

## **DEVELOPMENT OF STRUCTURAL PANELS FROM FLAX SHIVES, FIBER & HPDE**

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**Written for presentation at the  
CSBE/SCGAB 2006 Annual Conference  
Edmonton Alberta  
July 16 - 19, 2006**

### **Abstract**

Biocomposite panels are made from hemp stalk, flax shives or other wooden materials as the main part of the product and the other important part is binder. Some advantages of such biocomposite board are low density, insulation capacity against heat and moisture transfer, and biodegradability, as well as cheapness compared to concrete based materials. In this research, two binders based on HDPE (High Density Polyethylene) with two different percentages of Flax fiber (as reinforcement in order to improve the mechanical and physical properties of the product) were assessed for flax shives to make panel board. The flax fiber was chemically treated to improve the adhesion between the fiber and polymer. According to binder formulations, ingredients were mixed and extruded using double extruder. The extruded materials were pelletized and used as binder for flax shives at three ratios. For this purpose, compression molding technique was applied to enhance the efficiency of the binder. Biocomposite board quality was studied by doing physical and mechanical tests on the products including: tensile strength, bending strength, melt flow index, thermal properties and water absorption following ASTM standards.

**Keywords:** Biocomposite, flax shives, HDPE, extrusion, compression molding

## Introduction

In present scenario, petroleum resources are getting depleted and demand is increasing along with increase in hazardous effect on global environment. Therefore, there is a growing awareness for the integrated approach with global environmental factors, sustainability, industrial ecology, green chemistry and engineering for developing materials and product for the coming generation. In this new paradigm shift, composite materials are one of the best alternatives. In other words adding fiber and plastic together to form an eco-friendly composite. The more usage of natural fiber makes the product less hazardous and reduces usage of the plastic. However, fibers do possess high strength and stiffness but are difficult to use in load bearing applications by themselves due the fibrous stability. Therefore, a polymer matrix is needed to bind or hold the fiber in place.

A polymeric binder binds the fibers together to develop a desired biocomposite. There are various thermoplastic available to bind the fiber and other agri-residues like straws together. Waxes are often used as primary binder components. They have low melting point, good wetting behavior, short molecular chain, low viscosity and decompose with small volume change. However, its disadvantage is poor mechanical properties. High Density Polyethylene (HDPE) is good secondary binder component since it has high strength and can serve as backbone polymers during binding. Cheap and durable structural and construction materials based on agricultural residues can be good alternatives for traditional construction materials if they are constructed with proper ingredients. Biocomposite boards can be made from hemp stalk, flax shives or other naturally occurring materials as the main part of the product. Some advantages of such materials possess some properties including: high strength, low density, thermal insulation capacity, and potential biodegradability, as well as attractive economics compared to concrete based materials. Biocomposite can be made from hemp stalk, flax shives or other naturally occurring materials as the main or filler part of the product along with polymeric binder. Natural fibers are gaining importance to make bio-composites for various applications during last decade due to the desirable properties (Bledski et al. 1999). Major thrust of on going research nowadays is on flax, hemp, jute etc. Out of various natural fibres, flax is considered to be one of the strongest and widely available bast fiber in North America.

Flax is one of oldest fiber grown in North America as early as 1626. Flax (*Linum Usitatissimum*) is a dicotyledonous of the linacea family. The by-product after extracting oil from seed is used as animal fodder. The principal constituent of flax fibers is cellulose, with smaller amounts of hemicellulose, lignin, pectin, oils and waxes. Most of the flax growers in Canada are burning the flax straw since it is not easily reincorporated into the soil and they are unable to find a viable market for the flax straw. Only a small percentage of total crop residue produced annually is utilized in paper and pulp industry, consisting of the bast fibre which forms up to 25% of the stalk. However, the interior or core part, which is separated as shives during decortication and pulping, was mostly discarded in the past from papermaking applications (Jagannadh and Kolla, 1997).

In a composite, similar to wood fibres, flax shives are stress concentrators. The presence of hollow cells in the core cells can also affect wet strength and moisture absorption properties. On the other hand, flax shives are rigid and have extremely high modulus fibres. Pulped flax shives could be an important source of reinforcing fibre for paper and packaging products. Because flax shives and fibres are lower in lignin and higher in hemicellulose content compared to that of wood fibre, they can absorb moisture more easily than wood fibres. Surface sizing (eg: clay) is expected to play an important role in the economic utilization of flax shives.

Most of the research reviewed indicated that only a limited work had been done on using flax fiber with HDPE to form biopolymer binder. Flax fiber is used to reinforce the HDPE and reduce its usage. Overall goal of the research was to prepare and test a biopolymeric binder from HDPE plastic and flax fiber which can be used with flax shives or any other agriculture residues to manufacture cheap biodegradable biocomposite structural panel boards.

## Materials and Methods

The basic ingredient for the biopolymer binder was HDPE and flax fiber. Flax fiber was used as reinforcement to the HDPE plastic based biopolymeric binder. Flax fiber can withstand processing temperatures up to 250°C (Sreekala et al. 2000). To enhance the processability, 5 % paraffin wax was added while preparing the binder. Biocomposite structural panel were prepared by using two types of flax shives, 5-6 cm and 2 mm chopped shives.

ExxonMobil HDPE HD 8760.29 was used as basic binder ingredient. This material is economical, impact resistant, and provides a good moisture barrier. HDPE is compatible with a wide range of products including acids and caustics but is not compatible with solvents. HDPE is naturally translucent and flexible. HDPE is supplied flame-treated on a stock basis and lends itself readily to silk screen decoration. While HDPE provides good protection at below freezing temperatures, it cannot be used with products filled at over 160° F or products requiring a hermetic seal. Some of the important properties are enlisted below.

**Table 1. Important Properties of HDPE used**

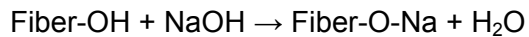
Polymer type	Density (g/cm <sup>3</sup> )	Melt Flow Index (g/10 min)	Tensile Strength (MPa)	Flexural Modulus (GPa)	Melting point (°C)	Crystallization Temperature, (°C)
ExxonMobil HDPE HD 8760.29	0.948	5.00	23.4	1.06	131	115.3

## Treatment of Fiber

It has been found out that flax fibres are more hydrophilic in nature than other green fibres, and properties such as density, electrical resistivity, ultimate tensile strength, initial modulus, etc., are related to the internal structure and chemical composition of fibres (Reddy et al. 2005). Fibre-matrix adhesion and its influence on composite properties have been the subjects of research for a long time. It is revealed from various literatures that to have the most desirable properties for the composites, it was necessary have an effective fibre-matrix adhesion. Therefore the flax fiber was subjected to alkaline treatment. This treatment is one of the most common treatments adopted to treat fiber before reinforcing any thermoplastic or thermoset polymer (Ray et al. 2001).

In this treatment, removal of hydrogen bonding in network structure is most important modification which increases the surface roughness. This treatment also removes certain amount of lignin, wax and oils covering the external surface of the fiber cell wall, depolymerizes the native cellulose structure and exposes the short length crystallites (Mohanty et al. 2001). This treatment directly influences cellulosic structure of plant fibers, degree of polymerization and removal of lignin and hemicellulosic compounds may lead to change in molecular orientation of the cellulose crystallites.

In this treatment, flax fiber was immersed in 5% NaOH solution for 3 hrs and then was washed thoroughly with distilled water. The reaction of sodium hydroxide with natural fiber is given by the following equations:



It is reported in earlier studies that this treatment also improves the mechanical, impact fatigue and dynamic mechanical behavior of the fiber-reinforced composite with saline treatment (Mohanty et al. 2001). Weyenberg et al. (2003) reported that the combination of alkali and dilute epoxy enhances the flexural properties.

### **Drying of Fiber for Biopolymeric Binder**

After treating the fiber, it is very necessary to dry the fiber to minimum moisture level for developed high best mechanical properties based composite with suitable polymer. Main focus is uniformly drying the fiber as drying of moist materials is a complicated process involving simultaneous, coupled heat and mass transfer phenomena, which occur inside the material dried (Yilbas et. al., 2003). Therefore, after treatment, the flax fiber was washed and then dried in two stages. First the fiber was dried at room temp for 12 h and then as Re-circulating cabinet type dryer was used to the dry the flax fiber for 24 h. The dryer has two small household type dehumidifiers and air was re-circulated through the fiber from bottom to top. Each unit has condenser and evaporator coils, which provides the heating and dehumidification for the process air of the dryer. The dehumidifiers provide direct removal of moisture with minimal heating of the drying air. The flax fiber was dried in the dryer at 50 °C to reduce the moisture content to 2 %.

### **Bio-polymeric Binder Preparation**

In the first stage size reduction of the flax fiber was done, it enables the fiber to disperse properly with the matrix. The cleaned and dried flax fiber was ground in three stages by the hammer mill (Glen Mills Inc, Clifton, NJ) and knife mill (Thomas Wiley Laboratory Mill, Thomas Scientific, USA).



**Figure1. Knife Mill**



**Figure 2. Tumble type Mixer**

To reinforce the HDPE, two percentages (5% and 10 % by weight) of ground flax fiber (Table 2) was added to prepare the biopolymeric binder. For each sample mixtures of HDPE and ground flax fibers were prepared by using a tumble type mixer (National Hardware, Dresden, Canada). For proper compounding, the blended material was fed into the twin-screw extruder (Werner & Pfleiderer Engineers, Ramsey, NJ, USA). In the twin-screw the barrel-to die temperature profile in range of 80-150 °C/176-302°F (four zone heating plus heated die), a screw speed of 150 rpm and six-hole strand die. For better processing of the material, 5 % paraffin wax was added to all of the samples before extrusion. The extruded strands were then pelletized using a Nelmor granulator (3/8"≈10 mm screen size, AEC Nelmor, South Attelboro Massachusetts) and these pellets were further ground to fine powder using the same Wiley mill described above. This fine powder is the prepared biopolymer binder for preparing biocomposite panels using flax shives.



**Figure 3. Twin Screw Extruder**

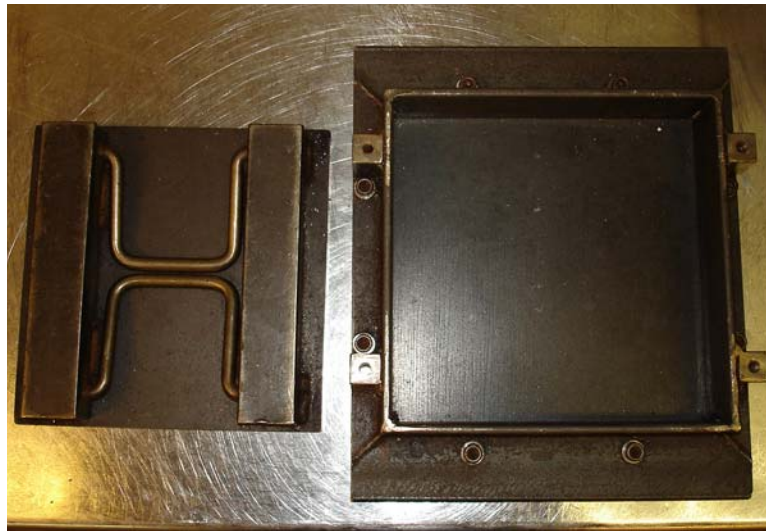
**Table 2. Different samples of Biopolymer Binder from HDPE and Flax Fiber**

Sample No.	Type of Plastic & Percentage	Flax Fiber Percentage ( weight %)	Paraffin Wax ( weight %)
1	HDPE 90 %	5	5
2	HDPE 85 %	10	5

### **Biocomposite Structural Board Preparation**

For studying the usages of the biopolymeric binder, biocomposite boards were prepared which can used in various household and other transportation applications. Saskatchewan has largest flax production in Canada, 881.4 tonnes in 2005 (<http://www.flaxcouncil.ca>). But still the flax straw is not properly utilized. Therefore to make a useful product involving flax straw, flax shives were selected as the base ingredient for the biocomposite board. For preparing biocomposite structural boards, six types of samples were made using the two different samples of biopolymer

binder with three percentages (35 %, 45 % and 55% by weight) of flax shives. Initially 5-6 cm length chopped flax shives were used for preparing the biocomposite. However after first two boards the flax shives size, due to problem in mixing the binder and the size of flax shives was further reduced and it was finely chopped using Wiley mill equipped with 2 mm sieve to get better aesthetic look and homogeneity during molding. A rectangular mold was designed in-house to mold a rectangular sample of 20 cm x 20 cm size. The mold was made up of mild steel angle frame on a 12.5 mm (1/2") removable backing plate.



**Figure 4. Rectangular Mould**

The flax shives and biopolymer binder are mixed thoroughly in a tumble mixer and fed into the designed mold. The mold is pressed by a heated press (Miller Machine Tools, J.B. Miller Machinery & Supply Co. Ltd, Toronto, Canada).



a) Flat Pates



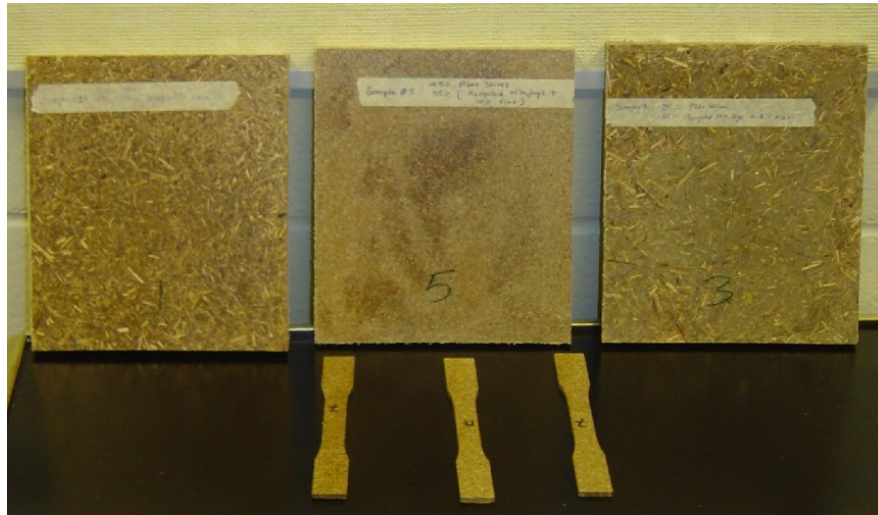
b) With mould in place

**Figure 5. Hydraulic Press used for Compression molding**

The temperature of lower and top plate of the compression molding machine was maintained at 170°C (338°F) and 8.62 MPa (1250 Psi) was applied throughout the residence period. The



residence period for molding of all the samples was 10 minutes. The samples were cured by the water cooling system of the molding machine.



**Figure 6. Molded Biocomposite sample with machined specimens for tensile test**

Altogether six biocomposite boards were prepared by the two different samples of binders having 5 and 10 % chemically treated flax fiber by weight respectively. The complete details of the biocomposite prepared are given below.

**Table 3. Different Biocomposite Board Samples Composition**

Sample No	Type of Biopolymer binder (weight %)	Flax Shives (Size & weight %)
HDPE5FF35FS	HDPE 85 % + Flax Fiber 5 %	} 65 % 5-6 cm shives 35 %
HDPE10FF35FS	HDPE 90 % + Flax Fiber 10 %	
HDPE5FF45FS	HDPE 90 % Flax Fiber 5 %	} 55 % 2 mm shives 45 %
HDPE10FF45FS	HDPE 85 % Flax Fiber 10 %	
HDPE5FF55FS	HDPE 85 % + Flax Fiber 5 %	} 45 % 2 mm shives 55 %
HDPE10FF55FS	HDPE 90 % + Flax Fiber 10 %	

To understand the processability behavior of the binder, the melt flow index and thermal analysis of the biopolymer binders was determined following ASTM standards. To study the efficacy of the biopolymeric binder, physical and mechanical properties of the biocomposite prepared by the flax shives and biopolymeric binder were determined by doing series of test like water absorption, tensile and 3-point bending tests following ASTM standards.

## Results and Discussion

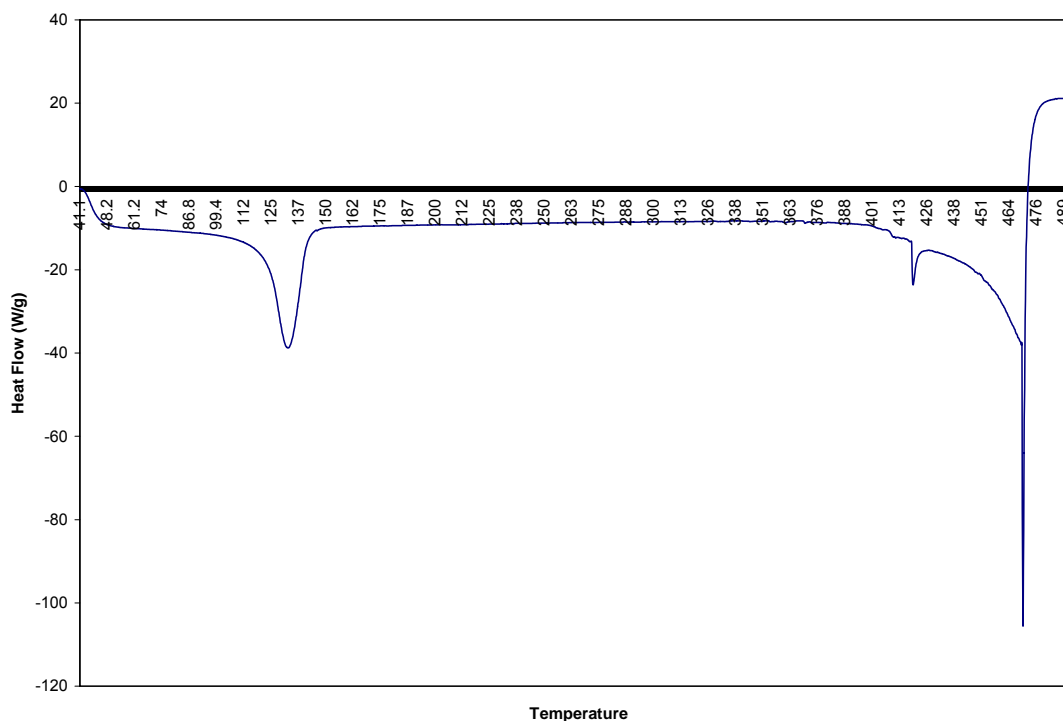
The flow characteristics of the biopolymeric binder was studied by measuring the MFI (Melt Flow Index), which describes the suitability of the polymer based composite for various forming processes. MFI as an indicator for viscosity of the binders made from different ratios of HDPE and flax fiber, are shown in table 4. These results indicate that fiber content has significant effect on MFI of the product. With increase in fiber content the viscosity of the binder increases, in

other words MFI decreases. Therefore the percentage of flax fiber was restricted to 10 % only during preparation of biopolymeric binder so that it does not adverse effect on processability.

**Table 4. Binders melt flow index measured at 190°C (374°F)**

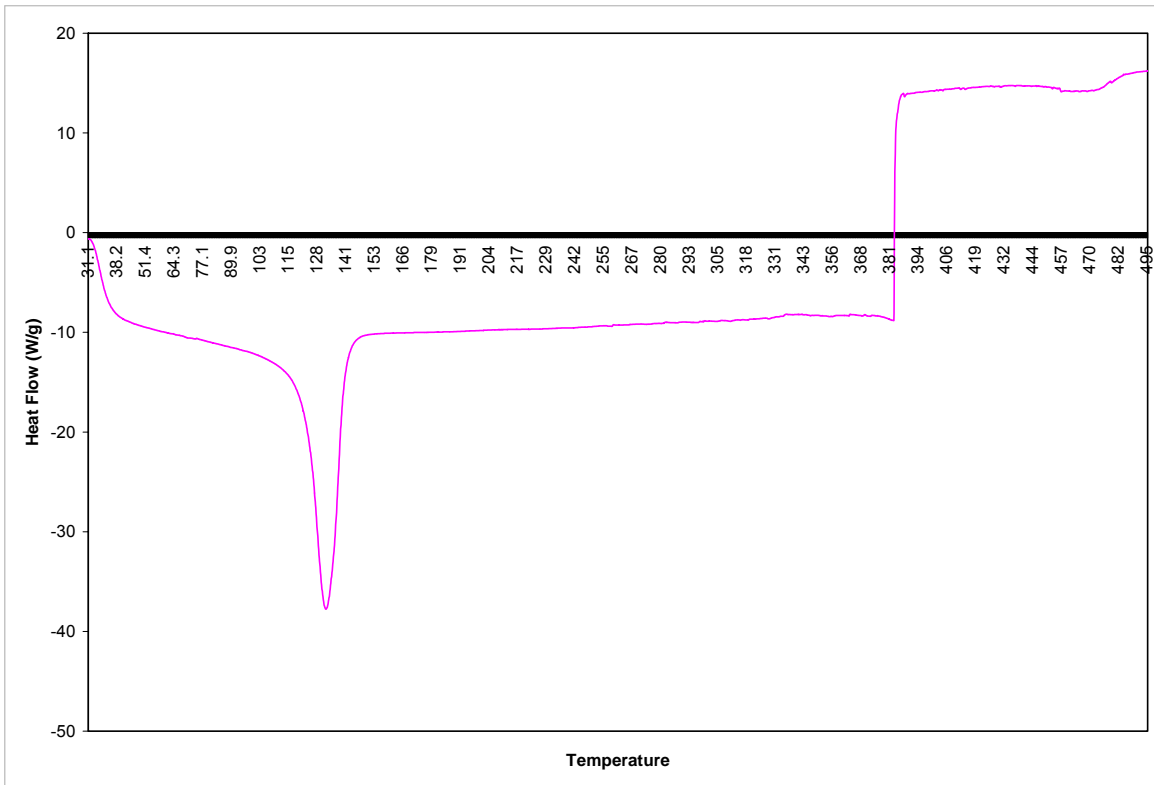
SI No.	Polymer/ Binder	MFI(g/10min)
1.	HDPE	5.00
2.	HDPE + 5%Fiber	4.813
3.	HDPE + 10%Fiber	4.490

To determine the processing parameters of the biopolymeric binders, thermal analysis was done by Differential Scanning Calorimetry. Analyzing the DSC thermograms showed that the melting range of biopolymer binder is displayed as an endothermic peak. DSC analysis enables the identification of chemical activity occurring in the fiber as heat is applied. DSC was used to determine not only the melting point ( $T_m$ ) but also the melting range of the polymer. The glass transition temperature ( $T_g$ ) could not be observed. The  $T_g$  of pure polyethylene is usually below  $-100^\circ\text{C}$ . Table 5 shows the melting points of the two different binders. Changing in the melting point temperature of the polymer due to the different percentage of flax fiber incorporation was observed. The increase of  $T_m$  may be attributed to the plasticization effect of the fiber that diffuses or dissolves into the polymer. The incorporation of 5 % & 10 % chemically treated flax fiber in HDPE increased the  $T_m$  of untreated fiber composites. The increased melting point of composites meant that thermal resistance increased as elucidated by the DSC method. The DSC Thermograms for the two types of binders are given below.



**Figure 7. DSC Thermogram of Biopolymer Binder (HDPE with 5 % Flax fiber)**





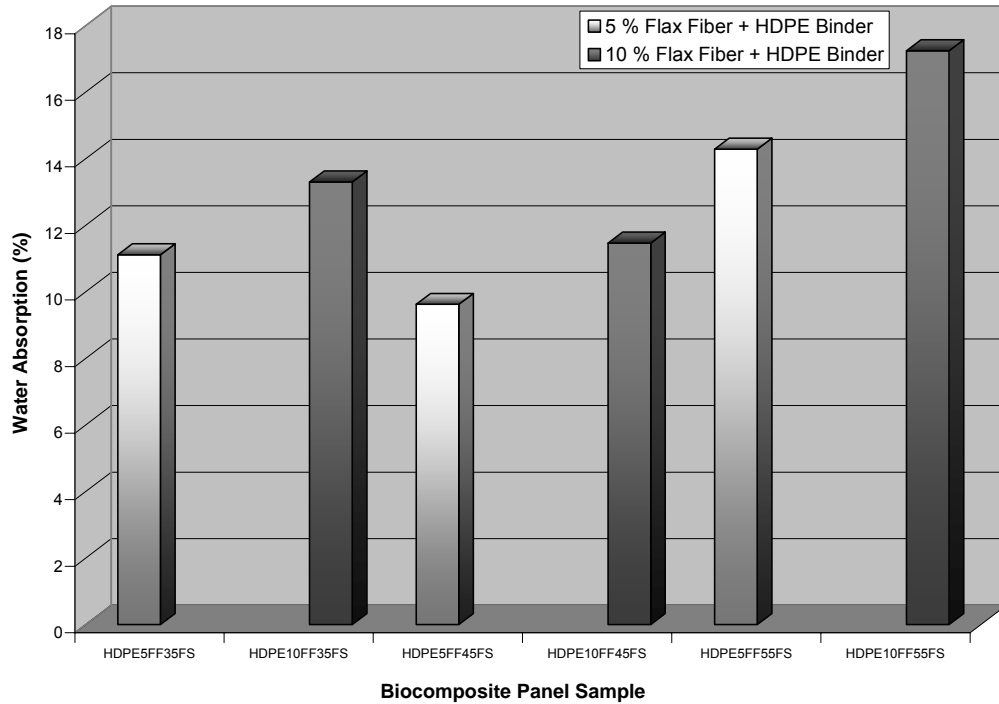
**Figure 8. DSC Thermogram of Biopolymer Binder (HDPE with 10 % Flax fiber)**

**Table 5. Melting Point of the Biopolymer Binders**

Binder	Melting Temp (° C)
HDPE	131
HDPE+ 5% Fiber	132.34
HDPE+ 10% Fiber	131.75

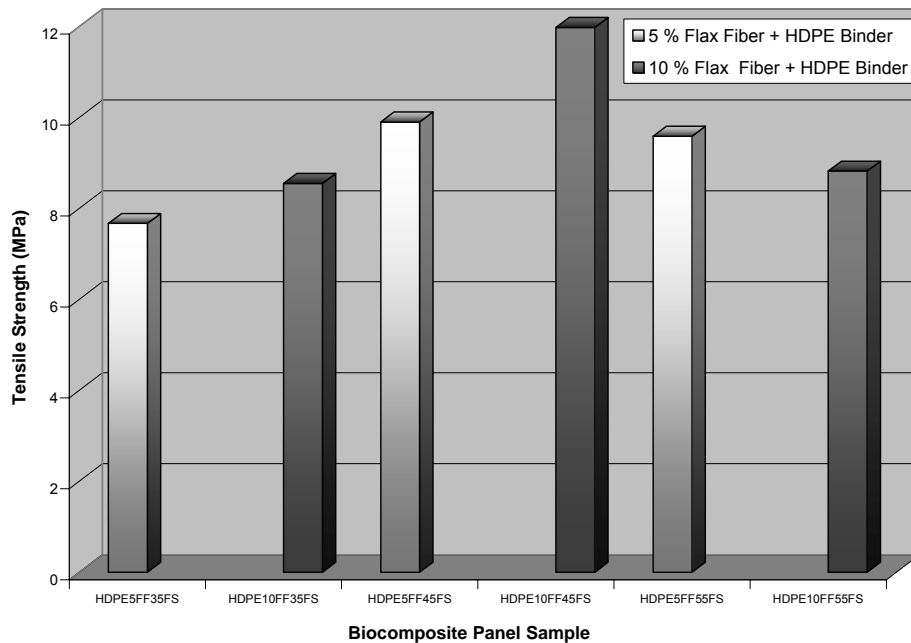
Flax fiber and shives are mainly composed of biopolymers which are hydrophilic materials. As it was found that the water absorption was too high with 35 % chopped shives of size 5-6 cm, the shives were further chopped by 2 mm screens.

It was observed that on increasing shive percentage from 35 to 45 after size reduction, water absorption was reduced. Therefore it can be inferred from this phenomenon that the reduction of shives size reduces the porosity of the board, in other words causes to being better packed in the polymeric matrix and reduces air spaces and consequently decreases capacity for absorbing water. But after further increase in shive percentage to 55 %, the water absorption increased.



**Figure 9. Water absorption characteristics of the Biocomposite Panel Boards**

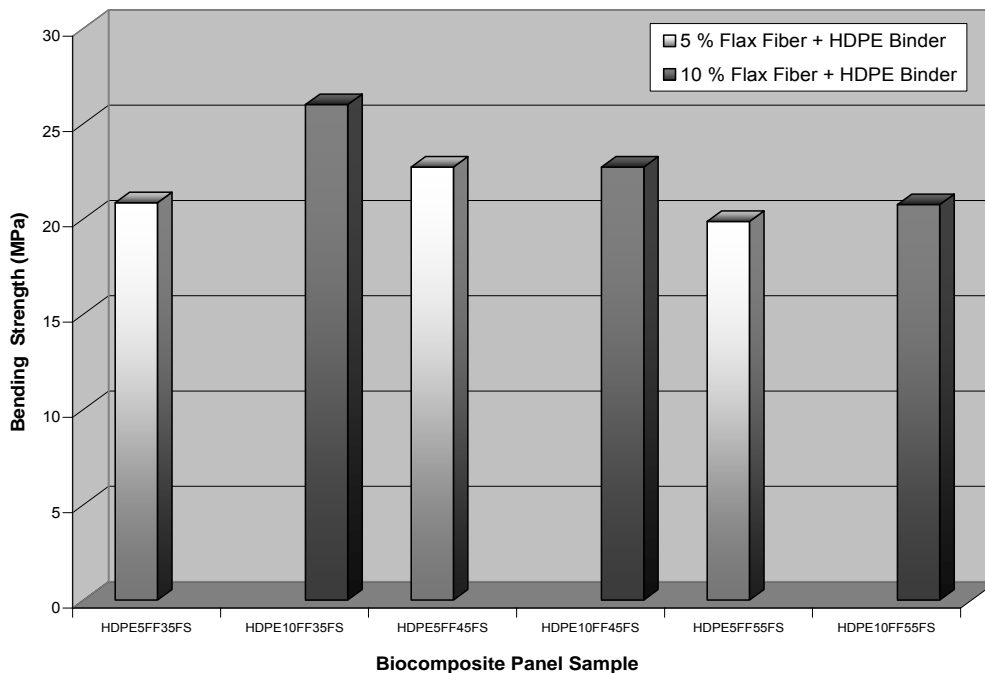
The tensile strength of biocomposite board prepared from both types of biopolymeric binders was found comparable. It was found on increasing the flax fiber content in the binder enhances the tensile strength. The tensile strength also increased with increase in percentage of flax shives but after 45% it decreased.



**Figure 10. Tensile Strength of the Biocomposite Panel Boards**

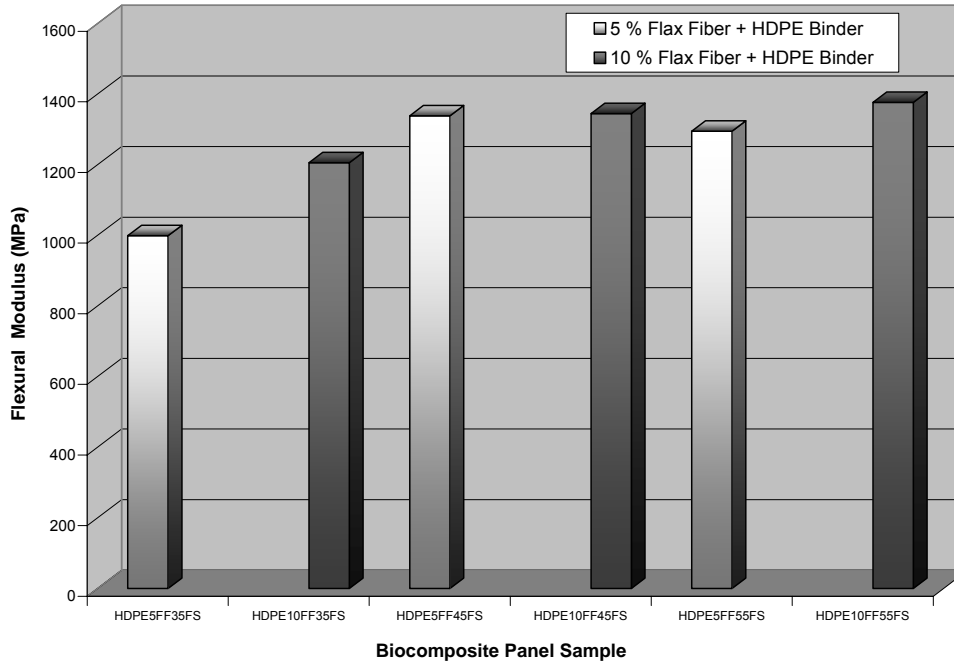
The highest tensile strength was observed by biocomposite made of biopolymeric binders having 10 % flax fiber based HDPE binder and 45% flax shives. Figure 10 shows the tensile characteristics of the different biocomposite board made out of HDPE based biopolymeric binder and flax shives.

Three-point bend (flexure) test results are shown in Figure 6. Figure 6 shows that the bending strength ranges from 19.87 MPa to 26 MPa.. Initially using flax shives of size 5-6 cm, the bending strength of 10% flax binder was more where as it decreases when the size of flax shives was reduced to 2 mm. The result pattern was not uniform which can be attributed to the random orientation of flax shives in the biocomposite However, it was observed that thee biocomposite boards with higher plastic content had higher strength.



**Figure 11. Bending Strength of the Biocomposite Panel Boards**

The flexural modulus increased when the size of shives was reduced to 2 mm from 5-6 cm and percentage was increased from 35 to 45. The increase in modulus was mainly because of the better mixing between reduced size shives and biopolymeric binder. However no consistent pattern was derived from the results as the shive orientation and mixing plays an important role in the board formulation. The flexural modulus varied between 999.13 MPa to 1376.6 MPa.



**Figure 12. Flexural modulus of the Biocomposite Panel Boards**

The density of the all the six types of boards prepared were found ranging between 0.8 -0.9 g/cm<sup>3</sup> (50-56 lb/cu ft). The little variability in densities can be attributed to the orientation of the flax shives in the samples.

## Conclusions

Flax fiber can be feasibly used as an alternative for reinforcing HDPE based biopolymeric binder for manufacturing biocomposite boards with flax shives or any other agricultural residue straw for various applications. The incorporation of flax fiber decreases the MFI and simultaneously increasing the thermal resistance of the biopolymeric binder by increasing the melting point ( $T_m$ ). The biopolymer was efficient in uniformly binding the different percentages of flax shives to form biocomposite panel board.

The biocomposite panel board prepared had a high rate of water absorption. However the size reduction of shives will mitigate this somewhat at inclusion rates less than 45%. But Still water absorption was found quite high at higher (55% or greater) inclusion rates of flax shives. Therefore, chemical treatment of the shives is suggested to reduce the water absorption depending upon the application for board is to be used. The tensile strength and flexural modulus increased with increase with fiber and shive content, but after 45 %, both properties started declining. However, the bending strength was higher for samples having higher percentage of the fiber.

This product can be used in panels for door & windows, furniture and in automotive applications. Further study on other mechanical properties and other agri-residues/straw should be conducted.

## Acknowledgements

The authors would like to thank Bio-fiber Ltd, The Saskatchewan Agriculture and Food - Agriculture Development Fund and SaskBET INC for their contributions in this research.

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