EFFECT OF FIBRE LOADING AND ALKALINE TREATMENT ON MECHANICAL AND SOIL BIODEGRADING OF LOCUST BEANS HUSK FILLED UNSATURATED POLYESTER COMPOSITES

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Abstract: The paper examines the effect of fibre loading and alkaline treatment on tensile strength, impact and soil biodegrading of locust beans husk filled unsaturated polyester composites. The fibre was collected from the fruit of locally available Parkia biglobosa tree. The composites were produced using several percentage fibre loadings (0, 10, 20, 30, 40 and 50 weight percent). Locust beans husk was used as the reinforcing fibres in unsaturated Polyester resin. The composites was produced by manual mixing of the Locust beans husk and the unsaturated polyester resin using a glass mold. The effect of fibre loading on the tensile strength, flexural strength, and impact strength of locust beans husk/unsaturated Polyester composites shows that the strength increases as the fibre loading increases from 10 wt % fibre loading to 30 wt % fibre loading and further incorporation of fibre beyond 30 wt % fibre loading results to decrease in strength. It was found that optimum values and significant improvement were at 30 wt % fibre loading. The alkaline treatment was found to be effective in improving the tensile, flexural and impact strength by 25.8 %, 98.2 % and 25.0 % respectively. During samples soil burial process, it was observed that the composites became more susceptible to degradation with increase in fibre loading from 10wt % to 50 wt % fibre loading. These properties relate positively with some reported properties for natural fibre reinforced polymer composites.

Keywords: Locust beans husk, soil burial, tensile strength, unsaturated polyester

1.0 Introduction

The extensive use of synthetic fibre reinforced polymer composites has a predisposition to decline because of high initial costs and, more importantly, their adverse environmental effect (Mohanty and Drzal, 2001; Mishra *et al.*, 2002). Today, the growing environmental awareness throughout the world has brought about the shift from synthetic fibres towards composites made from natural reinforcing constituents; natural fibre and natural particulate fillers which are more ecologically friendly (Wretfors and Svennerestedt, 2006).

Research has shown that large amounts of agriculturally produced wastes are generated yearly across Nigeria and the potential uses of these byproducts have not been effectively utilized. In recent time, the trend in developing a composite material using this byproduct as reinforcement has been a worthy area of interest because of their easy access, low cost and biodegradability. Composites developed by various researchers by combining natural fibres and synthetic fibres with epoxy, polyester, phenolic, poly vinylester, poly urethane resins, etc., are well well-known (Jawaid and Abdl, 2011; Sathishkumar *et al*., 2014). This research serves as a means of obtaining data on Locust beans husk filled thermoset resin composites materials and will probably lead to alternative materials in some areas of applications. Thus, the study aims to take the advantage of unutilized locust beans husk as filler in the preparation of composites.

Figure 1: Locust beans husk

2.0 Materials and Methods

Locust beans husk obtained from Parkia biglobosa tree was collected from Auta local farm in Chikun Local Government Area in Kaduna state, Nigeria, while commercial unsaturated polyester resin, cobalt naphthenate which serves as accelerator and Methyl Ethyl Ketone Peroxide (MEKP) serving as the hardener were procured from GZ industrial suppliers in Lagos state, Nigeria.

2.1 Fibre Preparation

Preliminary cleaning of the collected locust beans husk was done using water, the husk were then dried directly under the sun for 12 hours and prepared for pulverization. The pulverizing machine (Thomas-Wiley Laboratory Mill Model: 4) was used to pulverize the fibre into short fibre length.

2.2 Composite Preparation

The composites were fabricated with 0, 10, 20, 30, 40 and 50 weight percent (wt %) fibre loading. This was prepared by mixing the various ratios of the prepared fibres with the unsaturated polyester (UPE) resin made up of 100 % of each as in weight fractions and in the process, varying amount of both fibre and unsaturated polyester resin using electronic weighing balance. Component of each composite sample was obtained using the formulations shown in Table 1 and combination was done in weight percent (wt %) fibre loading. The mixing was achieved by mechanical mixing. 2 ml of cobalt naphthenate as accelerator and 2ml of Methyl Ethyl Ketone Peroxide (MEKP) as hardener were used. The mixing was poured into glass mould previously covered with aluminum foil serving as releasing agent. The sheets produced were stored at room temperature for 14 days for complete curing operation. The produced composites were thereafter shaped into various dimensions according to ASTM standards for various tests.

Table 1: Percentage combination of Locust beans husk and reinforcing Matrix (unsaturated polyester)

Fibre (wt %)	**0**	**10**	**20**	**30**	**40**	**50**
Matrix (wt%)	100	90	80	70	60	50

2.3 Tensile Strength Test

Instron Universal testing machine (3369 model) was used to carry out tensile test with maximum load of 10 KN as described in ASTM method D638. A cross speed of 2mm/min was used. Each tensile specimen was positioned in the Instron Universal tester and then subjected to tensile load. Five specimens for each composite were tested and statistical averages for each set of results were recorded.

2.4 Flexural Strength Test

Universal Testing Machine with maximum load of 100 KN and a cross head speed of 5 mm/min was used to carry out the three point bending test and maintained in accordance with ASTM D790-03 to measure the flexural strength and flexural modulus of the composites. The flexural strength (FS) and flexural modulus of the specimen was determined using equation 2.1 and 2.2 respectively.

Flexural Strength (F_s) = $3PL/2bt^2$ ………..………………………………………………… (2.1)

Flexural modulus (F_m) = $PL^3/4bwh^3$.. (2.2)

Where L is the span length of the sample in mm, P is the load applied in Newton or Pascal (pa), b and t are the width and the thickness of the specimen respectively in mm, while h is the specimen thickness (mm) and w indicates the depth of the deflection (mm).

2.5 Impact Strength Test

Pendulum-type hammer impact testing machine (Charpy Impact testing machine, capacity 15 J and 25 J, Serial Number 412-07-15269c, manufactured by Norwood instruments limited, Great Britain) was used. The specimen was supported as a horizontal simple beam and is broken by a single swing of the pendulum with the impact line midway between the supports. It is capable of measuring the impact energy expended in breaking a test specimen, the value of which was taken as equal to the difference between the initial potential energy in the pendulum and the energy remaining in the pendulum after breaking the test specimen.

2.6 Soil Burial

The biodegradability of Locust beans husk filled unsaturated polyester (UPE) composites were carried out by soil burial method to stimulate natural fibre composite. Rectangular samples with 25 mm x 25 mm x 3mm dimensions were oven dried until their weight become persistent (W_1). The samples were then buried in the soil at a depth of 20 cm from the surface of the soil for 75 days. The samples were carefully taken out of the soil for testing every 15 days and then washed with distilled water to remove the adhered sand particles from the surface of the samples and was then oven dried at 55 ^{0}C until their weights become constant (W_2). The percentage of weight loss (% WL) was calculated using equation 2.3.

$$\%WL = [(W_1 - W_2)/W_1] \times 100 \quad (2.3)$$

3.0 Results and Discussion

All assessments were carried out in agreement with ASTM standards. The results of the effect of the fibre loading on tensile strength, tensile modulus, flexural strength, flexural modulus, impact strength, and soil burial analysis of both treated and untreated locust beans husk reinforced unsaturated polyester composites are presented in Figures 1 to 5.

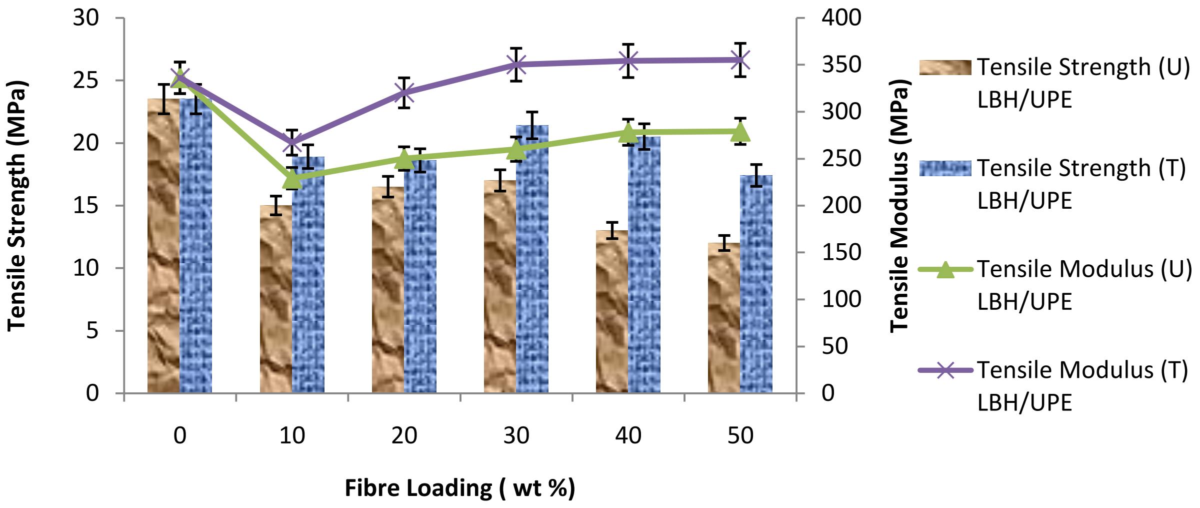

Figure1: Effect of fibre loading on the tensile strength and modulus of NaOH treated and untreated locust beans husk/unsaturated polyester (LBH/UPE) composites.

3.1 Tensile Properties

The results of the effect of fibre loading on the tensile strength and modulus of unsaturated polyester filled with locust beans husk composites is shown in Figure1. It can be observed that as the fibre loading increases from 10 wt % fibre loading to 30 wt % fibre loading, it shows a gradual increase in strength and subsequently decreases the tensile strength beyond 30 wt % fibre loading, indicating an initial gradual reinforcement. The decrease in tensile strength may be attributed to interruption in the stress transfer along the applied force and lack of significant interfacial interaction between fibre and the matrix resulting in poor wettability of the fibre by the matrix, hence weakening the fibre/matrix adhesion leaving fibre agglomeration and producing stress concentration points in the composites, the broken end of the short fibre form during tensile deformation was believed to induce crack in the matrix leading to reduction in tensile strength thereby signify the decrease (Shajan *et al.*, 2012).

The tensile strength of untreated LBH/UPE composite was at maximum and minimum values at 30 wt % fibre loading with 17 MPa and 12 MPa respectively, while that of the unfilled resin was found to be higher with 23.5 MPa. It was observed that the tensile strength of the unfilled resin was reduced to 6.5 MPa (38.2 % reduction) on addition of 30wt % fibre loading but show a decrease in tensile strength as the fibre loading increases from 30 wt % to 50 wt % fibre loading.

The study also shows that the tensile modulus increases as the fiber loading increases from 10 wt % fibre loading to 50 wt % fibre loading. It has been reported by many researchers that the modulus increases with increase in fibre loading (Raju *et al.*, 2012). According to Adams et al. (1969), the relative stiffness of a material is indicated by its modulus. Incorporating locust beans husk has improved the stiffness of the matrix since the modulus of the composite increases as locust beans husk fibre loading was increased. The modulus of untreated locust beans husk

(U LBH/UPE) composite was at maximum and minimum values at 50 wt % fibre loading and 10 wt % fibre loading with 279 MPa and 229 MPa respectively, while unfilled UPE resin was 336 MPaas shown in Figure 1. The increase in tensile modulus with increase in fibre loading is consistent with Nara et al. (2012), where it was reported that the tensile modulus, which is an indicator of load bearing capacity, increases with fibre loading. The effect of fibre loading on the tensile strength and modulus of sodium hydroxide (NaOH) treated locust beans husk/unsaturated polyester (T LBH/UPE) composites is shown in Figure1. It can be observed that the tensile strength increases from 10 wt % fibre to 30 wt % fibre loading indicating reinforcement and gradually decrease with further increase in fibre loading beyond 30 wt % fibre loading. The decrease might be attributed to poor dispersion of fibre in the matrix in addition to poor fibre-matrix interaction as established by Salma et al. (2010).

The maximum and minimum values were observed at 30 wt % fibre loading and 50 wt % fibre loading with 21.4 MPa and 16.5 MPa respectively. The high tensile strength of the samples due to sodium hydroxide treatment was an indication of improved interaction and stress transfer between the fibre and matrix. The result shows 29.7 % improvement at 30 wt % fibre loading.

The modulus of the treated samples also show a corresponding increase as the fibre loading increases. The modulus of treated locust beans husk/unsaturated polyester was at maximum and minimum values at 50 wt % and 10 wt % fibre loading with 355 MPa and 267 MPa respectively as against the neat resin valve of 336 MPa. It can be seen that treated samples gave more strength than untreated samples. It is expected that the treated samples will have more surface area interaction with the resin thereby improving the stress transfer process that will ultimately increase the tensile strength. The improved strength of the composite as a result of sodium hydroxide treatment can also be linked with reduction in lignin content of the fibre as lignocellulosic material are originally composed of cellulose in lignin matrix, reduction of the lignin in the fibre will enhance better binding between the resin and the fibre thereby enhancing the strength. The modulus increased as the fibre loading increases. However, it can be seen that the treated sample composites improve greatly at 50 wt % fibre loading more than that of the untreated composite samples. Modulus of locust beans husk/unsaturated polyester (T LBH/UPE) composite at 50 wt % was 355 MPa while that of untreated locust beans husk (U LBH/UPE) composite at the same fibre loading was 279 MPa. The improvement in the tensile modulus of the alkaline treated fibre is attributed to the fact that alkaline treatments increase the effective surface area and percentage crystalinity index of fibre due to the removal of natural cementing materials leading to a better packing of the cellulose chains, in most cases impacting positively on the mechanical properties of fibre reinforced composites (Santoe *et al.*, 2011).

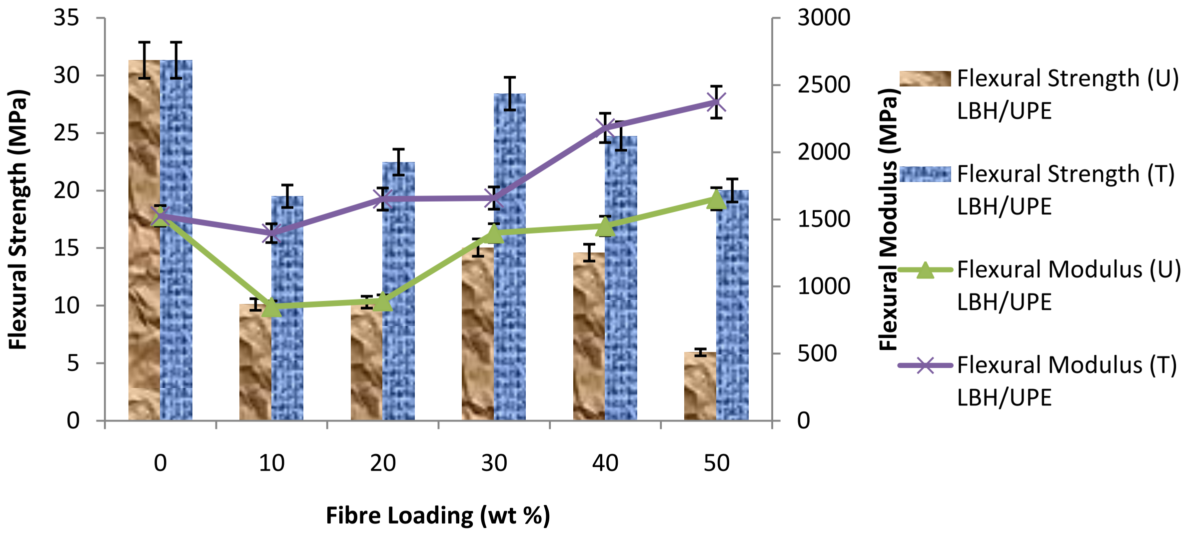

Figure 2: Effect of fibre loading on the flexural strength and modulus of NaOH treated and untreated locust beans husk/unsaturated polyester (LBH/UPE) composites.

3.2 Flexural properties

Figure 2 shows the relationship between the flexural strength and the modulus. It can be seen that an increase in fibre loading from 10 wt % to 30 wt % fibre loading produces a corresponding increase in the strength of the composite indicating that the composite presented limited plastic deformation with a tendency for rupture just beyond the elastic limit at 40 wt % fibre loading, a sudden fracture occurs indicating a brittle behavior in the strength of the composite. The flexural properties of the composites depend critically on the microstructure of the composite and the interfacial bonding between the reinforcement and the matrix (Subita and Pardeep, 2013), from the results, it was observed that the flexural strength of the unfilled unsaturated polyester resin was reduced by 67.7% on addition of 10 wt % fibre loading as shown in Figure 2, that is, increasing the fibre loading from 40 wt % to 50 wt % fibre loading and effect the strength to decrease by 31.5%. The decrease is due to agglomerate formation at higher fibre loading which is also observed in the tensile strength behavior (Sarojini, 2013). Similar results have been reported by other researchers that the flexural strength decreased after 40 wt% fibre loading (Raju *et al*., 2012).

The maximum and minimum flexural strength values for untreated LBH/UPE composites were obtained at 30 wt % fibre loading and 50 wt % fibre loading with 15.04 MPa and 5.94 MPa respectively on the other hand, the study shows that flexural modulus increases with an increase in fibre loading.

The relative stiffness of a material is indicated by the modulus (Adams *et al*., 1969). Figure 2 also shows the effect of fibre loading on the flexural strength and modulus of (NaOH) treated locust beans husk/unsaturated polyester composites. It can be seen that the flexural strength increases with increase in fibre loading from 10 wt % fibre loading to 30 wt % fibre loading and decreases from 40 wt % fibre loading to 50 wt % fibre loading. Increasing the fibre loading by 10 wt % fibre loading decreases the flexural strength of the unfilled unsaturated polyester resin by 37.7% as against 67.7 % obtained for untreated LBH/Unsaturated Polyester composites, which is an indication of improved interaction and better stress transfer between fibre and matrix. Maximum and minimum values were at 30 wt % and 50 wt % fibre loading respectively with 28.41MPa and 20 MPa respectively.

The treated samples show a corresponding increase in their modulus. The modulus of treated LBH/unsaturated Polyester (LBH/UPE) composite was at maximum and minimum values at 10 wt % and 50 wt % fibre loading respectively with 2371.7 MPa and 1396.6 MPa respectively as against unfilled resin value of 1525.2 MPa. It can be seen that the treated LBH/UPE composites exhibited higher flexural strength compared to their corresponding Untreated LBH/UPE composites. It is expected that the treated fibre shows better fibre/matrix interaction thereby improving the stress transfer efficiency (Screenivasan *et al*., 1996 and Kin *et al*., 2010).

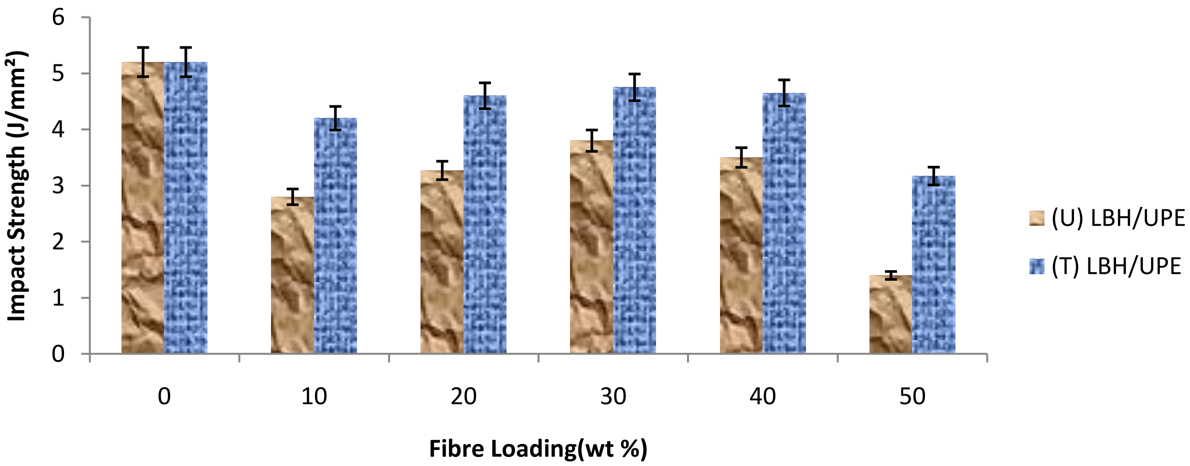

Figure 3: Effect of fibre loading on the Impact strength of NaOH treated and untreated locust beans husk/unsaturated polyester (LBH/UPE) composites.

3.3 Impact Strength Test

The results in Figure 3 show the effect of fibre loading on the impact strength of NaOH treated and untreated locust beans husk/Unsaturated Polyester (LBH/UPE) composites. It can be seen from the results that as the fibre loading increases from 10 wt % fibre loading to 30wt % fibre loading, the impact strength increases and beyond 30 wt % fibre loading the strength decreases progressively. Addition of 30 wt % fibre loading of locust beans husk into the unfilled Unsaturated Polyester resin decreases the impact strength by 33.3 %. The impact strengths of the unfilled resin reduce from 5.2 Jmm^{-2} to 3.8 Jmm^{-2} at 30 wt % fibre loading. The material under study also shows lower values of impact strength in which the minimum impact strength value of 1.4 Jmm^{-2} was recorded at 50 wt % fibre loading.

Figure 3 also show the results of the effect of fibre loading on the impact strength of treated locust beans husk/Unsaturated polyester composites. The results show similar trend, the impact strength increases as the fibre loading increases from 10 wt % to 30 wt % and gradually decreases at 40wt% fibre loading. The result shows that addition of 30wt % fibre loading into the unfilled unsaturated polyester resin decreases the impact strength by 9.5 %, reducing from 5.2 Jmm^{-2} to 4.75 Jmm^{-2}. The treated locust beans husk also shows lower values of impact strength in which the minimum impact strength value of 3.17 Jmm^{-2} was observed at 50 wt % fibre loading. It can be seen that the treated locust beans husk shows higher impact strength than the untreated locust beans husk/unsaturated polyester composites. The treatment disrupts hydrogen bonding in the network structure, thereby increasing surface roughness (Demir *et al.*, 2006 and Seki *et al.*, 2011).

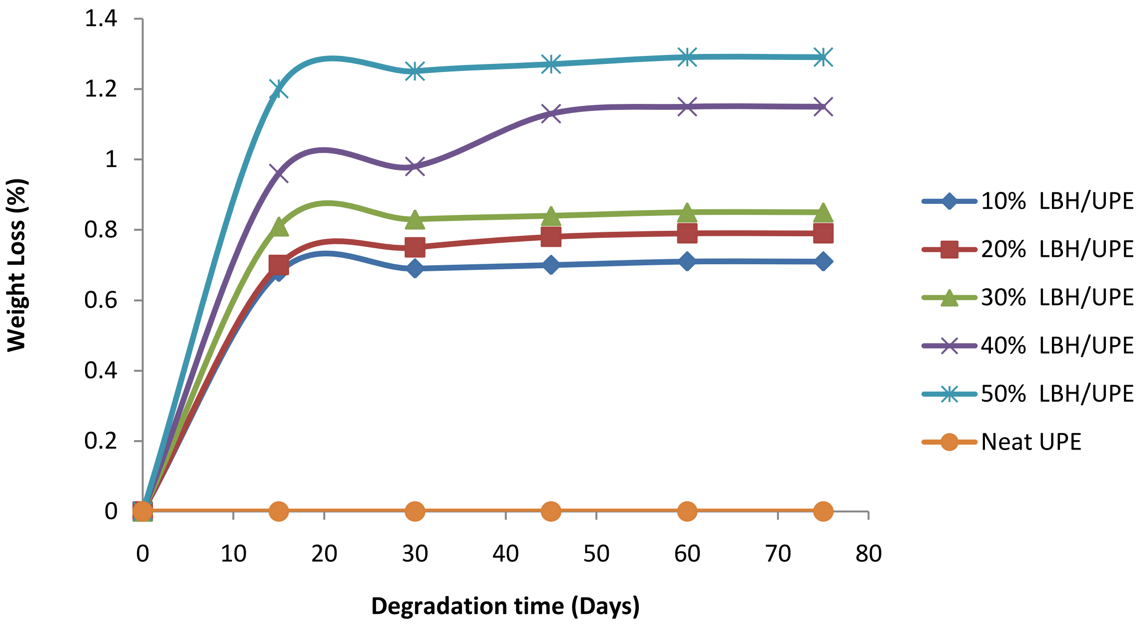

Figure 4: Effect of weight loss of untreated locust beans husk composite at varying fibre loadings as a function of degradation time.

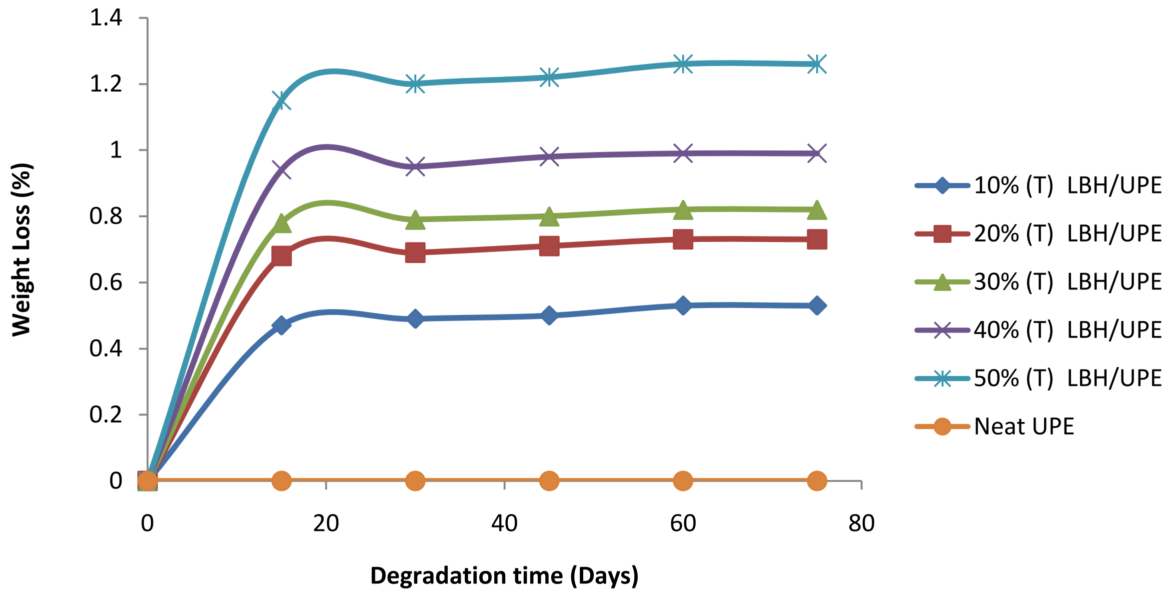

Figure 5: Soil burial on the weight loss of treated locust beans husk/UPE composites at varying fibre loadings as a function of degradation time.

Results in Figure 4 show the effect of soil burial on the weight loss of untreated locust beans husk composites at varying fibre loadings as a function of degradation time. The degradation of the samples was evaluated by measuring the weight loss of samples which were buried in the soil. The outcome was initially measured on a daily basis and results obtained was not significant, hence, the results was then measured at an interval of 15days over a span of 75days as depicted in Figures 4 and 5. Results show that an increase in fibre loading from 10wt% to 50wt% fibre loading produced an increase in percentage weight loss of untreated locust beans husk/UPE composite over the span of 75days. This may be attributed to low surface area of interaction with the resin thereby enhancing degradation leading to increase in weight lost. Water uptake gradually increases producing a corresponding weight loss until saturation occurs thereby producing a constant weight loss. The minimum percentage weight loss for untreated locust beans husk unsaturated polyester (LBH/UPE) composite was obtained at 10wt% fibre loading with 0.71% after 75days, while the maximum weight loss was obtained at 50wt% fibre loading with 1.29% after 75days in the soil. The increase in water absorption as fibre loading increases from 10wt% to 50wt% fibre loading causes the degradation of fibre–matrix interface region, resulting in reduction in the mechanical properties along with the change in dimensions of the buried untreated LBH/UPE composites.

Figure 5 shows the weight loss of treated locust beans husk/UPE composite at varying fibre loading as a function of degradation time. It can be seen that sample percentage weight loss increases as the fibre loading increases from 10wt% to 50wt% fibre loading as the degradation time increases. Results show that there was a gradual rise in percentage weight loss from initial day to day 30 indicated by the Plateau. The rate of weight loss gradually reduces until a constant rate of percentage weight loss was achieved in 75days. The treated LBH/UPE composite at

50wt % fibre loading shows higher percentage loss with 0.99% while the minimum was attained at 10wt% fibre loading with 0.53 %. The neat unsaturated Polyester resin serving as the control sample does not show any significant weight loss which is attributed to its hydrophobic nature. The low percentage weight loss of the treated samples was an indication of improved interaction between the locust beans husk and unsaturated polyester resin.

From the results, treated samples gave less percentage weight loss as the degradation time increases compared to untreated samples; this may be ascribed to the increase in the fibre surface roughness as a result of treatment resulting in better mechanical interlocking (Bledzki and Gassan, 1999).

4.0 Conclusion

The research work has successfully fabricated treated and untreated Locust beans husk filled unsaturated polyester composites and has served as a means of obtaining data on the fabricated composites. It was also found that optimum values and significant improvement were at 30 wt % fibre loading for both treated and untreated composites. Maximum and minimum tensile strength, flexural strength and impact strength values for treated and untreated locust beans husk filled unsaturated polyester composites were obtained at 30 and 50 wt % fibre loading with 21.4 MPa and 17 MPa (maximum and minumun tensile strength), 28.41 MPa and 15.04 MPa (maximum and minimum flexural strength) and 4.75 mpa and 3.8 mpa (maximum and minimum impact strength) respectively. The composition can be used for load bearing applications and can be found useful in humid environment due to its less water absorption capacity. In view of this, the approach is useful for the current effort towards research and development aimed at economic viable and practicable industrial end-use applications.

References

[1] Mohanty, A.K., and Drzal, L.T. (2001). Surface modification of natural Fibres and Performance of the Resulting Biocomposites. An overview–composite interface,Vol.8, No.5, pp. 313-343.

[2] Mishra, S., Misra, M., Tripathy, S.S., Nayak, S.K., Mohanty, A.K.J.(2001). Reinf. Plast. Comp., 20;4

[3] Wretfors, C., and Svennerstedt, B. (2006). Biofibre Technology used in Military Applications. An overview. JBT Rapport, No. 142, pp. 1-40.

[4] Jawaid, M., Abdul Khalil, H.P.S. (2011). Cellulosic/synthetic fibre reinforced Polymer Hybrid composites: A review. Carbohydr. Polym., 86, 1–18.

[5] Sathishkumar, T., Naveen., J., Satheeshkumar, S. (2014). Hybrid fibre reinforced polymer composites—A review.J. Reinf. Plast. Compos. 33; 454–471.

[6] Shajan, F., Vilaseca F., Llop M., Gironès J., Méndez J.A., and MutjeP. (2012). Chemical modification of jute fibres for the production of green-composites. *Journal of Hazard Materials.*144; 730-5.

[6] Raju, G.U., Kumarappa, S., Gaitonde, V.N., (2012). Mechanical and physical characterization of agricultural waste reinforced polymer composites *J. Mater. Environ. Sci. 3 (5):907-916.*

[7] Adams, D.F. Tsai S.W. (1969). The influence of random packing on transverse stiffness of unidirectional composites. *Journal of Composite Materials.* 3:368-381.

[8] Nara, R., Sultana, K., Ruhul, A., Khan, M.A.(2012). Bioplastiscs from renewable resources laboratory to market. *Abstract Paper American Chemistry*; 227:U279

[9] Salma, S., Fayeka M., Mahbub, H., and Azman H. (2010). Effect of Reinforcement and chemical treatment on the properties of jute-coir fibre reinforced hybrid polypropylene composites. *Fibres and polymers*. 15(5);1023-1028.

[10] Santos, P., and Pezzin, S. H. (2011). Mechanical properties of polypropylene reinforced with recycled-pet fibres. *Journal of Materials Processing Technology*143-144; 517-520

[11] Subita, B., and Pardeep K.V., (2013): "Effect of Graphite Filler on Mechanical Behaviour of Epoxy Composites" *International Journal of Engineering Technology and Advanced Engineering.*www.ijetae.com (ISSN 2250-2459, ISO 900:2008 Certified Journal, Volume 3,(2).

[12] Sarojini, S. (2013). "Synthesis and Characterisation of Graphene Based Unsaturated Polyester. Resin Composites''. Department of Advanced Material Process Technology Centre, Crompton Greaves Ltd., Kanjur Marg, Mumbai 400042, India. Transactions On Electrical And Electronics Materials, Vol.14, No. 2, pp 53-58.

[13] Screenivasan, S, Iyer, B.P., and Iyer, R. (1996). Influence of delignification and alkali treatment on the fibre structure of Coir fibre (cocos Nucifera). J. mater. sci., 31(3), 721-26.

[14] Kim, J.T., and Netravali, A.N. (2010). Mercerization of sisal fibres and effect of tension on mechanical properties of sisal fibre and fibre-reinforced composite. Compos. Part A-Appl. S., 14(9) 1245-52.

[15] Demir, H., Atikler, U., Balköse, D.,and Tıhmınlıoğlu, F. (2006). The effect of Fibre surface treatments on the tensile and water sorption properties of polypropylene–Luffa gourd fibre composites. Composites Part A: Applied Science and Manufacturing, 37(3), 447-456.

[16] Seki, Y., Sever, K., Erden, S., Sarikanat, M., Neser, G.,and Ozes,C.(2012). Characterization of Luffagourd fibres and the effect of water aging on the mechanical properties of its composite with polyester. Journal ofApplied Polymer Science, 123(4), 2330-2337.

[17] Bledzki, A.K, Gassan J., (1999). Prog. Polym. Sci., 24,221.