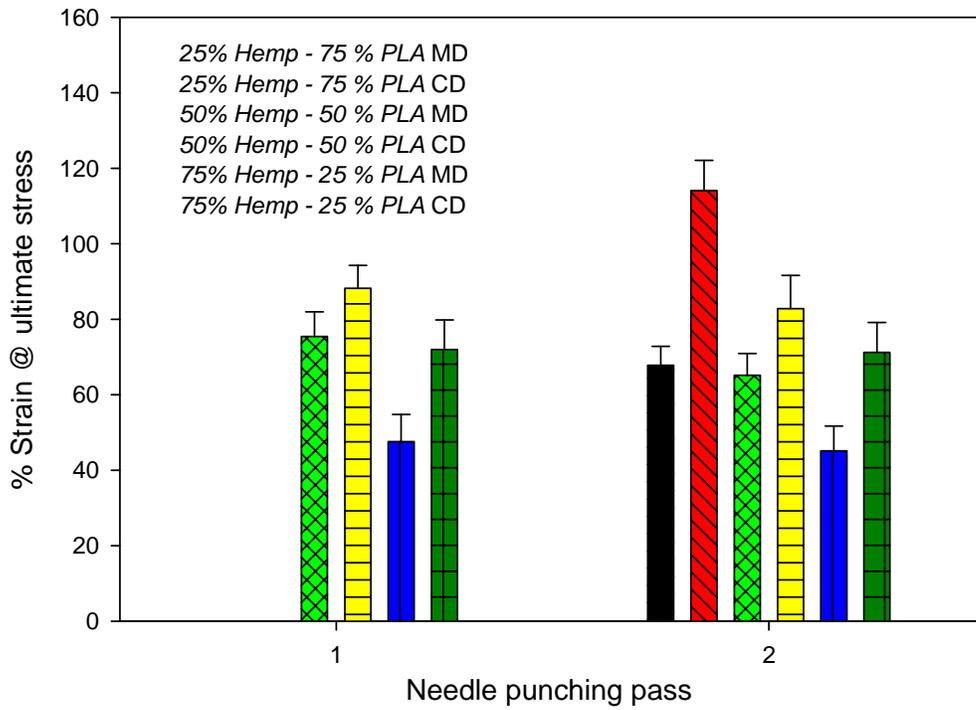


Figure 5.13 % Ultimate tensile strain Hemp-PLA fiber webs - NP

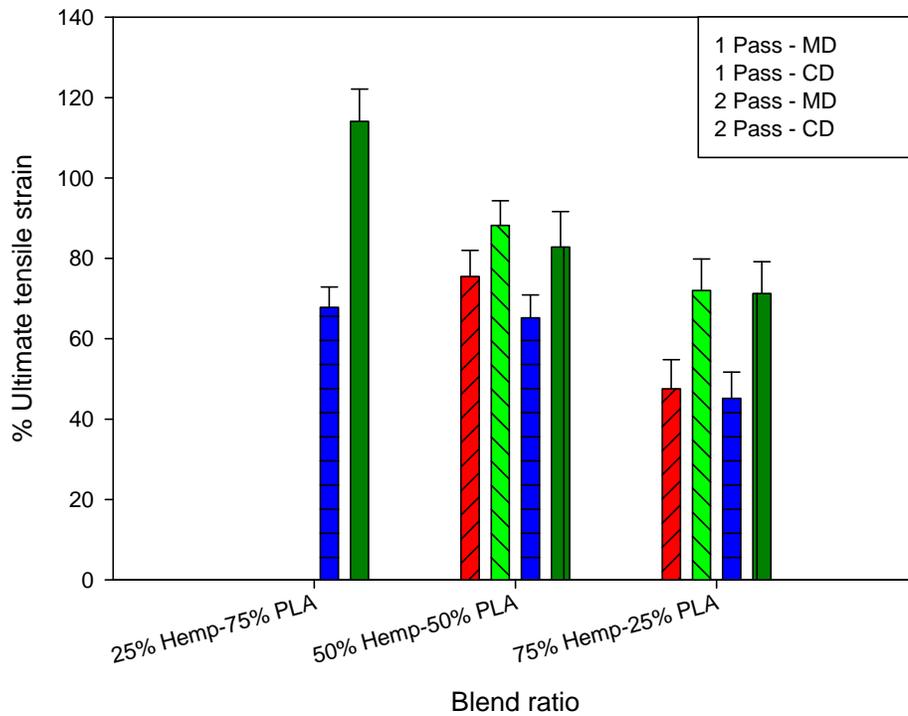


Figure 5.14 - % Ultimate tensile strain Hemp-PLA fiber webs - blend ratio.

Table 5.4 - Flexural properties of needle-punched web:

	Needle-punch Pass	Weight (g/m <sup>2</sup> )	Flexural rigidity (uN.m)				Bending Modulus (N/m <sup>2</sup> )			
			MD		CD		MD		CD	
			Mean	Std Dev	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
4 Pass – 1200 g/m <sup>2</sup>	4	1200			35362.94	13420.07			9057067.59	3077742.93
4 Pass – 695 g/m <sup>2</sup>	4	695	18681.93	2759.43	12450.84	5073.75	13626805.39	1724262.12	8390772.41	3045086.90
2 Pass – 1135 g/m <sup>2</sup>	2	1135			72376.67				8648193.95	
2 Pass – 634 g/m <sup>2</sup>	2	634	26004.39	5737.96	13658.83	3783.26	12494432.81	2420865.88	7453266.62	1993321.27
1 Pass – 1100 g/m <sup>2</sup>	1	1100			70146.24	8929.94			9287180.23	1145455.68
1 Pass – 708 g/m <sup>2</sup>	1	708	37911.77	12311.98	32621.15	7819.04	11996993.58	3660024.92	11105875.86	2148421.94
25-75 : Hemp – PLA	2	602	22364.39	6919.03	18525.80	6210.53	14927905.14	3463488.51	10791293.36	3401444.34
50-50 : Hemp – PLA	1	730	19667.18	4081.59	22017.46	5041.92	8565831.29	1466856.10	9385025.75	1696287.40
75-25 : Hemp – PLA	1	792	20595.99	4832.94	10640.00	1753.93	7036558.53	1183882.76	4038628.08	498851.28
50-50 : Hemp – PLA	2	650	25048.22	2417.75	27082.18	4776.79	19802303.60	1928755.83	20905755.75	2908235.91
75-25 : Hemp – PLA	2	718	40544.45	11051.97	27582.91	7898.91	28547074.46	6930466.95	20970142.85	3934046.19

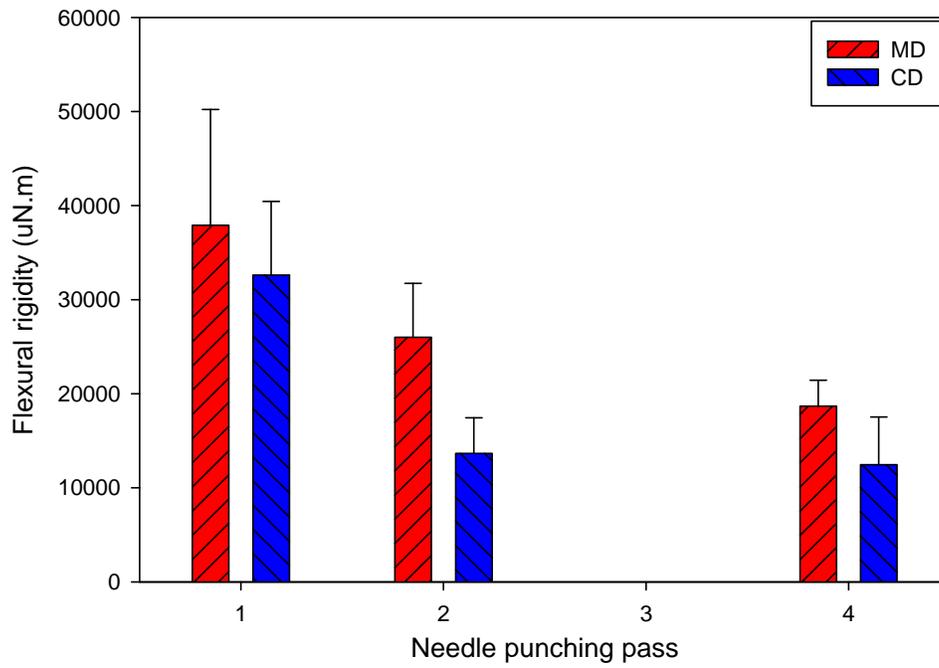


Figure 5.15 - Flexural rigidity 100% Hemp fiber webs  
Low web weight

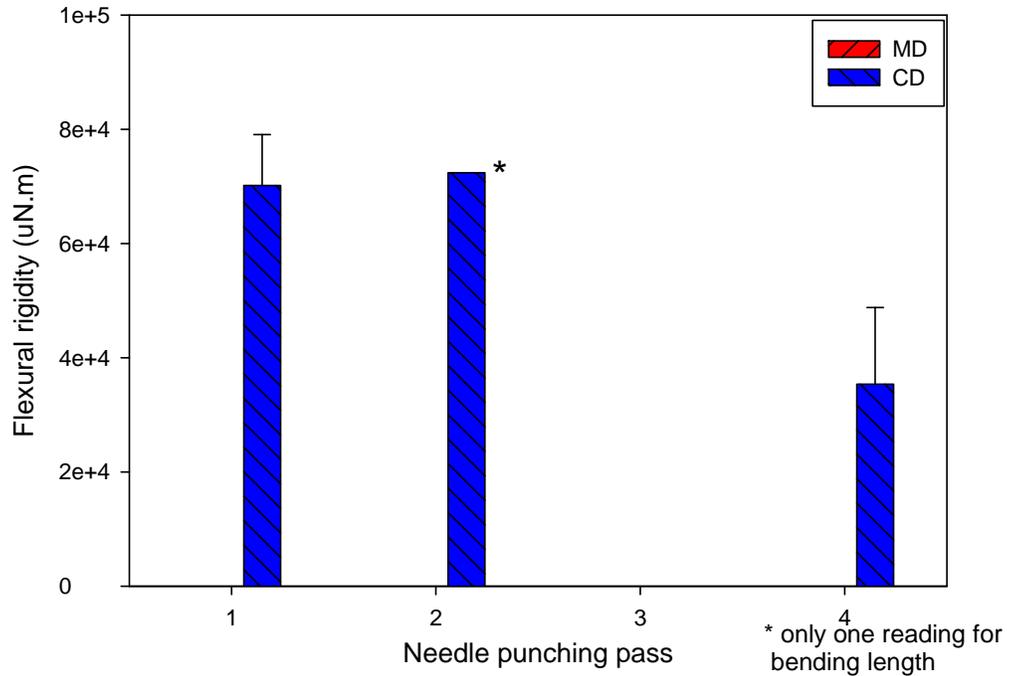


Figure 5.16 - Flexural rigidity 100% Hemp fiber webs  
High web weight

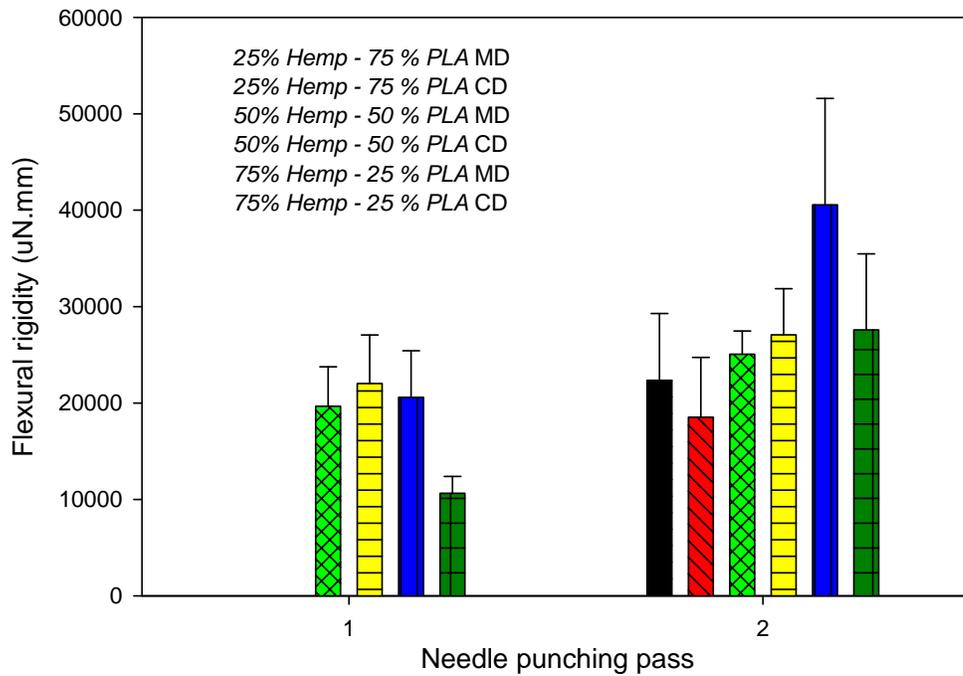


Figure 5.17 Flexural rigidity Hemp-PLA fiber webs - Needle punching pass

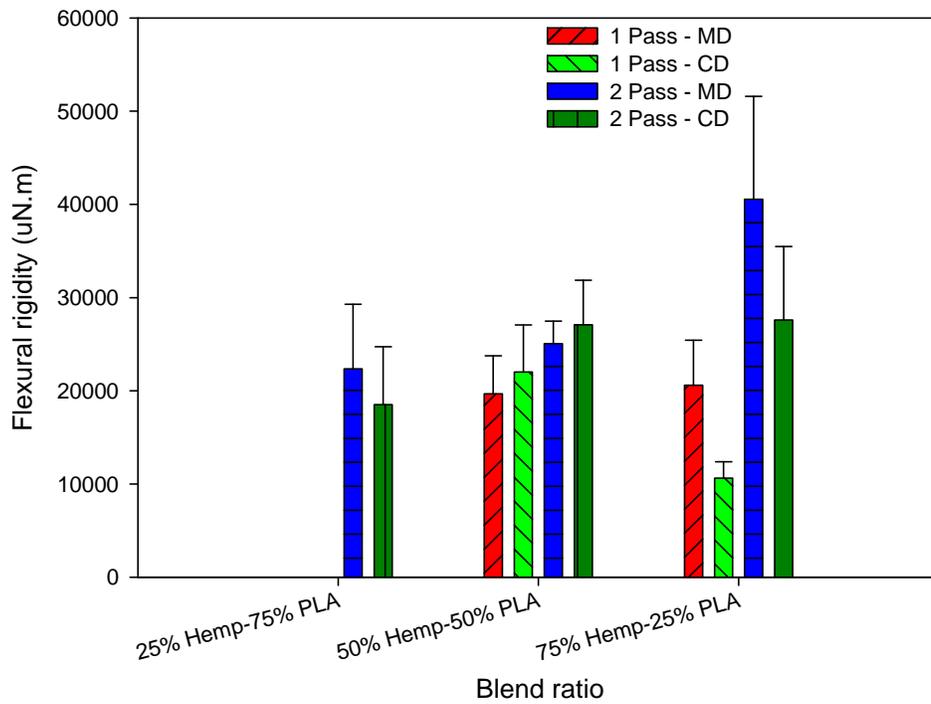


Figure 5.18 Flexural rigidity hemp-PLA fiber webs: blend ratio

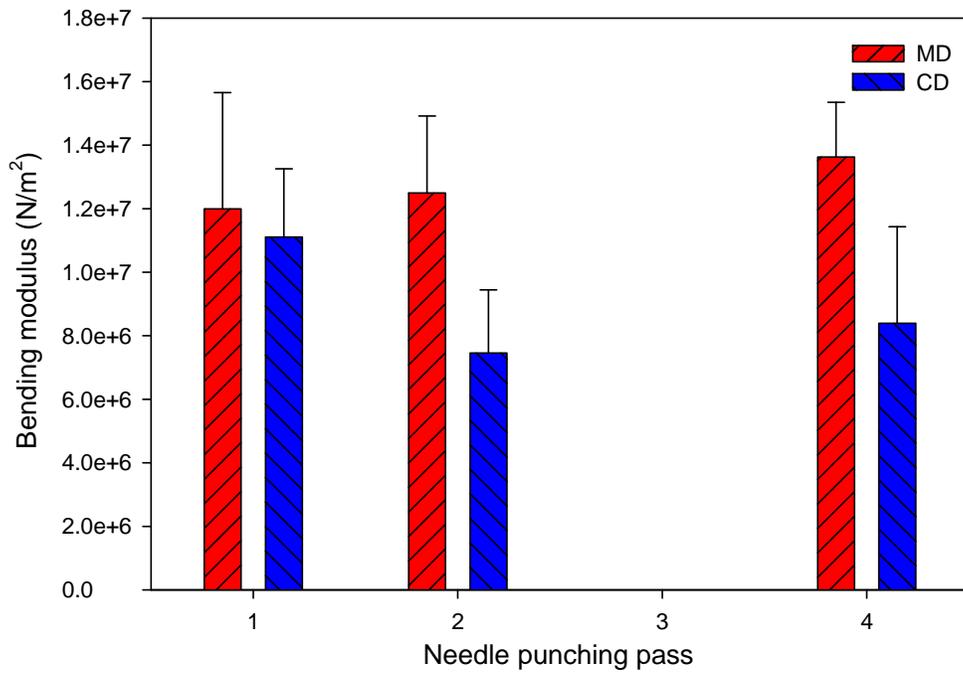


Figure 5.19 - Bending modulus 100% Hemp fiber webs  
Low web weight

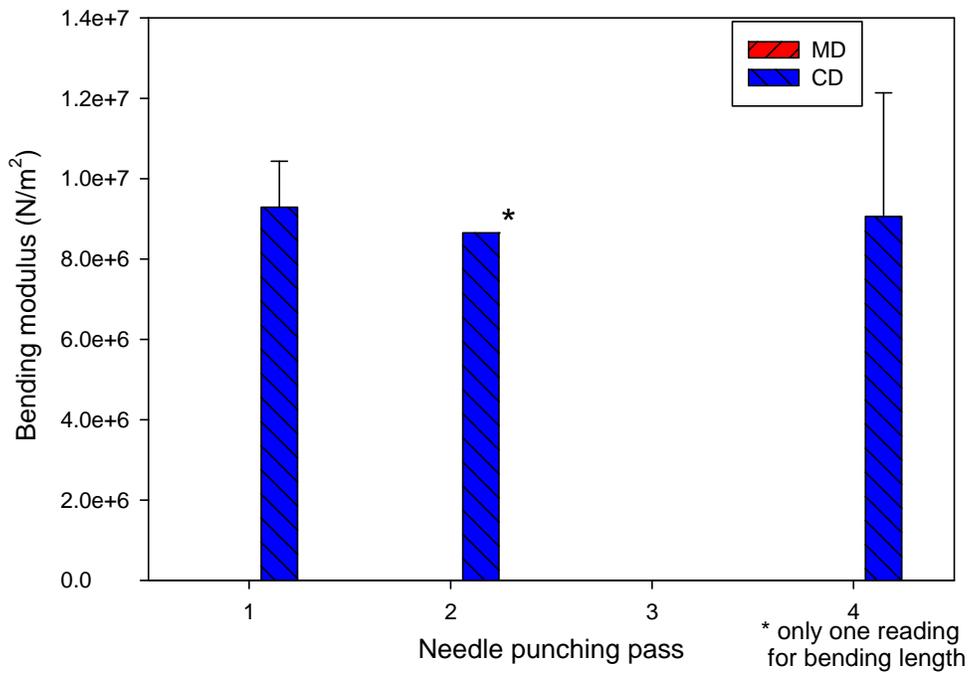


Figure 5.20 - Bending modulus 100% Hemp fiber webs  
High web weight

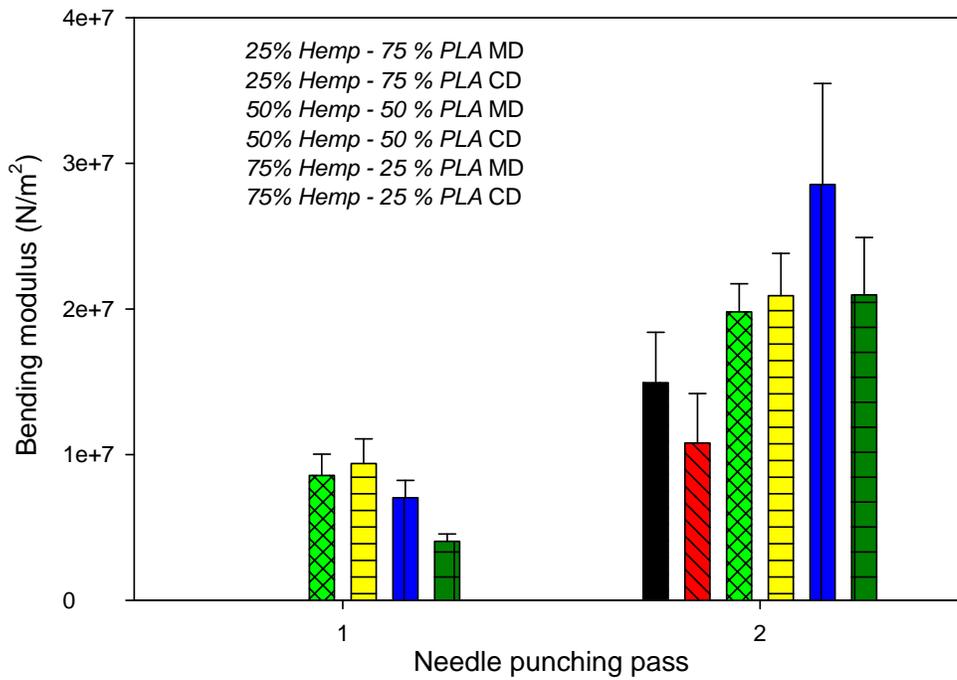


Figure 5.21 Bending modulus Hemp-PLA fiber webs - Needle punching pass

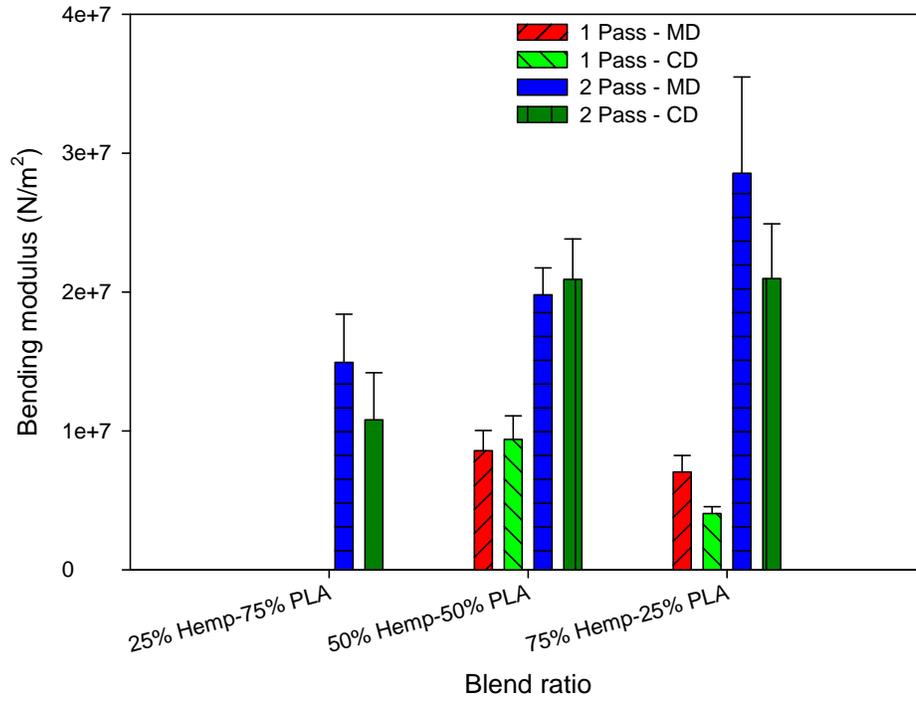


Figure 5.22 Bending modulus hemp-PLA fiber webs: blend ratio

### *5.2.3 Flexural properties:*

For hemp webs the flexural properties, in terms of both bending modulus and flexural rigidity, were dependant on the web weight. As the web weight increased the bending modulus increased. Some of the higher weight webs in machine direction could not even be characterized for the flexural properties. An increase in needle punch density, i.e. needle punch pass, within a weight group, initially leads to web compaction. However, further increase in the needle punching passes leads to higher fiber being broken and subsequently lower flexural rigidity.

For hemp – polylactic acid (PLA) fiber webs, flexural properties were affected by both blend ratio as well as number of needle punching pass. As the number of needle punch passes increased the webs became more compact and stiffer increasing its flexural rigidity and bending modulus. The increase in flexural rigidity with increasing hemp fiber percentage in the blend can be explained by inherent rigidity of the bast fibers as compared to the Polylactic acid (PLA) fibers.

### 5.3 Hemp – PLA composites characterization:

The idea behind fabricating the hemp – polylactic acid (PLA) fiber composite was the ease of taking the web anywhere and heat pressing it to form the composite in desired form and shape without the requirement of an expensive equipment or extensive process. From the prior knowledge by literature review, the maximum fiber volume fraction used for reinforcing the composite was around 60% for glass fibers used to reinforce epoxy matrix. Thus the fiber volume fraction has to be lower than 60% in order to achieve reinforcing via hemp fibers.

From literature the elastic moduli of the Polylactic acid (PLA) fibers<sup>[19]</sup> and hemp fibers<sup>[13,38]</sup> are 5.33 GPa and 42 GPa. These values along with appropriate fiber volume fraction can be used to calculate the elastic modulus of composite using modified rule of mixture<sup>[52]</sup> (ROM) formula as given below:

$$E_c = kE_fV_f + E_m(1-V_f)$$

Where,  $E_c$  is the composite elastic modulus,  $E_f$  and  $V_f$  are the modulus and the volume fraction of the fibers,  $E_m$  is the modulus of the matrix and  $k$  is the fiber orientation factor ( $k = 1$  in case of long fibers aligned in the direction of test; in case of short fibers that are randomly oriented  $k < 1$ , value of  $k = 3/8$  for planar random configuration<sup>[57]</sup>).

The composite modulus based on the fiber elastic modulus values, calculated using the modified rule of mixture for various blend ratio's is given in table below:

Table 5.5 Comparison between theoretical and experimental composite elastic modulus

Blend ratio	Composite elastic modulus – Modified Rule of mixture (MPa)	Maximum value for composite elastic modulus – Experimental (MPa)
25 % hemp – 75 % PLA	7852.5	1258
50 % hemp – 50 % PLA	10485.0	1321
75 % hemp – 25 % PLA	13117.5	669

It can be observed from Table 5.5 that the composite elastic modulus values achieved are very low as compared to the predicted values by the modified rule of mixture equation. It can be explained by the fact that in this case Polylactic acid (PLA) used to form matrix is in fiber form. The polymers used for spinning fibers usually have high molecular weight and thus lower melt viscosity. This hinders the flow of PLA resin formed by melting of the PLA fibers between and around the hemp fibers to form a uniform matrix. The process of nonwoven web formation via Scanfeed machine is not able to provide a very uniform web with proper blend of the hemp and PLA fibers. Thus when the composites are fabricated by hot pressing the nonwoven webs, via melting of polylactic acid (PLA) fiber, the fibers melt locally. These localized melt point then instead of acting as matrix to transfer stress to fibers may act as stress points deteriorating mechanical properties of the composites. Thus the idea of using the Polylactic acid (PLA) in the fiber form, to form the matrix around hemp fiber may not be suitable for fabricating composites. As the benefits of both the hemp fibers as well as PLA in matrix form cannot be harnessed together in forming a composite. Also, as listed in literature under best of circumstances, fibers can act as reinforcing elements only till about volume fraction of 60%. As can be seen from Figure 5.23 below there is no significant improvement in composite tensile properties with increasing the proportion of reinforcing hemp fibers above 25%. In fact the tensile properties for composites with high fiber volume fraction is similar to the needle punched nonwoven webs, especially for the composites fabricated at low processing temperature from webs that have higher needle punch density, i.e. 2 passes through needle punch.

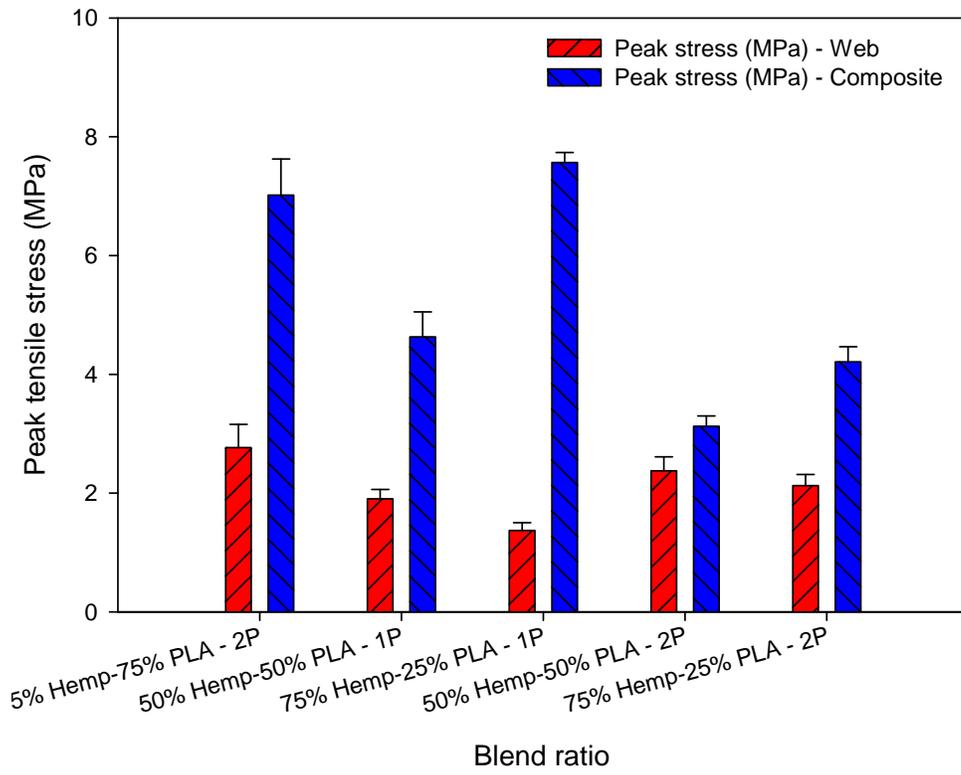


Figure 5.23 Comparison of web and composite properties

Table 5.6 Hemp-PLA composite tensile properties

	Hot press Temp (°C)	NP pass	Test Direction	Peak stress		% Strain		Modulus		Energy to Break	
				(MPa)	std dev		std dev	(Mpa)	std dev	(N-mm)	std dev
25% Hemp-75% PLA	190	2	MD	3.27	1.96	1.30	0.30	373.02	313.90	130.04	38.25
25% Hemp-75% PLA	200	2	MD	9.72	2.09	1.00	0.20	1258.59	368.36	188.92	64.01
25% Hemp-75% PLA	210	2	MD	9.60	2.52	1.10	0.10	1148.28	248.34	208.51	50.35
50% Hemp-50% PLA	190	1	MD	5.55	1.91	2.50	0.40	322.25	132.20	737.71	250.77
50% Hemp-50% PLA	200	1	MD	3.85	1.23	1.40	0.10	360.87	141.93	322.49	77.39
50% Hemp-50% PLA	210	1	MD	5.65	0.95	1.40	0.20	498.43	87.30	401.75	139.40
75% Hemp-25% PLA	190	1	MD	2.72	0.80	1.60	0.30	220.51	41.80	854.54	361.79
75% Hemp-25% PLA	200	1	MD	3.37	0.95	2.00	0.30	242.28	73.04	982.50	324.09
75% Hemp-25% PLA	210	1	MD	4.70	0.59	1.70	0.20	399.32	85.34	782.00	219.10
50% Hemp-50% PLA	190	2	MD	7.77	1.01	1.29	0.21	815.74	208.37	353.14	80.27
50% Hemp-50% PLA	200	2	MD	6.21	1.30	1.15	0.17	697.76	159.05	308.31	106.74
50% Hemp-50% PLA	210	2	MD	10.78	1.38	1.17	0.11	1182.91	77.37	376.63	80.95
75% Hemp-25% PLA	190	2	MD	3.49	0.75	1.30	0.25	382.25	77.10	581.68	270.09
75% Hemp-25% PLA	200	2	MD	5.35	0.85	1.13	0.22	594.75	78.86	518.38	256.18
75% Hemp-25% PLA	210	2	MD	5.84	0.82	1.13	0.13	669.88	67.39	488.04	172.14
25% Hemp-75% PLA	190	2	CD	2.96	1.08	1.10	0.20	372.70	152.08	106.25	26.78
25% Hemp-75% PLA	200	2	CD	8.63	1.72	1.00	0.20	1139.80	144.38	193.45	59.44
25% Hemp-75% PLA	210	2	CD	7.91	2.81	0.90	0.20	1169.10	355.07	151.38	61.03
50% Hemp-50% PLA	190	1	CD	5.92	1.19	1.40	0.10	564.83	109.92	441.51	172.06
50% Hemp-50% PLA	200	1	CD	3.12	0.78	1.40	0.20	298.13	61.68	277.43	152.64
50% Hemp-50% PLA	210	1	CD	3.69	1.62	1.70	0.30	313.87	201.91	360.29	89.65
75% Hemp-25% PLA	190	1	CD	2.30	0.88	2.00	0.60	214.92	61.66	819.72	523.07
75% Hemp-25% PLA	200	1	CD	2.31	0.51	2.90	1.00	163.80	47.80	1027.64	239.47
75% Hemp-25% PLA	210	1	CD	3.34	0.64	2.40	0.30	208.58	47.85	890.79	211.82
50% Hemp-50% PLA	190	2	CD	5.89	0.95	1.39	0.14	579.34	84.41	370.14	91.80
50% Hemp-50% PLA	200	2	CD	4.76	1.22	1.17	0.24	529.18	133.16	225.22	84.41
50% Hemp-50% PLA	210	2	CD	10.00	1.07	1.01	0.11	1321.37	84.72	318.91	76.53
75% Hemp-25% PLA	190	2	CD	3.02	0.26	2.06	0.26	261.70	49.24	885.22	203.07
75% Hemp-25% PLA	200	2	CD	2.80	0.65	1.61	0.22	273.29	51.22	686.41	178.18
75% Hemp-25% PLA	210	2	CD	4.74	1.01	1.26	0.18	543.98	43.65	403.14	148.70

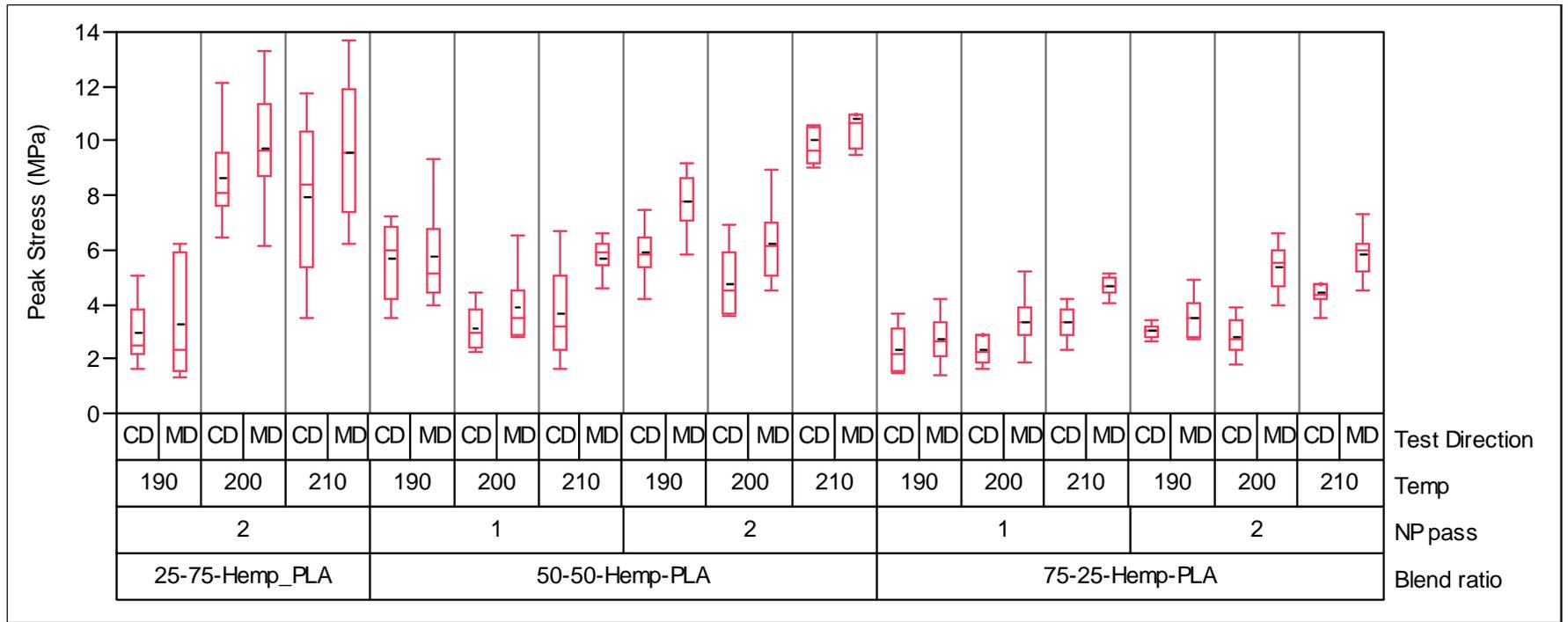


Figure 5.24 Peak stress Hemp-PLA composites

### *5.3.1 Tensile properties:*

Stress of the composites varied based on the blend ratio and hot press temperature, there was no significant effect of number of pass through needle punch machine. Strength of the composites increased with increasing percentage of Polylactic acid (PLA) fibers and increasing hot press temperature. Thus the composites having 25% hemp – 75 % PLA pressed at 200°C and 210°C had the highest strength.

The melting point of Polylactic acid (PLA) is around 170°C but from literature it could be determined that good operating window for hot pressing Polylactic acid (PLA) fibers was around 200 to 210°C. Thus our experiments corroborate that fact.

The effect of blend ratio could be explained by the fact that at low fiber volume fraction the fibers acts as filler and they do not contribute to composite mechanical properties. At high fiber volume fraction there is not enough matrix around all fibers and hence the stress is not distributed. In composite with 75% hemp fibers the proportion of PLA to form uniform matrix around the fibers is low leading to poor mechanical properties.

The strain of composites at varying processing conditions and with various blend proportion is shown in the graph below. Blend ratio does not significantly influence % strain at high processing temperatures. However, when the hot press temperature is 190°C Polylactic acid (PLA) fibers do not completely melt to form a uniform matrix around hemp fibers. Thus the hemp fibers are not bonded properly. This leads to composite failure due to fibers sliding past each other rather than fiber failure.

At high hot press temperatures of 200°C and 210°C the PLA melts completely. With the composites having high volume fraction of PLA, the melted polymer forms a uniform matrix, reinforcing matrix. This leads to composite with better tensile properties.

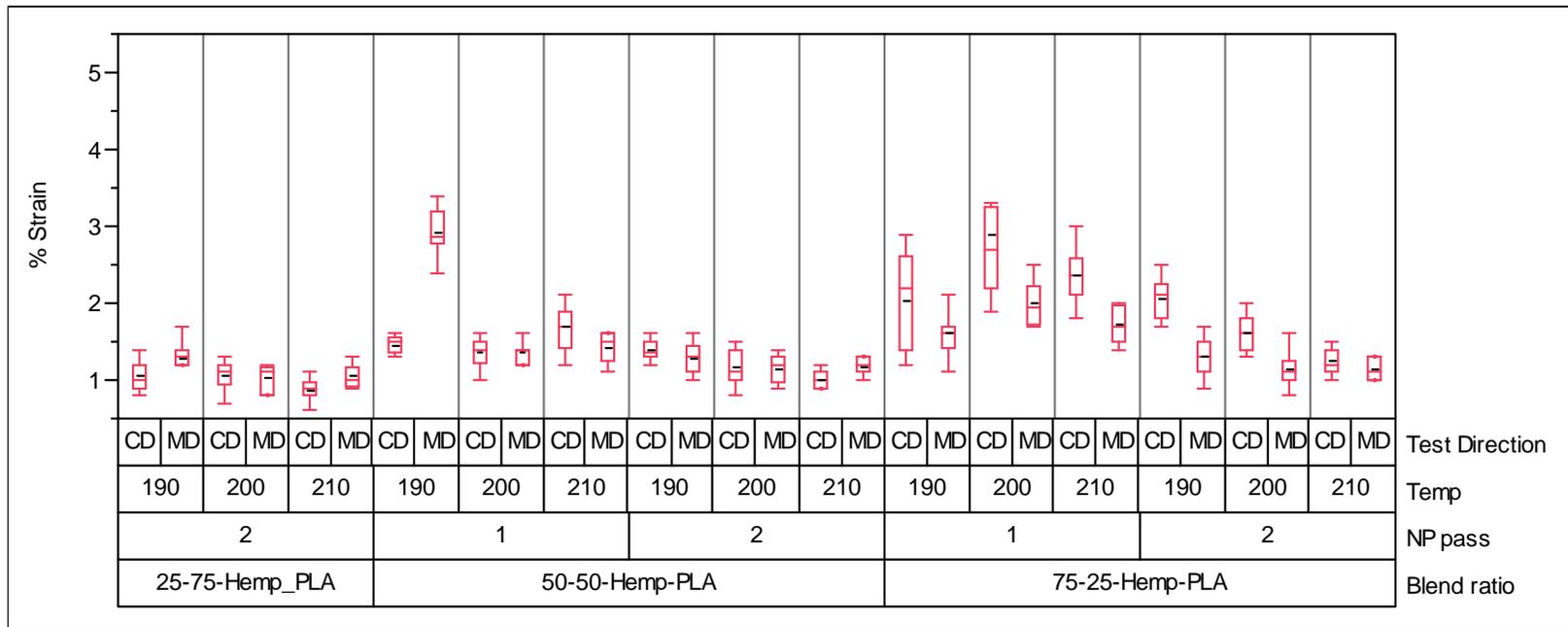


Figure 5.25 Peak tensile % strain Hemp-PLA composites

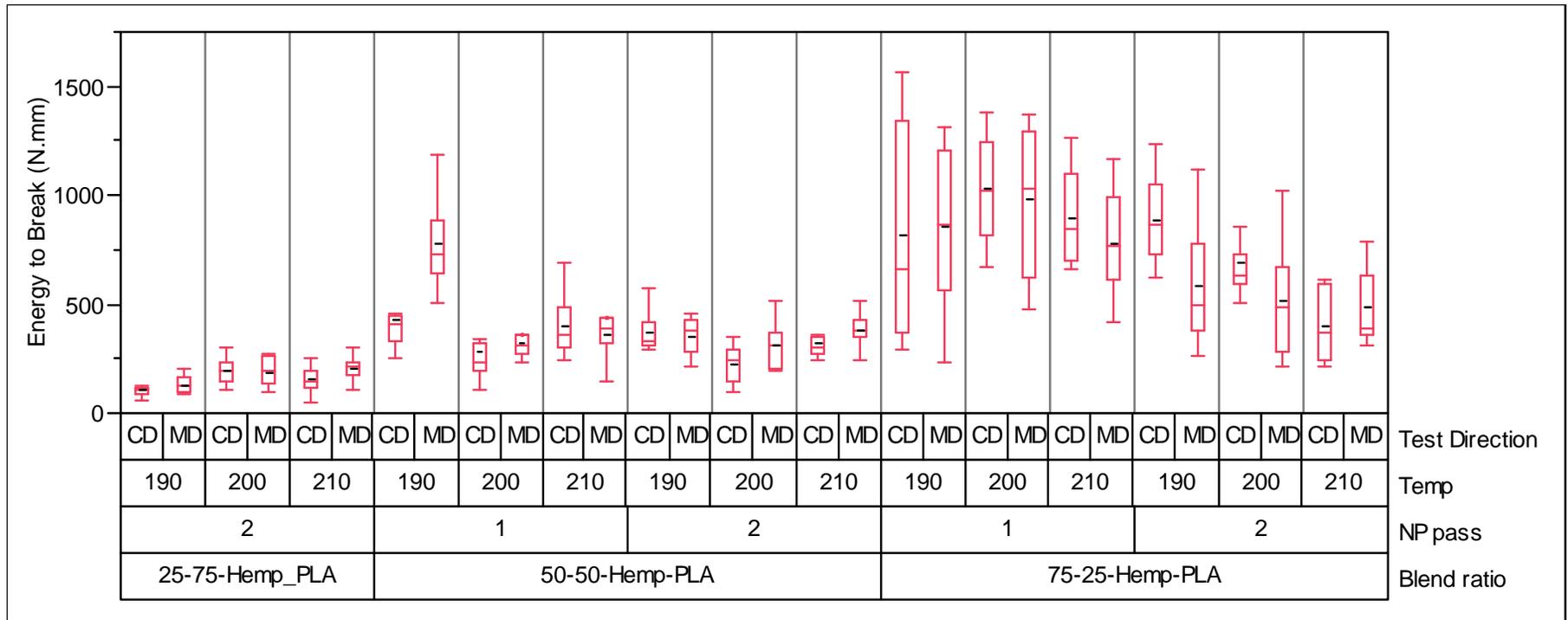


Figure 5.26 – Energy to break Hemp-PLA composites

Energy to break is calculated from area under the stress-strain curve. Thus it is not only influenced by how strong a composite is but also by how much is the strain before it breaks. The composites that were fabricated at lower temperature had high % strain that dominated the energy to break. The composites having 75% hemp fiber had higher % strain and moderate strength. Thus those composites had the highest energy to break. The composites having 75% PLA were strong, but since the Polylactic acid (PLA) melted and formed a matrix around the hemp fibers the composite failed at low extensions. This leads to lower energy used to break of the composite.

5.4 Hemp-Epoxy vinyl ester characterization:

5.4.1 Tensile properties:

Table 5.7 – Tensile properties of Hemp-Epoxy vinyl ester composites

	Peak load (N)		Peak Stress (MPa)		%Stn @ peak load		Energy to Break (N-mm)		Modulus (Mpa)		Thickness (mm)	
	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev	Mean	Std Dev
1P-1100g: MD*	3819.0	388.7	37.65	3.61	1.6	0.2	5139.49	1195.94	2939.76	185.29	3.933	0.125
1P-1100g: CD	3252.8	647.2	33.20	5.55	1.8	0.4	5191.82	1959.50	2508.33	375.68	3.835	0.165
2P-1135g: MD**	3892.9	147.8	38.44	2.48	1.6	0.1	5157.03	481.36	3084.65	223.04	3.995	0.164
2P-1135g: CD	2998.7	297.7	28.08	2.33	1.3	0.2	3289.83	765.24	2786.07	130.35	4.201	0.124
4P-1200g: MD	3518.6	426.5	34.68	4.06	2.1	0.1	6656.27	1225.10	2513.10	228.54	3.993	0.041
4P-1200g: CD	2539.5	281.8	24.44	1.84	1.8	0.3	4272.41	981.80	2222.95	92.19	4.083	0.193
1P-708g: MD	1809.3	249.9	27.07	4.05	1.1	0.2	169.44	485.91	2991.57	95.00	2.635	0.092
1P-708g: CD	2080.6	392.5	30.89	5.01	1.4	0.3	2531.22	979.83	2897.64	213.58	2.620	0.135
2P-634g: MD	1609.6	271.4	23.16	5.10	1.3	0.2	1701.18	572.86	2378.70	255.58	2.768	0.227
2P-634g: CD	1092.6	235.3	13.26	2.70	1.0	0.2	911.86	346.74	1628.04	190.25	3.243	0.226
4P-695g: MD	1718.5	597.4	23.60	8.59	1.2	0.3	1825.59	1133.37	2397.23	486.66	2.884	0.096
4P-695g: CD	964.4	261.9	12.10	3.08	0.8	0.2	648.91	366.86	1879.90	200.77	3.129	0.134

\* 1 out of 10 specimens exceeded load limit of the 1000 lb load cell

\*\* 3 out of 8 specimens exceeded load limit of the 1000 lb load cell

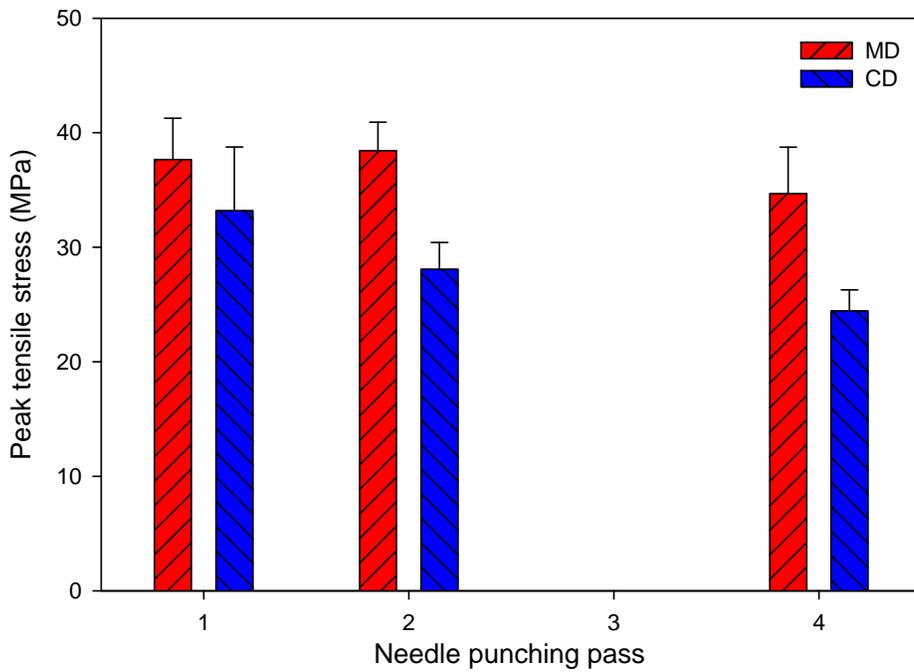


Figure 5.27 Peak tensile stress of Hemp-Vinyl ester composites Reinforced with high web weight

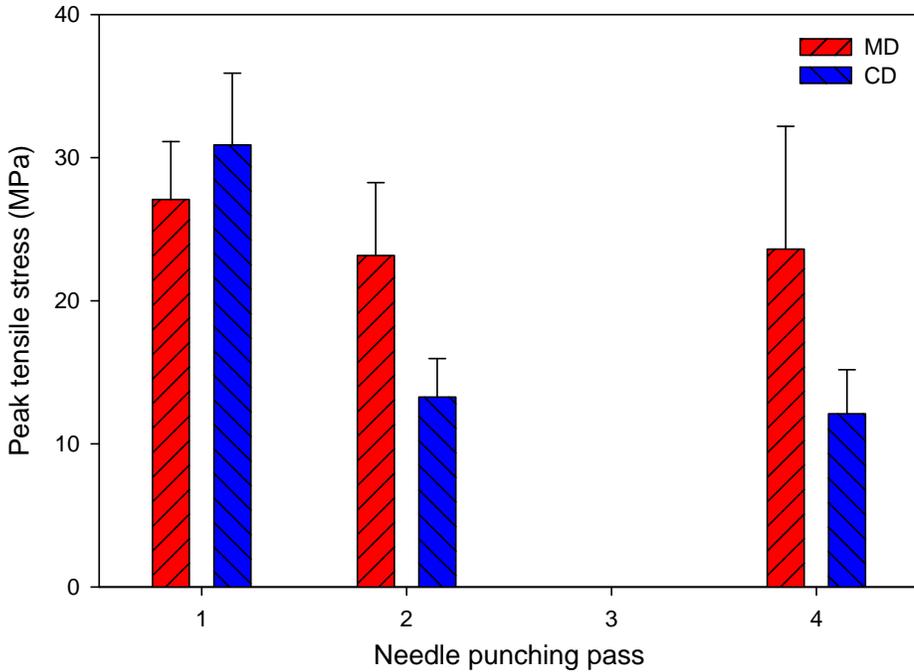


Figure 5.28 Peak tensile stress of Hemp-Vinyl ester composites Reinforced with low web weight

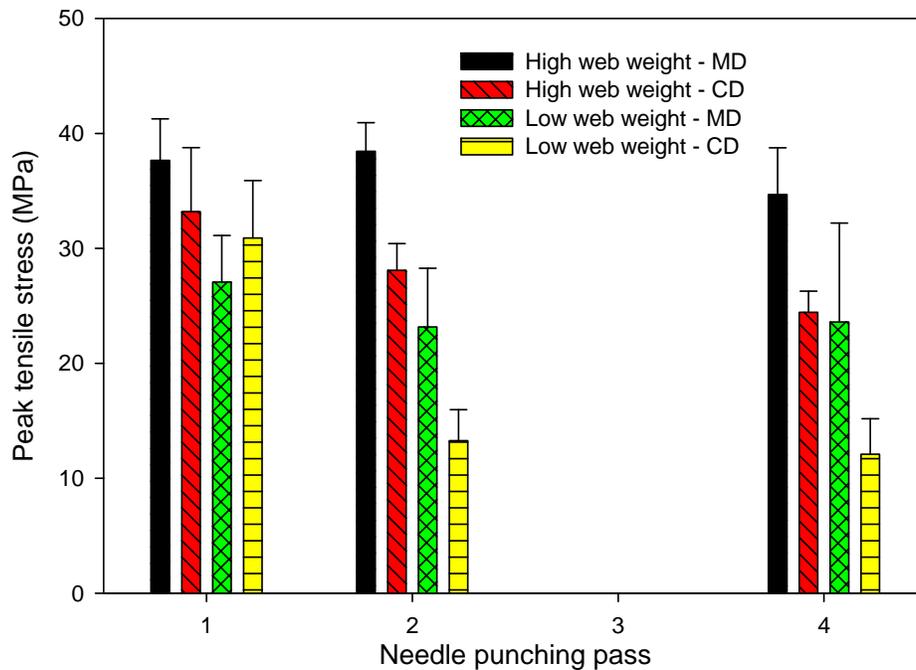


Figure 5.29 Peak tensile stress of Hemp-Vinyl ester composites  
Collated results

In the machine direction the tensile properties for the composites, within a weight group, are not significantly affected by number of passes through needle punch machine. However, as the web weight used for reinforcement increases the tensile strength of the composite increases.

The tensile properties of the hemp-vinyl ester composite in cross direction is affected by both passes through needle punching machine as well as the weight of the web.

With an increase in passes through the needle punch machine, the web becomes more compact, as verified by air permeability results, and becomes difficult to impregnate with resin. Also, with an increase in the needle punch density, the number of broken fibers in the web increases. The tensile properties of the web thus deteriorate with increasing needle punch density.

As the web weight increases, the number of fibers used for reinforcing the composite increases, increasing the load bearing element, thereby improving the tensile properties of the composite.

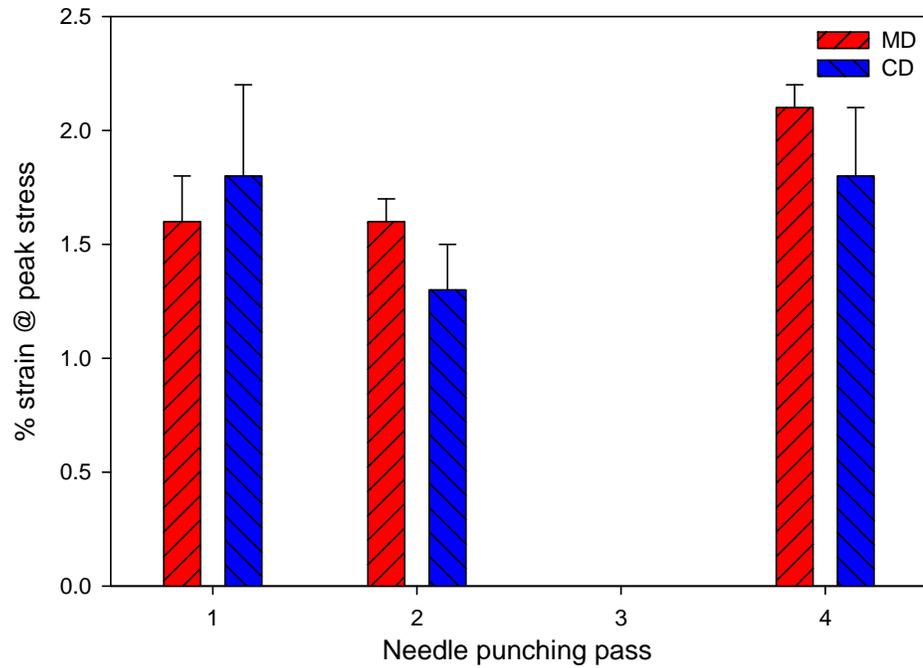


Figure 5.30 - % Strain @ Peak load of Hemp-Vinyl ester composites Reinforced with high web weight

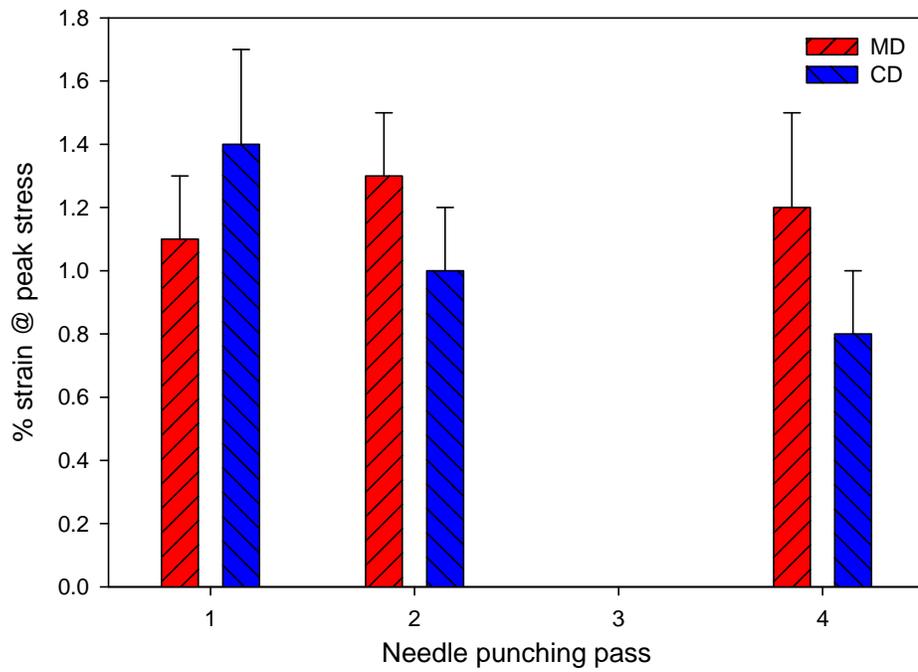


Figure 5.31 - % Strain @ Peak load of Hemp-Vinyl ester composites Reinforced with low web weight

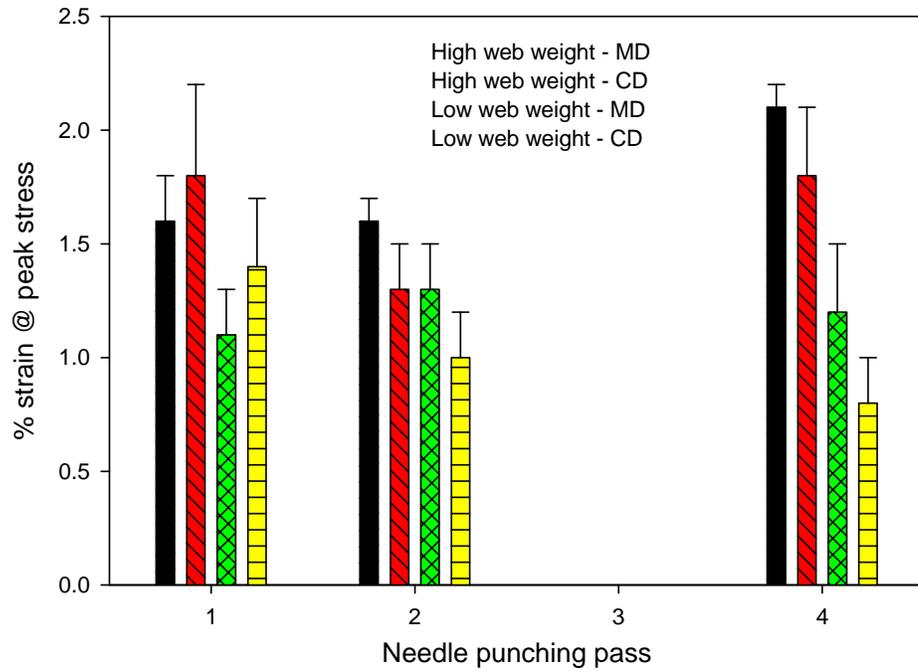


Figure 5.32 - % Strain @ Peak load of Hemp-Vinyl ester composites Collated results

In general the % strain at peak load is usually higher in cross-machine direction as compared to machine direction. Number of passes through the needle-punch machine does not significantly affect the % strain at peak load. As compared to hemp- poly(lactic acid) fiber (PLA) composite the % strain is lower for hemp-epoxy vinyl ester composite. For hemp-epoxy vinyl ester composite the failure is due to matrix failure and also due to vacuum assisted resin transfer molding the composites are compact and the fibers are well impregnated with resin reducing the % strain of composite.

The weight of the web affects the % strain at peak load. Composites reinforced with hemp fiber web of higher weight have more % strain at break as compared to the one's reinforced with lower web weight.

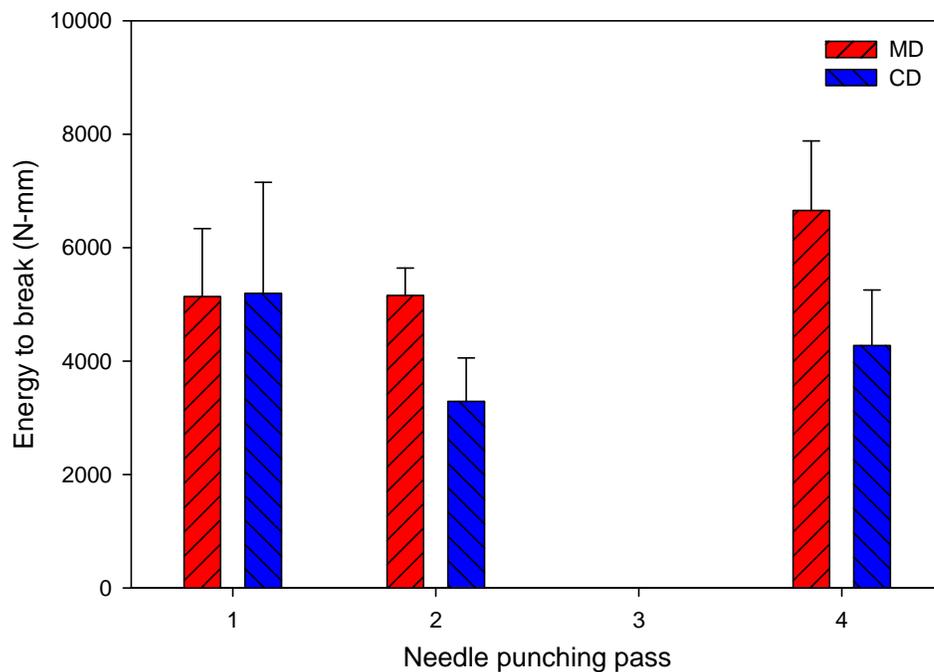


Figure 5.33 - Energy to break for Hemp-Vinyl ester composites Reinforced with high web weight

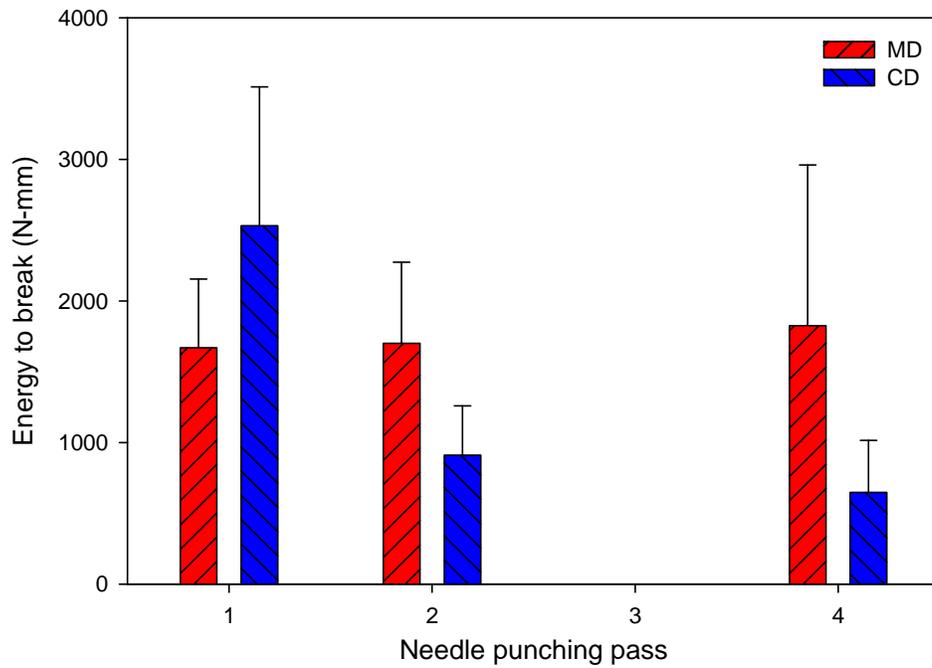


Figure 5.34 - Energy to break for Hemp-Vinyl ester composites Reinforced with low web weight

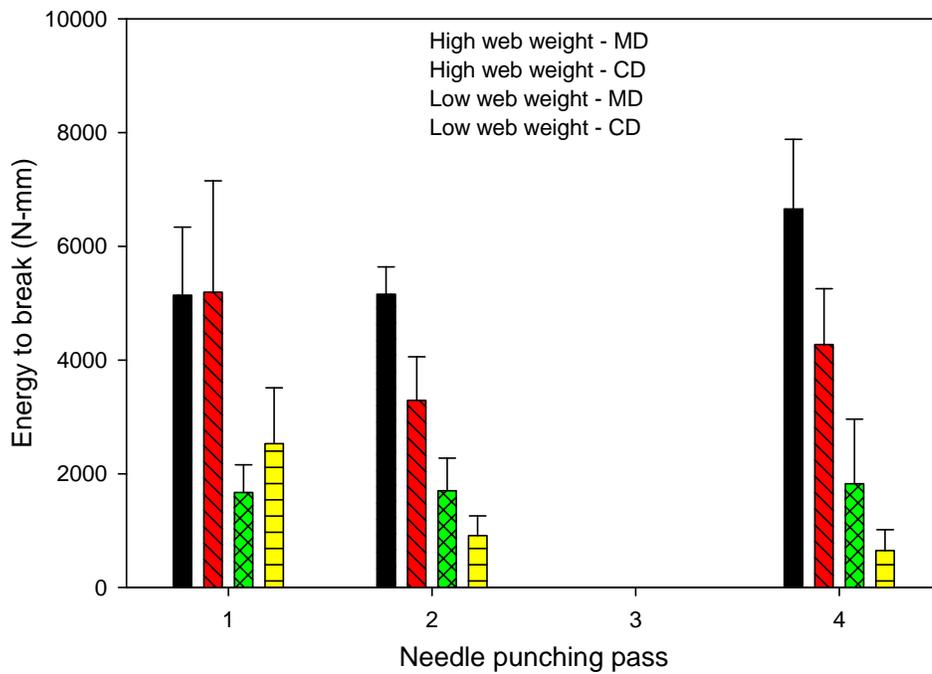


Figure 5.35 - Energy to break for Hemp-Vinyl ester composites Collated results

The energy to break of the hemp-epoxy vinyl ester composite is not significantly affected by the number of passes through needle-punching machine for both lower as well as higher web weight composites. This is as the stress as well as % strain to break, within the same web weight group, of the hemp-epoxy vinyl ester composites are not significantly affected by number of passes through needle-punching machine.

Web weight used for reinforcing epoxy vinyl ester composite affects the energy to break. The composites reinforced with higher web weights have more number of fibers reinforcing the composite and due to vacuum assisted resin transfer molding process the thickness of the composite is about the same. Thus composites reinforced with higher web weights provide better tensile properties.

5.4.2 Flexural properties:

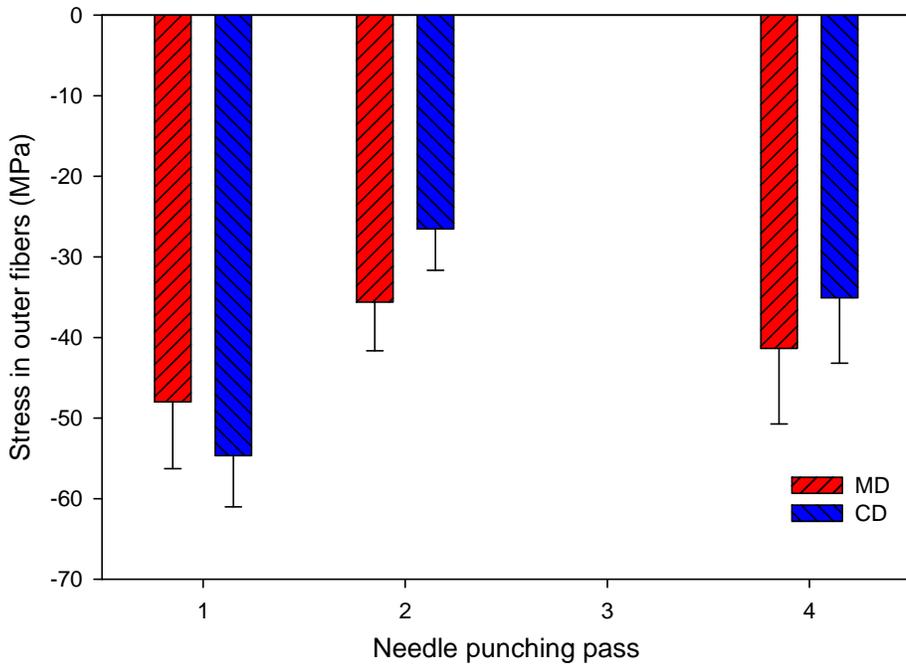


Figure 5.36 - Stress in outer fibers of Hemp-Epoxy vinyl ester composites Reinforced with high web weight

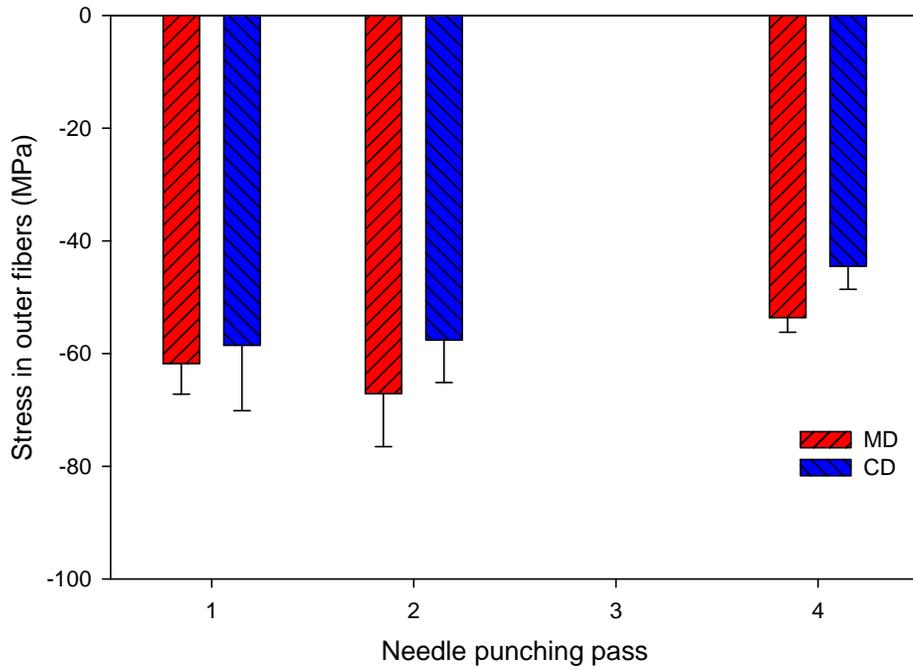


Figure 5.37 - Stress in outer fibers of Hemp-Epoxy vinyl ester composites Reinforced with low web weight

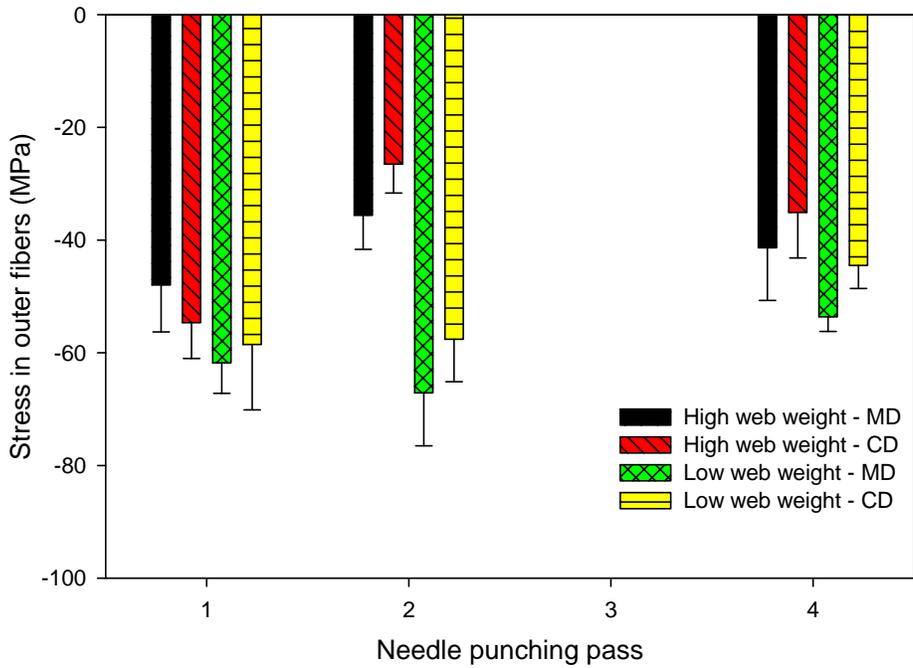


Figure 5.38 - Stress in outer fibers of Hemp-Epoxy vinyl ester composites Collated results

For flexural properties, the failure of the composite is dominated by properties of the fiber/matrix interface and unlike in tensile failure, is not due to fiber and/or matrix failure. The number of passes through needle punching machine, for composites reinforced with same web weight group, does not significantly affect the stress in outer fibers. This is as within a weight group almost the same number of fibers are present, thereby providing same area for fiber/matrix interface that is available for bearing the load. Thus the stress transferred to them from matrix is similar. However, as the web weight increases, the number of fibers and thus fiber surface area available for forming interface with matrix increases. Thus higher web weight group composites provides higher surface area for stress transfer that leads to composite being able to bear more load and thereby providing higher values for stress in outer fibers.

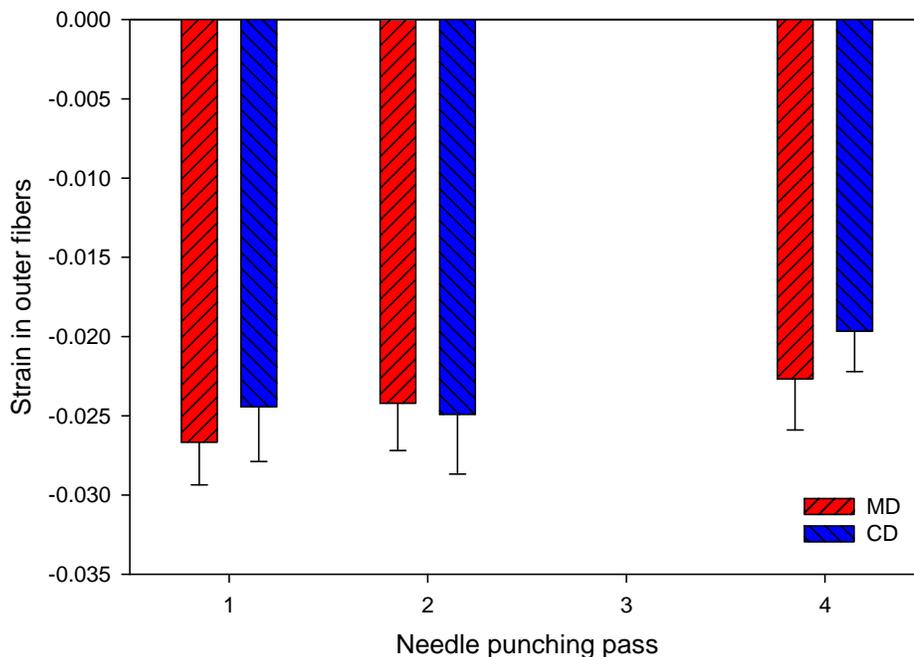


Figure 5.39 - Strain in outer fibers of Hemp-Epoxy vinyl ester composites Reinforced with high web weight

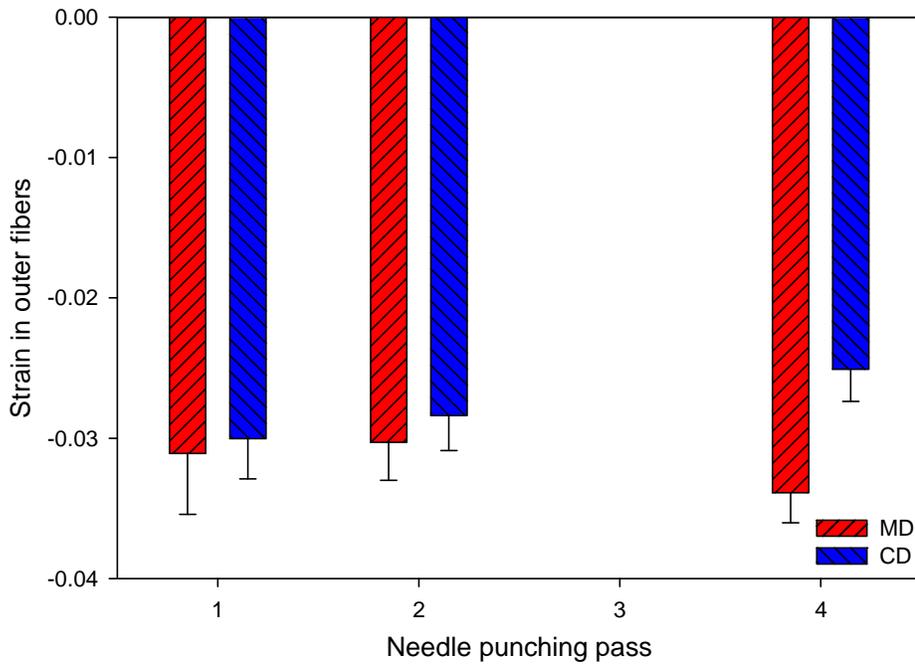


Figure 5.40 - Strain in outer fibers of Hemp-Epoxy vinyl ester composites Reinforced with low web weight

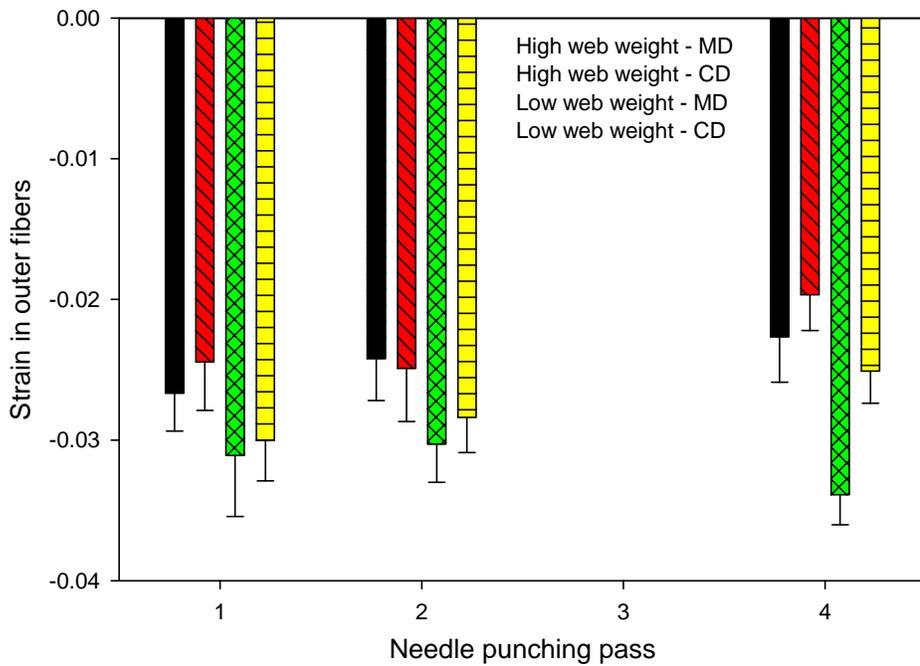


Figure 5.41 - Strain in outer fibers of Hemp-Epoxy vinyl ester composites Collated results

When testing flexural properties, Strain in outer fibers gradually decreases, however not significantly, as the number of passes through needle punch machine increases. Thus the process of making the web compact and provide more integrity does not seem to have significant effect on strain during testing flexural properties.

Weight of the web used for reinforcing affects the strain in outer fibers during flexural property evaluation. As the web weight used for reinforcing the epoxy vinyl ester resin is increased, the number of fibers and thereby available surface area for stress transfer increases. This leads to better distribution of load among more number of fibers allowing the composite to bear higher stress and in the process also allowing it to flex more than the lower weight web reinforced composites.

## *Chapter 6*

### CONCLUSIONS

Trials for fabricating the hemp – soy protein composites and then hemp – polylactic acid composites were a learning experience. Even though both the composites did not have properties as expected and did not compare anywhere close to the mechanical properties of hemp – epoxy vinyl ester composites. The experiments provided invaluable lessons that based on which the conclusions are drawn and discussed in this chapter.

Hemp is a bast fiber that has good mechanical properties and low % strain, whereas polylactic acid (PLA) has very high % strain for low stress values. Thus the main objective of this project was to fabricate composites, from hemp and polylactic acid (PLA) fiber, using nonwoven webs produced by needle punching Scanfeed batts; and compare their properties with the hemp – epoxy composites. Also, hemp fibers are flexible and provide ease of processing when compared to brittle fibers like carbon and glass.

The objective behind using thermoplastic PLA fibers as matrix was that the web could be fabricated in to a composite by hot pressing in to desired shape or form without requiring an extensive fabrication process. The PLA used in fiber form has higher molecular weight and melt viscosity, thereby the polymer when melted cannot flow easily in fine web pores to form a uniform matrix around the hemp fibers. Coupled with this the non-uniform nature of the nonwoven webs causes localized melting of the PLA fibers. These localized spots instead of acting as matrix around the fibers act as stress concentration points.

Utilization of 50% and 75% by volume fraction of hemp fibers for reinforcing the hemp-PLA composites does not provide improvement in composite properties. In these cases there is not enough polymer to flow around the hemp fibers and form uniform matrix.

Lower temperature (190°C) for hot pressing the hemp – PLA composites is not sufficient to completely melt the PLA polymer. This leads to improper and localized melting of PLA polymer forming composites that are similar in properties to the needle punched webs.

The composites fabricated from hemp – polylactic acid (PLA) fiber webs fare poorly for mechanical properties when compared to hemp – epoxy vinyl ester composites and cannot be used as load bearing composites.

Fabricating the hemp – PLA composites from needle punched webs is not the optimal process, for future work it is recommended to use PLA in polymer form or using different process methodology that ensure proper melting of the PLA polymer forming a uniform matrix around the reinforcing fibers.

Further conclusions drawn based on the results have been categorized based on the stages of processing of material, i.e. fiber, web and composites.

### *6.1 Fiber*

- As the fibers are cleaned and opened, although not significantly, fibers fineness and tenacity improves and variability reduces.

## 6.2 Hemp web

- Air permeability reduces as the web weight increases; but within a weight group there was no significant effect of number of passes through needle punch machine on air permeability.
- Tensile strength improved as the web weight increased.
- In lower web weight, tensile strength decreases as the number of passes through the needle punch machine increases.
- In higher web weight 2 passes through needle punch machine provides optimal tensile properties.
- % Strain at peak is not significantly affected by passes through needle punch machine or web weight. However, % strain is more in cross directions as compared to machine direction.
- Web weight as well as number of passes through needle punch machine changes flexural rigidity and bending modulus of hemp web. Web rigidity and bending modulus increases with increasing web weight and reducing the number of passes through needle punch machine.

## 6.3 Hemp- Polylactic acid fiber (PLA) web

- Air permeability decreases as the percentage of PLA fibers increase in the blend ratio.
- Within same web weight group as the number passes through needle-punch machine increases the air permeability increases.

- Number of passes through needle punch machine does not significantly affect the tensile strength
- Percentage of PLA fibers influence the tensile strength. As the proportion of PLA fibers increase, tensile strength increases.
- % Strain at peak stress is not significantly affected by number of passes through needle punch machine
- Blend ratio influences % strain at peak stress. Higher the proportion on PLA fibers, lower the % strain.
- With increase in the number of needle punch passes the webs became more compact and stiffer increasing its flexural rigidity and bending modulus.
- Blend ratio also influences the flexural properties. As the proportion of hemp fiber increases, web becomes stiffer and bending modulus increases.

#### *6.4 Hemp – Epoxy vinyl ester composite*

- For machine direction, tensile properties are influenced only by web weight used for reinforcing. As web weight increases the tensile strength improves.
- For cross direction, tensile properties are influenced both by number of passes through needle punch machine as well as web weight used for reinforcing. For same web weight group as the number of passes through needle punch machine increases tensile strength decreases. Whereas as the web weight increases the tensile strength increases.
- % Strain for the composites is not affected by number of passes through needle punch machine. However, as the web weight used for reinforcement

- Number of passes through needle punch machine, for same web weight, does not influence energy to break.
- Energy to break is however influenced by web weight used for reinforcing the composite.
- Stress and strain in outer fibers is not influenced by number of passes through needle punch machine for same web weight group.
- Stress and strain in outer fibers is influenced by web weight used for reinforcing the composite. Higher the web weight, better is the ability of the composite to bear flexural stress.

#### *6.5 Hemp – Polylactic acid fiber (PLA) composite*

- Tensile strength of composite improved as proportion of PLA fiber increased and at higher hot press temperature.
- Blend ratio does not influence % strain at high processing temperature. However, at lower hot press temperature, % Strain increased with increasing hemp fiber proportion.

## *Chapter 7*

### SUGGESTIONS FOR FUTURE WORK

This chapter provides suggestions for future work based on the results and conclusion drawn from the current project.

- The process used in the current project was very energy intensive and lead to brittle, non-uniform hemp – soy protein composites. Based on the learning experience from fabricating composites from hemp and soy protein isolate, it is suggested to utilize soy protein either in paste or other form that provides uniform matrix.
- Polylactic acid (PLA) when used in the fiber form, does not melt and form a uniform matrix around the hemp fibers. Thus it is recommended to use PLA as resin to achieve better mechanical properties for composites.
- The hemp – Polylactic acid (PLA) composites cannot be used as load bearing composites, however, they may be suitable for thermal, and acoustical insulation purpose. Thus a study should be conducted to characterize insulation properties of hemp – PLA composites.
- Study should be conducted for the biodegradability of the hemp – PLA composite along with the studying the effect of heat and humidity on the properties of the composite.
- Determine the feasibility of using the bast fibers to reinforce other resin for fabricating the composite that are biodegradable.

- If used for non-load bearing applications, composites may be treated with appropriate fire retardant finishes and then studied for the flammability of the hemp – Polylactic (PLA) composites.
- Form web using air lay or carding system, for reinforcing the composite keeping needle punch density and other composite fabricating parameters constant. Characterize and compare the properties of composite thus formed with the ones' fabricated by using scanfeed system.

## Chapter 8

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