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Processing and Evaluation of Cotton-based Composites for Automotive and Other Applications

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To the Graduate Council:

I am submitting herewith a thesis written by Manjeshwar G. Kamath entitled "Processing and Evaluation of Cotton-based Composites for Automotive and Other Applications." I have examined the final electronic copy of this thesis for form and content and recommend that it be accepted in partial fulfillment of the requirements for the degree of Master of Science, with a major in Polymer Engineering.

Gajanan S. Bhat, Major Professor

We have read this thesis and recommend its acceptance:

Larry C. Wadsworth, Roberto S. Benson

Accepted for the Council:
Carolyn R. Hodges

Vice Provost and Dean of the Graduate School

(Original signatures are on file with official student records.)

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Accepted for the Council:

Anne Mayhew

Vice Chancellor and
Dean of Graduate Studies

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Processing and Evaluation
of
Cotton-based Composites for Automotive and Other Applications

A Thesis
Presented for the
Master of Science
Degree

The University of Tennessee, Knoxville

Manjeshwar G. Kamath
December 2004

Dedication

This thesis is dedicated to my parents

Rama Kamath and **Meera Bai**

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Finally, I thank my spouse Lakshmi Kamath for her support and encouragement during my studies.

Abstract

Fiber reinforced composites (FRCs) have been used for a long time in structural and semi-structural applications. FRCs have the advantage of lightweight and best property performance compared to traditional materials such as metals. In several instances, disposability of such products becomes a major issue. There has been increasing demand for use of recyclable and or biodegradable composites for automotives, especially due to the recent European Union directives. With the growth of automobiles in the global market, and a simultaneous pressure to address the issue of sustainability, there is continual need for the incorporation of natural fiber based materials into automotives. The focus of this research has been to produce biodegradable cotton fiber-based composites that can be safely disposed off after their intended use without polluting the atmosphere, in an environmentally safe manner.

This research deals with cotton-based nonwovens using blends of cotton, flax, kenaf and a biodegradable thermoplastic fiber. Biomax®, Polylactic Acid (PLA), Polyvinyl Acetate (PVAc), and Eastar®bio-copolyester (PTAT) are the chosen thermoplastic fibers that could function as the binders, thus eliminating the use of any non-biodegradable synthetic fiber such as Polypropylene (PP) or a chemical binder. The process involves the fabrication of nonwovens from blends of fibers in different proportions made by air laying or carding to form webs, molding these webs into composites, and subsequent characterization of the composites for their properties such as tensile strength, flexural strength, and acoustic properties. Results from these studies addressing the structure and properties of the composites, contribution from individual constituents, with respect to their suitability for automotive applications are discussed.

Basic studies on structure and properties of fibers showed the ability of these natural fibers to form a good bond between thermoplastic polymer such as Eastar, Biomax, and Cellulose Acetate. Fiber bonding studies reinforced this observation. Comparison of Sandwich type composites with Fiber mix type composites showed that the bonding between natural fibers and the binder polymer is better when composites are made from mixed fiber webs. Furthermore, intimate blending is the key to make a composite with good properties.

Biodegradable composites were developed from air laid webs of natural fibers (cotton, flax, and kenaf) and binder fibers (Biomax, PLA, and PVAc) by thermal bonding in a hot press. It proved that blending of flax and kenaf increases the tensile strength of the cotton composites. Further, Three point bending test showed that PLA based cotton composites have slightly lower flexural strength compared to conventional PP. Adding about 10% kenaf or flax increases flexural strength substantially, indicating that kenaf and flax act like stiffeners. Acoustics properties of the composites measured by Four point Impedance Tube method showed that blending kenaf or flax increases noise absorption quality of cotton-PLA composites. Notched Izod impact tests showed that the impact strength of PLA and PLAbico binders is higher than that of PP. Moreover blending kenaf or flax increases the impact strength of the composites substantially. Impact strength increases as the composite thickness is raised keeping same basis weight.

Comparison of binders, Biomax, PLA, and PVAc fibers in a natural fiber composite showed that PVAc provides more tensile strength and elongation to the cotton or flax rich composite, where as PLA performs similarly in kenaf rich composites. Biomax performance is very close to that of PVAc. In other words, PVAc and Biomax form better composites with cotton and flax than PLA. If PVAc stands out for its superior performance in composites containing more cotton or flax, PLA stands out for the similar performance at lower curing temperature that reduces the bad odor in composites and has processing conditions close to conventional PP. The main advantage of Biomax is its lower cost compared to both PLA and PVAc.

Process optimization studies showed that there is an optimum bonding temperature and optimum-curing time for composites. Tensile strength increases as the curing pressure or basis weight/ thickness increases. Increase in tensile strength achieved by blending kenaf or flax (even at 10% level) is substantial. However there is marginal drop in elongation.

Further on the basis of these studies, it is expected that viable composite parts containing cotton and other natural fibers can be produced with a thermoplastic binder fiber, that are biodegradable and possess the required properties that are comparable to the traditional polypropylene based composites. Such composites are suitable for automotive and many other semi-structural applications.

Table of Contents

List of Tables

Table of Figures

I. INTRODUCTION

Evolution of Automotives:

The history and growth of transportation system shows an enlightening picture of how scientific, technological, social, and economic factors interplay to change the environment in this world [1]. Automotive growth is strongly related to the economic growth in the early part of this century [2]. Automobiles utilized diverse sources of energy like steam, electric, or internal combustion engine; developed from models of a cart or carriage; and moving parts such as wheels, sprockets, and chains from bicycle. By 1920, the four-wheeled vehicles with a steering wheel power from an internal combustion piston engine with spark plugs became popular. The wheels had tires, springs and brakes. Electric headlights allowed the driver to see at night. In 1925, the life of the car was about 25,000 miles, it grew to 40,000 by 1930, and today it is beyond 200,000 miles before it is salvaged or sent to the junkyard.

In the first quarter of 20th century, cars replaced personal conveying animals like horses and carts such as horse-drawn vehicles that were considered as the primary form of transportation. This is shown by the statistics of approximately three cars registered for every four households in 1925 indicating that one car was sufficient for one urban family. As women joined hand in earnings for the family, the growth in economy changed the need from one car per family to one car person of driving age today, thus a multi-car family became very common.

Automotives play an important role in the transportation of goods as well as people both individual and in groups. Manufacture of these vehicles, operation of the transport system, and construction of the infrastructure has impact on economical growth. The entire sector demands a substantial need for fabricated materials. Worldwide it is estimated that there are now more than 500 million passenger vehicles in use. At an average of 3000 lbs of materials for a passenger vehicle, for about 50 million new cars produced every year, the consumption of materials is in the tune of 70 million tons. In other words, this quantity has to be produced, used and ultimately be recycled or disposed.

Composites:

Composites are produced from two or more distinct materials to achieve the combination of their best properties. Generally the composites are made up of just two phases namely matrix and dispersed phases [3]. Matrix is the continuous phase that completely surrounds and holds the dispersed particles or fibers in place. If the particles constitute the dispersed phase, the product is called as particle- reinforced composite. These composites are mostly equiaxed as the particles behave the same in all directions. Improved strength is obtained by these tiny particles, which inhibit motion. The mechanism is due to the interactions on the atomic level. In a fiber-reinforced composite, the dispersed phase is made up of fibers, whose mechanical characteristics are enhanced by reinforcement action.

Generally, matrix is made of materials such as metal, ceramic, and polymer. The Polymer-matrix composites consist of matrix made up of polymer, reinforced with the fibers. Fibers possess a large length to diameter ratio and spatial orientation, which provides a potential for reinforcement efficiency. These fiber-reinforced polymer composites gained prominence as they substituted structural materials such as wood, metal due to their high strength, stiffness on a weight basis.

Properties of the fiber-reinforced composites are decided by the fiber diameter, length, orientation (parallel or random), surface roughness, level of consolidation, and level of adherence to the binders. The polymeric material of the matrix binds and holds the fibers together. When load is applied, it is transmitted and distributed to the fibers. Only the matrix sustains a small portion of the applied load, which is ductile in nature. Further, these matrix polymers protect the fibers from surface damage or environmental attacks and prevent crack propagation. Ease of fabrication, excellent properties and relatively lower cost make the composites very popular.

In addition, advanced composites have been produced from high performance carbon and aramid fibers. These are in use for special applications such as extra high strength, durability, and higher service temperatures. Glass fiber reinforced polymer composites are produced in large quantities and increasingly used in automotive, structural, and semi-structural applications. In automotives, they are used in an effort to decrease vehicle weight and gain fuel efficiencies.

More than forty automotive parts such as trunk liners, floor mats, package trays are made of nonwovens and their composites that contain synthetic fibers, which are not recyclable or biodegradable and pose difficulty in disposing at the end of their useful life. This has triggered a need for biodegradable composites. Enhanced biodegradability is achieved by replacing glass fibers with the cellulosic fibers such as cotton, kenaf, and flax. Such natural fiber reinforced composites are known as green composites [4]. Furthermore, these green composites are efficient sound absorbers and reduce noise in the automotives. However to achieve total biodegradability even binder should be made of biodegradable polymer.

Among fibers, cotton is a durable, breathable fiber with a soft feel and excellent absorbency. Products such as wipes made of bleached cotton webs bonded to the nonwoven fabric are attractive as they harvest these benefits of cotton [5]. In addition, there are advantages such as superior wet strength, ability to dry quickly, and biodegradability. It is possible to impart antimicrobial nature, flame retardancy, durability etc. by additional treatments. Small quantities of other natural fibers like kenaf, hemp, and jute are added to cotton to reinforce the product.

Recent innovation led to the composites consisting of two or more layers of different raw materials such as spunbond and or melt blown nonwovens [6]. Similarly composites produced by combination of technologies such as air laying, needle punching, spunlacing fibers followed by bonding to a nonwoven fabric. They are known as hybrid fleece. Extending the same principle there are composites made of fiber, film and paper. Eventually multi-layered multi-functional unique composites are entering the market serving the specific needs.

Newer or alternative materials are considered to achieve cost effectiveness, fuel efficiency, reduced emissions, increased safety, and always with a target on future ability to recycle or biodegrade. Fiber reinforced polymer (FRP) composite materials provide the automotive industry with a new range of performance materials. The major obstacle for the growth of polymer composites in automotive industry is the inexperience with these materials related to production rates, the joining techniques, response to automotive environments, crash tests, recycling or disposing methods and a small supplier base.

Lightweight construction, resulting in reductions in fuel consumption and emissions, has been a factor in the move to composites for all transport vehicles. The weight reduction has shown significant advantages in military and civilian applications. As an example, composite materials reduce aircraft empty weights and increase fuel efficiency that leads to smaller, lower-cost aircraft that use less fuel to perform a given task. For commercial airliners, this translates into simple economics, leading to increased sales. Where as for military aircrafts, composites reduce weight and thereby decrease life-cycle cost and fuel usage. Forty percent of the structural weight of the Fighter Jet 22 is expected to be from polymer composites, and we can expect that future transport aircrafts will also make higher use of composites.

Today's rising cost of fuel is a main factor in the move to utilize composites for all vehicles, and thereby achieve a lightweight construction with an ultimate goal of reduction in fuel consumption and emissions. About 25% reduction in the weight of the vehicle is equivalent to a savings of 250 million barrels of crude oil and carbon dioxide emissions of the tune of 220 billion pounds annually. Automobiles today have better fuel economy and safety features than it had 25 years ago. Still, there is enormous scope to improve. According to an estimate, that out of 10 gallons of fuel we pump to the car only 1.7 gallons goes in motion [1]. It would be more efficient if the vehicles were lighter. Future requirements must also look to reducing mass in the load carrying elements, with space frame structures receiving keen attention for volume production. Composites with oriented long fiber reinforcements and fiber volume content more than 50% are desired. Compared with glass fibers, thermoplastic fiber composites generate higher fiber volumes with a fast, clean processing behavior.

One way to achieve this is by replacing steel with composites without losing the performance and safety features. Composites are made of two or more materials are superior to their starting materials [7]. Generally, composites are strong, light and resistant to corrosion and wear. They have been used in space vehicles and military, and could play bigger role in automotives than they do today. Manufacturers accept this only if they are proven to be economical and perform better than the conventional materials. Auto manufacturers, federal agencies, and universities jointly formed a team with a target of this New Generation of Vehicles (NGV) to achieve 80 miles per gallon for a medium size sedan without sacrificing safety, performance, and affordability. To achieve cost reduction with good performance suitable blends of natural fibers and synthetic materials are attractive.

Fibers in Automotive Composites:

Durability of composites in automobiles is very important in its relation to the performance under conditions of wear and tear, small impacts, creep, fatigue, exposure to oils, food and cleaning agents, and variations in ambient temperature and humidity variations. Continuing pressure for better quality, assured performance at ever demanding cost reduction is the primary goal of a manufacturer of any automotive component. This encourages search for new materials and innovative approaches to satisfy the short term and long term needs. Polymers and composites are growing and demonstrating their potential to replace conventional materials. Aggressive strategy is necessary to bring them into the mainstream of applications in high volume automobile. The coming years will see dramatic changes in the level of their acceptance and the diversity of their use, as engineers gain control over the necessary design skills and wider adoption make them more cost effective. Moreover, increased importance of renewable resources for raw materials and recyclability or biodegradability of the product at the end of the useful life is demanding a shift from petroleum-based synthetics to agro based natural fibers in automotive applications. In addition, there exist technical advantages like strength, lightweight, and noise adsorption for natural fibers such as cotton, flax, kenaf, hemp, sisal etc.

It is a fact that automotive textiles are the growing markets in terms of quantity, quality and product variety [8,9,10]. On an average, each automobile utilizes fibers and non-woven based composites to the tune of 20 square meters. This trend is increasing due to the advantages of lightweight, high strength and day by day lowering cost of textile products. As many as 40 automotive components such as trunk and hood liners, floor mats, carpets and padding, speakers, package trays, door panels, and oil and air filters contain fabrics made of synthetic fibers [11,12,13]. Furthermore, these fiber-based composites can contribute greatly to the automotive manufacturer's final goal constituting weight reduction of 30% and cost reduction of 20% [14].

Increased social awareness of environmental problems posed by the non-degradable, non-recyclable contents of the salvaged automobiles is forcing automotive manufacturers to enhance the biodegradable content which is in favor of switching to natural fibers. If biodegradable fibers were chosen to substitute many of the existing composites, the finished products do not pose difficulty in disposing [15,16]. To accelerate this process of switching to recyclable and biodegradable constituents, legislation in US and Europe have issued specific directives on the end-of-life of vehicles [17] that promotes the use of environmentally safe products and reduces the landfills. The directive, which came into effect at the turn of this century, predetermines the deposition fraction of a vehicle to 15% for the year 2005, and then gradually reduced to 5% for the year 2015 [18].

Over the last century, increased use of fiber-reinforced composites (FRC) for various structural and semi-structural applications has lead to the development of varieties of synthetic fibers for such purposes. Nonwoven webs are one of such products popularly used in making composites for many applications since they possess a good blend of strength, lightweight, and flexibility compared to conventional materials

[19,20,21,22]. Figure 1 shows generalized tensile properties of composites when they are produced from natural fibers and synthetic binders.

Natural fibers are a good substitute [23] for reinforcing parts having large area and complex geometry such as door trims. Moreover, these composites meet crash safety requirements and favorable crash behavior of no sharp edges at the rupture point. These composites have an important inherent quality that provides excellent z-directional properties that minimizes delamination problem.

Formation of composites using flax fibers and biodegradable melt blown polymers as main components have been studied by Mueller and Krobjilowski [24,25,26,27]. These natural fiber based composites made of biodegradable melt blown fabrics as binder, possess many of the required properties that are comparable to the traditional polypropylene based composites.

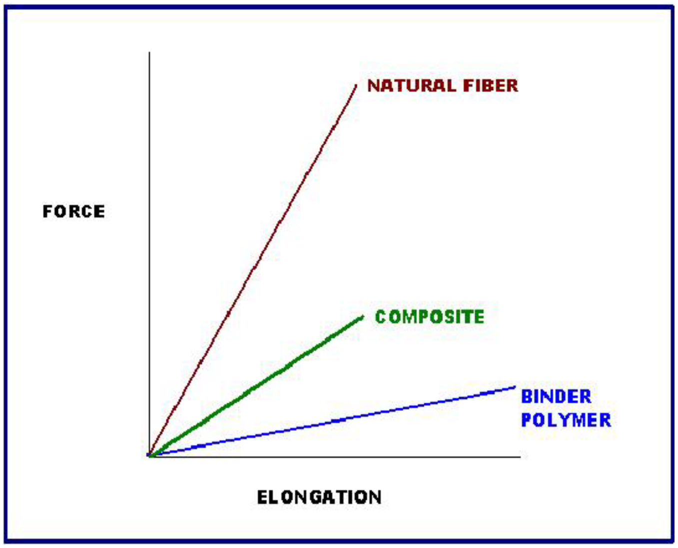

Figure 1 Composite Tensile Properties.

Further, flax fiber based composites are generally stronger, but somewhat brittle, due to the inherent nature of the fiber. Incorporation of cotton is likely to increase the impact resistance of these structures that will make such composites suitable for many more applications. It is evident from Mueller's research and from our studies, that biodegradable nonwovens can be produced from cotton and a biodegradable binder fiber. Extending this study by suitably combining cotton with a carefully selected biodegradable binder fiber in the right proportion, a nonwoven fabric can be produced, which in turn can be molded into required shape.

Natural Fibers:

First fibers known to mankind are the natural fibers that are found in nature or produced from naturally available materials from plant and animal sources. Various types of natural fibers are shown in Figure 2. Majority of the natural fibers are plant based cellulosic fibers.

Asbestos is an inorganic natural fiber, which was used as insulation material. Due to its carcinogenic nature, it is no more used without encapsulation. Among organic fibers, wool and silk are made of protein and are of animal origin. Wool is popularly used in warm clothing, whereas silk is used in expensive garments and gained popularity for its shine and producing bright colors on dyeing.

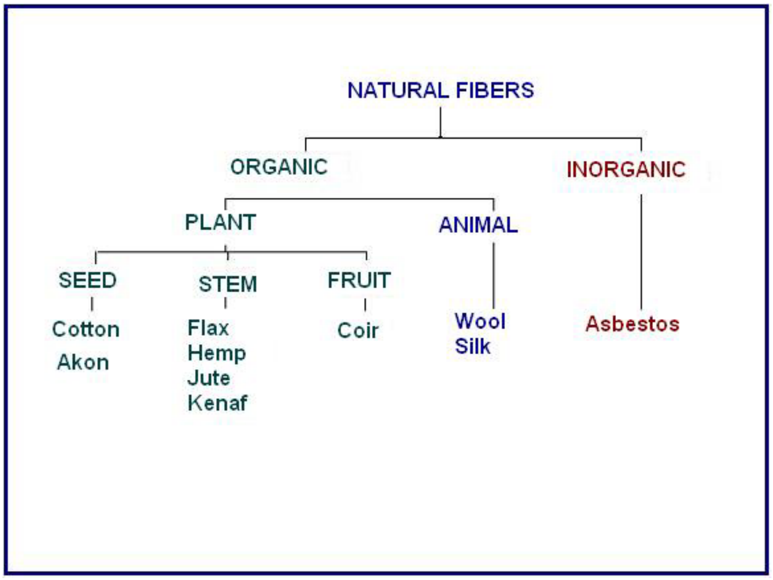

Figure 2 Types of Natural Fibers.

Cotton is a natural cellulosic fiber, well known for its excellent absorbency, comfort properties, and natural feel. In addition, biodegradable nature of cotton is an important quality that makes it an attractive and strong candidate in a situation, where waste disposal is becoming a major concern. Cotton is the seed fiber of the plant, *Gossypium hirsutum*. Flax with properties similar to that of cotton, but with better strength and modulus, is an alternative to cotton. Flax is a bast fiber from the plant, *Linum usitatisimum* [28] that grows 12 to 40 inches tall in temperate climate. Flax is known to have existed 10,000 years ago.

Kenaf is another bast fiber from a cane like plant, *Hibiscus cannabinna*, which grows 12 to 15 feet just in seven months. Compared to cotton and flax, kenaf entered the textile fiber market very recently. Initial interest in kenaf in the United States was as a domestic supply of cordage fiber as a jute substitute in the manufacture of rope, carpet backing, and burlap. Later, kenaf was identified as a very promising fiber source as a substitute for fiberglass and synthetic fiber. Today, kenaf is considered as a commercial crop in USA. Therefore, government encourages the commercial use of the kenaf.

The research has shown that when properly retted and treated kenaf after blending with cotton fibers can be used to produce yarns and fabrics. Besides vehicle weight reduction, ability of natural fiber based automotive composites to reduce noise level inside the vehicle is an important factor as far as the passenger is concerned and this will open an opportunity for a lot of new applications for such products. Automotive acoustics involve reduction of noise, vibration, and harshness. Considering this, custom-made products are developed since 1960 [29].

Binder Fibers/Polymers and Biodegradability:

In order to produce nonwovens from natural fibers, it is generally blended with a synthetic binder fiber or a polymeric chemical binder. At present, the most common synthetic binder fibers are polypropylene fibers and bicomponent fibers containing low melting polyethylene sheath and PET core. These binder fibers are not biodegradable, thus pose difficulty in disposal. Hence, biodegradable polymers and fibers are gaining importance. Commonly used binder fibers are shown in Figure 3. Many of the biodegradable synthetic fiber forming polymers are still at the developmental stage and very few have reached commercial production stage. Out of these, a few are easily available in the market.

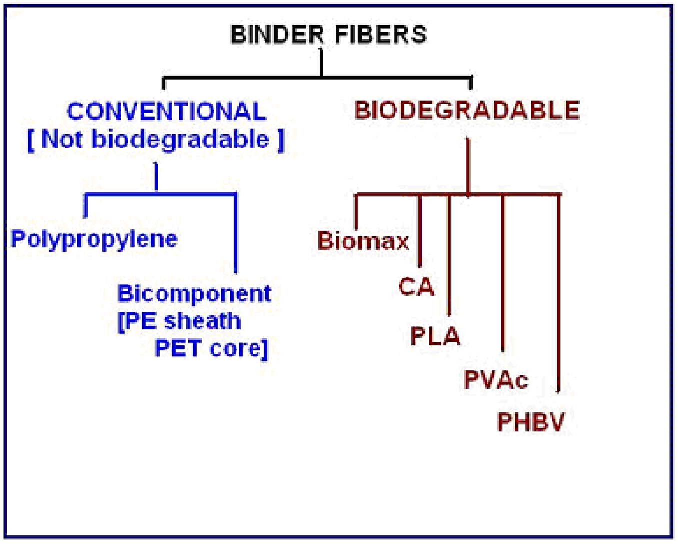

Figure 3 Types of Binder Fibers.

Cellulose acetate (CA) is a well known modified cellulosic fiber for its properties such as biodegradability, wettability, and liquid transport. In addition, it is made from cheaper renewable sources such as wood pulp or cotton linters. The thermoplastic nature of CA makes it a suitable binder fiber that can undergo thermal calendaring while producing nonwovens out of blends containing cotton and CA. These blends can produce good quality nonwovens and they are compostable at the end of their useful life [30]. It is also observed that the plasticized CA fibers have good thermal bonding ability and could achieve acceptable tensile properties.

Other promising candidates for thermoplastic and biodegradable binder fibers are the recently developed materials in the markets such as Eastar from Eastman Chemical Company [31], Ecoflex from BASF, PLA from Dow-Cargill [32], PHBV from Metabolix and Biomax from Dupont [33]. Generally, these contain chemicals that are tasty to the degrading organisms so that the final product of degradation is safe. Eastar, a biodegradable thermoplastic fiber produced by Eastman Chemical Company, is co-polyester with melting point of 120°C. It has been demonstrated that Eastar is easily bondable with cellulosics and it is completely biodegradable. Initial experimental studies in the laboratory showed promising results for using this biodegradable copolyester as a binder fiber [34].

Polylactic acid (PLA) is another biodegradable fiber that is produced from the cornstarch. PLA fiber has a melting temperature of 175°C and tensile properties comparable to that of polyester fibers. Similarly, Biomax is a hydro/biodegradable polyester, presently used in packaging such as sandwich wraps, has a melting point 200°C and it could be tried for bonding with cotton. As a melt spinnable fiber with a vegetable source, PLA has many of the advantages of both synthetic and natural fibers [35]. Beyond having a renewable raw material, it possesses biodegradability. However, it has poor abrasion resistance and bonding behavior compared with conventional binders. However, PLA is not yet made available in commercial quantities in the open market.

Recently P&G has published its ability to produce a biodegradable thermoplastic polymer and its fiber products with the trade name Nodax [36], which can be used as a binder. One more type of biodegradable binder fibers, Poly vinyl acetate (PVAc) are recently available for our research.

Considering all these, the current research on developing cotton based biodegradable composites for automobile applications, appears quite encouraging, promising and challenging. Since the use of thermal bonding technique does not involve any chemicals that pollute the surroundings, the process and the products of this research are totally safe to the environment.

A portion of the useful land is dedicated for the purpose of landfill. One of the major loads to the landfill is the trash produced by the population, which is of the tune of 7000 lb per year per family. By recycling, a preferred way, this burden can be reduced to a certain extent. Furthermore, if the remaining portion is biodegradable there is no need for a landfill. One way to achieve this is by making use of biodegradable polymers to make plastic cups, forks, spoons, snack bags and gum wrappers that are used in everyday life. DuPont scientists have researched and created a type of polymer that decomposes in compost and support plant life in the soil or do not harm the environment. They have overcome the cost and performance barriers to have a breakthrough in consumer applications.

Polyester, polyethylene terephthalate (PET), is a well-known economically and commercially available product. Biomax® is a modified hydro/biodegradable polyester. Proprietary monomers are incorporated into the polymer, creating sites that are susceptible to hydrolysis. At elevated temperatures, the large polymer molecules are cleaved by moisture into smaller molecules, which are then consumed by naturally occurring microbes and converted to carbon dioxide, water and biomass.

Biomax® has been designed such that it can be recycled, incinerated or sent to landfill for composting. Several tests have shown that it is friendly to the environment, promotes growth of plants, earthworms and microbes in the composting soil. This biodegradable polymer is readily available at present. Biomax® polymer can be used to make injection-molded parts, coatings for paper, thermoformed cups and trays, and films. With its diverse product properties, it is suitable for film applications, thermoformed packaging and injection-molded parts. In addition, it is versatile, very good for a variety of single-use products such as disposable biodegradable plates, bowls and sandwich wraps.

According to Dr. Otto Angleitner (DOA), a leader in the air lay market [37], new products are being developed that can be made blends of all natural and manmade fibers, including fiberglass, wood, coconut and even straw. These fiber webs are possible at both high and low basis weights in uniformly blended composites or as layered webs. Products can be used as molded, needled, insulation, automotive, high loft, geo-textiles, apparel, furnishings, mattresses, carpets, carpet fiber pads, fiber glass mats, filtration, etc.

Bonding methods that involve heated rolls or plates can be used for compression molding or bonding. Other bonding methods involve chemicals bonding or combination of both thermal and chemical methods. Generally, air doctors are used to obtain lightweights and can be configured for a desired pattern and moderate density. The thermal process bonds the fibers through heat energy. It is possible to use the thermoplastic synthetic fibers as the binder material in the natural fiber matrix.

Thermal bonding is an economical and environmentally safe technology, which also enhances the product performance. In this process, heat softens the surface of the fiber, and fibers in contact with each other will form strong bonds holding the fabric together. On cooling, the bonding points solidify and ensure sufficient product strengths. In thermal calendaring, the heat and pressure applied to weld the fiber webs together.

When it is done in a through-air oven bonding, the products are bulkier and overall bonding of the web is by the low melting fibers in it. Ultra-sonic bonding is one more method, not so common, where the molecules of the fibers held under an embossed roller are excited by high frequency energy generating internal heating, softening and bonding of the fibers.

The polymer composite system [38] consisting of cellulose acetate butyrate and lyocell, a high modulus, continuous, regenerated cellulose fiber exhibited good interfacial adhesion between the fiber and matrix in the composite materials. Interfacial adhesion was found to be substantial due to the relatively less fiber pullout after tensile failure in the unmodified fiber composites. This concept supports cotton, a similar fiber with suitable properties to form composite.

BioCycle [39] surveys resin producers, bag manufacturers, certifiers and composting agencies to provide a summary of biodegradable and degradable plastics. Symphony Environmental, a British company is producing fully degradable bin bags, carrier bags and other plastic bags from polyethylene, using new additive technology to reduce the plastic to carbon dioxide and water in just a few weeks. This allows one to feed the compost heap with the plastic bags too. Symphony's material is the first example of 100% degradable polyethylene.

The plastic, SPITEK, has mechanical properties and processing characteristics similar to polyethylene and goes for similar applications. It contains a patented additive at the level of 3% that acts as a catalyst for the degradation of polyethylene and jumpstarts the reaction that leads to degradation.

According to the supplier the SPITEK, material degrades in about 60 days if conditions are quite favorable, otherwise it might be as long as 6 years. The level of the proprietary additive determines the rate of degradation as well as the shelf life of the products. Factors favoring degradation are sunlight, heat, tear that initiates the process, which then continues even if the material is in landfill or under water.

The need for a fully degradable plastic is pressing since billions of pounds of plastic waste, including bags, boxes and containers are buried in landfill sites. China produces about 30 billion pounds, India 10 billion pounds and the UK 2 billion pounds, out of which nearly 2 billion pounds is waste polyethylene. Other disposal routes are possible for these materials, such as recycling and incineration. As the waste plastic is mixed up with other non-plastic materials and separation is expensive. Conventional polyethylene products can take longer than 100 years to degrade. This takes up valuable landfill space and prevents the composting of biodegradable materials packed with in it. Use of degradable bags would increase the capacity of landfill sites by about 25% speeding up composting.

Bayer's product BAK, a polyester amide biodegradable plastic that it claims is 100% biodegradable and recyclable, and has desired properties such as high tensile strength. The plastic got green credentials, as it does not have solvents, chlorine or any aromatics. BAK is a semi-crystalline, transparent thermoplastic material that breaks down into carbon dioxide, water and biomass in favorable conditions. Its degradation rate is comparable to that of other organic materials. The polymer is suitable for film blowing, extrusion, thermoforming, spinning, injection molding, blow molding, hot sealing and welding. The potential applications considered were in the field of horticulture, agriculture, and food wherein plastics is used in conjunction with compostable waste. Unfortunately, Bayer discontinued BAK production since 1996, as it is not economical.

Clearly there is a long way to go before these new degradable materials grow to an alarming level and make a significant impact in the landfill space available. This promotes use of degradable polymers to replace conventional polymers in traditional applications. Thus, consumers can expect to see more products with labels ‘full degradability or biodegradable’ in the future. Biomax is one such polymer picking up in the market since several tests were carried out by Dupont to prove that it is biodegradable, friendly to the environment, promotes growth of plants, and earthworms and microbes find it tasty in the composting soil.

Cost Factor:

The main delay for implementation of biodegradable materials is the cost factor. If a conventional trash bag costs less than 10cents, degradable bag costs 30 to 40cents and fully biodegradable one costs 80cents! So far, it is found difficult to prove to the community the worth of paying 8times more money on disposable that makes trash eco-friendly. Similarly, if common synthetic resin price is 50 cents per pound or film price is 100 cents per pound, it costs 500 cents per pound of biodegradable ethylene-vinyl alcohol copolymer film (EVOH). Metabolix has produced PHBV for the first time in a small-scale fermentation plant. It is looking for a joint venture to cut the current high costs of developmental work.

In order to reduce the cost and sell the product at a price nearly equal to conventional cup, Earth-shell makes biodegradable disposable cups out of starch, limestone, and a thin coating of Biomax biodegradable film. Similarly, using a blend of natural fibers and synthetic biodegradable fibers can reduce the cost of composites. The goal is to make composites versatile in nature could go for automobiles as well as similar other applications where the existing routes or alternate routes are not attractive due to high cost. If it is confirmed that all the materials involved in the manufacturing process are biodegradable in nature, there is no doubt about the biodegradability of the product composite.

II. EXPERIMENTS

Research was carried out by acquiring the samples of natural fibers and binder polymers from available sources within the institution as well as from the industries and organizations that can contribute to, and take advantage of this project. Experiments were carried out to produce composites using natural fibers, with cotton as the major component fiber and various binder polymers as matrix. Natural fibers such as kenaf and flax were also used along with cotton in the composites to derive certain advantages.

Properties of Fibers and Binders:

The properties of the natural fibers and binder materials that were used in this research were studied. Properties include fiber diameter, denier, density, fiber length, tenacity, elongation and melting point for binder fibers. Diameter was measured using the optical microscopic pictures. Denier was calculated from the weight of the fiber. Density gradient column was used to determine the density. Melting temperature was obtained from the DSC scan. Since fiber bundle behavior is important in composites, tensile test was carried out for a bundle of fibers containing 20filaments. Morphology of the fibers was studied using microscopic pictures of the surface and cross-section. Natural fibers investigated include cotton, kenaf and flax. The binder materials investigated include, Eastar and Eastar/PP bicomponent fiber provided by Eastman Chemical Company, biodegradable polyester and a bicomponent fiber from Dupont, biodegradable polyethylene film from Maverick, and cellulose acetate (CA) from Celanese Acetate. In the later stages of the research concentrated upon presently available type of binders namely: PLA, PVAc, and Biomax fibers. To get an idea about the melting behavior of various binder fibers, differential scanning calorimetric (DSC) scans were obtained using the Mettler 821 DSC system.

Fiber Bonding:

A preliminary study on thermal bonding behavior of binder fibers was studied at Sunoco Chemicals Fiber Laboratory at Pittsburgh using a Dynasco Hot Track Heat Sealer Model HTH-2. With a small quantity of fiber samples (50filaments), preliminary studies on fiber-to-fiber bonding were conducted with the precise control on process conditions such as hot plate temperature, contact time and pressure. Various fibers studied were Raw & bleached cotton, EastarPP bico, Biomax, PVAc and PLA.

Samples were tested for fiber denier using Vibromat M and tensile properties using Textechno Fategraph M Tester using 13mm(~half inch) gage length, 100 cN load cell and extension rate of 25 mm/min (1 inch/min). Bonding study was carried out using EastarPP binder fiber with cotton fibers.

Composite Sample Preparation:

For producing composite panels, a hydraulic hot press (Wabash) with capability to control heating of both upper and lower plates and applying pressure on the composite was used for all experiments. Initially composites were made from available fibers, film and melt blown webs using sandwich type construction as shown in Figure 4.

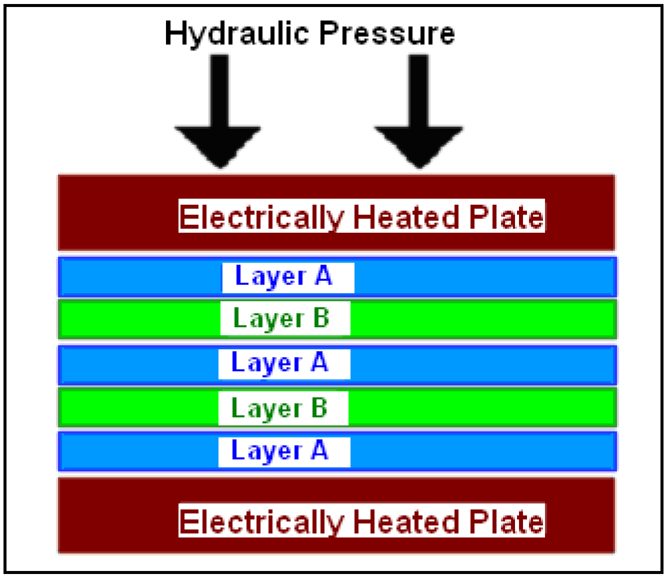

Figure 4 Schematic for the Preparation of Sandwich Type Composite.

Sandwich Type Composites:

In these sandwich type composites, the core consists of natural fiber (Layer B) and surfaces consist of binder polymers (Layer A) as shown in Figure 4. This is a simple type of construction involving fewer steps in preparation. When the sandwich is cured under pressure in the hot press, binder is expected to percolate into the natural fiber web and provide bonding with the surrounding fibers. Curing temperature was set based on the melting temperature range of the binders as observed from the DSC results.

Fiber Mix Composites:

Fiber Mix Type composite shown in Figure 5 is another type of construction consisting of fiber blends. A uniform blend of natural fibers and binder fibers was used to produce fiber mix composites wherein the composition is expected to be uniform throughout the product. When this mixed fiber mat is heated under pressure in the hot press, bonding takes place between binder fibers and the natural fibers. The design of experiments consisted of production of the composite samples under various process parameters like curing time, temp, and pressure for various compositions. Bonding conditions were set based on the melting temperature range as observed from the DSC results.

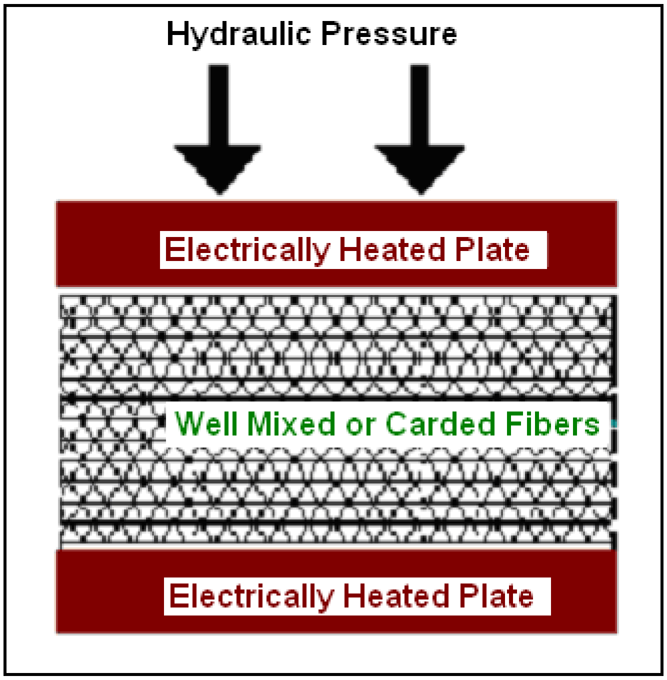

Figure 5 Schematic for the Preparation of Fiber Mix Type Composite.

Preparation of Fiber Mixture:

Fibers were well mixed by hand and a dry laid using air jets, or carded, where possible, to produce webs/fleece before making composites. Initially a few samples of composites were made to establish the procedure. Table 1 shows the various sets of mixed fibers produced. A Hollingsworth card (at Star Lab Inc. Knoxville) was used to make 12 inches wide webs from small amount (200 g) of fiber samples.

These mixed fiber webs were subjected to thermal bonding in the hot press at a temperature of about 20°C more than the melting point of the binder under 1 bar pressure and for 5 minutes duration. Both raw cotton and bleached cotton were tried in this set of trials. Later sets of experiments were conducted specifically with the raw cotton to keep the cost low and the various biodegradable binder fibers such as Biomax, PLA, and PVAc.

Table 1 Details of Fiber Mixture.

Sample No.	Binder	Cotton	Kenaf	Flax
1	50	50		
2	50	25	25	
3	50	25		25
4	50		50	
5	50			50

Initially Biomax was chosen as a biodegradable binder since it was available in commercial quantities. Since Biomax is a derivative of fiber grade polyester, an attempt has been made to produce biodegradable binder fiber starting from Biomax polymer supplied by Dupont at Foss Manufacturing Company, Hampton, NH. Later, in-house spinning facility was used to make fiber from polymer by melt spinning. Similarly, PLA fiber was also produced and used in the research. (Details of melt spinning of Biomax and PLA are given in Appendix 1) The fiber thus produced was used as binder fiber in making biodegradable nonwovens or composites. Cotton, flax and kenaf fibers were mixed with this Biomax binder fiber by air dispersion (dry laid) as per the experimental plan given in Table 1.

The composite samples produced in the experiments were analyzed for physical properties and structure under standard laboratory conditions [40]. The physical properties include weight and thickness, using TMI thickness tester. Tensile properties were determined using the United Tensile Tester. One inch wide, five inch long samples were clamped and stretched to break at uniform rate of elongation. The structural evaluation consists of tensile characteristics, failure mechanism and morphology. Scanning Electron Micrographs of the samples and the fractured samples provide additional information to understand the structure and correlate the properties. These photographs were obtained using the Hitachi and Leo 1525 surface scanning electron microscope in back scatter with Gemini column with system vacuum of ~$1.3\text{x}10^{-5}$ torr and at an acceleration voltage ~5 kV. SPI Module sputter coater was used to coat the samples (where necessary) with gold for 5 seconds at 20 mA plasma current to reduce charging while scanning.

Dry Laid Fiber Mix Composites:

More experiments were carried out in the Hot Press using natural fibers such as raw cotton, kenaf, and flax and binder finders. Binder fibers used are Biodegradable Eastar, semi-biodegradable Eastar-Polypropylene bicomponent (70:30) fiber, and non-biodegradable Dupont's PE-PET bicomponent fiber (50:50). Keeping the binder content 50%, the mixture of cotton, flax, and kenaf in the desired ratio prepared in the laboratory by hand mixing followed by air laying. This mixed web was subjected to thermal bonding at hot plate temperatures of 20°C above the melting point of the binder, 1 bar pressure and for 5 minutes duration. The purpose is to observe contribution of kenaf and flax fibers in the various steps involved in the cotton based composite processing such as uniformity of blending, the formation of composite web, its properties, thermal bonding behavior, and the mechanical properties of the finished product.

In the next stage of research, experiments were specifically confined to the biodegradable binder fibers like Biomax, PLA, and PVAc. Eastar, PCA, and Eastarbico are no more manufactured due to economic reasons. Among natural fibers cotton, kenaf, and flax were used in the experiments. Thus, the product is fully biodegradable. Procedure of blending was by hand mixing and air laying, similar to earlier sets. Binder content was maintained at 50% in all samples. Combinations of Natural fibers (Cotton, Kenaf, and Flax) are shown in Table 2 and Table 3. This evaluation was carried out for each type of biodegradable binder. Binder:Cotton (50:50) is used as a control.

Table 2 Fiber Mixtures with Varying Kenaf Content.

Sample No.	Binder	Cotton	Kenaf	Flax
1	50	50	0	0
2	50	40	10	0
3	50	25	25	0
4	50	10	40	0
5	50	0	50	0

Values in the table are in percent

Table 3 Fiber Mixtures with Varying Flax Content.

Sample No.	Binder	Cotton	Kenaf	Flax
1	50	50	0	0
2	50	40	0	10
3	50	25	0	25
4	50	10	0	40
5	50	00	0	50

Values in the table are in percent

Process Optimization (using Intimately Mixed Fiber):

Intimately mixed carded web of cotton (60:40) produced in commercial unit by blending cotton with binders such as PLA, PLAbico, and PP (control) was available in desirable quantity for experimentation. Uniformity of the web was quite superior to the previous samples prepared by hand mixing and air laying. Trials to arrive at optimum process conditions were carried out using these webs. The composite processing parameters such as curing time, temperature of the hot press, basis weight etc. were studied in relation to the finished product quality, tensile strength in specific. Later same samples were tested for acoustic properties as well as flexural and impact properties.

III CHARACTERIZATION METHODS

The samples produced in the experiments were analyzed for physical properties and structure after conditioning the samples for at least 24 hours under standard laboratory conditions, which is 21°C +/- 1°C and 65% +/- 1% relative humidity [40].

Basis Weight:

Composite samples were cut into a rectangular piece of 6"x 4" size and weighed. Basis weight is expressed as grams per square meter.

Diameter and Denier:

Fiber samples were examined under a Olympus optical microscope or Scanning Electron Microscope and diameter was measured using the image analysis software. Since the cross section is not circular in all fibers, the second largest dimension is reported as diameter. Using diameter data from 20 readings denier was calculated using density data. Fiber denier was also calculated by measuring the mass of a known length of manufactured fiber and then converting the mass to grams per 9000 meters.

Density:

Density of the fiber samples were measured using the Density Gradient Column made of NaBr-Water for density range 1.2 to 1.5 g/cc; Isopropanol-water for density range 0.8 to 1.0 g/cc. Literature values were taken for the samples that are out of these ranges.

Differential Scanning Calorimetry:

Differential Scanning Calorimetry of the polymers and fibers was carried out in a Mettler DSC821 and DSC822. The sample was placed in a 40 microliter volume standard aluminum crucibles with lid. In order to vent the air trapped three holes were made on the lid using a pin. Nitrogen at 200 milliliter per minute was used as the carrier gas in all the samples. Heating rate of 10°C per min was used while running the DSC. DSC scan was used to get an idea about the melting behavior of various binder fibers. Bicomponent fibers showed two melting peaks.

Thickness:

Using TMI tester, thickness of the composites was tested like a nonwoven fabric. It is determined by observing the linear distance that a movable plane is displaced from a parallel surface by the specimen while under a specified pressure. For thicker composites (more than a mm) micrometer was used to measure the thickness. Average of four readings were taken for evaluation.

Tensile Properties:

For natural fibers, individual fiber tenacity elongation values were from the literature [28]. Since fiber bundle behavior is important in composites, tensile test was carried out for a bundle of fibers containing 20 filaments with 1-inch gage length. Tensile properties of the fibers and composites were measured using United Tensile Tester with test conditions described in ASTM [40]. Composite samples (up to 100 lb breaking load) were tested as nonwoven fabrics where in samples were cut to 1inch width, 5inch length and tested at 3 inches gage length. Uniform extension rate of 1inch per minute is maintained in all cases. Average of four readings is taken for evaluation. In case of thicker composite samples (breaking load more than 100 lb up to 5000 lb), MTI Pheonix Tensile Tester Model 386 was used. Average of four readings was used in the analysis.

Flexural Strength:

The flexural strength of a composite is its ability to resist bending under load. For composites, bending load is typically measured at 5% strain of the outer surface. A 3-point bending set up (Figure 6) was mounted on MTI Phoenix Tensile Tester and bending test is carried out according to ASTM D178.

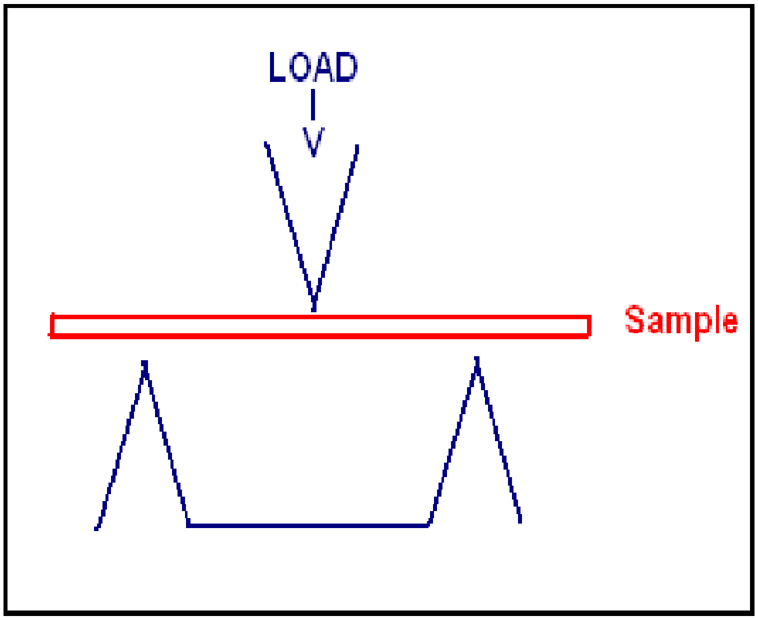

Figure 6 Three-Point Bending Test Setup.

A 5 inches long, 1inch wide specimen is placed on 3 point setup as shown in Figure 6 with 3”gage length (distance between lower pins) and test conducted at 0.07 in per minute compression rate. The bending load at 5% strain is entered. Average of four readings is used in the evaluation.

Surface and Cross-section Pictures:

Scanning Electron Micrographs (SEM) were obtained using the Hitachi and Leo 1525 surface scanning electron microscope in back scatter with Gemini column with system vacuum of ~$1.3x10^{-5}$ torr and at an acceleration voltage ~5 kV. SPI Module sputter coater was used to coat the samples (if necessary) with gold for 5 seconds at 20 mA plasma current to reduce charging while scanning. Sometimes, pictures were taken with the Olympus optical microscope (up to 40 magnification level). However, SEM pictures were taken for higher magnification and better clarity. Depending on the purpose different the magnification levels were chosen. Fiber surface, cross-section, uniformity, fiber pull out, melt flow over the fiber etc. were observed the SEM pictures. For cross-section, the samples were carefully placed on a cutting board and cut by a sharp blade with one hit to get a neat cut. Minimum of two specimens of the same sample were placed on the platform and observed. For surface observation, a thin wafer of the sample is placed on the platform. A few pictures (e.g., fiber samples) were taken by tilting the sample holding platform by about 30° where in both surface and cross-section could be seen in one frame. Samples after the tensile test (fractured) were prepared similar to cross-section, but fiber pullout focused while scanning.

Acoustics:

Generally, composites consist of randomly laid fibers bonded together. These fibers have ability to absorb sound energy. The ability to absorb sound waves depends on the dissipation of sound energy when it passes through the composites. The samples were sent to University of Bremen, Germany for acoustic testing in the Impedance Tube (Figure 7) and results were analyzed. The ratio of amount of absorbed energy to the original incident energy is known as absorption coefficient. Similarly, the ratio of the pressure amplitude of the reflected wave to the incident wave is known as reflection coefficient. Furthermore, acoustic analysis is done through software involving a set of calculations to arrive at the ratio of the surface sound pressure to the sound particle velocity through the surface is called as acoustic impedance. Similarly the ratio of the sound particle velocity through the surface to the surface sound pressure is known as admittance and the ratio of the airborne sound power incident on the partition to the sound power transmitted and exited on the other side is called as transmission loss.

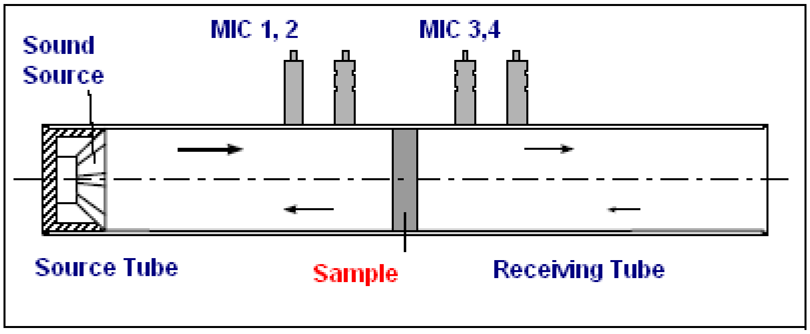

Figure 7 Schematic Picture of Impedance Tube.

The impedance tube consists of a sound source or loudspeaker at one end of the impedance tube and a sample placed in a holder at the other end. The loudspeaker generates sound waves of various frequencies that travel in the source tube. As it hits the sample, a part of the wave reflected back into the source tube, a part absorbed by the material, and part passing through the material into the receiving tube. On further travel, sound waves hit the end of the receiving tube where a part is reflected and some exit the tube.

The sound pressure is measured at four fixed points (two in the source tube and two in the receiving tube) and calculation is done through software that uses a complex transfer function using a four-channel digital frequency data to determine the transmission loss of the material. The usable frequency range depends on the diameter of the tube and the spacing between the microphone positions. This method is described in both ISO 10534–2 and ASTME 1050. Impedance Tube Kit Type 4206 [41] at uses a 100 mm diameter tube for frequency range 50 Hz to 1600 Hz and 29 mm diameter tube for frequency range 500 Hz – 6400 Hz.

Impact Strength:

At early stages, depending on the application and the customer's preference either falling-weight or pendulum impact test method was used by the material scientists and engineers. The pendulum method is increasingly popular now that uses notched specimens. It is known as notched-Izod impact test [42]. For automotive parts, ASTM D3763 covers the impact test method. Specimen size for Izod testing is 2.5 x 0.5 inch and sample thickness typically 0.125 to 0.5inch. Specimens are notched using Tinius Olsen specimen notcher Model 892 and conditioned at standard laboratory temperature and humidity before testing. Tinius Olsen Impact Tester Model 899 (Figure 8) was used for testing the composite samples. It consists of a base, a pendulum of single-arm design, and a striker rod (also called a hammer). The mass and the drop height determine the potential energy of the hammer and built in software calculates impact strength that is expressed in ft.lbf per inch. The thickness of the specimen is included in the calculations. Samples were taken at random. Average of four readings was used for evaluation .

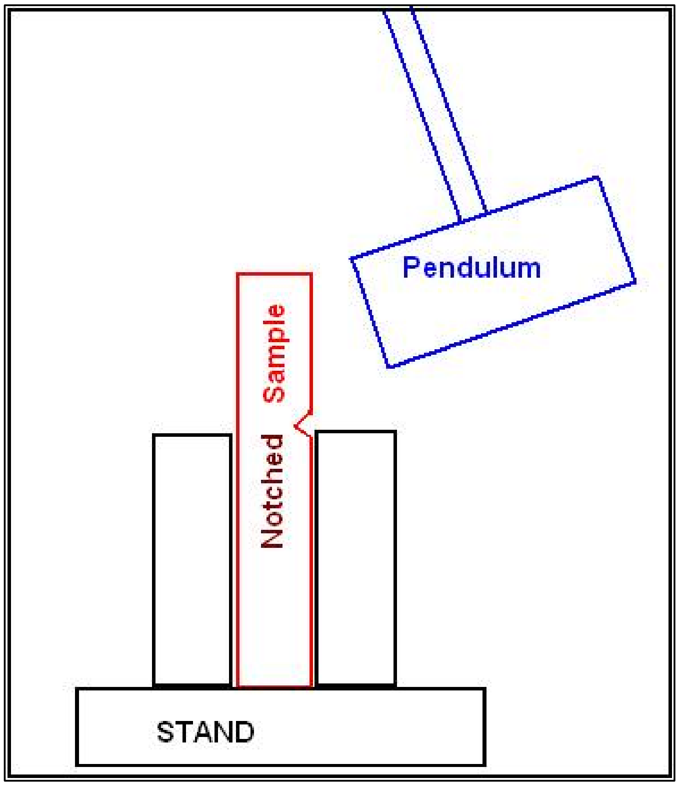

Figure 8 Schematic of Notched Izod Impact Test by Falling Pendulum method

IV. RESULTS AND DISCUSSIONS

Natural Fibers:

Properties of the natural fibers are presented in Table 4. It can be seen that cotton is a fine fiber with convoluted surface. Cotton is the most popular natural fiber for its natural soft and cool feel, comfort, and moisture absorption properties. Whereas kenaf and flax fibers are coarser than cotton, possess higher tenacity and lower elongation compared to cotton. Kenaf and flax surfaces are rough as they are bast fibers. These fibers are used to make ropes and cords. Later improvement in processing led to finer textures and thereafter blending with cotton gained importance. Kenaf exhibits lower apparent density due to several pores and voids in its structure. Equilibrium moisture level is about 7% for all these natural fibers. The cellulose content is the highest in cotton and has higher elongation at break compared to bast fibers.

Table 4 Properties of Natural Fibers

		NATURAL FIBERS		
		Cotton	Kenaf	Flax
Diameter *	micron	9 -27	18-37	10-25
Denier		0.7 -2.3	2.4 - 3.8	0.8 - 3.1
Fiber length	inch	0.5 - 1.5	0.5 - 4	0.5 - 5
Tenacity	gpd	2.2 - 3	4 - 11	3-11
Specific Gravity		1.54	#	1.51
Moisture**	%	7	7	7
Cellulose content**	%	80 - 90	60- 64	75- 79
Elongation at failure	%	6 - 8	4 - 5	3 - 5

* As fibers are not circular, the diameter corresponds to the second largest dimension.

** Values are from literature # Apparent density 0.31 g/cc due to high porosity

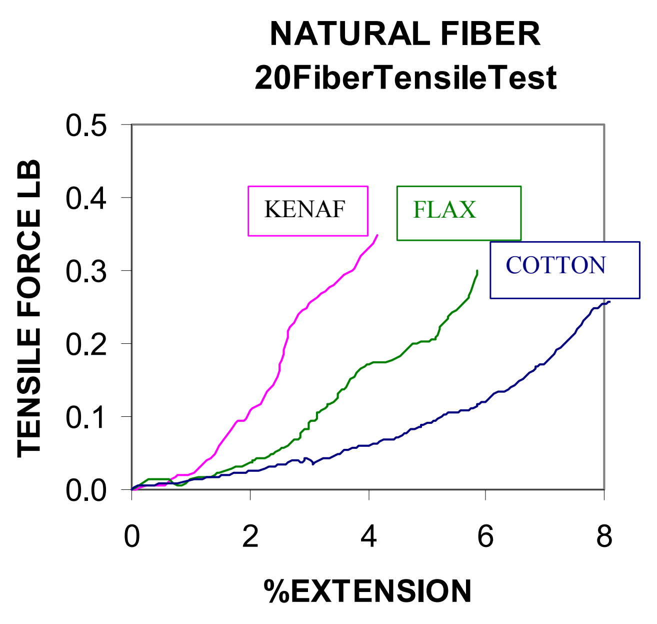

Figure 9 Tensile Properties of Natural Fiber Bundle.

Tensile properties of the bundle of 20 fibers are presented in Figure 9. Individual fiber strength as well as interaction among fibers plays a role in the bundle strength. Surface roughness or convolutions reduce slippage while tensile testing. It can be seen that bast fibers such as kenaf and flax fibers are coarser than cotton, exhibited higher fiber bundle tenacity and lower elongation compared to cotton. Cotton has surface convolutions (Figure 10 & Figure 11) and bean like cross section. Kenaf exhibited highest tenacity due to the higher tenacity of individual kenaf fibers as well as the surface roughness (Figure 12 & Figure 13). Flax properties are close to that of kenaf than cotton.

SEM PICTURES OF COTTON
(at Magnification 400)

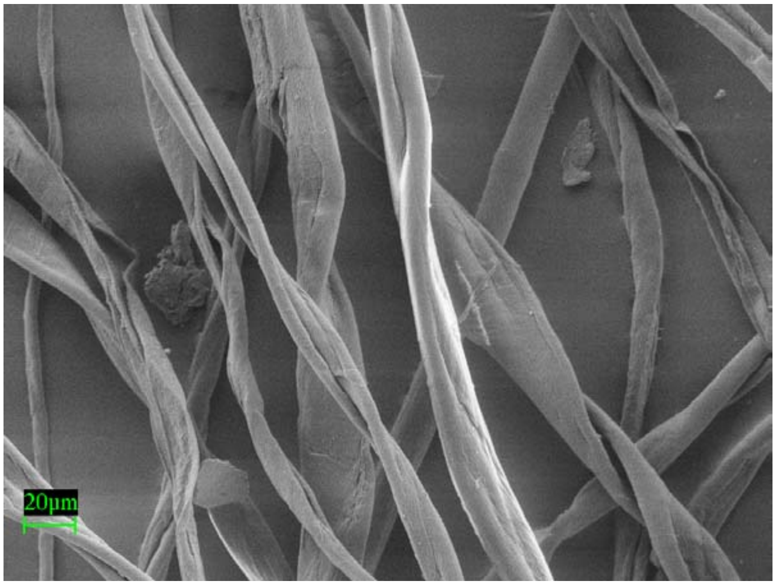

Figure 10 SEM Photo of Cotton Fiber Surface.
(showing convolutions)

Figure 11 SEM Photo of Cotton Fiber Cross-section.

SEM PICTURES OF KENAF

(at Magnification 400)

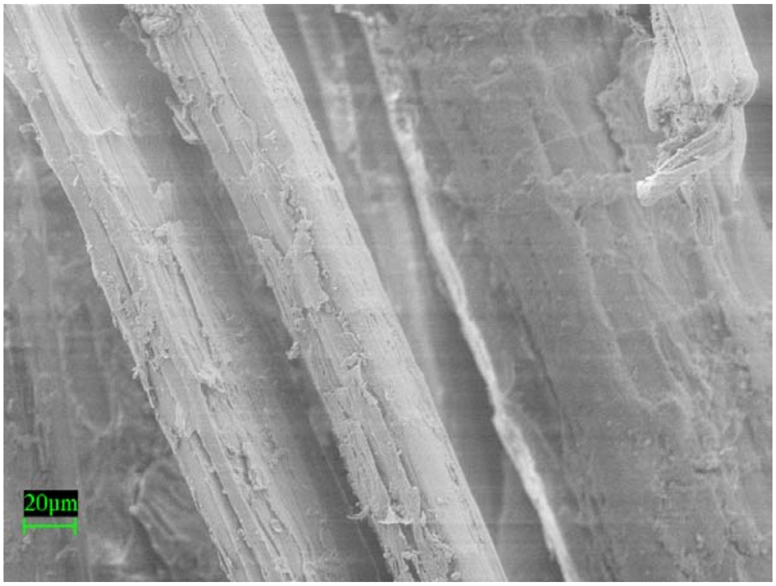

Figure 12 SEM Photo of Kenaf Fiber Surface.
(shows surface roughness)

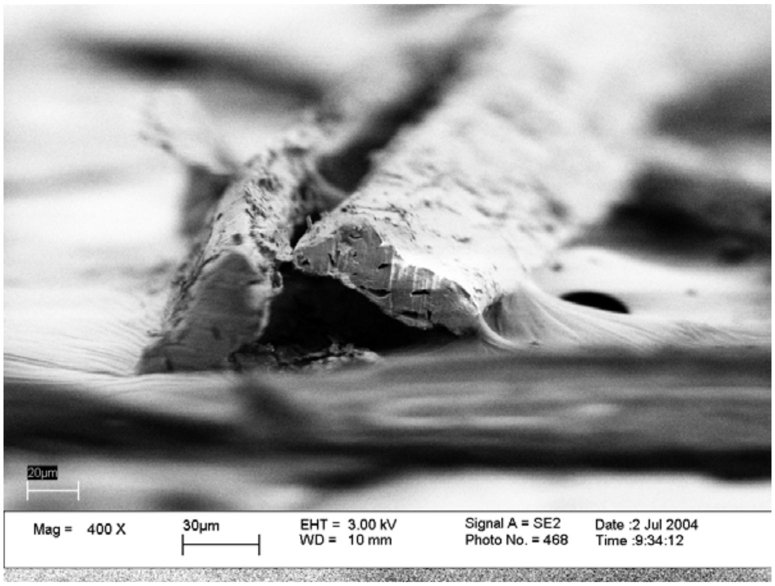

Figure 13 SEM Photo of Cross-section of Kenaf.
(showing pores)

Flax exhibited properties in between that of cotton and flax. Flax surface (Figure 14 & Figure 15) is smoother than kenaf. Generally, flax fibers are longer than cotton and kenaf. When these natural fibers are incorporated in the cotton composite, they lay at random in all three dimensional orientation. When subjected to mechanical stress the surface convolutions and rough points act like ratchets in action that gives rise to the flexibility and elongation. Bast fiber surface roughness greatly depends on the process of retting and after treatment. The pores in kenaf are responsible for the lower density and better sound absorption nature.

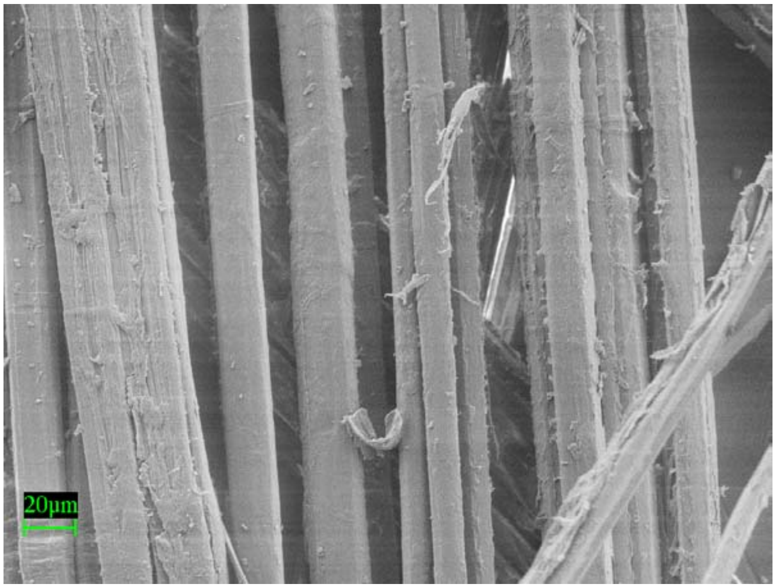

Figure 14 SEM Photo of Fiber Surface of Flax.
(shows rough surface)

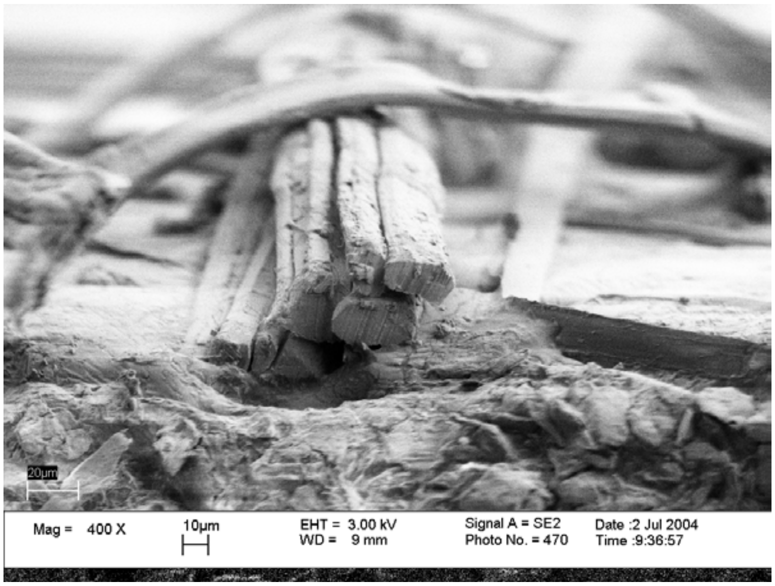

Figure 15 SEM Photo of Flax fiber cross-section.

Binder Fibers:

Properties of the binder fibers are summarized in Table 5. It can be seen that PLA, PVAc and Biomax fibers are coarser and have higher specific gravity compared to conventional PP binder. PLA exhibited higher tenacity and lower elongation compared to PVAc and Biomax. Moisture level is slightly higher than PP for all biodegradable binders. However, it is negligible compared to the moisture in natural fibers. Melting point of PLA is very close to that of PP around 170°C, thus can be a good substitute binder that is biodegradable, where as PVAc and Biomax have higher melting point of about 200°C. Moreover, higher temperature causes damage to cotton leading to yellowing and some odor. Longer the duration more the odor is. All the web samples were dried in the oven for about 2 hours at 90°C under vacuum to remove moisture before subjecting to thermal bonding.

Table 5 Properties of Binder Fibers

		BINDER FIBERS		
		Biomax	PVAc	PLA
Diameter (average)	micron	42	31.5	37.5
Denier		5.2	6.8	12.5
Staple length	inch	0'.5 - 2	2	0.5 - 2
Crimps	/inch	12	18	0
Tenacity	gpd	2.2	2.5	3.8
Specific Gravity		1.38	1.28	1.26
Moisture	%	0.6	1.5	1.8
Elongation at failure	%	6 - 10	6 - 10	2 -6
Melting Point (DSC)	C	201	199	171

*As fibers are not circular, the diameter corresponds to the second largest dimension

Results from the DSC scans of different binder fibers are shown in Figure 16 through Figure 17. As expected DSC of bicomponant fibers (Figure 17) show two melting points. Generally, sheath has lower melting polymer. Sheath polymer bonds the surrounding material where as core reinforces the product.

Tensile properties of the bundle of 20 fibers are presented in Figure 19. It can be seen that PLA fiber bundle is coarser, possess higher tenacity but lower elongation compared to PVAc and Biomax.

From the detailed observation of SEM pictures of fibers, it can be seen that PLA fibers have round cross section (Figure 20 & Figure 21). At the same time, only PVAc fibers that were used have trilobal cross-section (Figure 22 & Figure 23) that imparts more surface for binding. Biomax fibers produced (Figure 24 & Figure 25) were round in cross section. Some of the properties of Biomax & PLA fibers produced in our laboratory are reported in Appendix 1.

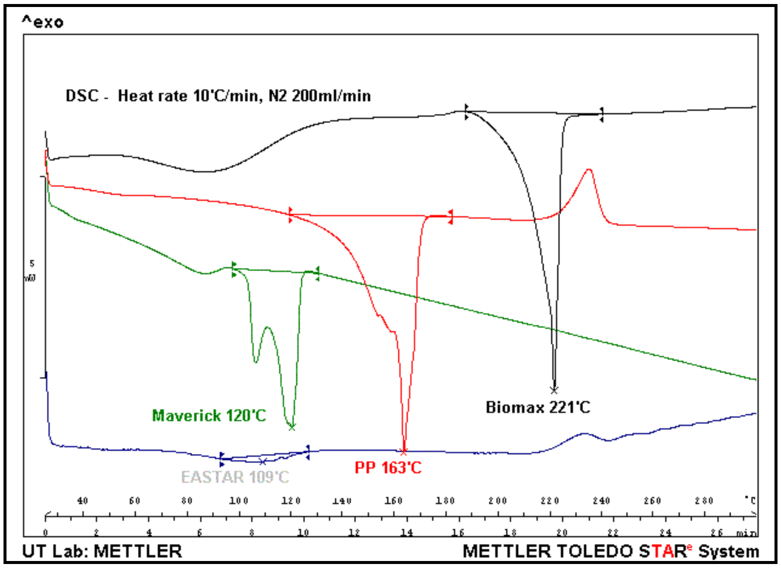

Figure 16 DSC of Binders: PE, PP, Biomax, & EastarPP.

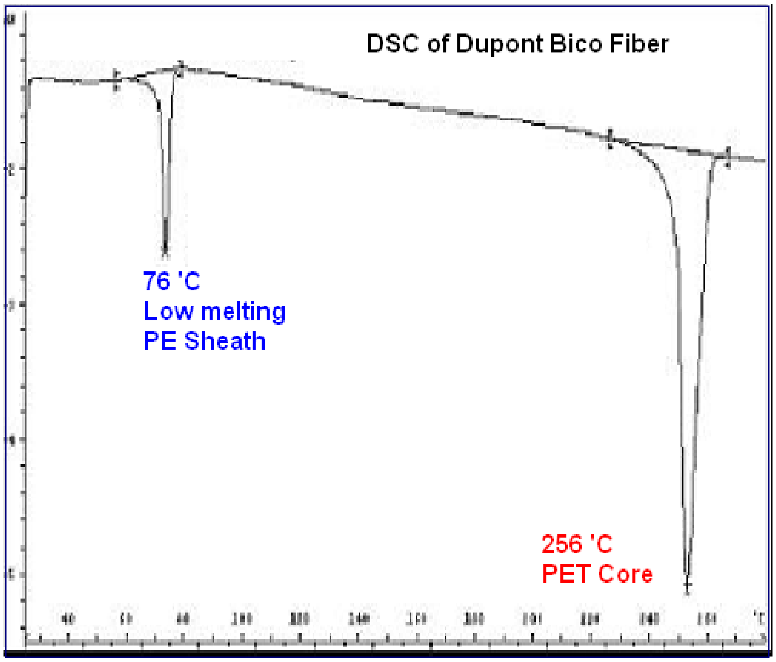

Figure 17 DSC of bicomponent fiber (PE-PET).

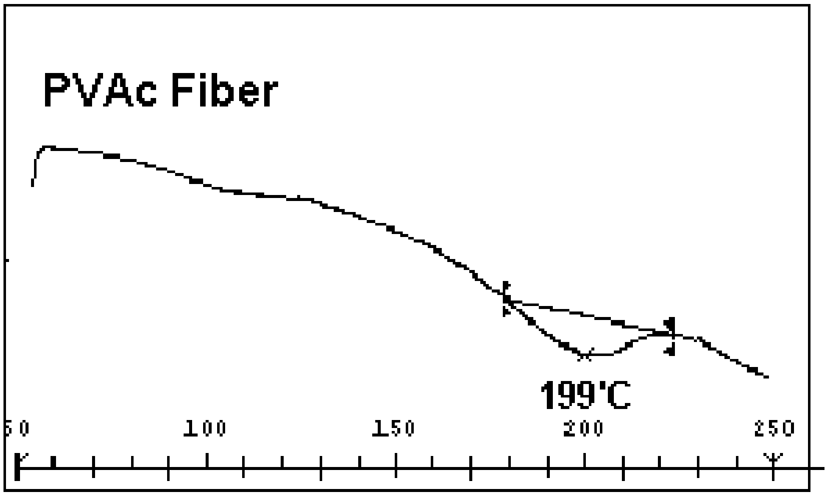

Figure 18 DSC Scan of PVAc fiber.

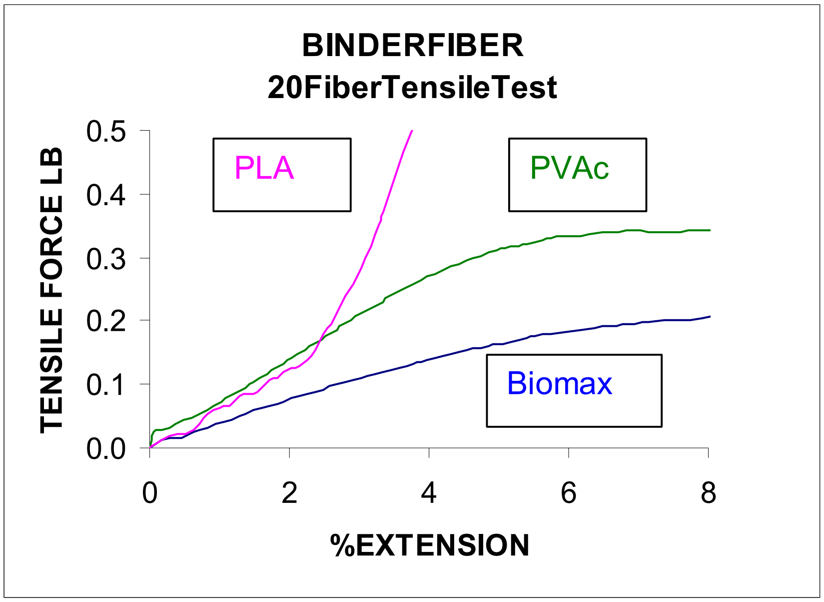

Figure 19 Tensile Properties of Binder Fiber Bundle.
(20filament bundle)

SEM PICTURES OF PLA
(at Magnification 400)

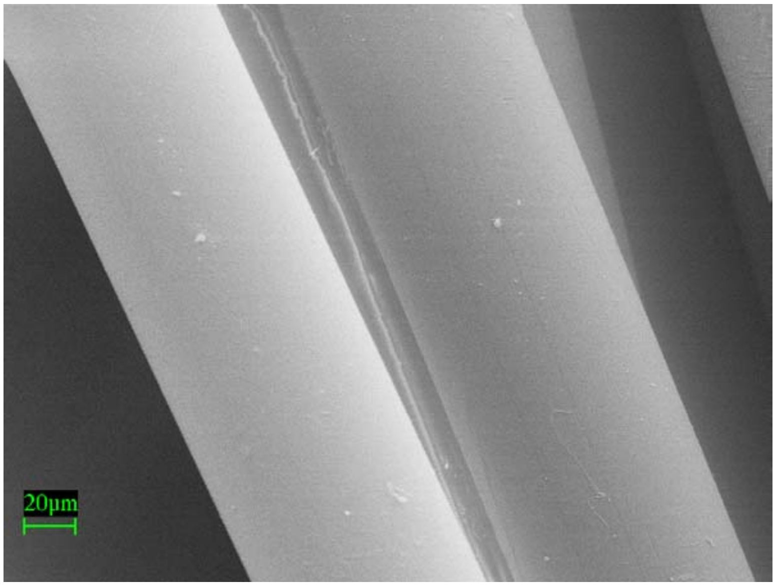

Figure 20 SEM Photo of PLA fiber surface.

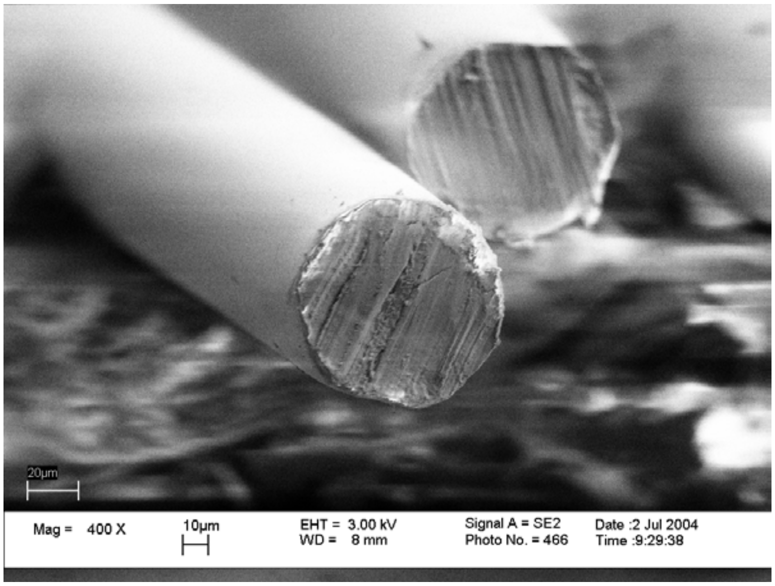

Figure 21 SEM Photo of PLA fiber cross-section.

SEM PICTURES OF PVAc
(at Magnification 400)

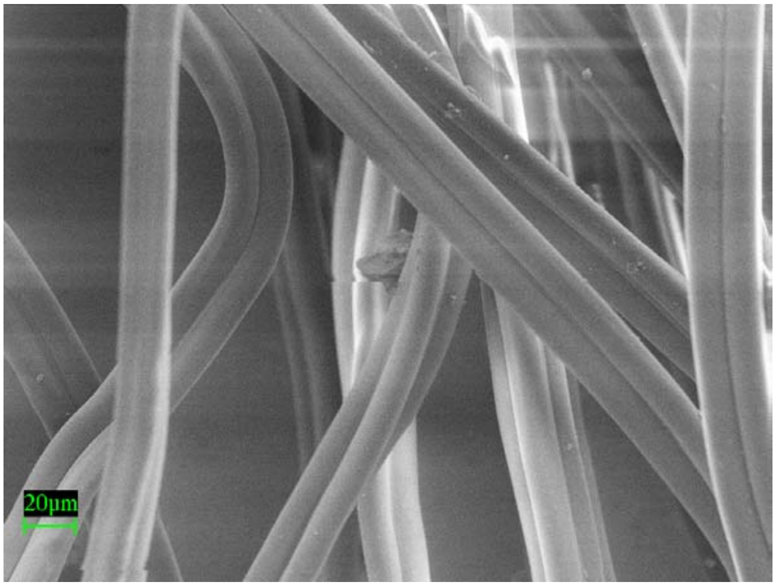

Figure 22 SEM Photo of PVAc fiber surface.

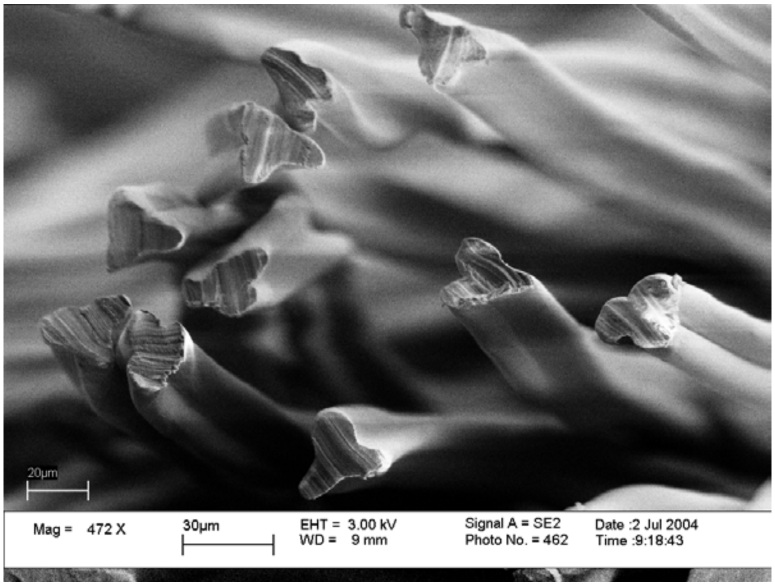

Figure 23 SEM Photo of PVAc fiber cross-section (Trilobal).

SEM PICTURES OF BIOMAX
(at Magnification 400)

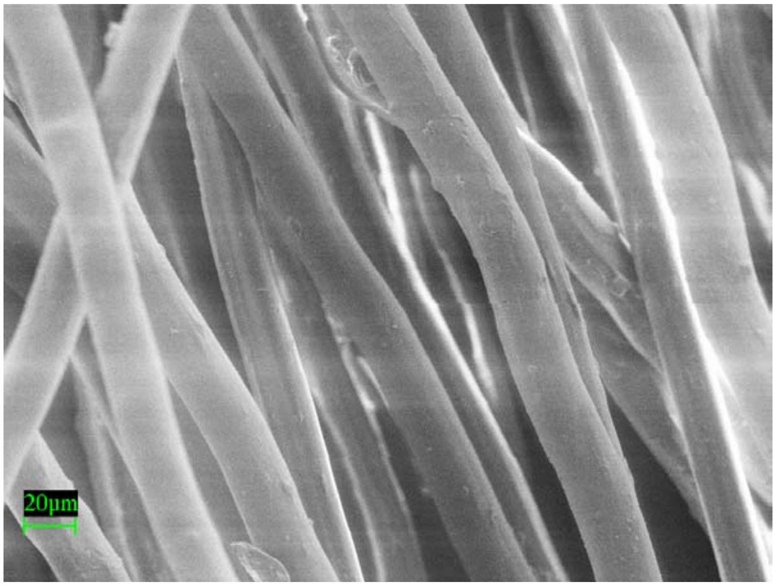

Figure 24 SEM Photo of Biomax fiber surface.

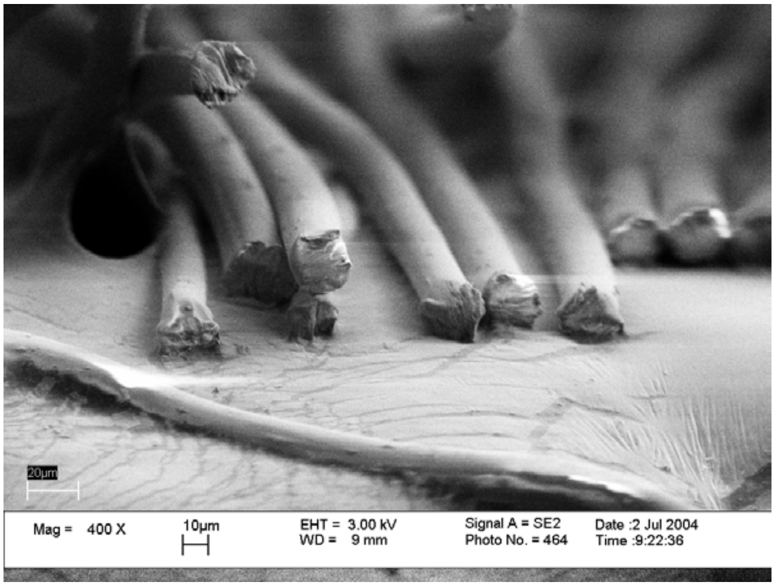

Figure 25 SEM Photo of Biomax fiber cross-section.

Fiber Bonding:

Results of the preliminary studies on thermal bonding behavior of binder fibers using Dynasco Hot Track Heat Sealer shown in Table 6 indicate that bond strength of raw cotton with EastarPP is more than that of the bleached cotton. Table 7 indicates optimum bonding temperature of various binder fibers that provides maximum bond strength. These tests were carried out using small number of fibers (50) and the results are used as guidelines for further experiments. Similarly the relation between the melting temperature and the bonding temperature was also observed .

Table 6 Bond Strength.

ID	**Bond Strength (g)**
EastarPP bonded with Raw Cotton	176
EastarPP bonded with Bleached Cotton	104

Bonding carried out at 90°C, 20 millisec, and 200 psi.

Table 7 Optimum Bonding Temperatures of Binder Fibers.

Fiber ID	**Denier**	**Bonding Temp °C**
Biomax	3	160
Eastar-PP bico	4	130
PE-PET bico	4	115
PLA	10	140

Comparison of Sandwich Type and Fiber Mix Type Composites:

The results from tensile testing of various samples in the initial test runs are summarized in Figure 26. Most of the samples had a comparable basis weight in the range of 180 to 220 gsm. In all the other cases, the web thickness was in the range of 400-600 micron, at least about a third of the original web thickness. These tests are carried out to understand the art of making composites and properties of various components present in it. The natural fibers and the binders were chosen at random and used in these trials. Tests results were standardized for 200gsm and reported.

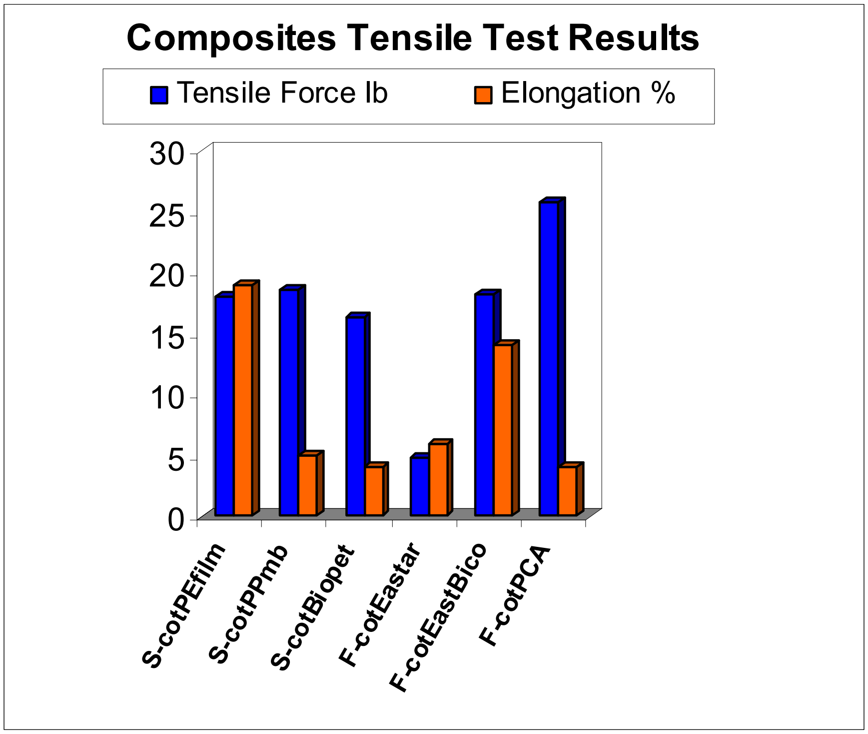

Figure 26: Comparison of Tensile Properties of Sandwich (S) type composites with Fibermix (F) type composites.

As evident from the data, though the sandwich type (S) composites showed increase in tensile strength, tensile failure occurs with separation of its layers (Figure 27) indicating poor bonding between surfaces and the core. In most of the sandwich type samples, there was no intimate mixing between cotton and binder fibers and it was not possible to obtain good consolidation of the webs. In addition, when pellets, film or melt blown webs were used, there was not sufficient flow to uniformly spread the matrix resin all around the webs. That is why strength of the webs was not as high as expected. This lack of bonding throughout the cross section can be seen from the pictures given in Figure 27. On both top and bottom there is melting of the binder and composite formation due to the flow of the polymer.

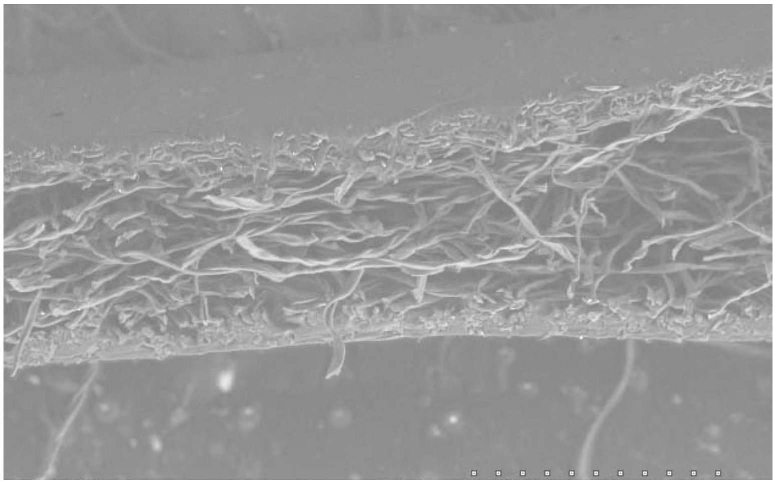

Figure 27 SEM Picture of Sandwich type Composites.
(Shows layer separation)

However, the internal layers or places in between are not well consolidated and it can easily delaminate. Even using multiple layers of binder and fibers did not show any improvements in the performance. This is due to the fact that in spite of allowing sufficient time under the processing temperature and pressure, there is not enough flow of the binder resin into and around the matrix fibers to form a good bonding interface. This observation suggests that if properly processed, it is possible to produce good composites. In fact, the strength and elongation values for these composites are reasonable.

Composites formed out of Cotton and Eastar had poor tensile properties due to the fact that there was not enough binder fiber in the webs to produce a continuous matrix. However, wherever there was binder fiber, it did melt and flow around the natural fibers. On the other hand, Cotton/EastarPP webs showed very high strength. This indicates that the Eastar polymer binds very well with the cellulosic fiber, and is a promising candidate for such products or processes. The SEM photograph (Figure 28) of Cotton/EastarPP (70:30) shows that there are still regions that do not have sufficient binder. This explains why the 70:30 blend has lower strength values compared to that of composites formed with 50% EastarPP binder fiber. Again, this being a bicomponent fiber, and the actual binder being only Eastar, the net binder component was only 15% of the total system.

In fiber mix (F) type of composites, the well-mixed fibers behave as one single material until the binders fall apart from the cotton fibers or fibers break at the final stage of fracture. On the other hand, the mixed fiber composites showed good bonding between cotton fibers and the binder fibers all through the cross section, thus resulting in increased tensile strength.

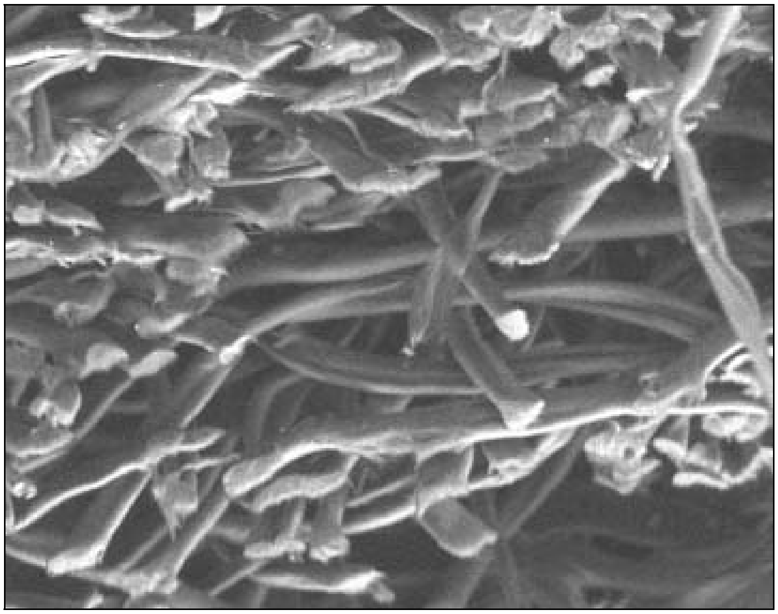

Figure 28 SEM Picture of Cotton Eastar/PP(70/30)Bico Sandwich.

PCA-Cotton showed excellent tensile properties and good bonding between fibers (Figure 29). During tensile testing, initially entire composite takes the load, then, gradually the load is taken by cotton fibers until it breaks. This can be clearly seen in the SEM picture as cotton fibers jetting out of the fractured sample (Figure 30), and the same is schematically shown in Figure 31. Moreover, the poor strength of Eastar-cotton fiber mix can be attributed to the non-uniformity of the web due to the difficulty in mixing of the two fibers and due to the loss of binder fiber during carding. Generally, the fibers and binders are placed in an Aluminum foil and hot pressed. In case of Eastar, teflon sheets were used to overcome the problem of the sample sticking to the foil. Thus, Eastar has excellent bonding quality, but it is difficult to distribute evenly in the product.

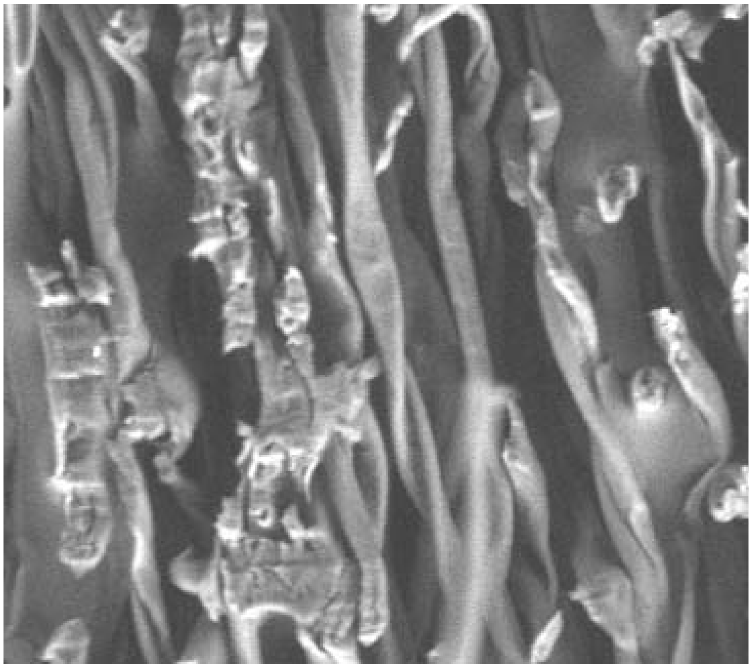

Figure 29 SEM Photo of the Cross section of Cotton/PCA mixed fiber composite.

(at Magnification 800X)

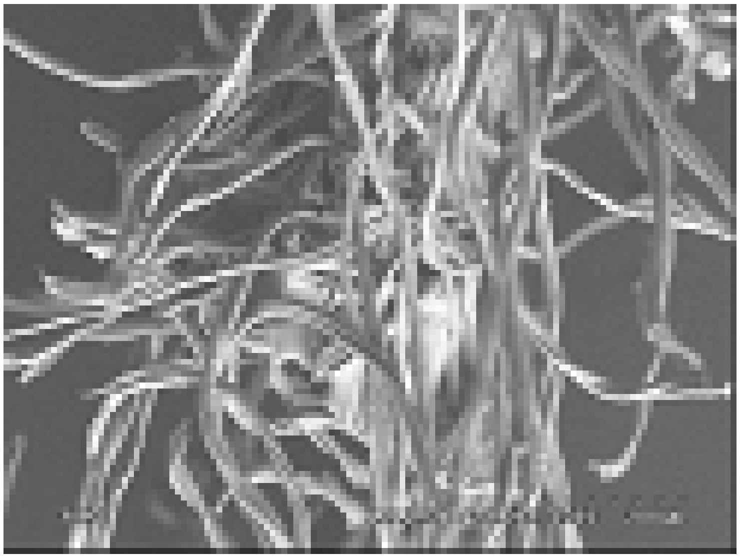

Figure 30 SEM Photo of Cotton/PCA composite after tensile fracture.

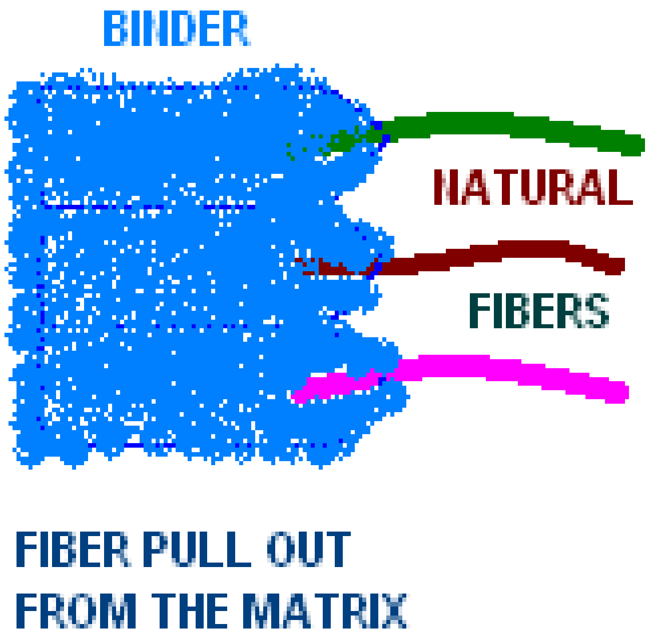

Figure 31 Schematic of Fiber Pull-out during Tensile Fracture

Dry Laid Fiber Mix Composites:

Effect of binder fiber, Dupont-Bico was evaluated using well-carded webs. Although this is not a biodegradable fiber, it is good for comparison and for molding at lower temperature. All the composites were processed under identical conditions and the product had about 400 gsm basis weight. The tensile properties for these samples are shown in Figure 32. Both raw and bleached cottons show good tensile properties, although bleached cotton indicated a drop in elongation. Further bleached cotton has attractive color (whiteness). Raw cotton was used in all further experiments since it is economical as desired in automotive industries. In fact lower grades of cotton and recycled cotton materials are widely used to lower the cost and are becoming popularity.

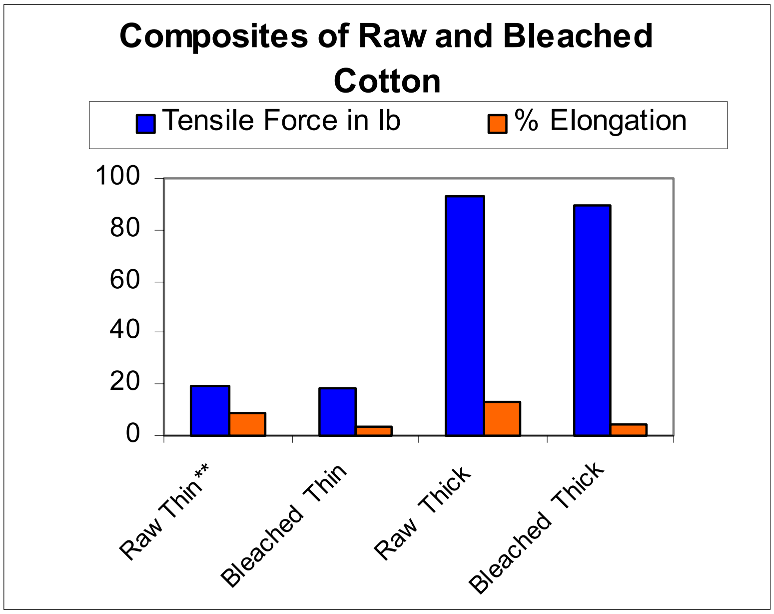

Figure 32 Composites of Raw and Bleached cotton.

With flax and kenaf fibers, it was difficult to produce carded webs, as they were not suitable for the carding machine that we used. However, composites were prepared to understand the bonding between the binder fiber and these natural fiber blends. The fibers were well mixed using an opener, air laid and then webs were formed by hand. These webs were consolidated using the hot press. Results of these composites involving Flax and Kenaf made from hand mixed fibers are presented in Figure 33. As seen in the SEM pictures, Figure 34 and Figure 35 there is a good flow of binder around the cellulosic fibers. It appears that the adhesion between the natural fibers and the binder fibers is quite good.

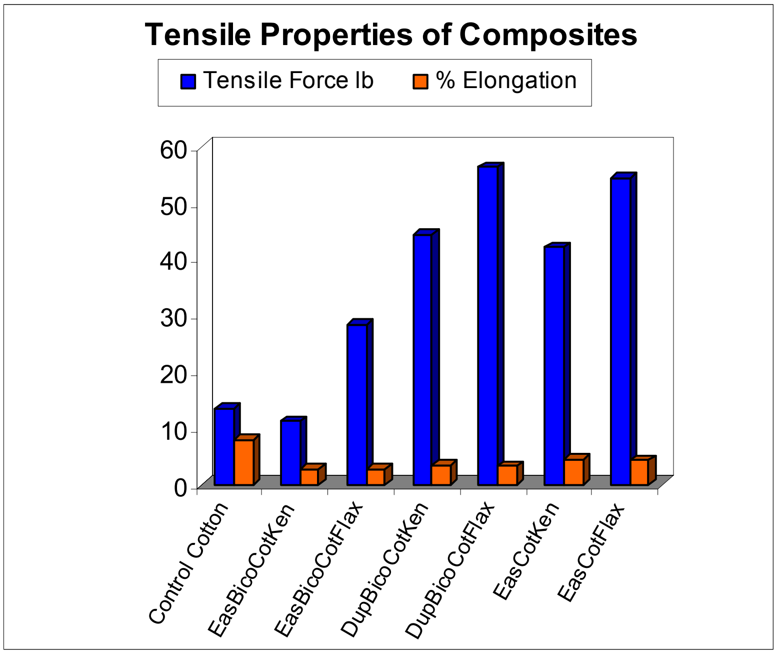

Figure 33 Tensile properties of Dry laid composites.

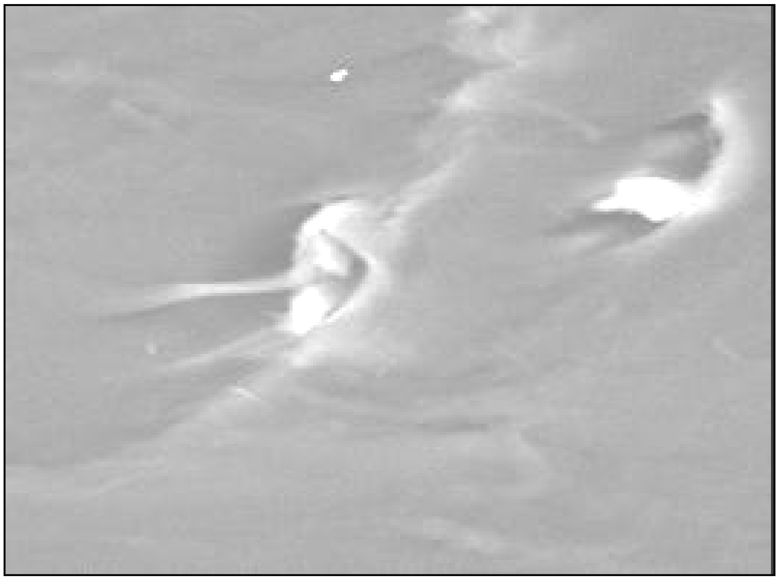

Figure 34 SEM Pictures of Cotton-Flax-Eastar Composites.
(Exhibiting Good Bonding)

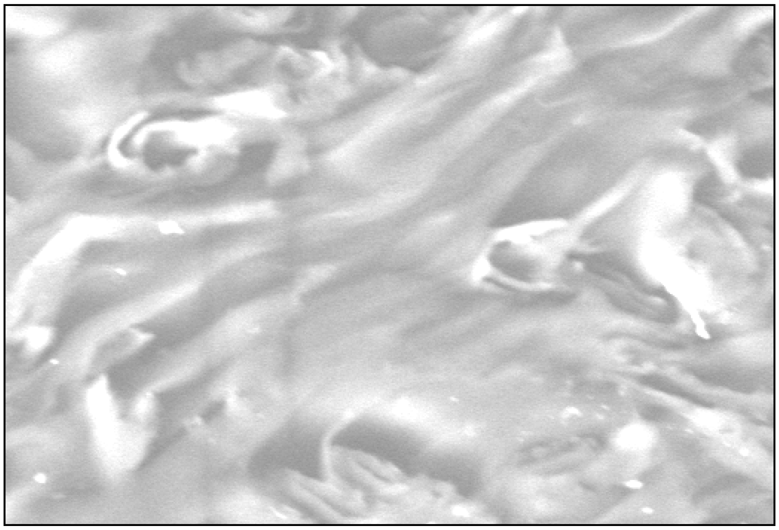

Figure 35 SEM Photo of Cotton-Kenaf-Eastar Composite.

Biodegradable Composites (Dry Laid, Fiber Mix Type):

These composites are made from natural fibers such as cotton, kenaf, and flax using biodegradable binders such as Biomax, PLA, and PVAc. Substantial increase in tensile strength and slight reduction in extension is noticed when flax or kenaf is present in the blend {Biomax in Figure 36 -38, PLA in Figure 42-44,and PVAc in Figure 45-47}. The SEM photographs {Figure 39 through Figure 41, Figure 48 through Figure 50} show strong bonding of the binder fibers with the natural fibers in the composites. As desired the melting and flow of the binder fiber over the cellulosic fibers takes place to form good bond. This is true for all the three binder fibers. Moreover, it can be derived from Figure 37, Figure 38, Figure 43, Figure 44, Figure 46, and Figure 47 that as percentage of kenaf or flax increases the tensile strength increases and the elongation decreases. This trend is nearly same with all three binders.

Composites using Biomax Binder:

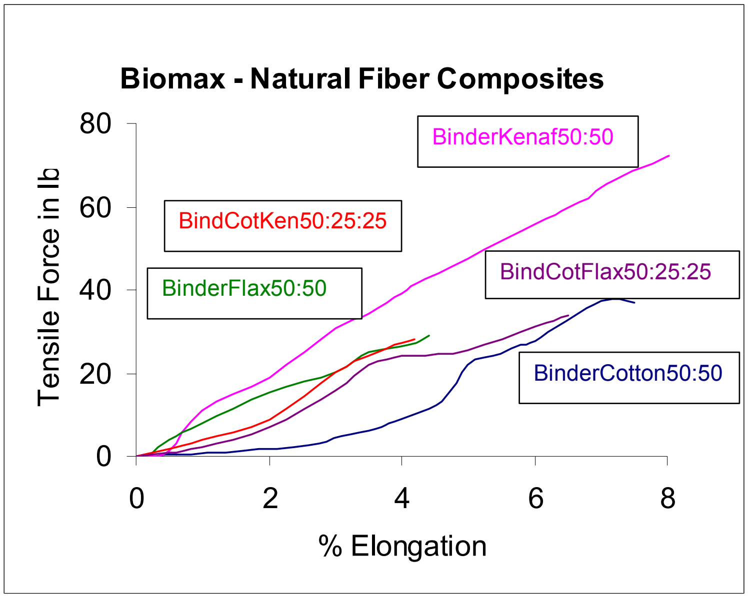

Figure 36 Tensile Properties of Biomax Natural Fiber Composites.

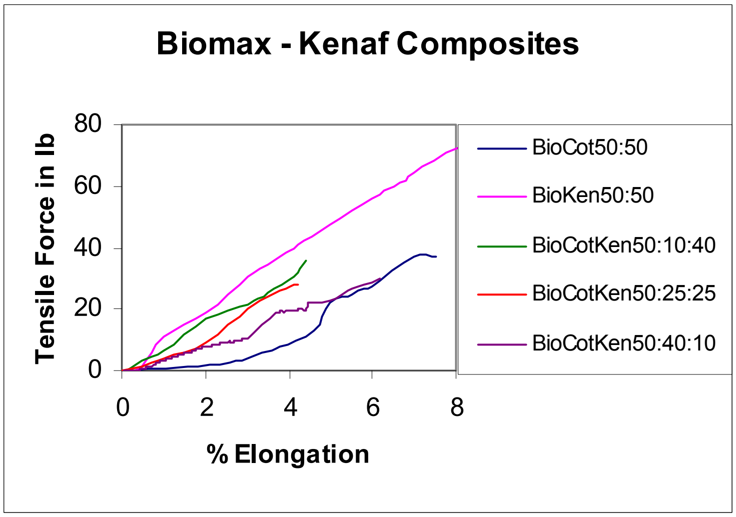

Figure 37 Tensile Properties of Biomax -Kenaf Composites.

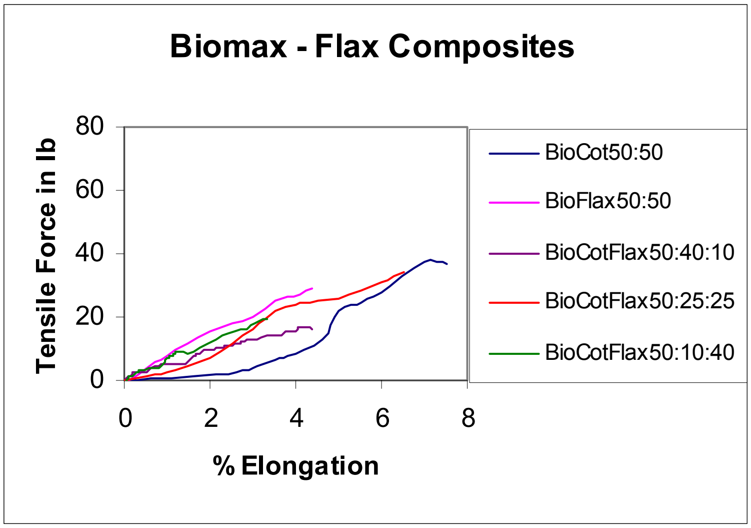

Figure 38 Tensile Properties of Biomax -Flax composites.

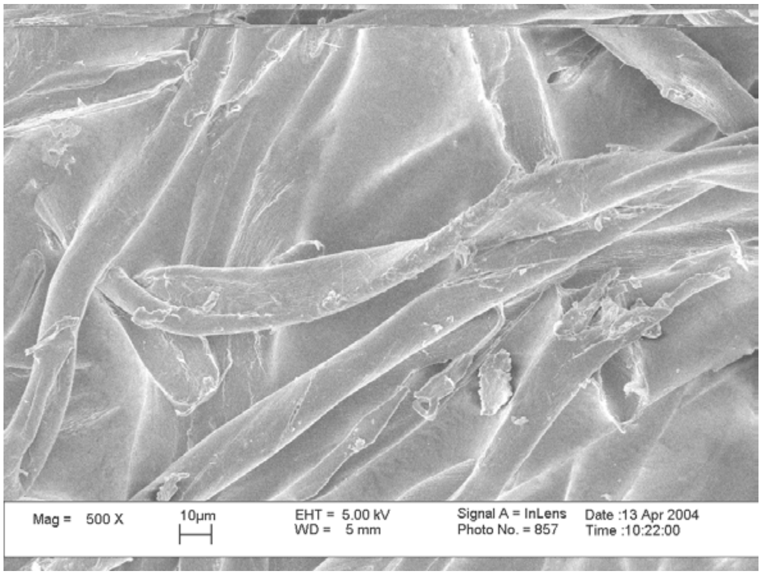

Figure 39 SEM Photo of Biomax-Cotton composite.
At Magnification 500X

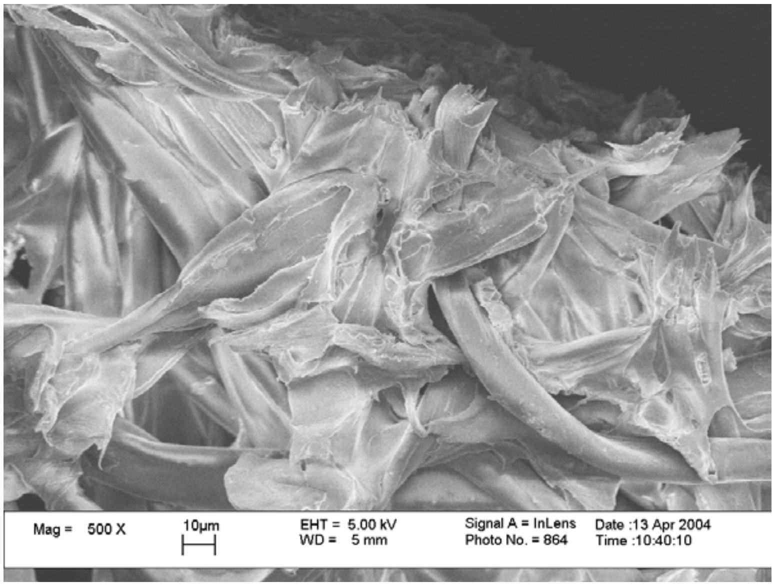

Figure 40 SEM Photo of Biomax-Cotton-Flax composite. At Magnification 500X

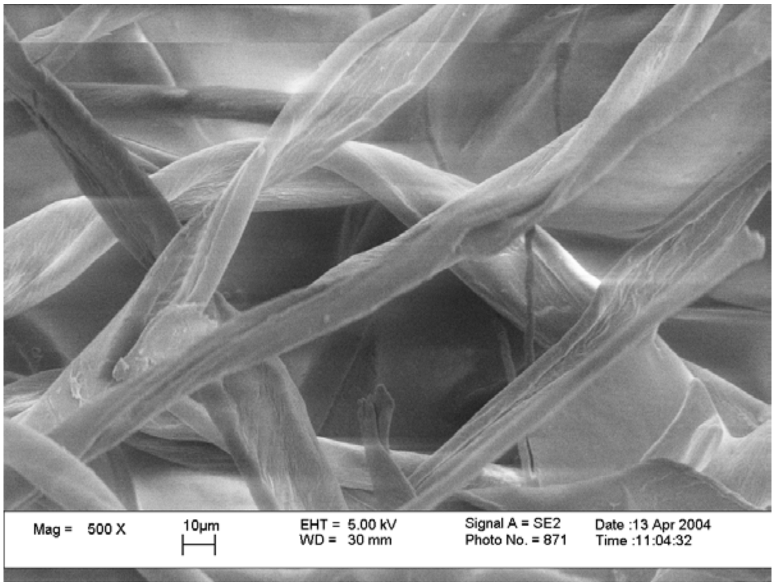

Figure 41 SEM Photo of Biomax-Cotton-Kenaf composite. At Magnification 500X

Composites using PLA Binder:

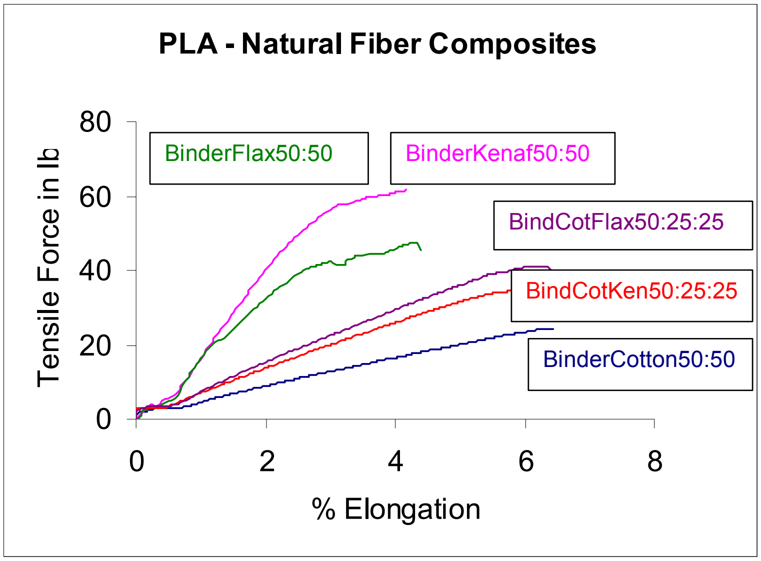

Figure 42 Tensile Properties of PLA Natural Fiber Composites.

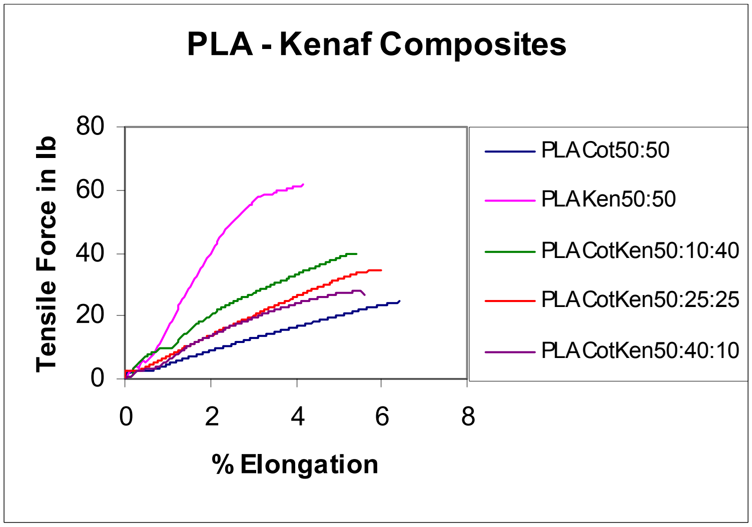

Figure 43 Tensile Properties of PLA-Kenaf Composites.

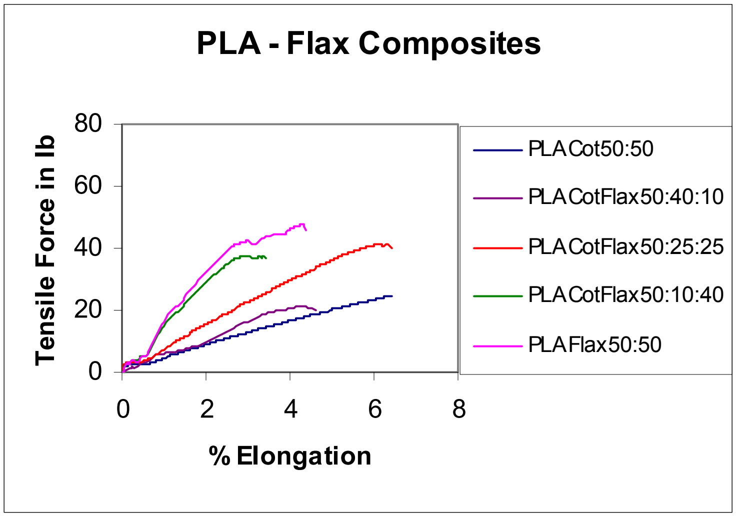

Figure 44 Tensile Properties of PLA-Flax composites.

Composites using PVAc Binder:

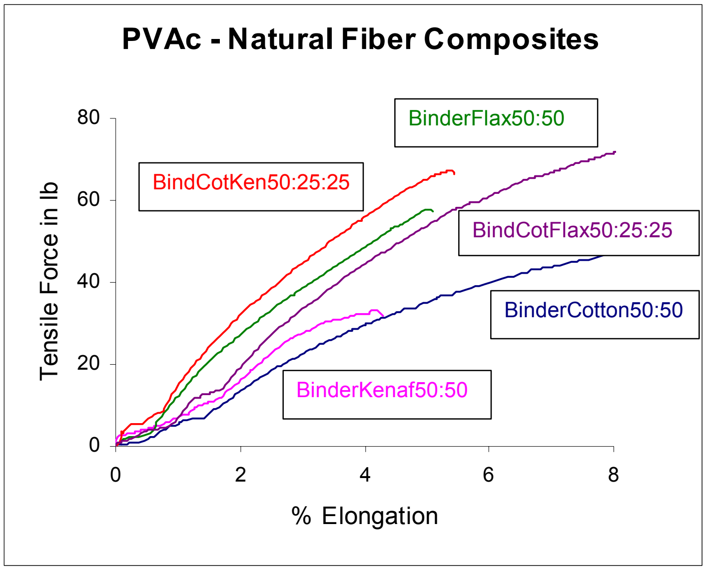

Figure 45 Tensile Properties of PVAc Natural Fiber Composite.

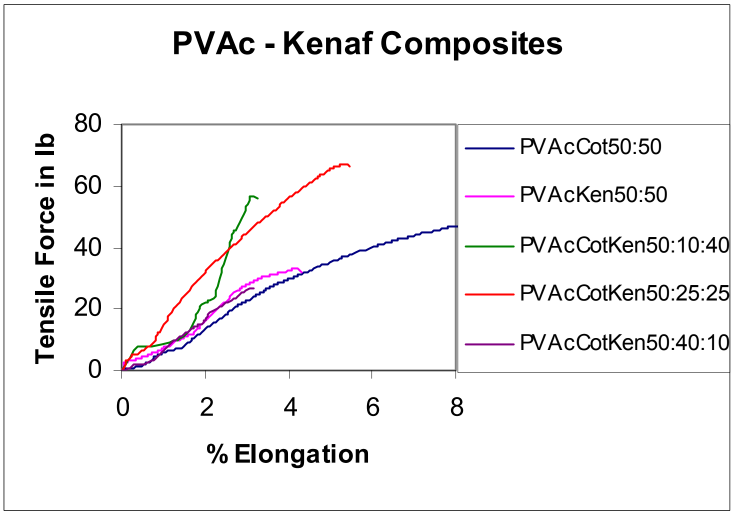

Figure 46 Tensile Properties of PVAc - Kenaf Composites.

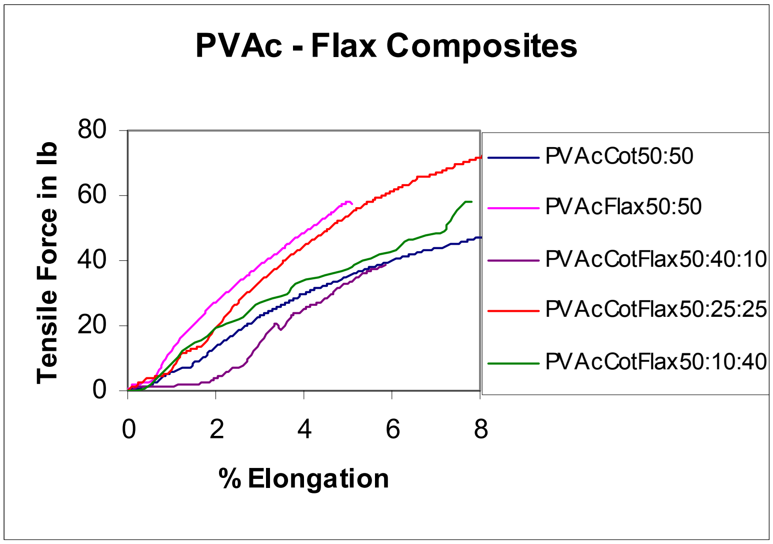

Figure 47 Tensile Properties of PVAc - Flax composites.

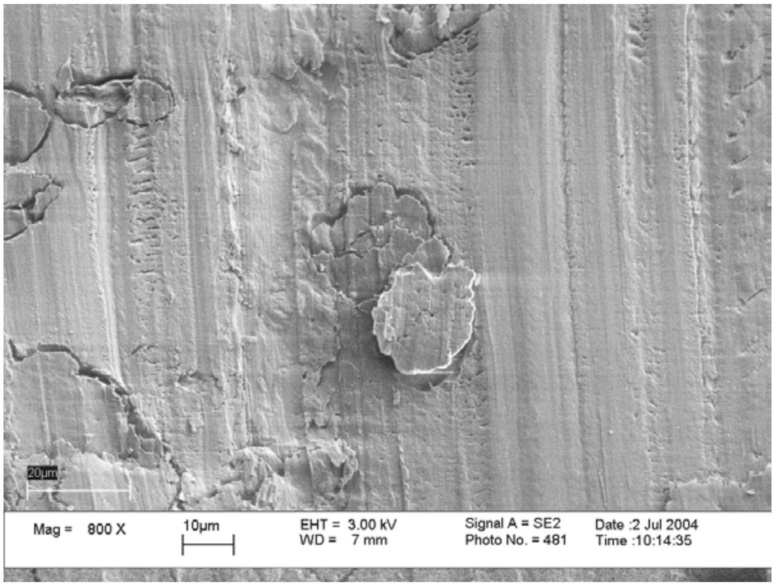

Figure 48 SEM Photo of Flax PVAc Composite cross-section. At Magnification 800X

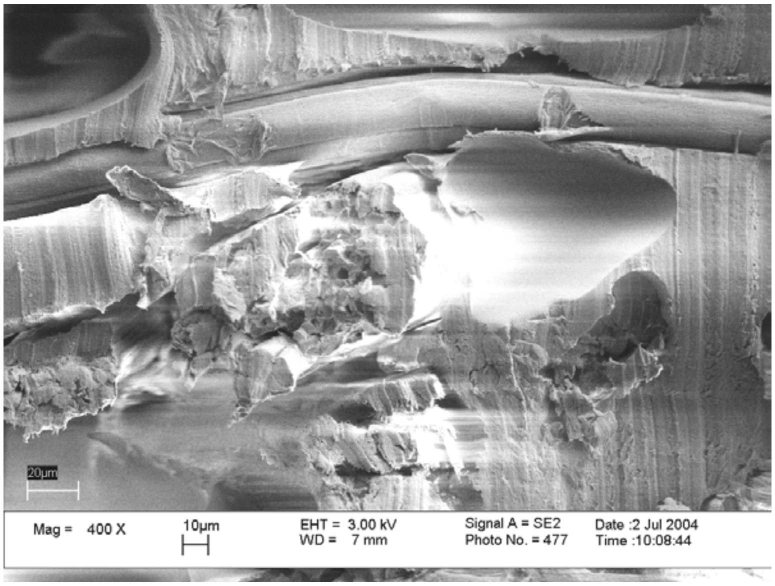

Figure 49 SEM Photo of Kenaf PVAc composite cross-section. At Magnification 400X

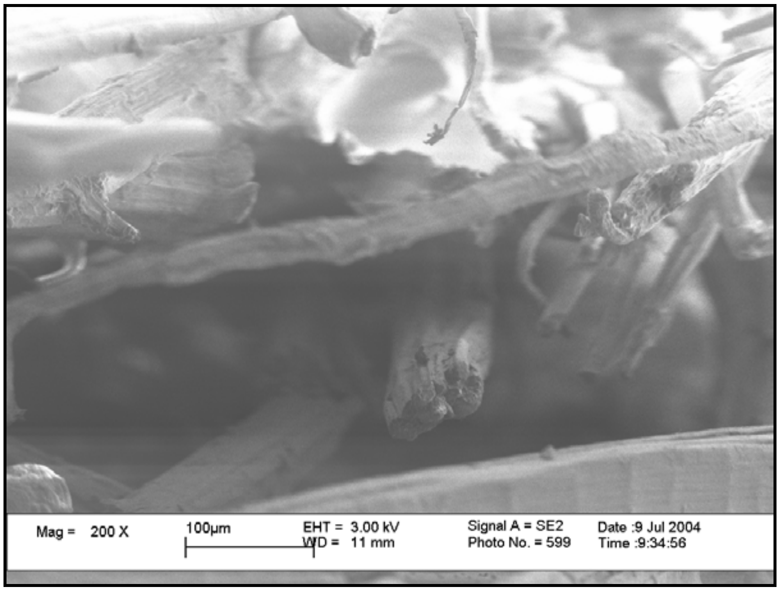

Figure 50 SEM Photo of Kenaf PVAc composite (tensile fractured). At Magnification 200X

Comparison of Binders:

In Binder:Cotton (50:50) composites (Figure 51) PVAc provides more tensile strength and elongation than PLA. Biomax performs very close to that of PVAc. In other words, PVAc and Biomax form better composites with cotton than PLA. In Binder:Kenaf (50:50) composites (Figure 52) PLA results in higher tensile strength and elongation of the composite than with PVAc. Biomax performance is comparable to that of PLA. In other words, PLA and Biomax form better composites with kenaf than PVAc.In Binder:Flax (50:50) composites (Figure 53) PVAc provides higher tensile strength and elongation to the composite than Biomax. PLA performs very close to PVAc. In other words, PVAc and PLA form better composites with cotton than Biomax.

PVAc is recommended based on its superior performance in cotton rich composites as far as tensile properties are concerned. PVAc and Biomax performed better than PLA in cotton rich composites. PVAc / Biomax composites exhibit slight odor (undesired) due to the thermal degradation in the process. Both PVAc / Biomax have higher melting point (~200°C) compared to PLA(~170°C). PLA composites do not have that odor due to the fact that PLA has lower melting point. Further PLA is an agro based product and is based on renewable resources. As of now, since Biomax is manufactured along with commodity grade PET, it is priced low and is preferred for many applications. We have established the process of making fiber from Biomax polymer using conventional melt spinning equipment. Thus, Biomax fibers may be preferred for the lower cost compared to PLA and PVAc.

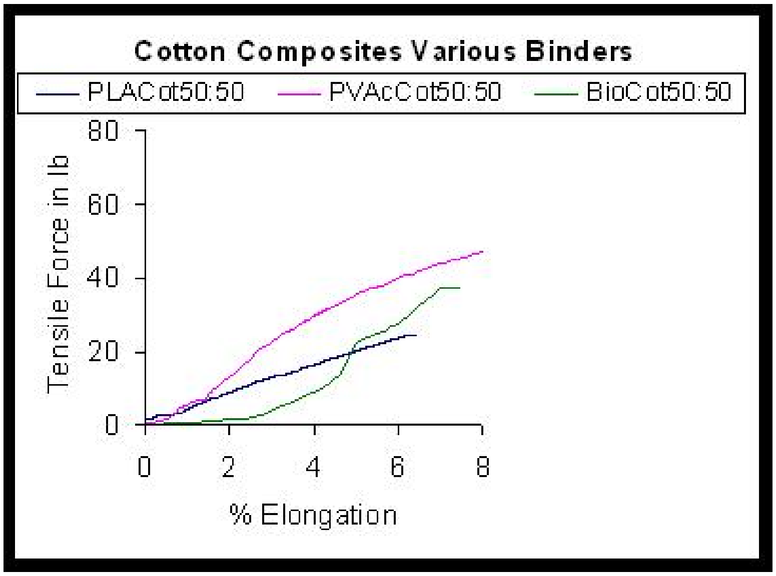

Figure 51 Comparison of Tensile Properties of Binders in Cotton composites.

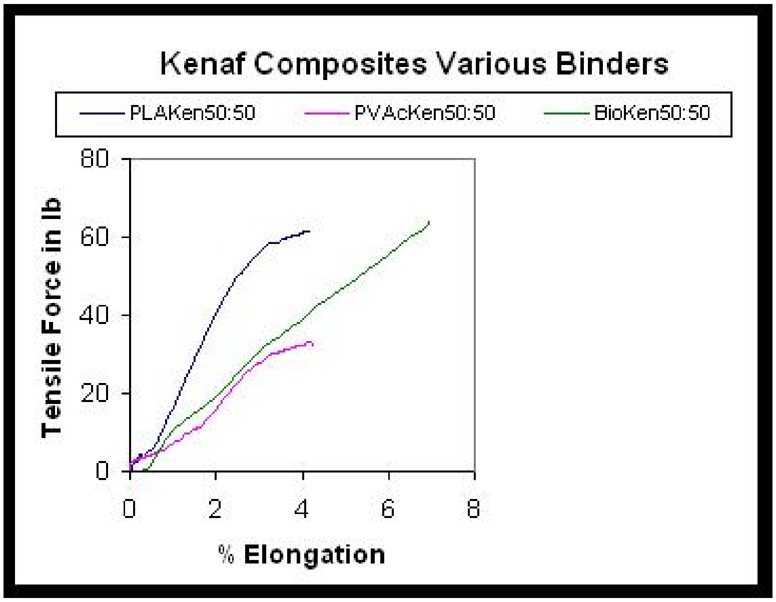

Figure 52 Comparison of Tensile Properties of Binders in Kenaf Composites.

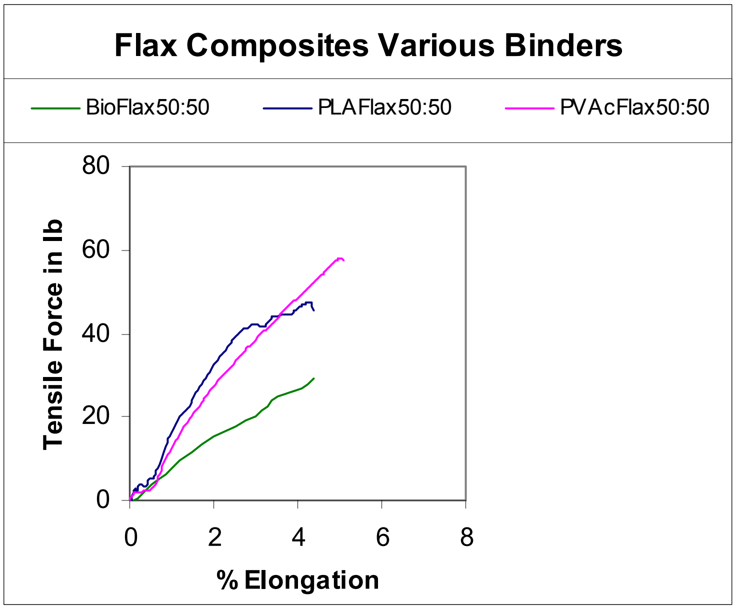

Figure 53 Comparison of Tensile Properties of Binders in Flax Composites.

Process Optimization:

At this stage of research, we acquired intimately mixed carded webs of cotton (60:40) with binders such as PLA, PLAbico, and PP (control). These webs with uniform composition are suitable for process optimization for making composites. Composites of 1200gsm basis weight were made from the intimately mixed carded webs. Comparison of the performance of the three binders was carried out based on the results of the tensile tests (as shown in Figure 54). Composites with PLA binder showed higher strength compared with PLAbico. It is interesting to note that both PLA and PLAbico binders performed better than conventional PP.

Other parameters involved in composite processing, such as curing time, temperature of the hot press, basis weight etc. were also studied in relation to the finished product quality. Results (Figure 55) indicate that to obtain maximum tensile strength the temperature for bonding is 20°C above melting point of the binder. Strength of the composites increased with the increase in basis weight (Figure 56) or curing pressure (Figure 57). Similarly, Figure 58 shows optimum-curing time of 4minutes for 400gsm webs. Morphology of the web was studied by taking SEM pictures of the samples (Figure 59) and cross-section of the composites after the tensile fracture (Figure 60).

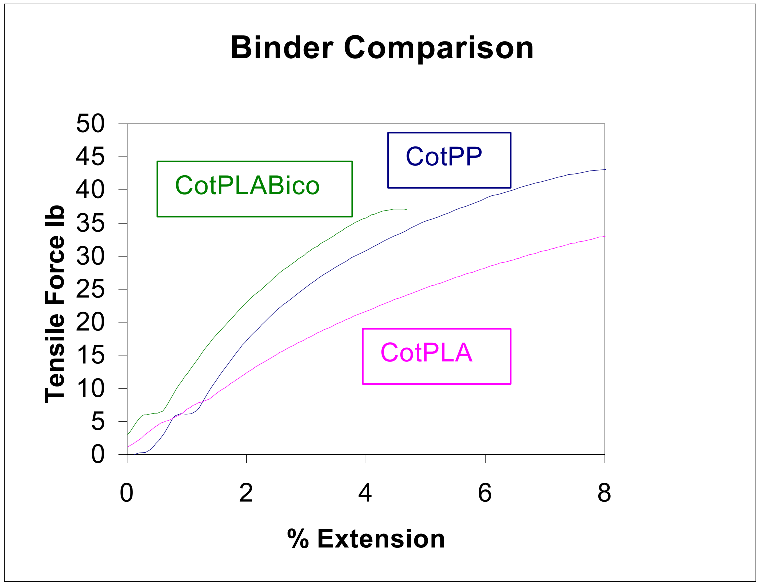

Figure 54 Comparison of various binders in intimately mixed fiber composites.

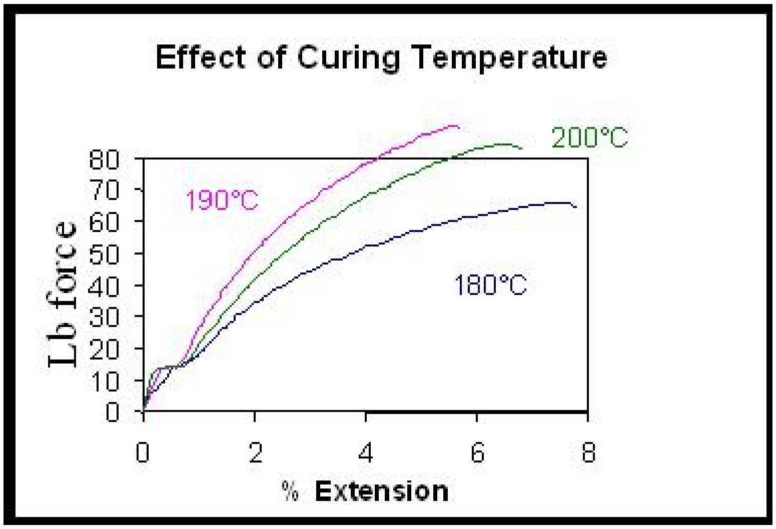

Figure 55 Optimization of Curing Temp in °C

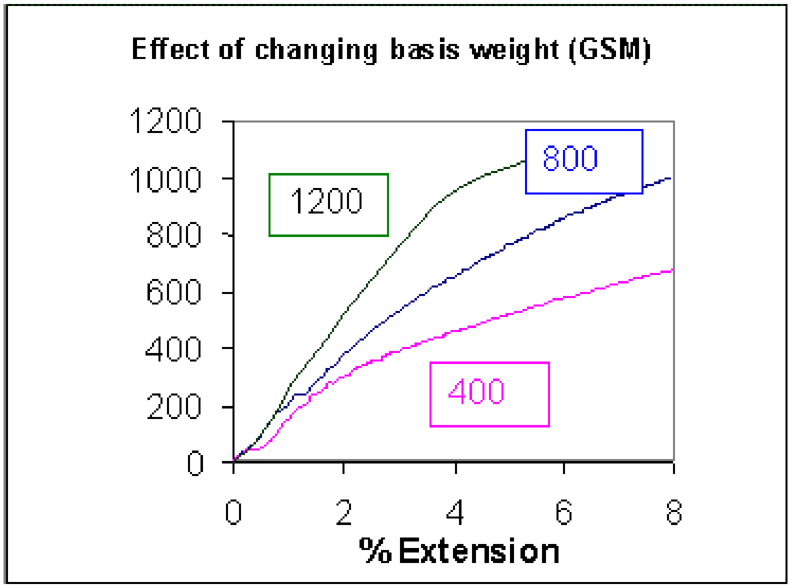

Figure 56 Effect of Basis weight or Thickness of Composite (Tensile force in PSI vs % Extension)

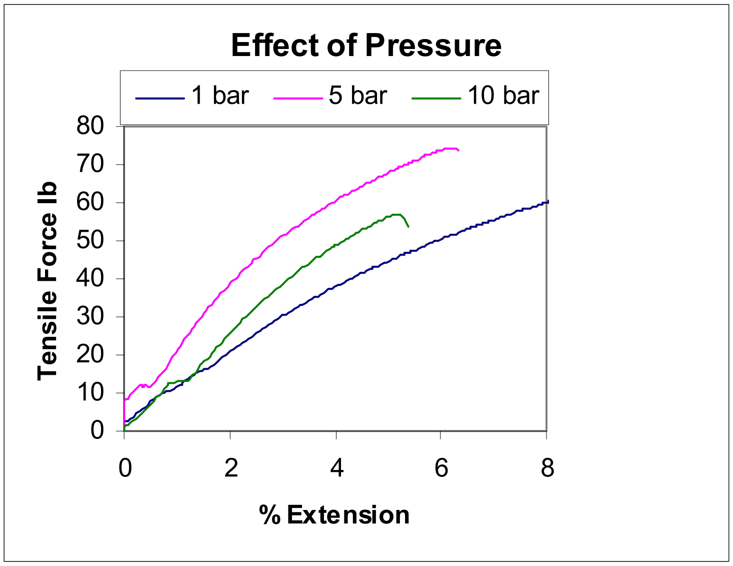

Figure 57 Optimization of Pressure

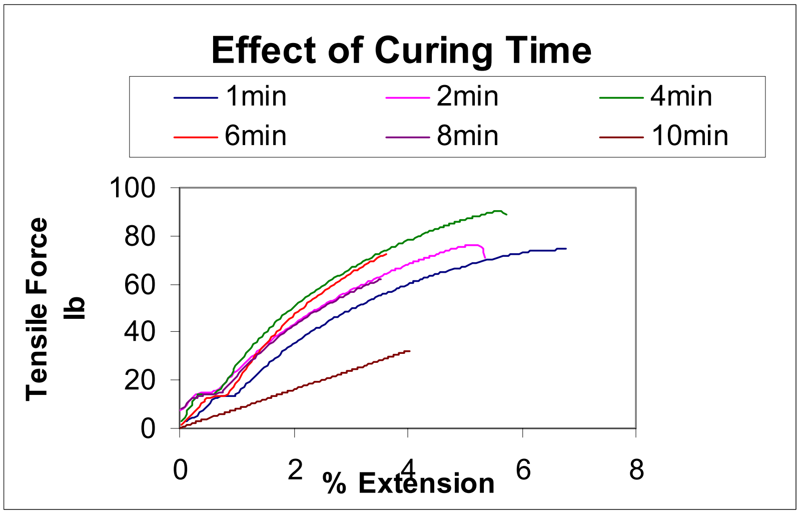

Figure 58 Optimization of curing time

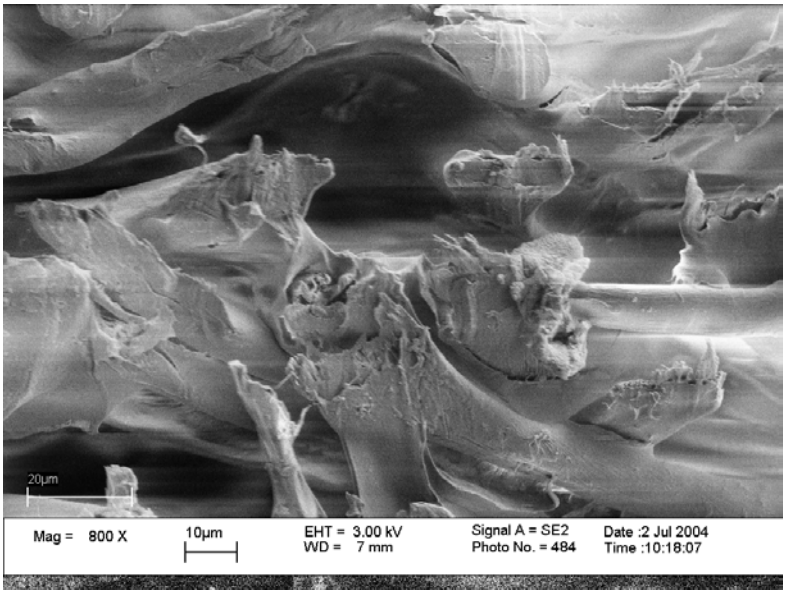

Figure 59 SEM Photo of Cotton-PLA card composite cross-section At Magnification 800X

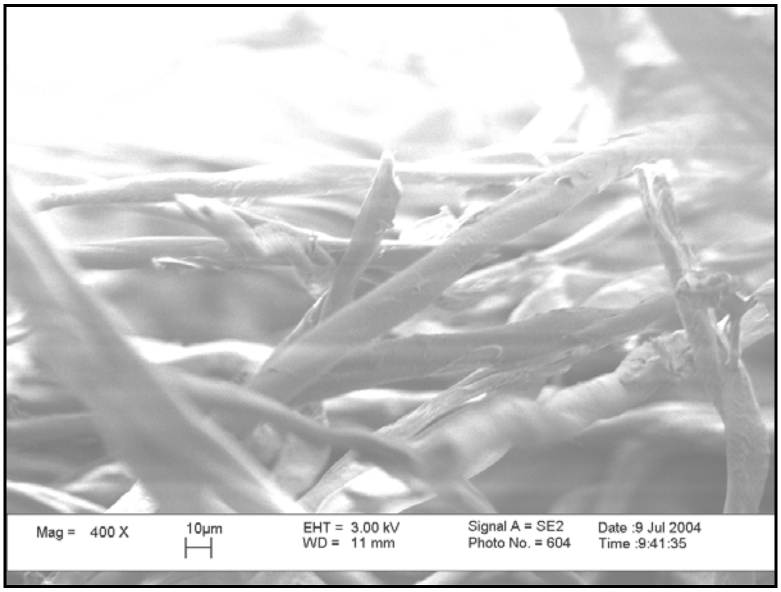

Figure 60 SEM Photo of Cotton-PLAcard composite tensile fractured. At Magnification 400X

Effect of Blending Kenaf or Flax on Tensile Properties:

Effect of blending kenaf or flax on the tensile strength and extension of intimately mixed composites of 1200 GSM (about 0.25"thick) are shown in Figure 61 and Figure 62 for PLA, and in Figure 63 and Figure 64 for PLAbico. PP binder is used for comparison (as a control). It can be seen that even at 10% level both kenaf and flax increase the tensile strength of the composites. Strength increase observed with kenaf is marginally higher than that observed with flax. This is true in case of both the binders (PLA and PLAbico). However, the effect of blending kenaf or flax on extension of the composite is marginal.

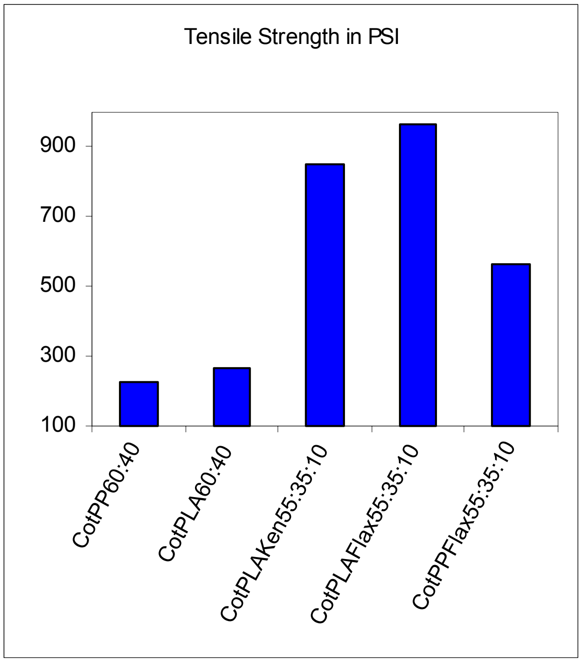

Figure 61 Tensile strength of PLA composites (Intimately mixed fiber).

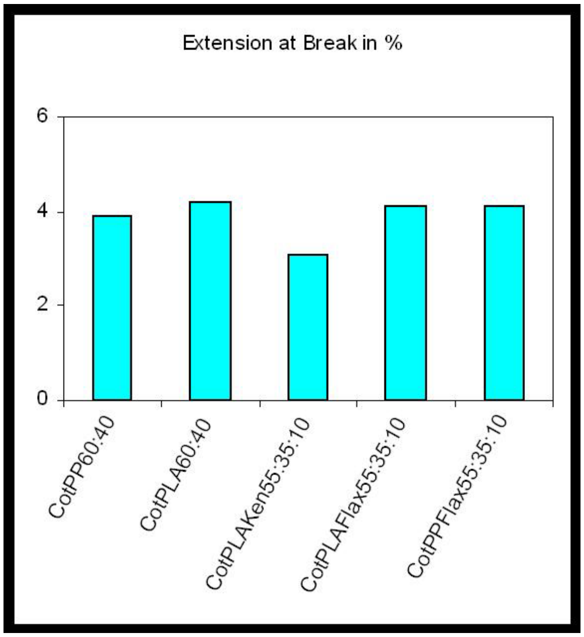

Figure 62 Extension at Break in % for PLA composites (Intimately mixed fiber).

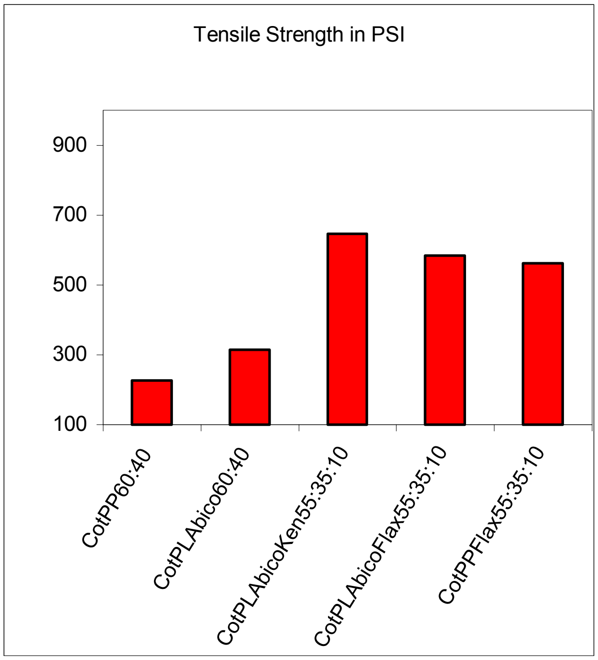

Figure 63 Tensile strength for PLAbico Composites (Intimately mixed fiber).

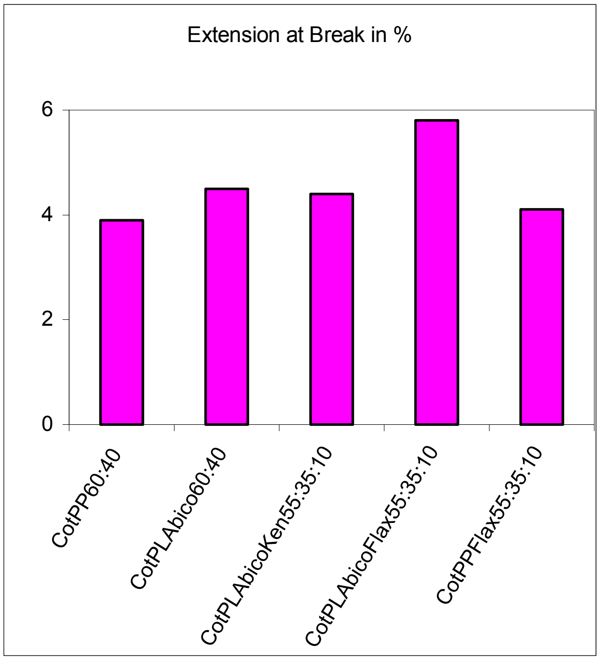

Figure 64 Extension at break in % for PLAbico Composites.

Effect of Blending Kenaf or Flax on Flexural Properties:

Flexural strength of the composites made of intimately mixed fibers is shown in the Figure 65 and Figure 66. It is the ability of the product to bend under load. In three point bending test the load is at the center when the sample is supported from the ends. It can be seen that composites with PLA are comparable with PP, whereas PLAbico binders provide more flexibility. Generally, cotton composites have low flexural strength. Kenaf and Flax add stiffness to the cotton based composites and thus lead to higher flexural strength in case of all binders. Moreover, it can be seen that kenaf provides more stiffness to the cotton composite than flax. Web of same basis weight is hot pressed at different pressures to obtain composites of varying thickness. As seen in Figure 67, Flexural strength increases with increase in consolidation that reduces the thickness of the composite.

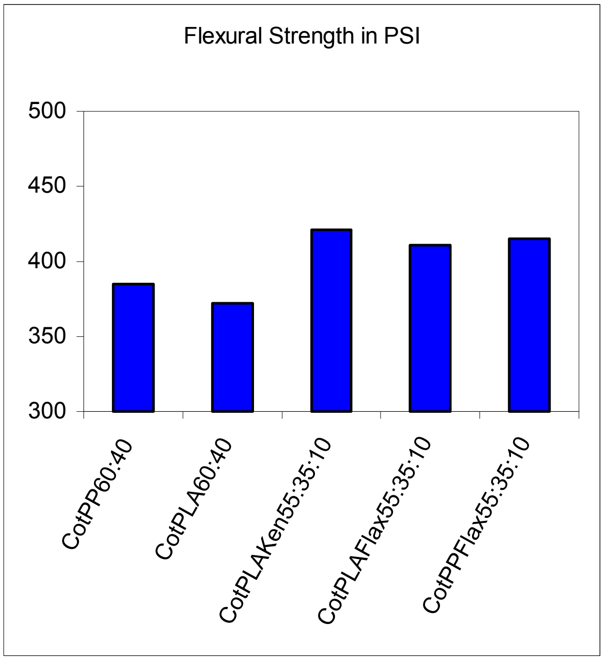

Figure 65 Flexural Strength of PLA Composites (Intimately mixed fiber).

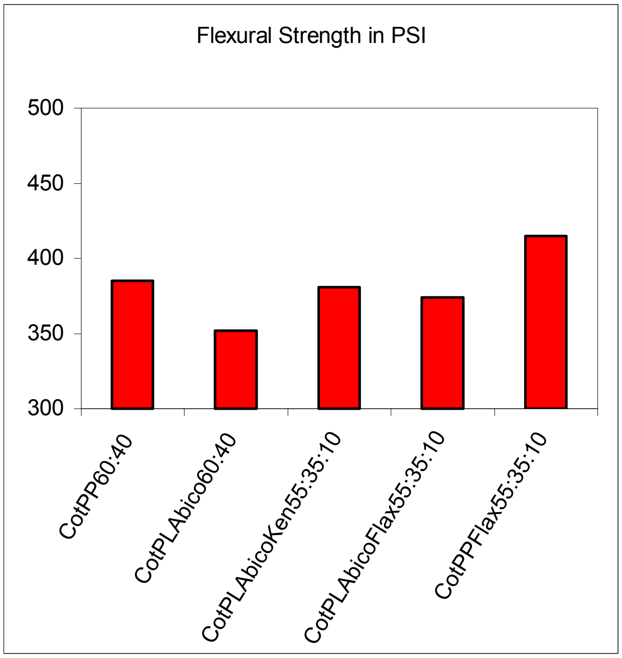

Figure 66 Flexural Strength of PLAbico composites (Intimately mixed fiber).

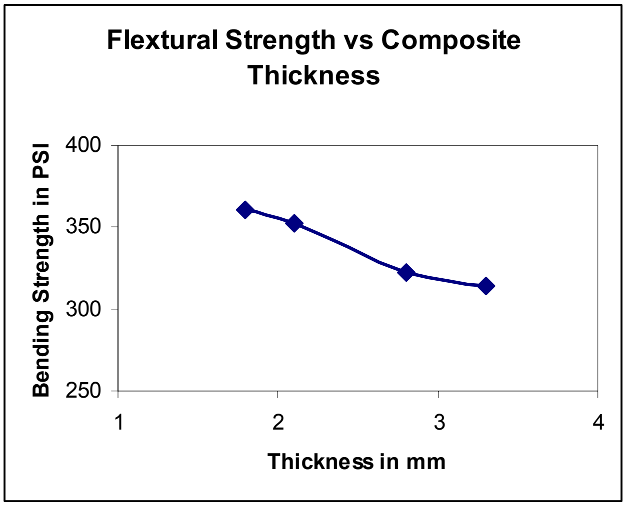

Figure 67 Effect of Composite Thickness on Flexural Strength (CottonPLAbico).

Effect of Blending Kenaf or Flax on Acoustic Properties:

Acoustic properties of the composites made of intimately mixed fibers are shown in the

Figure 68 through Figure 80. In four-point impedance tube test method, the sound waves hit and go through the sample placed at the center. Acoustic properties are known to depend on fiber laying pattern and extent of consolidation [43] within the product. The results have been interpreted based on the assumption that fiber lay is random and uniform in all samples.

Adsorption coefficient:

Experimental results (Figure 68) showed that PP fiber based cotton composites absorb sound better than PLA fibers. Whereas, by blending flax (~10%) both PLA and PLA bico performance is improved and is better than that of PP (Figure 69). In majority of the samples, blending kenaf and flax has shown improved acoustic properties (Figure 69, Figure 70, and Figure 71). Absorption increases as the frequency goes higher. This is possibly due to the inherent nature of these natural fibers and due to the surface and pores in their structure. By reversing the face of the sample, there is no substantial change in the acoustic performance (Figure 71 vs Figure 72), as the properties do not depend on direction in a random mixture.

Reflection coefficient:

Experimental results showed that PP fiber based cotton composites reflect sound better than PLA fibers (Figure 73). Blending flax has shown (Figure 74) to improve reflection coefficient of cotton/PLA composites, whereas change was marginal in case of kenaf.

Absorption Coefficient:

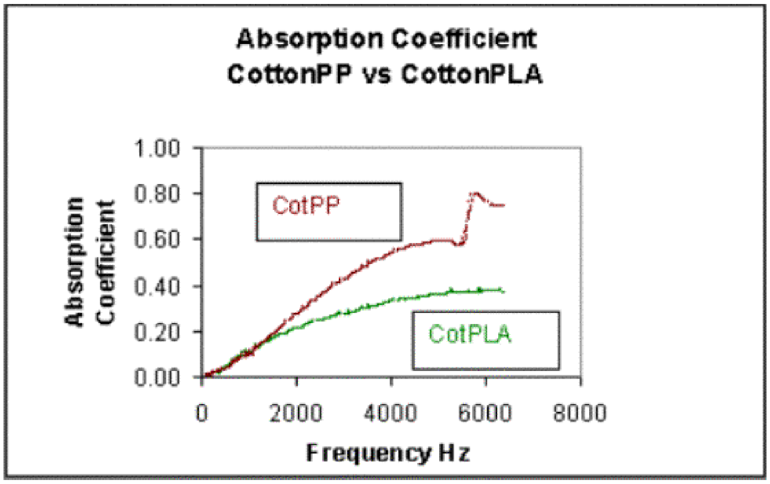

Figure 68 Absorption coefficient of CottonPP vs CottonPLA Composites.

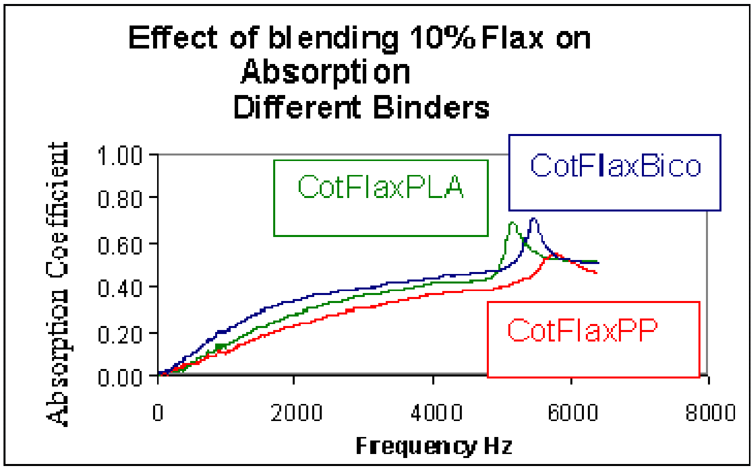

Figure 69 Effect of Flax on Absorption Coefficient of Cotton Composites

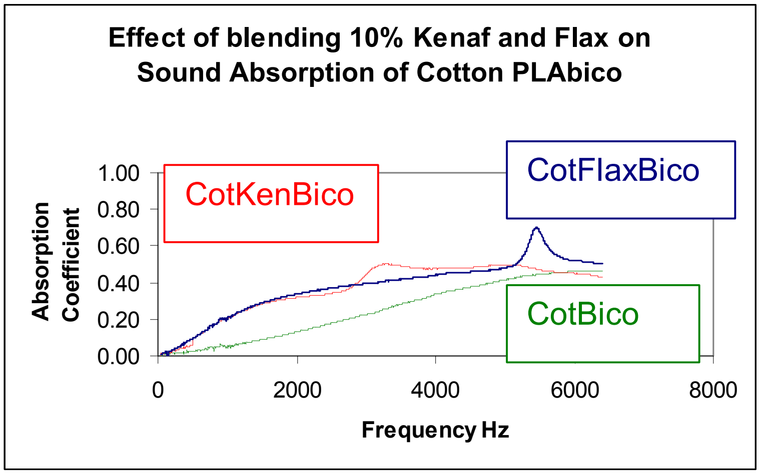

Figure 70 Effect of Blending Kenaf and Flax on Sound Absorption of Cotton PLAbico Composites.

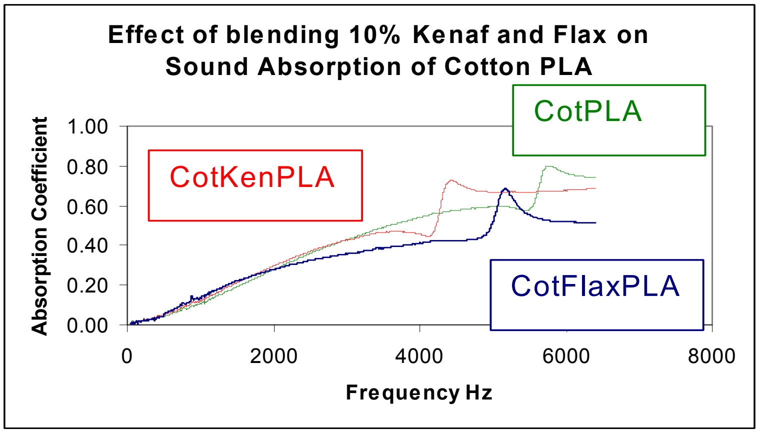

Figure 71 Effect of Blending Kenaf and Flax on Sound Absorption of Cotton PLA Composites

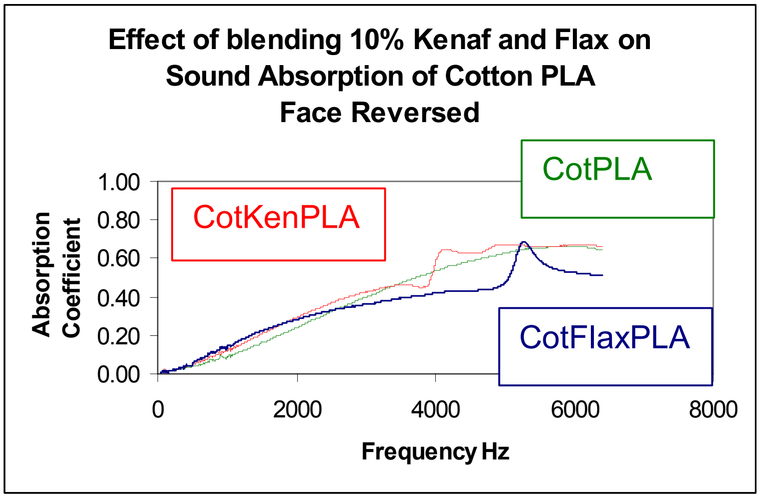

Figure 72 Effect of Blending Kenaf and Flax on Sound Absorption of CottonPLA Composites. (Face Reversed)

Reflection Coefficient:

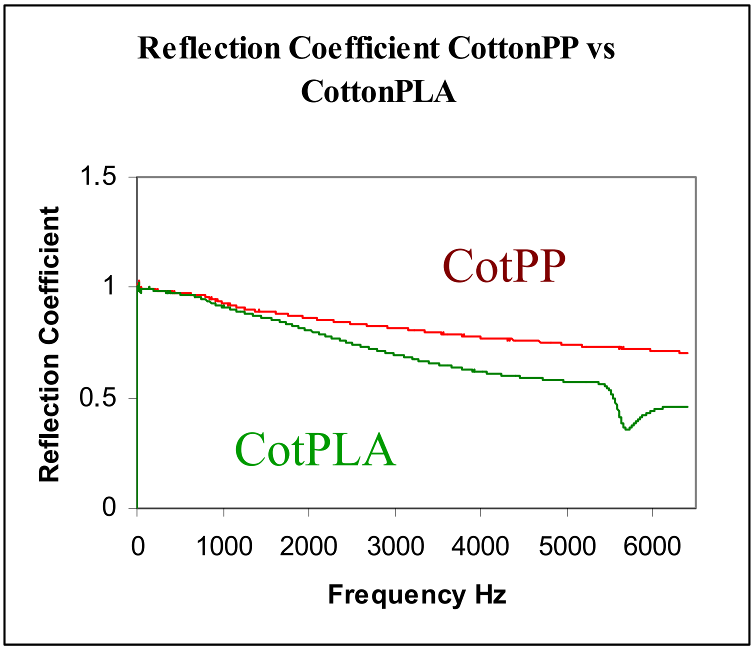

Figure 73 Reflection Coefficient of CottonPP vs Cotton PLA Composites.

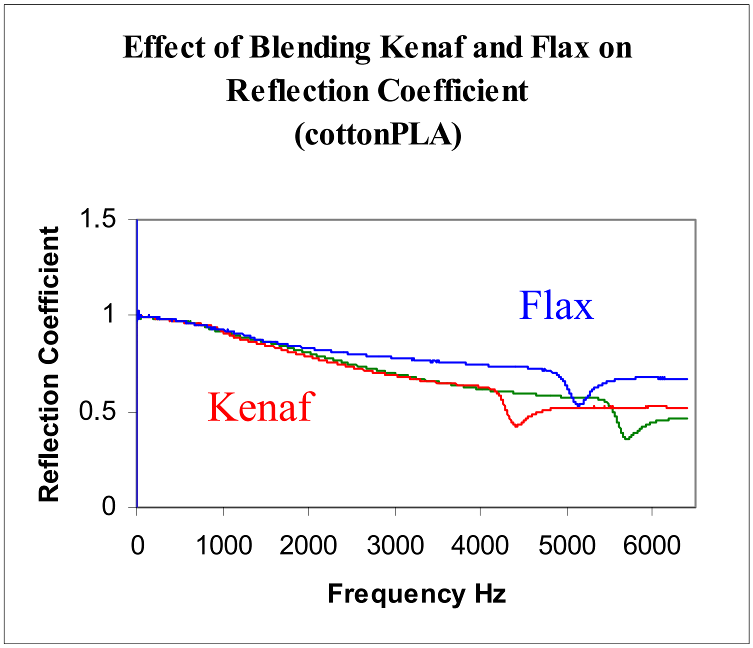

Figure 74 Effect of Blending Kenaf and Flax on Reflection Coefficient.

Admittance Ratio:

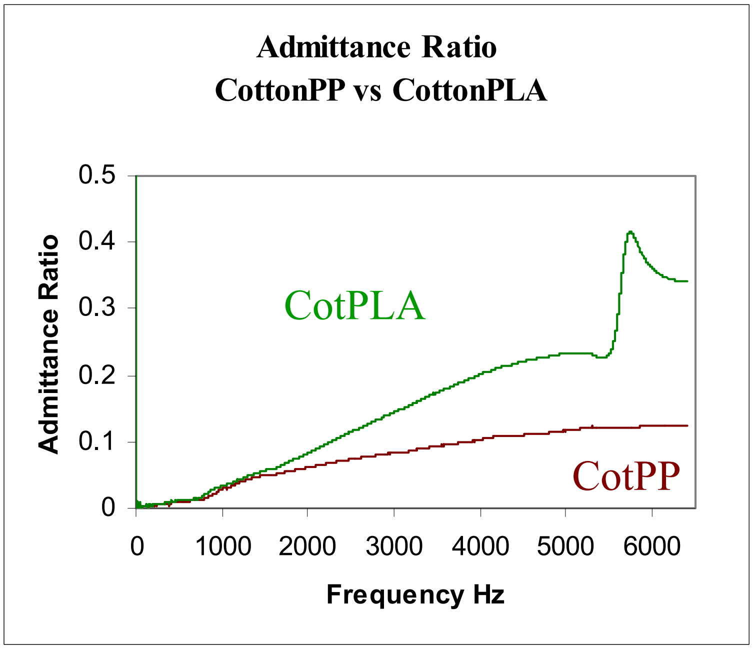

Figure 75 Admittance Ratio of CottonPP vs CottonPLA Composites.

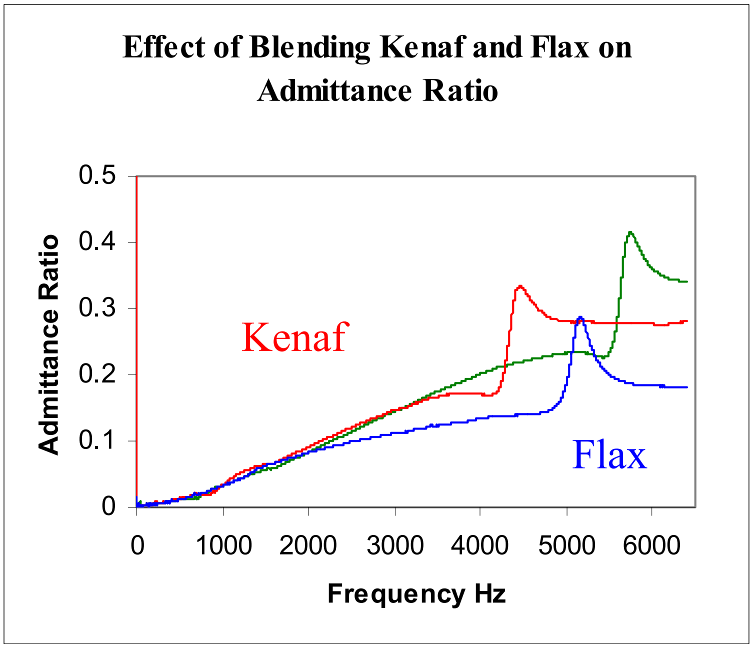

Figure 76 Effect of Blending Kenaf and Flax on Admittance Ratio of CottonPLA Composites.

Impedance Ratio:

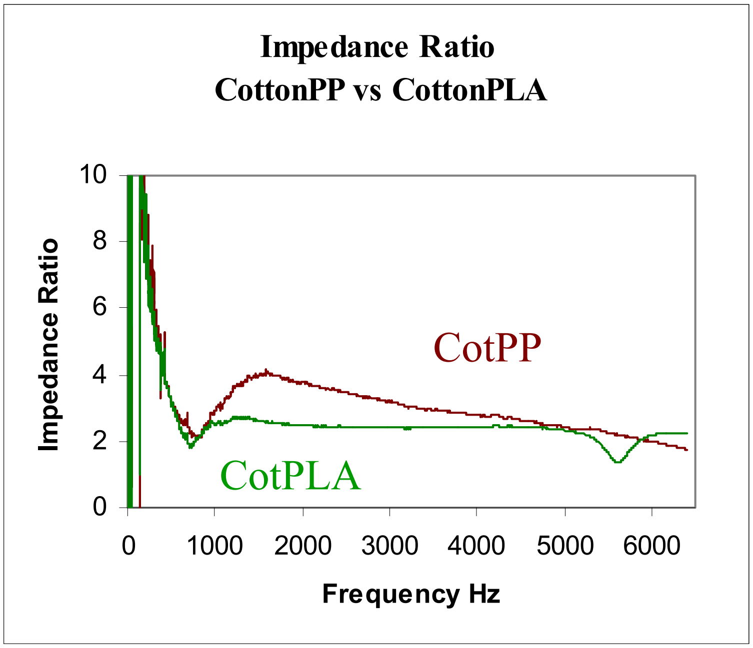

Figure 77 Impedance Ratio of CottonPP vs CottonPLA Composites.

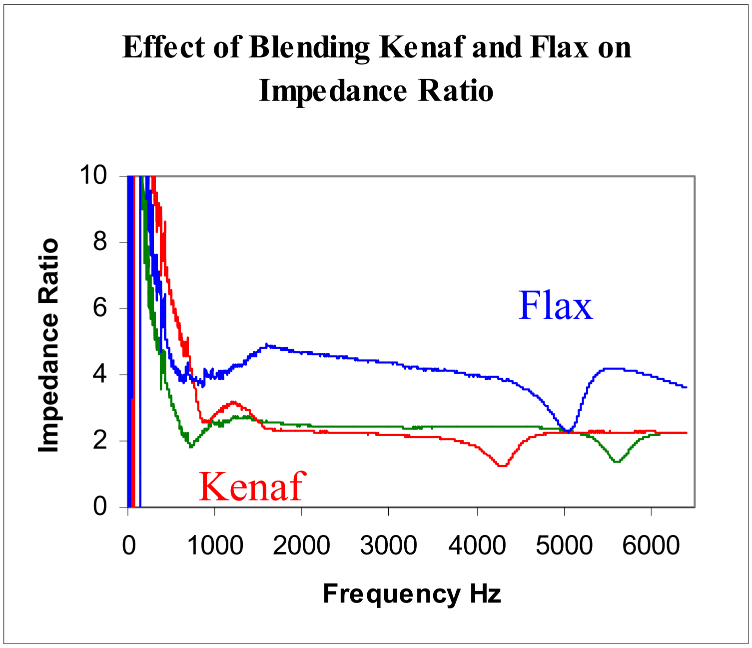

Figure 78 Effect of Blending Kenaf and Flax on Impedance Ratio of Cotton PLA Composites.

Transmission Loss (dB):

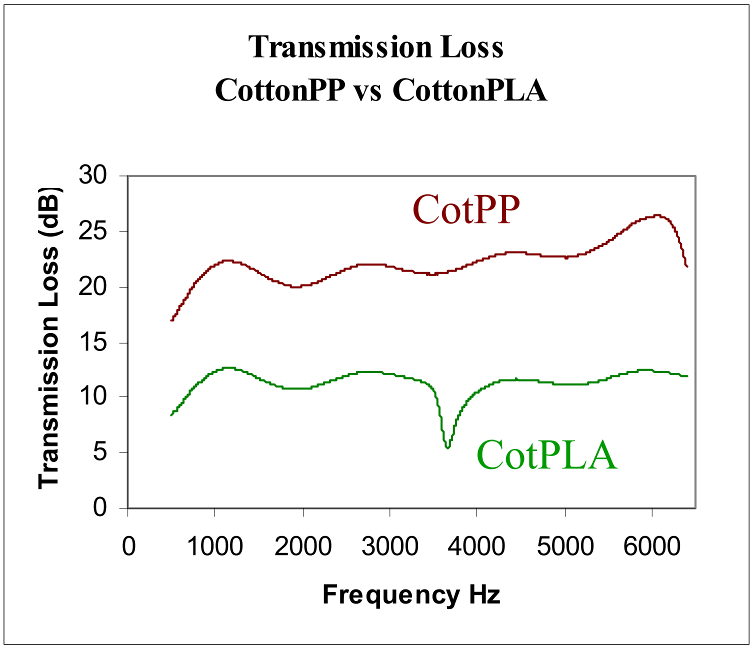

Figure 79 Transmission Loss CottonPP vs CottonPLA Composites.

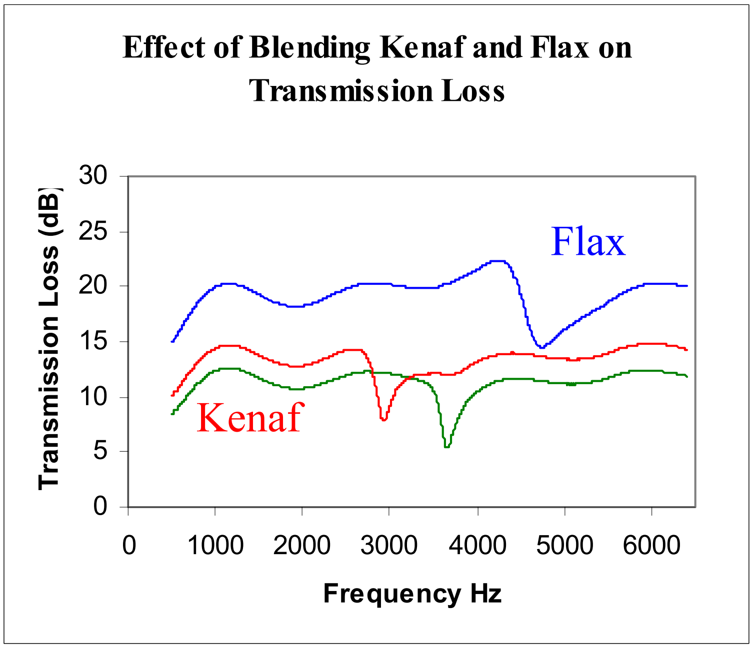

Figure 80 Effect of Blending Kenaf and Flax on Transmission Loss of CottonPLA Composites.

Admittance Ratio:

Experimental results showed that PP fiber based cotton composites has lower admittance ratio compared to PLA fibers (Figure 75). Blending flax or kenaf (Figure 76) has shown to reduce admittance ration of cottonPLA composites, whereas change was marginal in case of kenaf.

Impedance Ratio:

Experimental results showed that PP fiber based cotton composites have higher impedance ratio compared to PLA fibers (Figure 77). Blending flax or kenaf (Figure 78) has shown to increase impedance ratio of cotton PLA composites at lower frequencies upto 1000. At higher frequencies flax continue to exhibit higher impedance ratio, whereas change was marginal in case of kenaf.

Transmission Loss (dB):

Experimental results showed that PP fiber based cotton composites have higher transmission loss compared to PLA fibers (Figure 79). Blending flax or kenaf (Figure 80) has shown to increase transmission loss of cottonPLA composites.

Effect of Blending Kenaf or Flax on Impact Properties:

Acoustic properties tested four-point impedance tube test method is known to depend on fiber laying pattern and extent of consolidation within the product. Expecting random and uniform fiber lay in all the samples the results show that PP fiber based cotton composites absorb sound better than PLA fibers. As seen from Figure 81 and Figure 82, impact strength of PLA and PLAbico binders is higher than that of PP. Moreover blending kenaf or flax (~10%) increases the impact strength of the composites

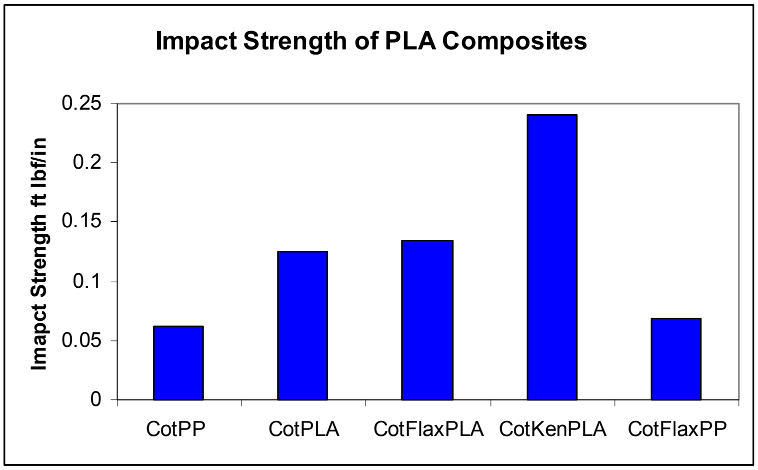

Figure 81 Impact Strength of CottonPLA Composites & Effect of Blending Kenaf and Flax.

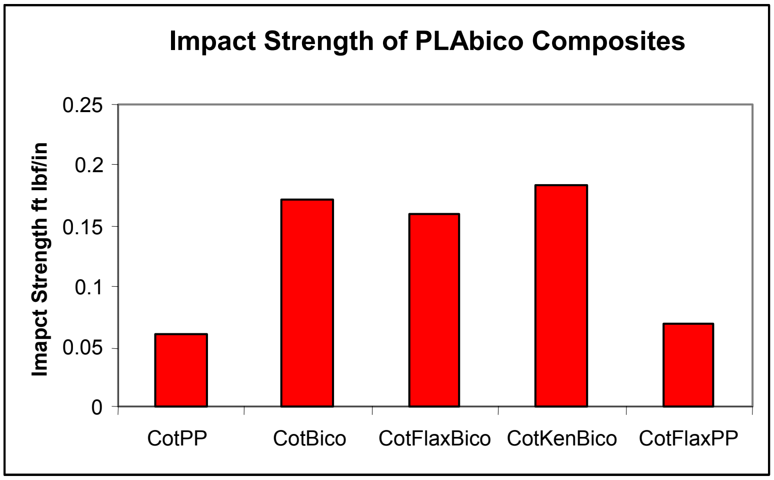

Figure 82 Impact strength of CottonPLAbico Composites & Effect of Blending Kenaf and Flax.

substantially. Impact strength increases as the composite thickness is raised keeping same basis weight (Figure 83).

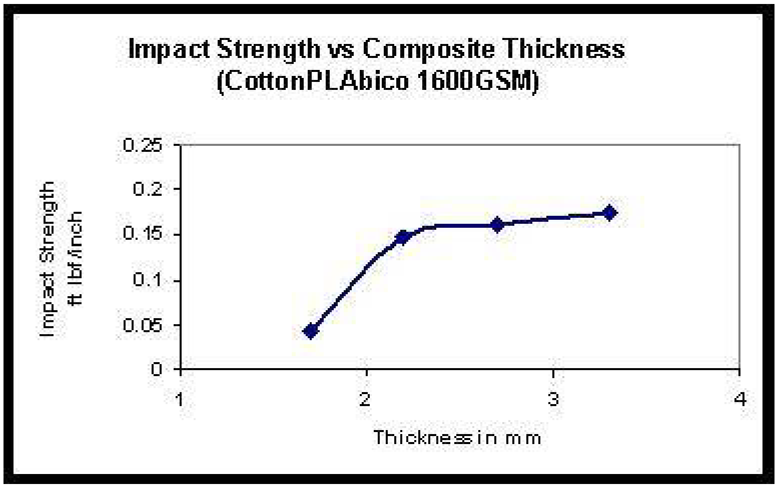

Figure 83 Impact Strength vs Composite Thickness. (CottonPLAbico at 1600GSM)

V. CONCLUSIONS

1. Initial studies on structure and properties of fibers showed the ability of these natural cellulosic fibers like Cotton, Kenaf, and Flax to form a good bond between thermoplastic polymer such as Eastar, Biomax, and Cellulose acetate. Preliminary results of fiber bonding studies done on a Dynasco heat sealer further reinforced this observation.

2. Comparison of sandwich type composites with fiber mix type composites proved that the bonding between cotton or natural fibers and the binder polymer is better when composites are made from mixed-fiber carded webs. Further, intimate blending of the binder fibers with the natural fibers is the key to making a composite with good properties. Carding produced more uniform webs and composites resulting in improved in tensile properties.

3. Biodegradable binder fibers can be produced by melt spinning of polymers such as Biomax and PLA. Such fibers have good physical properties and processability. It is possible to make a dry laid web using natural fibers such as cotton, flax, and kenaf; and binder fiber by air laying. These dry laid webs can be thermally bonded using the hot press to form composites.

4. Flax and kenaf provide substantial increase in tensile strength in the blend of Biomax and cotton. There is strong bonding of the binder fibers with the natural cellulosic fibers in the composites. The melting and flow of the binder fiber over the

cellulosic fibers takes place and appears to form good bond. The observation is true with most of the biodegradable binder fibers investigated in this study.

5. Comparison of Binders -Biomax, PLA, and PVAc with Natural Fibers in 50/50 composition indicated that:

(a) In Binder:Cotton composites, PVAc provide more tensile strength and elongation to the composite than PLA. Biomax performs very close to that of PVAc. Biomax and PVAc better composites with cotton than PLA.

(b) In Binder:Kenaf composites, PLA provides more tensile strength and elongation to the composite than PVAc. Biomax performance is comparable to that of PLA. Biomax and PLA form better composites with kenaf than PVAc.

(c) In Binder:Flax composites, PVAc provides more tensile strength and elongation to the composite than Biomax. PLA performs very close to PVAc. PLA and PVAc form better composites with flax than Biomax.

6. PVAc is recommended based on its superior performance in composites having greater cotton content. Although PVAc and Biomax performed better in cotton-rich composites, since both have higher melting point (~200°C), it causes degradation during consolidation and that generates slight odor in the composites. Considering this aspect, PLA is recommended since its melting temperature of 170°C and composite has fewer odors. Moreover, PLA performance is closer to that of conventional binder PP. Biomax is recommended due to its lower cost compared to both PLA and PVAc.

7. Process optimization results obtained from the composites made of intimately mixed carded web of cotton (60:40) with binders such as PLA, PLAbico, and PP (control) in relation to the finished product quality suggested that:

(a) Optimum temperature for bonding in a hot press is 20°C above melting point of the binder.

(b) Optimum-curing time is about four minutes for 400 gsm web.

(c) Composites strength increases with the increase in curing pressure or basis weight/ thickness.

(d) Cotton composites (1200GSM) with PLA binder showed higher strength compared with PLAbico. It is interesting to note that both PLA and PLAbico binders performed better than conventional PP.

(e) The increase in tensile strength by the addition of kenaf or flax (at 10% level) is substantial. This is true in case of all binders. However there is a marginal drop in elongation.

8. The three point bending test showed that PLA and PLABico based cotton composites have slightly lower flexural strength compared to conventional PP. Flexural strength increases with increase in consolidation that reduces the thickness of the composite. Adding 10% kenaf or flax increases flexural strength substantially. This shows kenaf and flax work like stiffeners. As the thickness increases flexural strength decreases (if basis weight is maintained same). In other words, flexural strength increases due to consolidation.

9. Acoustic properties tested four-point impedance tube test method are known to depend on fiber laying pattern and extent of consolidation within the product. Conclusions are drawn based on the assumption that random and uniform fiber lay in all the samples.

(a) Sound absorption: results show that PP fiber based cotton composites absorb sound better than PLA fibers. Whereas, blending flax (~10%), performance of both PLA and PLA bico is superior to PP. In majority of the samples, blending with kenaf and flax has shown improved acoustic properties. Absorption increases as the frequency goes higher. By reversing the face of the sample, there is no substantial change in the acoustic performance, as the properties do not depend on direction in a random mixture.

(b) Sound reflection: PP fiber based cotton composites reflect sound better than PLA fibers. Blending flax has shown to improve reflection coefficient of cottonPLA composites, whereas change was marginal in case of kenaf.

(c) Sound admittance & impedance: PP fiber based cotton composites has lower admittance ratio compared to PLA fibers. Blending flax or kenaf has shown to reduce admittance ration of cottonPLA composites, whereas change was marginal in case of kenaf. PP fiber based cotton composites has higher impedance ratio compared to PLA fibers. Blending flax or kenaf has shown to increase impedance ratio of cottonPLA composites at lower frequencies upto 1000. At higher frequencies flax continue to exhibit higher impedance ratio, whereas change was marginal in case of kenaf.

(d) Sound transmittance loss (dB): PP fiber based cotton composites have higher transmission loss compared to PLA fibers. Blending flax or kenaf has shown to increase transmission loss of cottonPLA composites

10) Notched Izod impact test results showed that the impact strength of PLA and PLAbico binders is higher than that of PP. Moreover blending kenaf or flax (~10%) increases the impact strength of the composites substantially. Impact strength increases as the composite thickness is raised keeping same basis weight.

Finally, on the basis of these studies, it is expected that viable composite parts containing cotton and other natural fibers can be produced with a thermoplastic binder fiber, that are biodegradable and possess the required properties that are comparable to the traditional polypropylene based composites. Such composites are suitable for automotive and many other semi-structural applications.

VI. FUTURE DIRECTIONS

In order to commercialize these composites for automotive and other applications it is suggested to conduct further research work in the following directions:

1. Influence of fiber lay, consolidation & composite composition on acoustic properties.
2. Process optimization to reduce odor of the composites by processing at lower temperature (by using binders such as PLA having lower M.Pt.170°C).
3. Study the effect of Needle punching of webs before thermal bonding.
4. Use of other air lay systems such as Randowebber to make composites.
5. Use of cheaper raw materials such as cotton waste, carpet waste for composites.
6. Development of flame retardant composites.

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APPENDIX

A REPORT ON MELT SPINNING OF BIOMAX & PLA

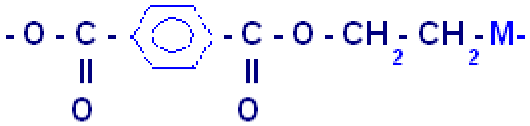
Abstract:

Recently research activities in the field of biodegradable products are growing fast as environmental activists are demanding the use of biodegradable products that can be disposed of in an environmentally friendly way at the end of their useful life. Typical products of interest, Biomax and PLA fibers are discussed in this section. Biomax® and PLA have been developed such that they can be recycled, incinerated or sent to landfill for composting. Biomax has been used in single use products such as bowls, plates, spoons, forks, and wraps for sandwich-containing Biomax is presently seen in the market. It is possible to produce Biomax and PLA fibers by melt spinning. To reduce degradation proper drying of polymer granules before spinning is necessary. Further, there is some optimum spinning melt temperature where the spinning performance is acceptable and degradation is lower. To produce stronger fibers it is necessary to go for higher spinning speed and or draw the as-spun fibers. The fibers thus produced are suitable as binder fibers for making nonwoven fiber based composites.

INTRODUCTION

Environmentalists are increasingly concerned about disposability of the products at the end of the useful life. If products are not reusable or recyclable, they need to be biodegradable/compostable, so that they can be disposed of in an environmentally friendly way. In a community, a portion of the useful land is dedicated for the purpose of landfill. One of the major loads to the landfill is the trash produced by the population, which is of the tune of 7000 lb per year per family. By recycling, a preferred way, load can be reduced to a certain extent. Furthermore, if the remaining portion is biodegradable there is no need for a landfill. One way to achieve this is by making use of biodegradable polymers to make plastic cups, forks, spoons, snack bags and gum wrappers that are used in everyday life. DuPont scientists have researched and created such a polymer that decomposes in compost and supports plant life in the soil or do not harm the environment. They have overcome the cost and performance barriers to have a breakthrough in consumer applications.

Polyester, polyethylene terephthalate (PET), is a well-known economically and commercially available product. Biomax® is a modified hydro/biodegradable polyester. Proprietary monomers are incorporated into the polymer, creating sites that are susceptible to hydrolysis. At elevated temperatures, the large polymer molecules are cleaved by moisture into smaller molecules, which are then consumed by naturally occurring microbes and converted to carbon dioxide, water and biomass.



Biomax Formula with M representing proprietary monomer part

Biomax® has been designed such that it can be recycled, incinerated or sent to landfill for composting. Several tests have shown that it is friendly to the environment, promotes growth of plants, earthworms and microbes in the composting soil. This biodegradable polymer is readily available at present.

Biomax® polymer can be used to make injection-molded parts, coatings for paper, thermoformed cups and trays, and films. With its diverse product properties, it is suitable for film applications, thermoformed packaging and injection-molded parts. In addition, it is versatile, very good for a variety of single-use products such as disposable biodegradable plates, bowls and sandwich wraps.

Initially Biomax was chosen as a biodegradable binder fiber for composites since it was available in commercial quantities. It is readily available in the market and is cost competitive too. Biomax has properties of low melting polyester and it can be processed in the same equipments that are used for PET by operating at a lower temperature. Dupont supplied Biomax polymer since they do not make fiber. Biomax has been used to make injection-molded parts, coatings for paper, thermoformed cups and trays, and films where it has exhibited superior barrier properties. A single use product such as bowls, plates, spoons, forks, and wraps for sandwich-containing Biomax is presently seen in the market. Since Biomax is a derivative of fiber grade polyester, an attempt has been made to produce biodegradable binder fiber starting from Biomax polymer at Foss Manufacturing Company, Hampton, NH. The fiber thus produced was used as binder fiber in making biodegradable nonwovens or composites. More quantity of Biomax fiber was produced in house using the Fourne Melt Spinning equipment. Similarly, PLA fibers were produced in the same set up by melt spinning of PLA polymer.

Polylactic acid (PLA) is another biodegradable fiber that is produced from the cornstarch. PLA fiber has a melting temperature of 171°C and tensile properties comparable to that of polyester fibers. As a melt spinnable fiber with a vegetable source, PLA has many of the advantages of both synthetic and natural fibers. Beyond having a renewable raw material, it possesses biodegradability. However, it has poor abrasion resistance and bonding behavior compared with conventional binders. However, PLA is not yet made available in commercial quantities in the open market.

EXPERIMENTS

Dupont supplied the commercial grade Biomax polymer for all our experiments. Polymer properties such as density, intrinsic viscosity (I.V.), DSC, and rheology data were obtained. I.V was measured using a 60/40 (W/W) mixture of Phenol/1,1,2,2 - Tetrachloroethane Polymer Characterization solvent. Samples were prepared in a 1% solution and analyzed at 25^{o} C.

Polymer drying was carried out in the laboratory oven. Preliminary spinning conditions were obtained based on the melt indexer and DSC results. Further, spinning was carried out at Foss Manufacturing as well as in house using Fourne melt spinning equipment. Fincor drawing equipments were used to draw as spun fiber. Both asspun fiber and drawn fiber were tested for tensile properties, denier, and observed under optical microscope.

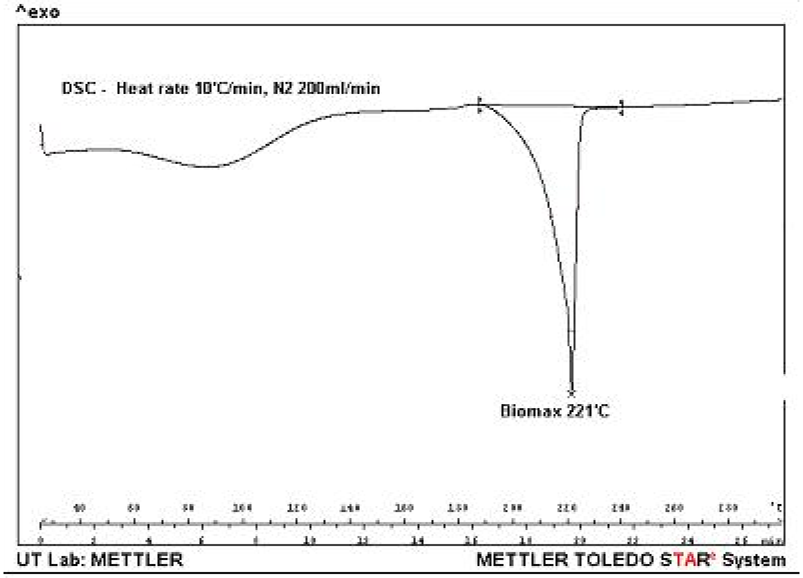


Figure 84 DSC of Biomax Chips

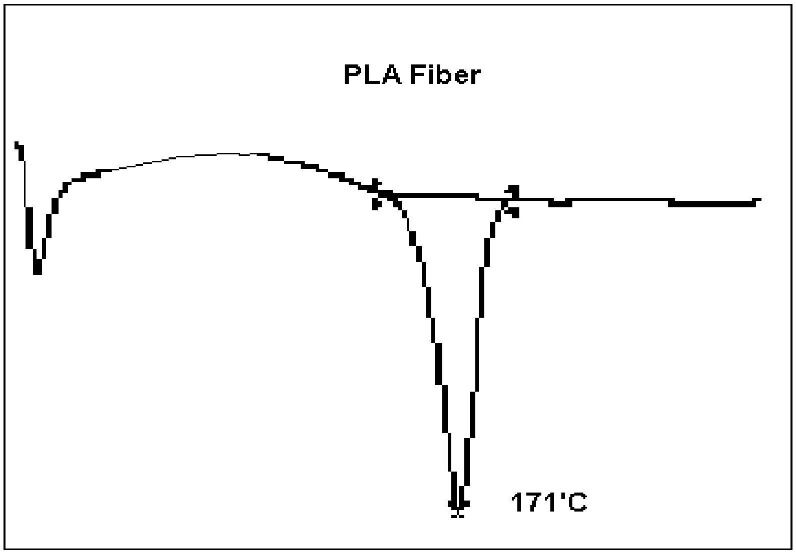


Figure 85 DSC picture of PLA fiber

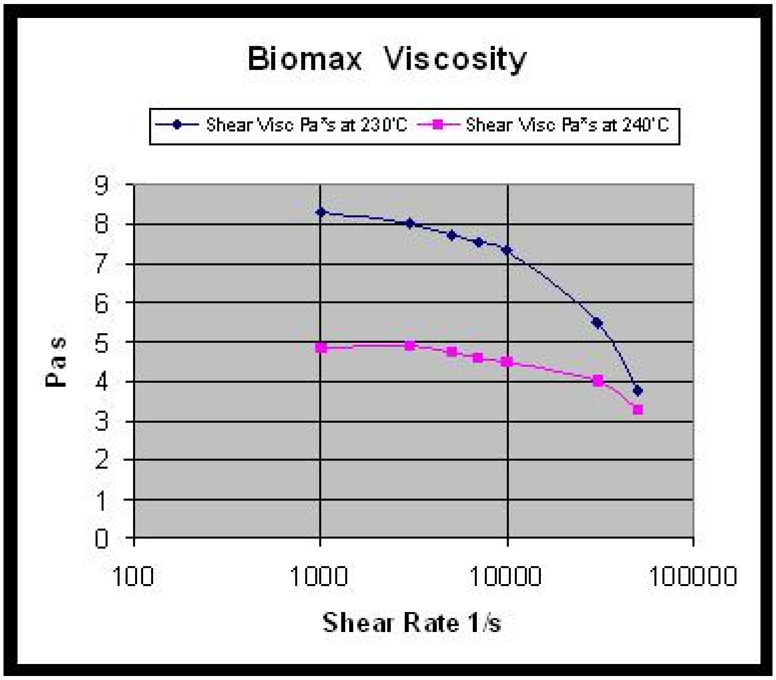


Figure 86 Biomax polymer shear viscosity

RESULTS AND DISCUSSIONS

Biomax DSC data (Figure 84) indicates onset of melting at 190°C and peak at 202°C where as PLA melting peak is at 171°C (Figure 85). This shows melt spinning can be carried out at about 230 to 240°C. Melt viscosity data for Biomax is shown in Figure 86. The polymer rheology shows shear thinning behavior like other polyesters, and a strong dependence on temperature. The viscosity drops with increasing shear and temperature. Moreover, preliminary experimentation with melt flow indexer indicated that at 225 ^{o}C, the polymer melt flows smoothly to form fibers. In addition, the viscosity data suggests that at comparable temperatures, the polymer is likely to melt and flow better during the molding process.

Biomax fiber was successfully produced by melt spinning the polymer obtained from Dupont. Initially polymer was dried at 100°C and under vacuum for 6 hours to remove the moisture and spun without any delay to avoid moisture regain. Spinning process conditions and the quality of the products are shown in Table 8. It can be seen the higher the speeds, finer the filament, and the strength increase. Drawing can reduce denier and elongation of the as spun fiber.

Table 8 Melt Spinning Biomax & PLA.

SPINNING PROCESS CONDITIONS

		Biomax			PLA
	Unit	Foss	UTK-Fourne Trial -1	UTK-Fourne Trial -2	UTK-Fourne Trial -1
Number of Spinneret Holes		500	1	12	12
Melt Temperature	deg C	240	240	240	208
Melt Pressure	psi	~1000	~1000	~1000	~1000
Melt Output per hole	g/min	0.3	1.9	0.42	0.42
Take up winding speed	m/min	175	450	615	1000
AS SPUN FIBER QUALITY					
As spun Denier		16	24	6	4
Elongation (peak)	%	747	358	164	358
Tenacity	g/den	0.2	0.24	0.83	2.2

As Spun yarn from Foss spinning was drawn at 2.5 draw ratio, crimped in the steam heated stuffer box, and was used in composite samples. Drawn crimped tow was cut to about 1 inch staple lengths and used as binder fiber. Later in order to optimize the process the same as spun tow was drawn at UT facility with preheating and two-stage drawing to obtain finer denier fibers. Properties of the As Spun filament and final fiber are given in Table 9. Preheating the as spun Biomax fiber to about 65°C helps in drawing.

Table 9 Biomax As Spun & Drawn Fiber Properties.

ID	Denier per	Break Force	Break Elong	Peak Force	Peak Elong	Remarks
	Filament	lb	%	lb	%	
AsSpun Tow*	15.9	0.76	747	3.83	726	
Drawn& Crimped *	7.7	1.16	87	5.86	35.5	poor run
DS1**	1.8	0.97	55	5	15	poor run
DS2**	4	2.5	121	12	81.8	better run
Note:						
* Produced at Foss Manufacturing Pilot facility.						
** DS1 and DS2 are fiber after the 2 stage draw at UT facility after preheat AsSpun tow to 65°C						

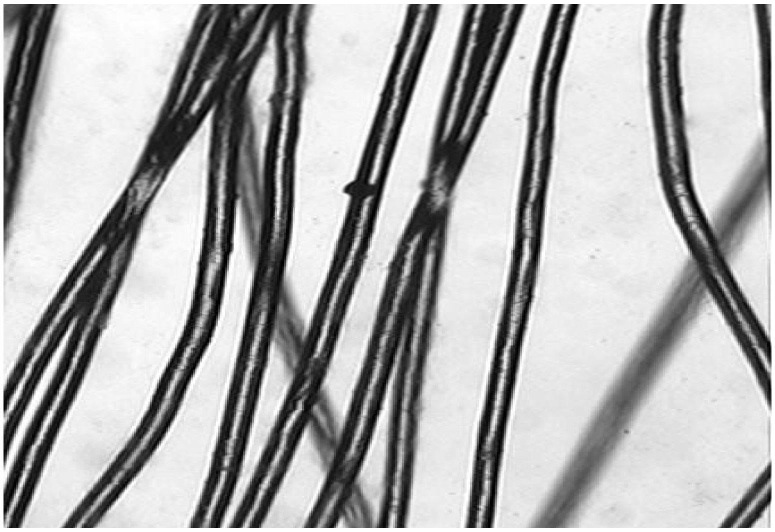


Figure 87 Microscopic Picture of Biomax As Spun Fiber (Dia 92 micron)

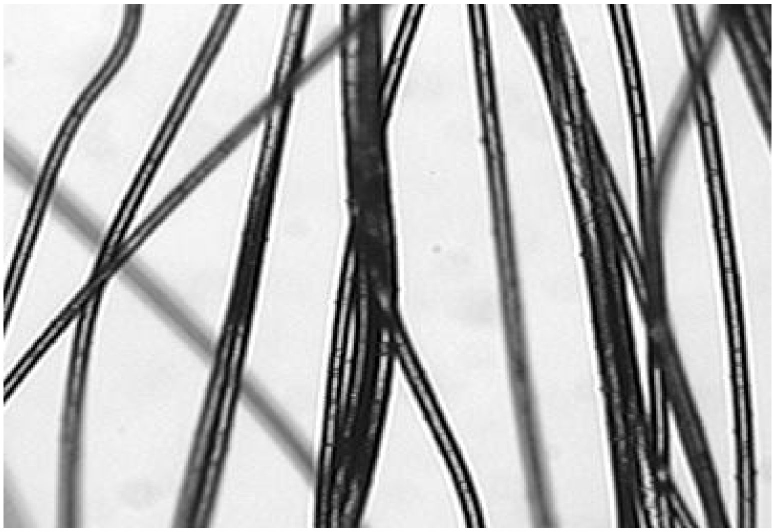


Figure 88 Microscopic Picture of Biomax Drawn Fiber (Dia 40micron)

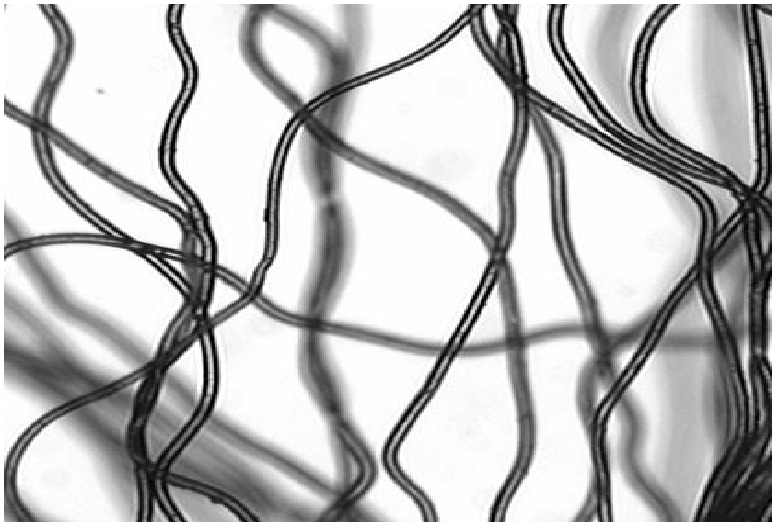


Figure 89 Microscopic Picture of Biomax fiber after drawing and crimping.

Comparing optical microscopic picture of Biomax fibers asspun (Figure 87) with the drawn fiber (Figure 88) shows reduction in fiber diameter. The drawn fiber was crimped in the steam-heated crimper. Figure 89 shows the crimps on the final fiber used in making composites.

I.V. analysis shows that initial I.V of Biomax chips was 0.655, it dropped to 0.461 after spinning and further dropped to 0.439 after drawing and crimping. This means Biomax is susceptible to thermal degradation and loses viscosity quite fast. Hence, it is necessary to dry the chips and spin at practically low temperatures.

CONCLUSIONS

It is possible to produce fibers by melt spinning Biomax or PLA. To reduce degradation, it is necessary to dry the polymer granules properly before spinning. Further optimum spinning melt temperature is in the range 230 to 240°C for Biomax and 205 to 210°C for PLA, where the spinning performance is acceptable and degradation is lower. To produce stronger fibers it is necessary to go for higher spinning speeds and or to draw the as spun fiber. The fibers thus produced are suitable as binder fibers for making nonwoven fiber based composites.

VITA

Manjeshwar G. Kamath received B.S. degree in Chemical Engineering from Karnataka Regional Engineering College, Suratkal, India. After serving several years in the industries manufacturing synthetic polymers, fibers, filaments, films and bottles in India he immigrated to USA in the specialty occupation of developing novel polymers and fibers. He is a licensed Professional Engineer in The State of Tennessee since 2000. In April of 2003, he entered the University of Tennessee, Graduate School of Engineering.