Effect of Silane Coupling Agent on the Tensile Properties of Rice Husk Flour (RHF) Polyester Composite.

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ABSTRACT

To determine, the effect of a silane coupling agent on an agricultural waste materials use as reinforcing fillers in thermosetting polymer composite. Polyester, as the matrix, and rice husk flour (RHF), as the filler, were used to prepare a particle-reinforced composite in order to determine testing data for the physical and tensile properties of the composite for both silane and NaOH treated filler according to the filler loading in respect to thermosetting polymer. In the sample preparation, five levels of filler loading (5, 10, 20, 30, and 40 wt. %) were designed, for silane, NaOH, and untreated filler. Test result shows that tensile strengths, modulus, maximum load, and micro-hardness of the composites show appreciable improvement with the use of coupling agent on the filler.

(Keywords: polyester composites, silane, rice husk flour, RHF)

INTRODUCTION

Recent investigations of polymer-based composite materials have opened new routes for polymer formulations and have allowed the manufacture of new products with optimal properties for special applications [10, 5]. In most cases, these composites improve the product design and reduce the material and energy consumption. Using natural filler to reinforce the composite materials offers the following benefit in comparison with mineral filler [7, 15], strong and rigid, light weight, environmental friendly, economical, renewable, and abundant resource. However, they have the disadvantage of degradation by moisture, poor surface adhesion to hydrophobic polymers, non-uniform filler sizes, and not being suitable for high temperature application, among others [1].

The improved performance of polymers and their composites in industrial and structural applications by the addition of filler materials has shown great promise and so has lately been a subject of considerable interest. Various kinds of polymers and polymer matrix composites reinforced with metal particles have a wide range of industrial applications such as in heaters, electrodes [19], and composites with thermal durability at high temperature [2]. These engineering composites are desired due to their low density, high corrosion resistance, ease of fabrication, and low cost [1, 7, 13]. Similarly, ceramic filled polymer composites have been the subject of extensive research in the last two decades. The inclusion of inorganic fillers into polymers for commercial applications is primarily aimed at the cost reduction and stiffness improvement [11, 21]. Along with fiber reinforced composites, the composites made with particulate fillers have been found to perform well in many real-operational conditions.

Studies are ongoing to find ways to use lignocellulosic filler in place of synthetic fillers. These natural fillers are especially being sought since the production of composites using natural substances as reinforcing fillers is not only inexpensive but also able to minimize the environmental pollution caused by the characteristic biodegradability [18], enabling these composites to play an important role in resolving future environmental problems. The need for materials that are non-toxic to the human body and have appropriate characteristics for specific purposes is ever increasing due to the lack of resources and increasing levels of environmental pollution. Thus, research is proceeding to develop composites using various recycled wastes [21], especially in developing composites using most agro-wastes as reinforcing fillers in polymers.

The chemical composition of different organic filler and fiber [14] are show in Table 1. As shown in the Table, Rice Husk Flour (RHF) is a Special lignocellulosic material. Although it contains the typical components of a standard lignocellulosic material, its lignin and hemicellulosic contents are lower than wood flour, whereas the cellulose content is similar. For this reason, the rice husk filler can be processed at higher temperatures than wood, which has thermal stability problems at temperatures above 200°C, [14].

The convenience of these composites lies in the fact that the ingredients are obtained easily from natural wastes and hence the composites can be made relatively easily. They can be used to resolve environmental problems and to produce products with various physical properties and effective functions. Lignocellulosic materials as reinforcing fillers in plastics, in place of the previously used inorganic substances and synthetic fibers, offer a major benefit in terms of environmental protection. The benefits offered by lignocellulosic materials include making the final product light [10], decreasing the wear of the machinery used, low cost, biodegradability [18], and absence of residues or toxic byproducts.

A number of natural occurring fillers and fiber in composite have been studied in the past. These include wood fillers [4] wheat straw, almond husk, ash rice husk [9, 20], pineapple leaf [16], sisal , coir, and rice husk [6]. These fillers introduce some advantages compared to traditional inorganic fillers, including their renewable nature, low density, nonabrasive properties, reasonable strength, and stiffness [17].

Luo and Netravali [13] studied the tensile and flexural properties of pineapple fiber. Belmeres *et al*. [1] studied sisal, henequen, and palm fiber, and found that they have similar physical, chemical, and tensile properties,. Fuad *et al.* [3] investigated wood base filler derive from oil palm wood flour. Lhalid et al. [12] also studied epoxy composite reinforced with the use of cotton fiber along with glass fiber.

MATERIALS AND METHODS

Experimental

Rice husk was obtained from a local rice mill dump, in southwest Nigeria. The rice husk was wash, dried and grounded to fine particle, and later sieve with BS/ISO 3310 into particle size of 53 µm. and dried in an oven at 105^{o}C to reduce moisture content. This was used for all the different percentage compositions of RHF that were used for the Composite preparation.

Sample Preparation

A commercially available Unsaturated Polyester Resin with Cobalt Octoate as Accelerator and Methyl Ethyl Ketone Peroxide (MEKP) as Catalyst was used as matrix for the composites. After the resin and filler has being thoroughly mixed, the mixture was cast onto the cavity of a steel mold, previously coated with a mould releasing agent and allowed to cure. Composites with amounts of RHF, ranging from 5, 10, 20, 30, and 40 wt. %, for untreated, NaOH, and Silane treated samples were manufactured.

Table 1: Dry Chemical Composition of Different Organic Filler and Fiber.

Organic filler	Cellulose (%)	Hemicelluloses (%)	Lignin (%)	Silica (%)	Moisture (%)
Pine (soft wood)	44.0	27.0	28.0	-	25
Yellow birch (hardwood)	47.0	31.0	21.0	-	25
Jute	73.2	13.6	13.4	-	10
Wheat straw	48.8	35.4	17.1	-	18
Rice husk	45.0	19.0	19.5	15	14

Sodium Hydroxide Treatment

200ml of 5% NaOH was used to soak 100g of Rice Husk Filler in a 600ml beaker about half an hour in order to activate the OH groups of the cellulose and lignin in the filler. Filler were then washed many times in distilled water and finally dried, in an oven at 105^{0}C to a constant weight. This was used to prepare the composite.

Silane Treatment

The structure of silane coupling agents, triethoxy vinyl silane (Alfa Aesar Chemical Co. Ltd. USA) is shown in Figure 1.

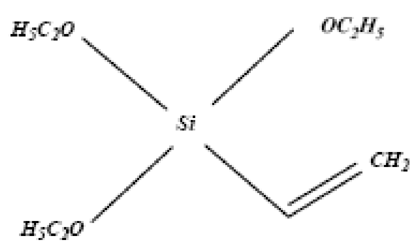

Figure 1: Structure of Silane Coupling Agents (tri ethoxy vinyl silane).

The filler were dipped in alcohol water mixture (60:40) containing 5% tri ethoxy vinyl silane coupling agent. The pH of the solution was maintained between 3.5 and 4, using the METREPAK Phydrion buffers and pH indicator strips, the mixture was allow to stand for one hour after which the filler were washed in double distilled water and dried in the oven.

Alkoxy silanes are able to form bonds with hydroxyl groups. Silanes undergo hydrolysis, condensation and the bond formation stage. By reacting with hydroxyl group of fillers, Silanols can form polysiloxane structures by reaction with hydroxyl group [22].

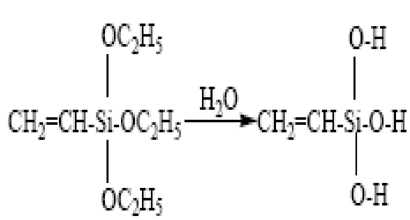

Figure 2: Hydrolysis of Silane [22].

The possible reactions are shown in Figures 2 . In presence of moisture, hydrolysable alkoxy group leads to the formation of silanols [22].

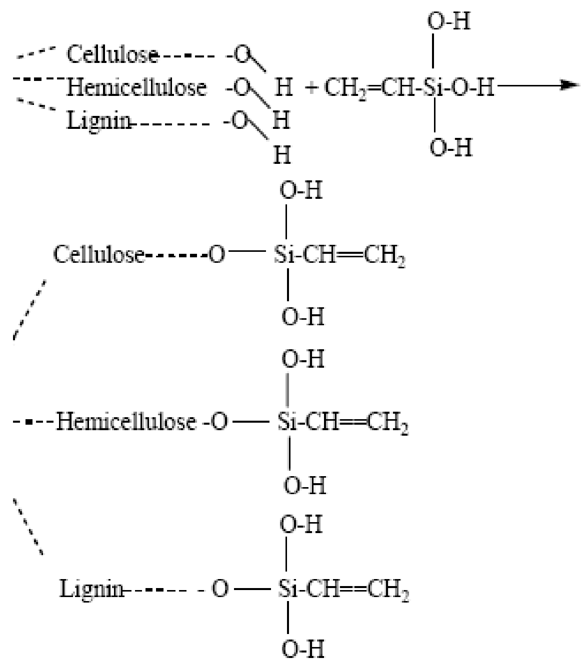

Figure 3: Hypothetical Reaction of Filler and Silane.

Mechanical Testing

The prepared sampled were analyzed using a universal Instron testing machine model 3369, in accordance with ASTM Test Method D638-03, and micro hardness testing was carried out on the sample using a Leco Micro Hardness Tester, model LM-700AT, in accordance with ASTM E384.

RESULTS AND DISCUSSION

The tensile strengths of the composites made of Rice Husk filler (RHF) and polyester matrix at different filler loadings are shown in Figure. 4. The tests were conducted at room temperature. Tensile strengths of RHF–PE composites slightly decreased with increasing filler loading.

As the filler loading increased, there is increase in the interfacial area, the worsening interfacial bonding between filler (hydrophilic) and matrix polymer (hydrophobic) decreased the tensile strength, which nevertheless remained within acceptable levels [18] it was however observed that Treated filler shows superior tensile strength than untreated fillers.

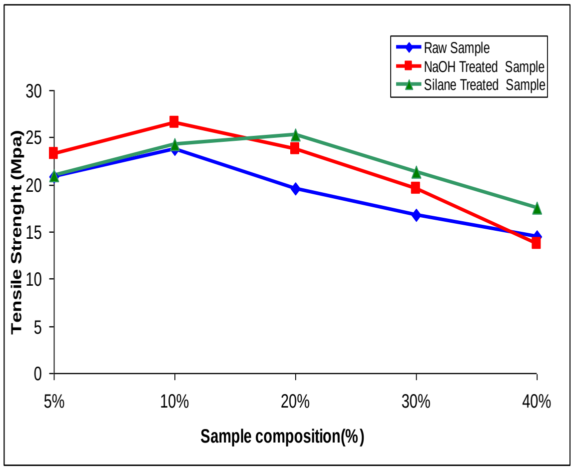

Figure 4: Tensile Test Result for Treated and Untreated Sample.

Modulus test result are shown in Figure 5, for irregularly shape fillers, the strength of the composites decreases due to the inability of the filler to support stresses transferred from the polymer matrix. Treated filler composite shows higher modulus compare to untreated filler composite.

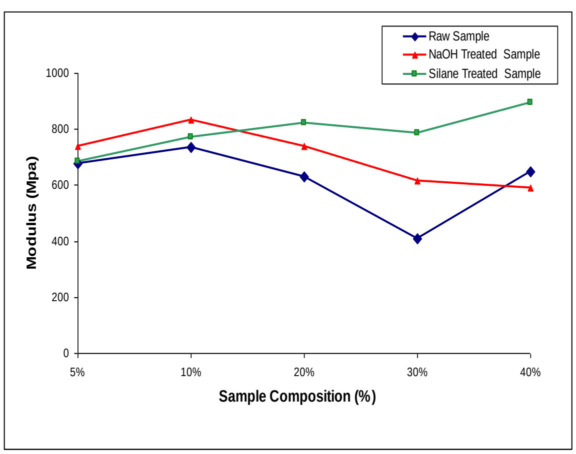

Figure 5: Modulus Test Result for Treated and Untreated Sample.

From Figure 6, Maximum Load of the raw RHF-PE composite show slight increase at 10% filler loading and there afterward decrease gradually as filler loading increases, however treated filler (NaOH and Silane) shows superior strength compare to untreated fillers.

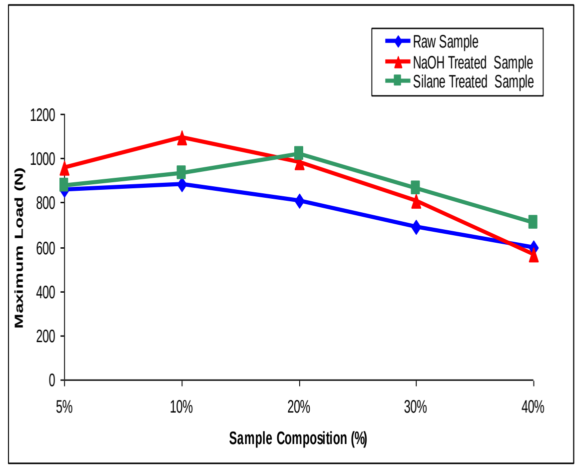

Figure 6: Maximum Load Test Result for Treated and Untreated Sample.

Micro hardness test result are shown in Figure 7,there was gradual increase in hardness as filler level increase from 5% to 10% for both treated and untreated filler, after which the value decreases gradually as filler loading increases. Treated filler composite however shows significant improvement in these properties.

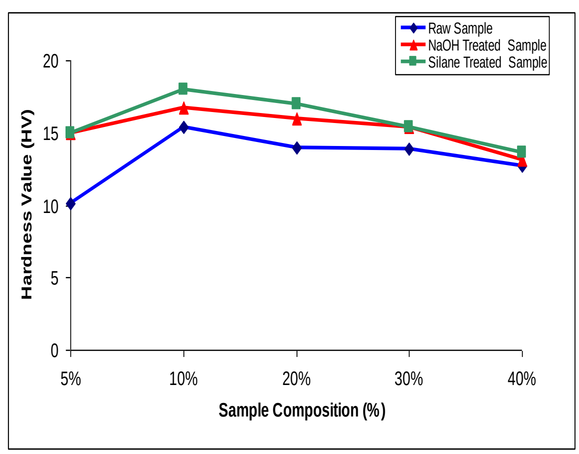

Figure 7: Micro-Hardness Test Result for Treated and Untreated Sample.

CONCLUSIONS

Silane and sodium hydroxide treatment on rice husk filler in polyester composite leads to a higher tensile strength compare to the low tensile strength recorded in the untreated fillers composite.

Modulus, maximum load and micro-hardness test results show increase value with treated fillers, Silane treated filler composite, however, showed superior properties over NaOH treated filler composite at above 10% filler loading. Thus the mechanical properties of rice husk flour-Polyester composite has been enhance with the use of surface treatment/coupling agent.

REFERENCES

1. Belmares, H., Barrera, A., and Monjaras, M.1983. "New Composite Materials from Natural Hard Fibers. Part 2. Fatigue Studies and a Novel Fatigue Degradation Model". *Industrial Engineering Chemical Product Research and Development*. 22:643–652.

2. Bucknall, C.B. 1977). *Toughened Plastics*. Applied Science: London, UK.

3. Fuad, M.Y.A., Rahmad, S., and Azlan, M.R.N. 1998. "Filler Content Determination of Bio-Based Thermoplastics Composites by Thermo gravimetric Analysis". *Proceedings of the Fourth International Conference on Advances in Materials and Processing Technologies*. Kuala Lumpur. 268–275.

4. Gattenholm, P., Berlisson, H., and Mathiasson, A. 1993. "The Effect of Chemical Composition of Interphase, On Dispersion of Cellulosic Fiber in Polymer.I.PVC-Coated Cellulose in Polystyrene". *Journal Applied Polymer Science*. 49(2):197-208.

5. George, J., Sreekala, M.S., and Thomas, S. 2001. "A Review on Interface Modification and Characterization of Natural Fiber Reinforced Plastic Composites". *Polymer Engineering & Science*. 41:1471–1485.

6. Han-Seung Yang, Hyun-Joong, Kim, Jungil Son, Hee-Jun Park, Bum-Jae Lee, and Taek-Sung Hwang. 2003. "Rice-Husk Flour Filled Polypropylene Composites; Mechanical and Morphological Study". *Composite Structures.* 63 (2004):305–312.

7. Herrara-Franco, P.A., Valadez-Gonzalez, and Cervantes, U.C.M. 1997. "Development and Characterization of a HDPE–Sand–Natural Fiber Composite". *Composites Part B: Engineering*. 28B(3):331–343.

8. Ismail, H, Edyham, M.R., and Wirjosentono, B. 2002. "Bamboo Fibre Filled Natural Rubber Composites: The Effects of Filler Loading and Bonding Agent". *Polymer Testing*. 21(2):139–144.

9. Ismail, H., Mega, L., and Abdul-Khalil, H.P.S. 2001 "Effect of a Silane Coupling Agent on the Properties of White Rice Husk Ash Polypropylene/ Natural Rubber Composite". *Polymer International*. 50(5):606-611.

10. Jacobson, R.E., Caulfield, D.F., Rowell, R.M., and Sanadi, A.R. 1995. "Recent Developments in Annual Growth Lignocellulosics as Reinforcing Fillers in Thermoplastics". *Proceedings of 2nd Biomass Conference of the Americas: Energy, Environment, Agriculture, and Industry.* August 21–24, Portland, OR. National Renewable Energy Laboratory: Golden, CO. 1171–1180.

11. Karnani, R., Krishnan, M., and Narayan, R. 1997. "Biofiber-Reinforced Polypropylene Composites". *Polymer Engineering & Science*. 37:476–483.

12. Khalid, A.A., Sahari, B., and Khalid, Y.A. 1998 "Environmental Effects on the Progressive Crushing of Cotton and Glass Fiber/Epoxy Composite Cones". *Proceedings of the Fourth International Conference on Advances in Materials and Processing Technologies*. Kuala Lumpur. 680–689.

13. Luo, S. and Netravali, A.N. 1999. "Mechanical and Thermal Properties of Environmentally Friendly "Green" Composites Made from Pineapple Leaf Fibers and Poly (hydroxybutyrate-co-valerate) Resin". *Polymer Composites*. 20(3):367–378.

14. Martí-Ferrer, F., Vilaplana, F., Ribes-Greus, A., Benedito-Borrás, A., and Sanz-Box, C. 2006. 'Flour Rice Husk as Filler in Block Copolymer Polypropylene: Effect of Different Coupling Agents". *Journal of Applied Polymer Science.* 99: 1823–1831.

15. Maulida, A., Nasir, M., and Khalil, H.P.S.A. 2000. "Hybrid Composites Based on Natural Fiber". *Proceedings of Symposium on Polymeric Materials*. Penang, 1-2 June 2000. USM Press, Penang. 216–219.

16. Mishra, S., Hamzah, H., Murshidi, J.A., and Deraman, M. 2002. "Chemical Modification on Lignocellulosic Polymeric Oil Palm Empty Fruit Bunch for Advance Material". *Advances in Polymer Tech*. 20(4): 289-295.

17. Neus Anglès, M., Salvadó, J., and Dufresne, A. 1999. "Steam-Exploded Residual Softwood-Filled

Polypropylene Composites". *Journal of Applied Polymer Science*. 74:1962–1977.

18. Premalal, H.G.., Ismail, B., and Baharin, H. 2002. "A Comparison of the Mechanical Properties of Rice Husk Powder Filled Polypropylene Composites with Talc Filled Polypropylene Composites". *Polymer Testing*. 21(7):833–839.

19. Riffle, J.S., Yilgor, I., Banthia, A.K., Tran, C., Wilkes, G.L., and McGrath, J.E. 1983. *Epoxy Resin Chemistry*. R.S. Bauer (ed.). ACS Symposium Series no. 201. American Chemical Society. Washington, DC. 21

20. Saroja Devi, M., Murugesan, V., Rengaraj, K., and Anand, P. 1998. "Utilization of Flyash as Filler for Unsaturated Polyester Resin". *Journal of Applied Polymer Science*. 69(7):1385-1391.

21. Shangjin, H., Keyu, S., Jie, B., Zengkun, Z., Liang, L., Zongjie, D., and Baolong, Z. 2001. "Studies on the Properties of Epoxy Resins Modified with Chain-Extended Ureas". *Journal of Polymer*. 42:9641–9647.

22. Son, J.I., Kim, H.J., and Lee, P.W. 2001. "Role of Paper Sludge Particle Size and Extrusion Temperature on Performance of Paper Sludge Thermoplastic Polymer Composites". *Journal of Applied Polymer Science*. 82(11):2709–2718.

23. Sreekala, M.S., Kumaran, M.G., Joseph, S., and Jacob, M. 2000. "Oil Palm Fibers Reinforced Phenol Formaldehyde Composites: Influence of Fibers Surface Modifications on the Mechanical Performance". *Applied Composite Materials.* 7:295-329.

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