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The Effects of Various Chemical Treatments of Flax (*Linum usitatissimum*) Fiber on Mechanical Properties of Their Biocomposites

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ABSTRACT

Measurement of mechanical properties of biocomposites is a good method for evaluating their effectiveness of adhesion between fiber and polymer matrix. In this research, the effects of four different chemical treatments of flax fiber on some mechanical properties of their biocomposites was investigated. Initially, the flax fiber was soaked in alkaline, silane, benzoyle and peroxide solution and the fiber were dried in an air-cabinet drier at 70°C. After grinding, each group were separately mixed with HDPE powder at a ratio of 10% flax fiber and 90% HDPE. From these mixture, composite plates were prepared through extruding, pelleting, and rotational molding. The resulting composites were tested for their various mechanical properties using tensile tests. The test results indicated the maximum strain was 6.22%, maximum supported load at yield point was 582 N, maximum stress at yield pint was 20.26 MPa and maximum modulus of elasticity was 467.75 MPa all for alkaline treatment. It was found that all tested mechanical properties for HDPE were significantly lower than the composites made from fiber containing biocomposites. However there was no significant difference between the mechanical strength of composites produced from various chemical treatments.

INTRODUCTION

Engineering composites are traditionally manufactured using a polymer matrix and synthetic fibers such as glass or carbon fibers for reinforcement. The increasing ecological and environmental concerns, together have caused the manufacturers and researchers seek alternatives for synthetic fibers (Bledzki and Gassan, 1999). Some advantages of natural fibers over that of synthetic fibers are: low density, low cost, relatively low energy consumption for production, recyclability, and biodegradability (Mohanty *et al.*, 2000).

In the past decade, the low cost and abundance of the natural fibers, created a new interest in utilization of these fibers as potential replacement for synthetic fibers in production of composite materials. Natural fiber reinforced composites, known as "biocomposites", are already used in automotive industry and there is an increasing demand for their usage in construction industries (Mohanty *et al.*, 2001). Using natural fibers for reinforcing composites has increased in recent years. Naturally reinforced composites consist of a polymer matrix a natural fiber is used for their reinforcement. These biocomposites are used in various industries as a replacement for conventional industrial parts. Some examples of potential usage of biocomposites include door panels, instrumental panels, and package trays (Gurunathan *et al.*, 2015).

Flax fiber, which is a renewable resource and possess relatively high mechanical properties, as compared with other natural fibers and it is considered as a potential replacement for glass fibers as reinforcing agent for composite materials (Arbelaiz *et al.*, 2005). More than half of the Canada flax crop is grown in Saskatchewan, and the remainder grown in provinces of Manitoba and Alberta (Panigrahi *et al.*, 2002).

The flax fiber coating consists of a complex heterogeneous polymer including cellulose, hemicellulose, and lignin. Cellulose is a hydrophilic polymer of D-glucopyranose units, which causes flax fiber surface to have hydrophilic properties and when this fiber is used to reinforce hydrophobic matrices causes a poor interface and poor resistance to moisture absorption (Le Duigou *et al.*, 2008). Cellulose fibers absorb water which causes swelling of the fibers. When these fiber are used for reinforcing composite causes micro-cracks in them and consequently degrades mechanical properties of the composites. Moisture absorption problem can be greatly reduced by treating these fibers with a suitable chemical reagent. Some chemical treatments also activate hydroxyl groups and introduce new moieties that can effectively interlock with the matrix (Assarar *et al.*, 2011).

Many researchers have sought various chemical treatments of flax fiber to modifying their coating properties. Alkaline treatment is one of the most popular chemical treatments for reducing hydrophilic properties of natural fibers (Baley *et al.*, 2006; Van de Weyenberg *et al.*, 2003). In alkaline treatment, sodium hydroxide (NaOH) is used to remove the hydrogen bonding in the network structure of natural polymers, thereby increasing their surface roughness (Biagiotti *et al.*, 2004). This treatment removes a certain amount of lignin, wax and oils covering the external surface of the fiber cell wall, depolymerizes cellulose and exposes the short length crystallites. The alkali treatment

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of fibers have shown to have significant effect on the mechanical behavior of composites mixed with fibers. Silane coupling agents are also effective in modifying natural fiber-polymer matrix interface (Wang *et al.*, 2003).

There have been many studies on utilization of flax (*Linum usitatissimum* L.) fibers as a reinforcing agent for bicomposite production in thermoplastic industries (Panigrahi *et al.*, 2002). Flax fiber, besides being environmentally friendly, has also proved to enhance mechanical properties of composites. The main disadvantage of flax fiber as well as other natural fibers is their hydrophilic nature that causes a weak bonding with hydrophobic polymers. This limitation is reduced through chemical modification of surface of the fibers.

The ultimate mechanical properties of biocomposites depend to a great extent on the adhesion between the reinforcing fibers and surrounding matrix ((Bos *et al.*, 2002). The adhesion between the two materials is a function of several factors among which are surface roughness and surface coating and both can be enhanced by special chemical treatments (Bos *et al.*, 2006). Measurement of engineering properties of the composites is a good method for evaluating the effectiveness of adhesion between fiber and polymer matrix due the applied chemical treatments (Aliotta *et al.*, 2019).

In this research flax fiber was treated with different chemicals including silane, benzoyl chloride and dicumyl peroxide. The treated fibers are processed in similar manufacturing steps to produce rotational molded biocomposites. The strength and optical properties of biocomposites were measured and compared to evaluate the effectiveness of the chemical treatments.

MATERIALS AND METHODS

Fiber Preparation and Chemical Treatments

Flax fibers, derived from linseed flax grown in Saskatchewan and decorticated on a standard scotching mill at Durafiber in Canora, SK, Canada, were used for these experiments. The fibers were first washed thoroughly with commercial detergent and dried in an air oven at 70ºC for 24 h. For alkaline treatment, the fibers were soaked in a 5% NaOH solution for about 30 minutes in order to activate the OH groups of the cellulose and lignin in the fiber. Fibers were soaked in 5% NaOH for about half an hour in order to activate the OH groups of the cellulose and lignin in the fiber. The fibers treated in this way were used as chemically untreated fiber. For silane treatment, treated fibers were dipped in an alcohol water mixture (60:40) containing triethoxyvinylsilane coupling agent. The pH of the solution was maintained between 3.5 and 4, using the METREPAK Phydrion buffers and pH indicator strips. Fibers were washed in double distilled water and dried in the oven at 80°C for 24 h.

For benzoyl treatment the pre-treated fibers were suspended in 10% NaOH solution and agitated with benzoyl chloride. The mixture was kept for 15 min, filtered, washed thoroughly with water and dried between filter papers. The isolated fibers were then soaked in ethanol for 1 h to remove the benzoyl chloride and finally was washed with water and dried in the oven at 80°C for 24 h. For the peroxide treatment fibers were coated with dicumyl peroxide from acetone solution after alkali pre-treatments. Saturated solution of the peroxide in acetone was used. Soaking of the fibers in the solution was conducted at a temperature of 70°C for 30 min. High temperatures were favored for decomposition with the peroxide. All chemically treated fibers were washed with distilled water and placed in an oven to remove their moisture, at 80°C for 24 h.

Biocomposites Preparation

The treated and untreated fibers were separately ground by the grinding mill (Falling Number, Huddinge, Sweden). Each type of chemically treated and untreated flax fibers were mixed with thermoplastic powder (HDPE) with a weight proportion of 90% HDPE and 10% fiber. Each type of blend was fed into an extruder (Dynisco, Franklin, MA) using a barrel to die temperature profile of 175°C with a screw speed of 100 rpm (Siaotong *et al.*, 2010). Extruded strands were then palletized and ground using a grinding mill (Retsch GmbH 5657 HAAN, West Germany). The ground product was fed into rotational molding machine to make 30 × 30 × 0.3 cm plates.

Tensile Test

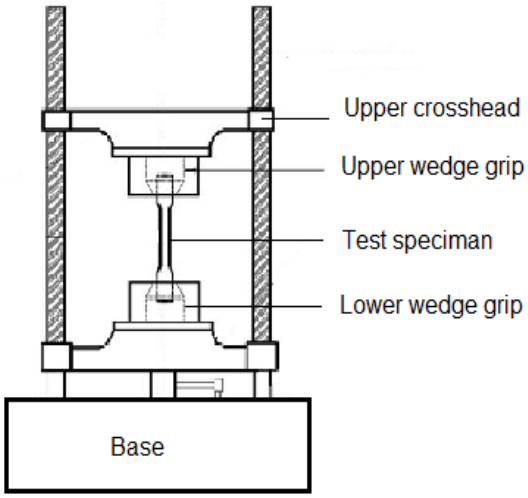

Fig. 1. The schematic of tensile test at a crosshead speed of 5 mm/min

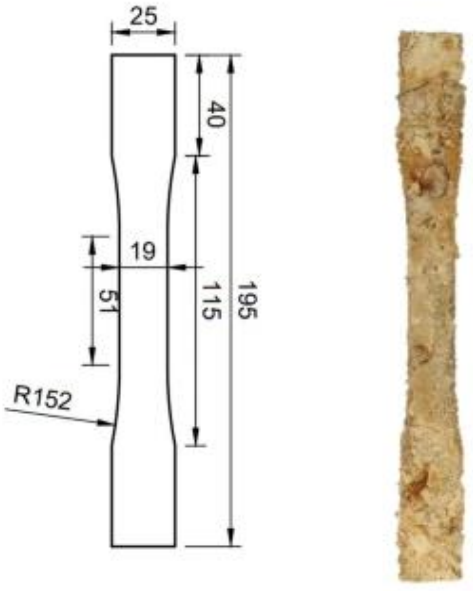

Fig. 2. The dimensions (mm) and a typical picture of tesing specimen used for tensile tests.

The mechanical strength of the prepared plates was evaluated by tensile test using ASTM standard test method of D638 for plastic materials. This test specifies methods for testing the tensile strength of plastics materials and for calculating their mechanical properties. An Instron Universal testing machine (SATEC Systems, Inc., Grove City, PA) was used to perform the tensile test at a crosshead speed of 5 mm/min. The tensile tests were conducted at standard laboratory atmosphere of 23 °C and 50% relative humidity. A schematic diagram for testing is given in Fig. 1. Each test was repeated five times. The applied force and displacement at yield point was recorded and accordingly the tensile strength, and modulus of elasticity and yield strain were calculated. For each test, six testing specimens with the dimensions specified in Fig. 2 were used. The collected data were tabulated and graphed using Minitab software. One way ANOVA test was performed on yield strength data to investigate if there was any significant difference between the mean of yield strength of the biocomposites prepared from chemically treated, untreated and pure HDPE. The comparison of the means was performed using Tukey method.

RESULTS AND DISCUSSION

A summary of the mechanical test results are presented in Table1. A general look at this table indicates that the composites made from pure HDPE have lower mechanical properties than thoes composites made from chemically treated fibers. The mechanical properties of the composites made form chemically treated fibers are very close to each other but the alkaline treatment have higher average values. The modulus of elasticity calculated for the treatments are presented in the last column of Table 1-. The highest module was obtained for was for composites from untreated fiber. Thus by adding flax fiber to the HDPE the resulting biocomposites had significantly higher modulus of elasticity. Comparing the average strain for different composites indicates that the strain of various composites ranged of 6.04% to 6.22% which indicates that yield displacement was almost the same for all of them.

Among the biocomposites the benzoyl treated fiber resulted the lowest modulus of elasticity. The untreated fiber resulted higher modulus of elasticity, which indicates that chemical treatment does not necessarily increases the mechanical properties, however chemical treatment produces more uniform composites with more flexibility

Table 1. The average values for various mechanical properties of composites.

Treatment	Yield strain (%)	Yield load (N)	Yield stress (MPa)	Modulus of elasticity (MPa)
Alkailne	6.22	582	2026	467.5
Peroxide	6.05	558	19.46	444.3
Silane	5.99	579	18.44	425.3
Benzoyl	6.04	488	19.99	386.9
HDPE	3.48	214	12.40	188.75

To investigate if there was any significant differences between treatments, one way ANOVA test was performed on the raw data. The results of ANOVA on modulus of elasticity, are presented in Tables 2. The table indicates a significant difference between treatment means. To investigate which means was different from others, Tucky comparison test was performed and the results are shown graphically in Fig. 3. This figure indicates that the mean of modulus of elasticity for pure HDPE composite is significantly different from other bio composites made from chemically treated fibers. Modulus of elasticity indicates is a measure that indicates the stiffness of a material. Higher-modulus materials exhibiting less deformation under load compared to low-modulus materials. Since the modulus of elasticity for HDPE is lower than the other treatments, it indicates the composite made from HDPE is softer and has more tendency for deformation than other composites. This can be associated with uniform matrix of the pure HDPE composite.

Table 2. ANOVA test results for modulus of elasticity of biocmoposite made from fibers with different chemical treatments.

Source of Variation	Degrees of Freedom	Sum of Squares	Mean Squares	F Value	*P* value
Treatments	4	302575	75644	25.75	0.00
Error	25	73444	2938		
Total	29	376018			

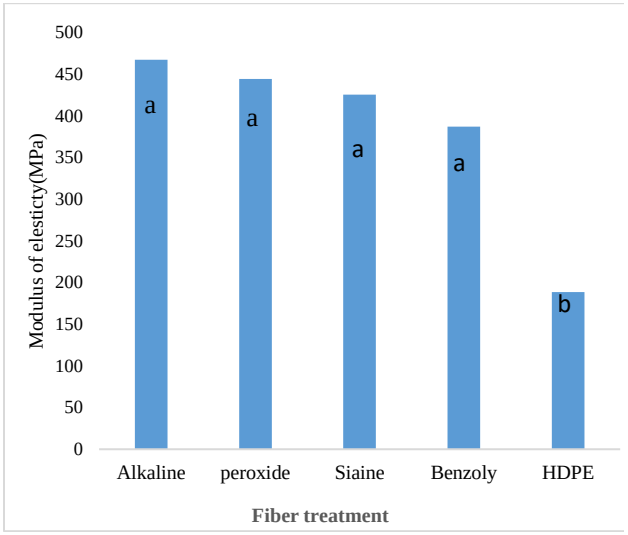

Fig. 3. The average values of modulus of elasticity of composites obtained from various treatments.

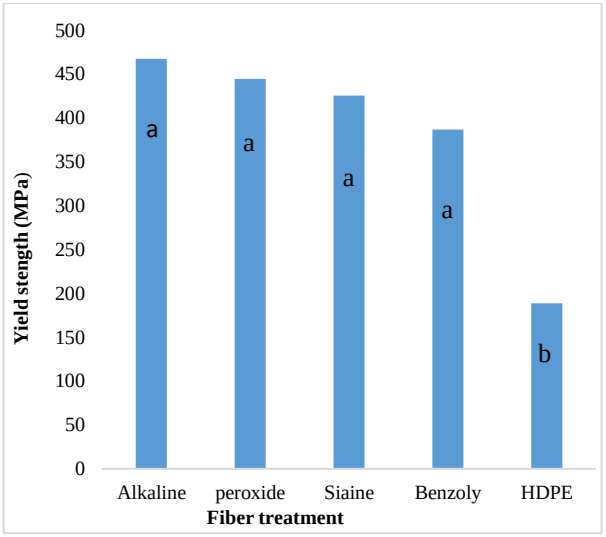

Fig. 4. The average values yield strength of composites obtained from various treatments.

The results of ANOVA on yield stress, are presented in Tables 3. The table indicates significant difference between treatment means. To investigate which means was different from others, Tucky comparison test was performed and the results are shown graphically in Figs. 4. This figure indicates that the mean of yield stress for pure HDPE composite is significantly different from other bio composites made from chemically treated fibers. Yield stress, marking the transition from elastic to plastic behavior, is the minimum stress at which a solid will undergo permanent deformation or plastic flow without a significant increase in the load or external force. Higher yield stress for a material indicates that it is more elastic. Since the Yield stress for HDPE is lower than other treatments, it indicates the composite made from HDPE deform more quickly than other composites. This can be associated with degree of plasticity of HDPE. When HDPE is mixed with fibers its stress yield increases.

The results of ANOVA on strain at yield point, are presented in Tables 4. The table indicates significant difference between treatment means. To investigate which means was different from others, Tucky comparison test was performed and the results are shown graphically in Figs. 5. This figure indicates that the mean of strain at yield point for pure HDPE composite is significantly different from other biocomposites made from chemically treated fibers. Strain at yield point is the maximum stress that a material can withstand while being stretched or pulled before breaking. It may be dependent on factors, such as the preparation of the specimen, the presence of surface defects, and the temperature of the test. Since the yield strain of HDPE composite is lower than other treatments, it indicates the composite made from HDPE is prone to breaking than other composites. The higher strain for composites made from chemically treated fibers can be associated with interlocking of fiber and HDPE mixture.

Table 3. ANOVA test results for yield strength of biocmoposite made from fibers with different chemical treatments.

Source of Variation	Degrees of Freedom	Sum of Squares	Mean Squares	F Value	*P* value
Treatments	4	231.30	57.83	13.73	0.00
Error	25	105.29	4.21		
Total	29	336.59			

Table 4. ANOVA test results for strain of biocmoposite made from fibers with different chemical treatments.

Source of Variation	**Degrees of Freedom**	**Sum of Squares**	**Mean Squares**	**F Value**	***P* value**
Treatments	4	34.03	8.51	77.31	0.00
Error	25	2.75	0.11		
Total	29	36.78			

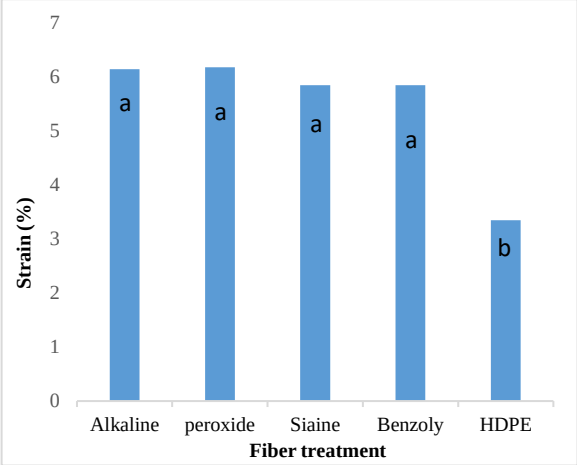

Fig. 5. The average values of strain of composites obtained from various treatments.

CONCLUSIONS

In this research composites plates was prepared from chemically treated flax fiber mixed with HDPE powder. The mechanical properties of theses composites were determined and were statistically compared. The mechanical test results on prepared composite specimen and ANOVA investigation resulted the following conclusions:

- The average mechanical test results obtained for fiber containing composites were significantly higher than the composites made from pure HDPE.
- Among the chemical treatment, alkaline gave higher mechanical properties values.
- There were no significant difference between the means of mechanical properties of the composites prepared from different chemically treated fibers, thus both economic and environmental aspects of using chemical must be considered.

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