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DEVELOPMENT OF BIODEGRADABLE COMPOSITE MATERIALS BY USING JUTE FIBRE REINFORCEMENT

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ABSTRACT

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Jute reinforced PLA (polylactic acid) fibre composites have been fabricated by using woven jute fabrics from Bangladesh. The area density of the fabrics was 158.38 g/m^2 and the thickness was o.86 mm. The staple length and linear density of the PLA fibres were 37.6 mm and 2.13 dtex respectively. Initially, a nonwoven fabric was produced by using jute fabric as the base fabric and laying a web of PLA fibres on it by using an Automatex carding and cross-lapping machine. The nonwoven cross-laid web was then needled into the jute woven fabric on the needle punching machine. The nonwoven base fabrics were cut into 17 x 17 cm sizes. Eight layers of the nonwoven base fabric were combined together to form a composite by using a hot press machine at 160℃ and 25 kg/cm^2 pressure for 2.5 minutes. The fabric layers were arranged at different angles to make the fabricated composite isotropic. The 3-point bending tests, tensile tests, impact tests and ageing tests were carried out on the finished composites by following the standard test methods and equipment. In the 3-point bending test, the applied load was 20N and the average deflection was 2.02 mm. In the tensile testing, the maximum average load was 3500N for a cross-sectional area of 20 mm x 2.88 mm and the tensile strength was 60.77 MPa. In the impact test, the allowable impact load was 2400N and the energy was 16.37 joules. The ageing of the composites was carried out by using different ageing media (water, salt water, acidic and alkaline solutions) and the biodegradability aspects of the finished composites were investigated.

***Key words**: jute, PLA fibre, composite, biodegradable, ageing*

INTRODUCTION

Jute is a biodegradable and environmentally friendly natural bast fibre grown mainly in India and Bangladesh. It is a widely used natural fibre because of its superior mechanical properties and biodegradability (Ochi 2008; Liu *et al.* 2009; Uddin *et al.* 1997; Kumar *et al.* 2005 and Plackett *et al.* 2003). Earlier, jute was used as hessian, sacking, carpet backing etc. Now-a-days jute is used in many different and diversified ways, such as fine yarn, jute wool, fine fabrics, blankets, different types of bags, jute geotextiles, jute composites etc. Jute composites are the most important amongst them. The thermoplastic polymer based composites are becoming more popular in many applications due to the possibility of combining the toughness of thermoplastic polymers with high stiffness and strength of the reinforcing fibres, and their ultimate properties depend on the characteristics of both the matrix and the fibres, as well as on the adhesion strength at the interface. Moreover, these composites can be easily manufactured by using standard techniques, such as injection moulding and hot press compression (Khandker 2006). In jute composites, the jute is used as the reinforcement material and the other may be used as the matrix material. Composites may be made from jute/polyester, jute/ acrylic, jute/ polypropylene, jute/PLA (polylactic acid) fibres etc. Jute/PLA composites are unique and novel due to their biodegradable nature. Jute is a natural plant fibre and PLA fibre is made from corn. A number of different research projects have been carried out in this field by various researchers.

The world production of jute, kenaf and allied fibres was 3,460,500 tonnes in 2011-2012. The Jute fibre is cheap, durable and has some superior mechanical properties, such as high modulus, high strength and low creep. Fibres reinforced composite materials are used in various fields of engineering materials. These include aerospace, automotive, construction, consumer goods, energy, marine, medical, offshore etc. (Brady and Brady, 2007). Generally, carbon or glass fibres are used as the reinforcing material but these are not biodegradable. The world is very much concerned about the environmental issues. So, biodegradability of any material is of great interest to the scientists. In this context, the jute fibre can be used together with the biodegradable polymer matrix as the reinforcement medium. The aim of the study is to utilize jute fibres in the composite materials. It is hoped that this research programme will enhance the understanding and benefits of using jute fibres as the reinforcement medium for composites and consequently will increase the usage of this special natural fibre in composite materials.

In this study, plain woven jute fabric of 158.38 g/m^2 was used as the reinforcing fabric. The base fabric was produced from the reinforcing fabric by needle punching polylactic acid fibre webs into this fabric. This base fabric was used as the substrate for the manufacture of thermoplastic composites. The fibre mass proportion of the composites was set at the carding and needle punching process. Natural fibre (jute) mass proportion of the composites was set at 35%. The PLA fibres of 37.6 mm staple length and 2.13dtex linear density were used as the matrix material for the fabrication of the composites. The mechanical properties of the finished composites were determined. These were 3-point bending test, tensile test and impact test. In addition, the ageing tests were performed for the determination of the extent of the biodegradability of these composites.

MATERIALS AND METHODS

Base Material and Matrix

Plain woven jute fabric was used as the reinforcing fabric for the production of base fabrics. The area density of the fabric was 158.38 g/m^2, the thickness was 0.86 mm and the bulk density was 0.0184 g/cm^3. The biodegradable thermoplastic polylactic acid (PLA) fibres were used as the matrix for the production of thermoplastic composites. The matrix fibres were needle punched into the reinforcing jute fibre to form the base fabric or substrate for the composites.

Production of Base Fabric

Nonwoven base fabric was produced by using polylactic acid (PLA) fibre cross-laid web lying on top of the jute fabric for making jute /PLA composite. 2 Kg of PLA fibre and 2 meters of the jute fabric were taken for making this nonwoven base fabric. The width of the jute fabric was 1 meter. Two pieces of the nonwoven base fabric were produced. At first, PLA fibres were opened manually. The fibres were spread on the lattice of the carding machine also manually. Automatex Laboratory Nonwoven Line, Nuova Automatex, Italy was used for the production of the base fabric. The line consists of a carding machine, cross lapper, needle loom and a thermal bonding unit. But in these experiments, the thermal bonding unit was not used.

The fibres are separated and oriented on the carding machine and converted into a single layer web. The basic function of the cross lapper is to accept a light weight fibre web and produce a heavier web by laying the lightweight web in layers, across the width of the delivery lattice. The jute fabric was fed underneath the cross lapper and it advanced slowly with the help of roller of the cross-lapper. The fibres were laid on top of the jute fabric in multiple layers to make it heavier. The cross-laid web and the jute fabric are mechanically bonded together by the needle loom. The purpose of needling is to intertwine the fibres and the jute fabric to achieve the cohesion of the fibre in the fabric, which improves the mechanical properties of the fabrics, especially the delamination resistance of the layers in the composites is significantly improved.

The conical needles were supplied by Foster Needles, USA. The length of the needle was 8.89cm, which was selected due the aimed higher needle penetration depth required during the base fabric production.

This novel method of manufacturing composites materials decreases the delaminating tendencies of the composite and improves the reinforcement material-matrix interface. At the end of the needle punching process, the jute/PLA base fabric was produced with a proportion of 35% and 65% respectively. This specific technique of the production of the base fabric for composite manufacture used in this work has been patented (Anand 2013).

Preparation of Composite

At first, the base jute/PLA nonwoven fabrics were cut according to the template size. The size of the template was 17cmx17cm. Eight layers of the nonwoven base fabric were arranged in 0, 90, +45, -45, -45, +45, 90, 0 degrees orientations to make the final composite an isotropic. The area density of the specimens were determined. The average area density of the specimen of eight layers was 107 gms for template size. The composite material was produced by using the hot press machine. The specimen was secured between two steel plates and was inserted into the hot press machine (TH&J Daniels Ltd, Stroud, England). The specimen was kept in the machine for 2.5 minute at 160℃ temperature and 25 kg/cm^2 pressure. After the hot press, the specimen was cooled for 2.5 minutes at a pressure of 24.2 MPa on a 10.16 cm diameter ram. Then the specimen was removed from the steel plate and its edge was cut by a band saw machine. The average area density and thickness of the fabricated composites were 3,328 g/m^2 and 2.88 mm respectively.

Mechanical Properties of Composites

In order to fully characterize the mechanical properties of the composite, the 3-point bending, tensile and impact tests were performed and fully evaluated.

3- Point Bending Testing:

Instron series 4303 tensile testing machine was used for performing the 3-point bending tests. The applied load was 20N, the specimen length under bending was 10 cm and the width was 2 cm. The load was applied on the specimen at a speed of 2 mm/min. At a load of 20N, the machine was stopped at the highest deflection. Three tests per specimen were performed and the average values were recorded. All the data related to the test was recorded and stored in a computer connected to the Instron tensile testing machine. In this test, at the highest load of 20N, the average deflection was 2.02 mm.

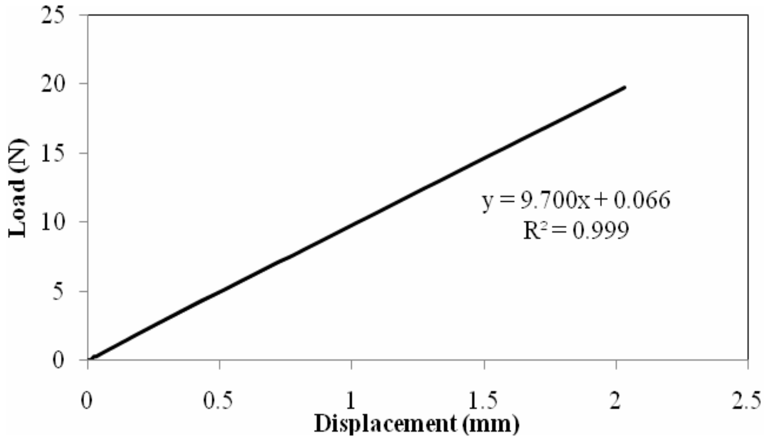

Fig. 1. Average Load - Displacement of Bending (Jute/ PLA Composites)

Tensile Testing:

For the determination of the tensile strength, 3 specimens for each direction of the fabricated jute/PLA composites were taken, the directions were 0, 90 and 45 degrees. The specimen's sizes were 16 cm x 2 cm and the thickness was 2.88 mm. The gauge length during testing was 10 cm. Instron 3369 machine was used for performing the tests. In every case, the average of the three values was recorded. For performing the tests in each direction, the composite specimens used were different for each direction. So, the maximum load values were slightly different. At 0 degree direction, the maximum load was 3620N and the breaking extension 2.15%, at 90 degree direction the maximum load was 3779N and the breaking extension was 2.18% and at 45 degree direction the maximum load was 3103N and the breaking extension was 1.72%.

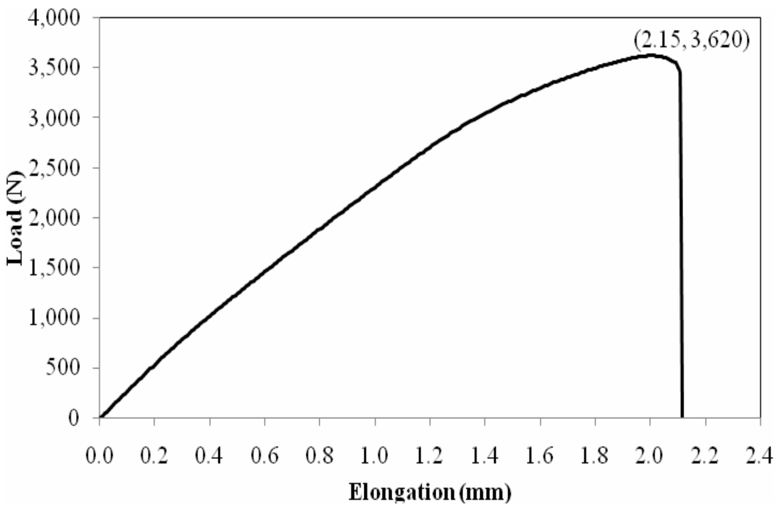

Fig. 2. Average Load-Elongation Curve (Jute/PLA Composite) at 0 Degree Direction (Machine Direction)

Impact Testing:

The Impact testing is the testing of an object's ability to withstand high velocity loading. An impact test is a test for determining the energy absorbed in fracturing a test piece at high velocity. Generally, it occurs when one object strikes another object at a relatively high speed.

Two specimens of the fabricated jute/PLA composites, measuring 10cmx10cm each specimen, were taken for testing the impact loading. The Dynatup Impact Tester was used for performing the tests. The mass of the impulse load used was 2.60 kg, the diameter of the striking spindle was 16 mm and the heights used were 200 mm and 400 mm. At 200 mm height the specimen was able to withstand the energy, but at 400 mm height, with the same applied load, the specimen was fractured due to the increase of the total energy (i.e. potential plus kinetic energy).

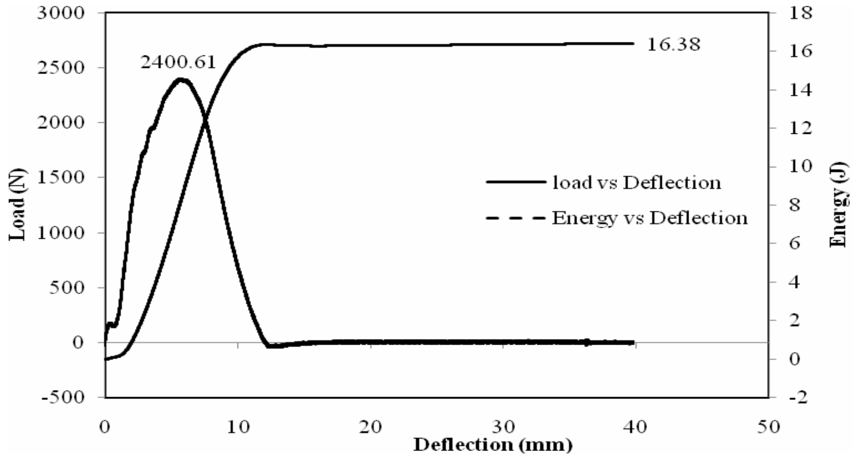

Fig. 3. Impact Test of Jute/PLA Composite (at 200mm Height and a Velocity of 1.99m/s)

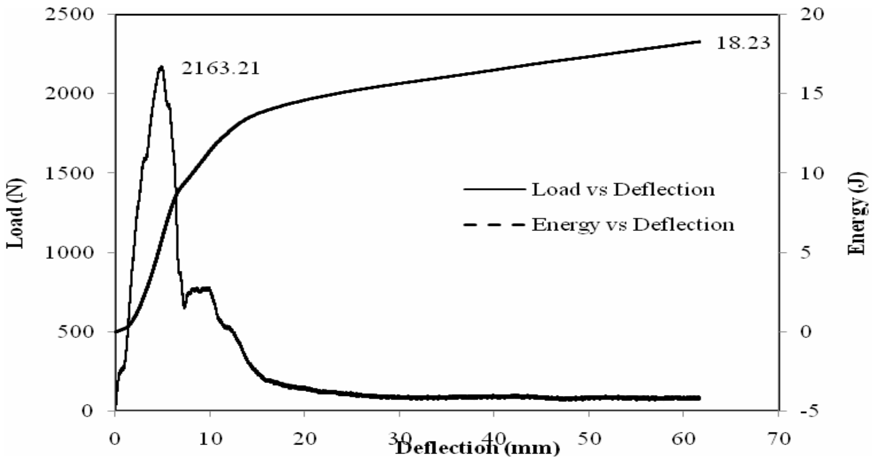

Fig. 4. Impact Test of Jute/PLA Composite (at 400mm Height and a Velocity of 2.701m/s)

The fabricated composite was able to withstand an energy of16.38 joule, but it was punctured at an impact energy of 18.23 joules.

Ageing Properties of Composites

During the ageing testing of these composite materials, the loss in the area density and the tensile strength of the composites in certain conditions and agents were considered in order to determine their ageing properties. The composite plates were cut 30 mm x 40 mm rectangular specimens for the ageing tests. The ageing was determined by measuring the mass loss of the specimens in different ageing conditions. For the determination of the loss of strength of the composites, the specimens were cut 20 mm wide and the gauge length was 10 cm, which were the same as the earlier tensile strength testing of the composites.

The ageing process was applied to the composites in four different conditions. These were; water; salt water; acidic; and alkaline states. Composite specimens were aged upto 30 days and 60 days under standard atmospheric conditions. The ageing procedure of the composites is given in Table 1.

Table 1. Ageing Procedure of Composites

Ageing condition	Agent used	Quantity
Water	Distilled Water	100%
Salt Water	Nacl	10%
Acidic (pH4)	Acitic Acid	Just a drop for 250 ml of water
Alkaline (pH10)	Sodium Hydrogen Carbonate	8 g/l

After 30 days and 60 days of ageing in different conditions, the specimens were taken out from their ageing medium, weighted in wet condition and then dried. The conditions used for the drying process were 50° for 24 hours. After drying, the specimens were kept in a desiccator for 24 hours. The water absorption characteristics of the composites were evaluated by the relative increase in the mass, defined by M_t, according to the following equation:

$$M_t = \frac{W_t - W_o}{W_o} \text{ X } 100\ (\%)$$

Where, W_o is the mass of the un aged specimen and W_t is the mass of the aged specimen (before drying, in wet condition)

The mass loss W_{loss} (%) was calculated by using the following formula:

$$\text{Mass loss } (\%) = \left[\frac{\text{initial mass (g)} - \text{final mass (g)}}{\text{initial mass (g)}}\right] \text{ X } 100$$

Where, initial mass and final mass are the masses of the specimen before ageing and after ageing and drying. Similarly, the strength loss was calculated using the following formula:

$$\text{Strength loss } (\%) = \left[\frac{\text{initial strength (N)} - \text{final strength (N)}}{\text{initial strength (N)}}\right] \text{ X } 100$$

Where, the initial and the final strengths are the strengths of specimen before and after ageing and drying.

Table 2. Ageing Test Results of Jute/PLA Composites in Different Ageing Conditions

Ageing condition	Water uptake (%) after		Mass loss (%) after		Tensile strength loss (%) after	
	30 days	60 days	30 days	60 days	30 days	60 days
Water	12.70	19.50	2.7	3	24.19	37.07
Salt water	10.26	25.38	0	0	34.53	38.78
Acidic	16.07	22.02	0	1.6	42.20	47.79
Alkaline	26.84	30.10	0	0	40.41	58.59

The novel characteristic of the fabricated composites is their biodegradability. The biodegradability tests were conducted by following the ageing tests. The ageing tests of the fabricated composites were carried out in four different ageing media (water, salt water, acidic, and alkaline). After 30 days and 60 days of ageing, the composites have absorbed water in every medium due to their hygroscopic character and in each case the uptake of water in the composite increased with the increase in the ageing time. For example, during ageing in water, the water uptake was 12.70% after 30 days and increased to 19.50% after 60 days of ageing.

After 30 days of ageing, the mass loss occurred only in water by 2.7%, but after 60 days of ageing, the mass loss increased in water to 3.0% and also in the acidic medium, the mass loss was 1.6%. The loss in the tensile strength of these composites was significant after ageing for 30 and 60 days in all four media. The composites lost from 24.19% to 42.2% strength after 30 days, but these values were much higher after 60 days of ageing, which ranged from 37.07% in water to 58.59% in the alkaline medium. The full ageing test results are given in Table 2.

RESULTS AND DISCUSSION

The jute/PLA composites comprising of 35% jute and 65% PLA fibres by weight were manufactured by using a novel technique. The area density and thickness of the composites were 3.328 kg/m^2 and 2.88 mm respectively. The 3-point bending tests, tensile tests at 0°, 90°and 45° directions, and impact tests at 200 mm and 400 mm heights with 2.60 kg loading were performed on the fabricated composites. The ageing tests were also carried out for ascertaining the ageing characteristics of the fabricated composites in four different ageing media and conditions.

In the 3-point bending tests, the applied load was 20N and the speed of load application was 2 mm/min, which yielded an average deflection of 2.02 mm. The deflection increased linearly as the loading was increased. The curve followed a linear regression equation of $y = 9.7004x + 0.0665$ and a value of $R^2 = 0.9999$ was obtained, which confirms the high accuracy of the linear relationship.

The tensile tests were carried out in three different directions of the fabricated composite, at 0°, 90°, 45° directions. The breaking loads and breaking extensions were 3620N, 3779N and 3103N, and 2.15%, 2.18% and 1.72% respectively for the above three directions. These values are similar to those found in other reported publications, in which the natural fibre composites were tested and analysed (Ku *et al.* 2011). The average tensile strength of the fabricated composite was 60.77MPa, but Bax and Mussing reported a tensile strength of 54.15Mpa in their flax/PLA composite of 30% fibre content (Bax and Mussing, 2008).

The impact loading is very important for any type of composite material. For the determination of the impact loading, Dyanatup Impact Tester (Instron series 9250) was used. The applied load was 2.6 kg and the heights used were 200 mm and 400 mm respectively. At the application of the loading from 200 mm height, the specimen was not damaged, but when loaded from the 400 mm height, it was fully punctured. When striking from 400 mm height, the highest load recorded was 2163.21N, with a deflection of 62 mm, a velocity of 2.70 m/sec and the total absorbed energy was 18.22 joules. On the other hand, at the application of loading from 200 mm, the highest load was 2400.6 N, with a deflection of 40 mm, a velocity 1.99 m/sec and the total absorbed energy was 16.37 joules. The impact properties determined in the work are very similar to those found in other natural-fibre reinforced composites. The allowable impact energy of the fabricated composite was 81.45kj/m^2 (drop mass impact), but Bax and Mussing reported an impact energy of11.3kj/m^2 in their flax/PLA composite of 30% fibre content (charpy impact) (Bax and Mussing, 2008).

From the ageing test results, it was shown that the mass loss of the composites occurred in water and acidic media, but in other media there was no mass loss up to 60 days. The reason is that during ageing for 30 days in water and 60 days in acidic conditions, the microstructure of the composite materials began to decompose. The other two media (salt water and alkaline) resisted the growth of the bacteria in the composites that are responsible for the decomposition of the composite materials during that period. Significant losses in the tensile strengths of these composites have been obtained during the ageing periods in different ageing media. The ageing characteristics of these fabricated composites confirm their biodegradability characteristics, which is one of the novelties of jute/PLA composites (Liu *et al.* 2009; Uddin *et al.* 1997; Kumar *et al.* 2005 and Plackett *et al.* 2003).

CONCLUSION

The Jute fabric reinforced PLA composites can be manufactured at 160° temperature and 25 kg/cm^2 pressure for 2.5 minute by using a hot press method. In 3-point bending tests, at 20N load for specimen size of 10 cm x 2 cm, the maximum deflection was 2.02 mm. The average tensile strength of these composites was 60 MPa. These composites can withstand 16.37 joules of impact energy under the impulse area of 2 cm^2. The ageing tests have proved that these natural fibre reinforced composites are biodegradable over time. The jute/PLA composites can be used in the applications where biodegradability is a desirable property for the eco-friendly disposability of the composites.

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