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EFFECT OF FILLER BLEND COMPOSITION ON THE MECHANICAL PROPERTIES OF HDPE/CASSAVA/WOOD FLOUR COMPOSITES

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ABSTRACT

The maximum tensile strength observed in the composite was 8.813Mpa with total filler content of 30% of cassava and wood flour while the unfilled high-density polyethylene (HDPE) was 12.231Mpa. Strength values of 8.317MPa, 7.572MPa and 8.59MPa were obtained for composites with total filler content of 15%, 35 % and 40% respectively.

The highest elongation at break of the composite was observed in sample with 30% cassava flour. 50% wood flour filler content exhibited the least break elongation. Above 40% total filler content, the Young's modulus decreases swiftly to 0. The results showed that 15% wood flour only and 40% total filler improved significantly the modulus (2% Secant). Viable compression moulded plastics can be formulated from cassava and wood flours to form composites with high-density polyethylene for applications in furniture and allied industries.

KEY Words: *composites; high-density polyethylene (HDPE); cassava flour; wood flour.*

1. INTRODUCTION

Research and development of biocomposite materials is a growing interest to scientists and engineers. Thermoplastic composites containing wood fillers are growing in use, because they are lightweight, strong, and sturdy (Bledzki and Gassan, 1999). During the last decade, extensive efforts have been made on the use of wood fibre as fillers or reinforcers for thermoplastic matrices (Liao, Huang and Cong, 1997, Lai, Yeh, Wang, Chan, Shen and Hsiao, 2001, Kazayawoko, Balatineez and Matuana, 1999, Myers, Chahyadi, Coberly and Ermer, 1991, Takase and Shiraishi, 1998, Joly, Kofman

and Gauthier, 1996, Matuana, Woodhams, Balatineez and Park, 1998, Matuana, Balatineez and Park, 1998, Matuana, Park, and Balatineez, 1997, Mengeloglu, Matuana, and King, 2000). Wood fibres offer many advantages over the most commonly used inorganic materials including a lower cost on a volumetric basis, higher specific strength and modulus, flexibility during processing, etc (Liao, Huang and Cong, 1997, Lai, Yeh, Wang, Chan, Shen and Hsiao, 2001, Kazayawoko, Balatineez and Matuana, 1999, Myers, Chahyadi, Coberly and Ermer, 1991, Takase and Shiraishi, 1998, Matuana, Balatineez and Park, 1998, Matuana, Park, and Balatineez, 1997). Effects of filler blend composition on electrical and mechanical properties of conductive EVA composites have been reported in the scientific literature (Das, Chaki, Khastgir and Chakraborty, 2000). The work described in this paper is part of an ongoing research programme that is aimed at producing composites of low strength-low cost applications, such as furniture, from discarded wood dusts and agricultural wastes which can be one possible route to solve the environmental problem posed by the large quantities of these wastes in cassava processing sites and saw mills.

Various theoretical models have been used to predict the mechanical properties of HDPE/cassava/wood composites and a comparison with experimental data has been undertaken.

Experimental

Materials and Methods

High-density polyethylene HDPE, (Elpene, with melt flow index of 0.923g/10min and density of 0.9445g/cm^3), was supplied by Eleme Petrochemicals Company Ltd (EPCL), Port Harcourt, Rivers State, Nigeria. Cassava and wood flour with a maximum particle size 50□m obtained locally were oven dried for 24hrs at 130^{o}C.

Reagent glycerol (Fisher Scientific, Pittsburgh, PA.) was of analytical grade and used as received.

Table 1.0: Formulation of the samples*

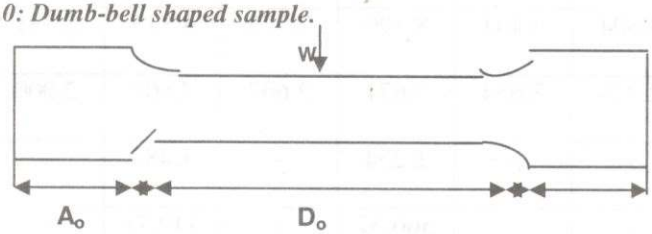
ID	1	2	3	4	5	6	7	8(unfilled)
Cassava flour(%).	0	70	70	35	25	30	0	0
Wood flour(%)	15	5	10	5	10	0	50	0
HDPE(%)	85	25	20	60	65	70	50	100

** The composite formulations were based on ANOVA technique.*

Composite Preparation

The components of the composites were weighed using the Toshiba analytical balance. The ground powder was oven dried at 80°C for 10mins. The components of each composite batch were obtained using the formulations in Table 1.0 and combinations were done in %wt. For each batch, 4%wt of glycerol based on the oven-dry weight of the fillers was added and thoroughly mixed using a kitchen size Binatone household blender. The Composites were prepared using Carver Hydraulic press Model 'C' with varying weight percentage of cassava and wood flour fillers at moulding temperature of 130°C and 150°C for rectangular and dumb-bell samples respectively. Higher temperatures gave unacceptable products due primarily to thermal degradation. Typical dimensions of specimen samples were 5×2.5×0.2cm.

Figure 1.0: Dumb-bell shaped sample.



A_o

Thickness = 0.2 ± 0.1cm

W_o = width section = 2.5 ± 0.1cm

D_o = gauge length = 5.0 ± 0.1cm

A_o = gripping length = 2.0 ± 0.1cm

Measurements

The Density of the samples was obtained using an automatic Toyoseiki Densimeter. Water absorption testing was carried out according to ASTM D 570-98 procedure. Rectangular specimens of dimension 5×2.5x 0.2cm were used. The composites samples were first weighed to the nearest 0.001g. To measure the water absorption of the composites, all the samples were completely immersed in water for 52hrs and 70days respectively at room temperature. Excess water on the surface of the samples was wiped out before weighing again. The hardness test was carried out according to ASTM D2240-97 specification.

Tensile tests were carried out at 23°C using Instron-5565 in accordance with ASTM D5323 specifications. The pressure of the Instron clamp was set at 5.8 bars; distance between clamps was 30mm; speed of Instron was 50mm/min.

Results and Discussion

Table 2.0: Density, Water absorption and Mechanical properties of unfilled HDPE and HDPE/Cassava/Wood composites

ID/Properties	1	2	3	4	5	6	7	8(unfilled)
Density (g/cm^3)	0.993	1.258	1.285	1.100	1.078	1.067	0.955	0.9455
Water absorbed (%) after 52hrs soaking	0	19.050	17.020	3.570	3.700	3.570	16.000	0
Water absorbed (%) after 70days soaking	0	19.050	17.020	3.570	3.700	3.570	24.000	0
Tensile strength, MPa	8.317	3.804	3.464	8.590	7.572	8.813	5.731	12.231
Break elongation, %	2.335	2.229	3.654	3.671	2.667	7.607	2.000	13.191
Yield elongation, %	-	-	-	1.254	-	1.487	-	1.573
Modulus of elasticity, MPa	-	-	-	566.377	-	449.772	-	654.493
Modulus (Secant 2%),MPa	415.808	186.618	163.563	379.97	312.991	342.308	286.309	515.668
Shore A hardness	98	95	85	96	95	97	95	62(D)

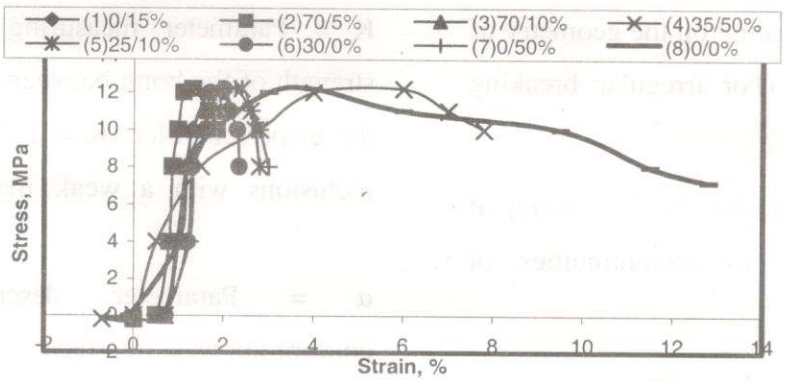


Fig. 2.0: Stress-Strain curves of HDPE/Cassava/Wood composites

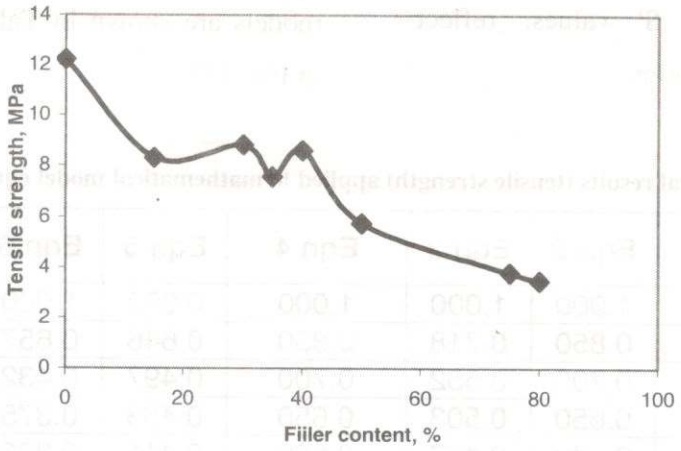


Fig.3.0: Tensile strength of HDPE/Cassava/Wood composites versus filler content

The tensile strengths of the composites were analyzed using the following mathematical models for monodispersed fillers (Ghosh and Maiti, 1996, Bigg, 1987) as in equations (1) to (6)

$$\frac{\sigma_c}{\sigma_p} = (1 - K\Phi^b) \quad (1)$$

$$\frac{\sigma_c}{\sigma_p} = (1 - \Phi) \quad (2)$$

$$\frac{\sigma_c}{\sigma_p} = (1 - \Phi^{2/3}) \quad (3)$$

$$\frac{\sigma_c}{\sigma_p} = (1 - \Phi).S \quad (4)$$

$$\frac{\sigma_c}{\sigma_p} = (1 - \Phi^{2/3}).S' \quad (5)$$

$$\frac{\sigma_c}{\sigma_p} = \exp(-\alpha\Phi) \quad (6)$$

where;

σ_c = Tensile strength of composite

σ_p = Tensile strength of unfilled polymer

Φ = Filler Content (Volume fraction)

b = parameter depending on the geometry of the breaking area (For irregular breaking area, b = 2/3)

S = parameter reflecting the weakening of the structure, due to discontinuities of charge transfer

S' = Parameter depending on stress concentrations. For S' = 1 means no stress concentrations; lower S' values, reflect higher stress concentrations

K = Parameter measuring the adhesive strength of the bond between the matrix and the dispersed filler (K = 1.21 for spherical inclusions with a weak matrix adhesion)

α = Parameter describing stress concentrations at interface.

The experimental results applied to the models are shown in Table 3.0 and plotted in Fig. 4.0.

Table 3.0: Experimental results (tensile strength) applied to mathematical model equations (1 –6)

% Filler content	Eqn 1	Eqn 2	Eqn 3	Eqn 4	Eqn 5	Eqn 6	Exptal
0	1.000	1.000	1.000	1.000	0.900	1.000	1.000
15	0.718	0.850	0.718	0.850	0.646	0.657	0.680
30	0.552	0.700	0.552	0.700	0.497	0.432	0.721
35	0.503	0.650	0.503	0.650	0.453	0.375	0.615
40	0.457	0.600	0.457	0.600	0.411	0.326	0.702
50	0.370	0.500	0.370	0.500	0.333	0.247	0.469
75	0.175	0.250	0.175	0.250	0.157	0.122	0.311
80	0.138	0.200	0.138	0.200	0.124	0.106	0.282

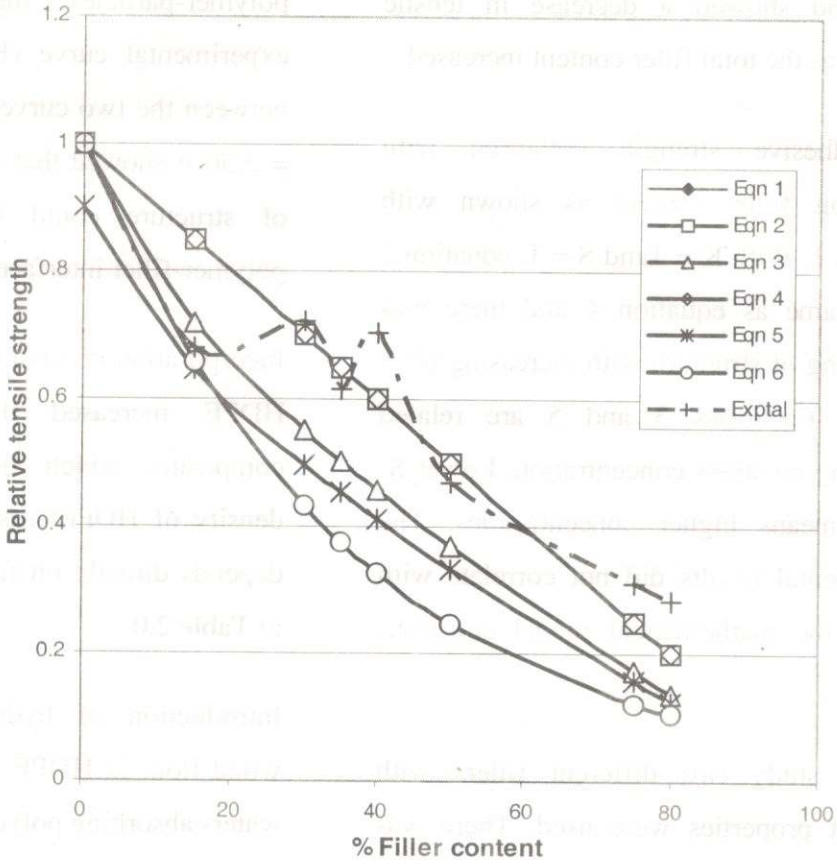


Fig.4.0: Relative break elongation for HDPE/Cassava/wood composites as a function of % filler content. Experimental curves are plotted as dashed lines and the corresponding theoretical models [described by eqs (1-6)] are plotted as full lines

The trend showed a decrease in tensile strength as the total filler content increased.

The adhesive strength collapsed with increasing filler content as shown with equation 1. With K = 1and S = 1, equation 2 is the same as equation 4 and there was weakening of structure with increasing filler content. Equations 3 and 5 are related indicating no stress concentration. Lower S' values means higher concentrations. The experimental results did not correlate with any of the mathematical model equations (1–6).

In this study two different fillers with different properties were used. There was downward reduction of tensile strength but occasional jump due to synergetic effects of the fillers. The reduction in tensile properties, with addition of more fillers may be explained by the fact that there was weak adhesion between the polymer and the fillers. Furthermore the interface is established by non-bonding van der waals forces between the polymer and the fillers, and there are no chemical bonds or other strong interactions to strengthen the interfacial adhesion. This lack of adhesion was observed during tensile tests, when the sample fracture always started from the polymer-particles interface. Since the experimental curve (Fig. 5.0) was placed between the two curves of S' = 0.90 and S' = 3.50 it showed that significant weakening of structure could be expected at the polymer-filler interfaces.

Incorporation of cassava and wood flours to HDPE increased the density of the composite, which becomes stiffer. The density of HDPE/cassava/wood composites depends directly on filler content as shown in Table 2.0

Introduction of hydrophilic cassava and wood flour in HDPE resin matrix produced water- absorbing polymer composite.

The water absorption behaviour exhibited by this system showed an increase as the proportion of cassava flour filler content was increased.

However, wood flour content of 50% of the polymer matrix water absorption was only 16% and 24% after 52hrs and 70days respectively. Longer immersion times could lead to disintegration of highly filled samples.

Cassava and wood flour fillers dramatically reduce the water resistance of unfilled

HDPE, which does not biodegrade. This ability to absorb water by this composite will lead to disintegration of the composite before biodegradation of the bio-components takes place leaving the synthetic polymer matrix untouched.

The hardness of the composites was reasonable except where 20% of the resin matrix was used.

The tensile tests of the composites yielded tensile strength, elongation at break, Young's modulus and modulus (Secant 2%) from the stress-strain curves presented in Fig. 2.0 and Table 2.0. The stress-strain curves of the composites are viscoelastic and similar to those of amorphous polymers.

Linear elastic behaviour was observed at low strains, which deviated from linearity at higher strains. The strain to failure was 7.61% for 30/70 Cassava flour/HDPE content of the composite and 2% for 50/50 Wood/HDPE content as shown in Table 2.0 and Fig. 2.0. The tensile strength of HDPE/Cassava/Wood composite decreased continuously with the increase in filler content. The maximum tensile strength observed by the composite was 8.813MPa, for 30% Cassava flour filler content. Values of 8.590MPa, 8.317Mpa and 7.572MPa were obtained for total filler contents of 40%, 15%, and 35% respectively. The cassava flour thus provided more strength to the composite than the wood flour. There were higher strengths for 35% Cassava flour and 5% wood flour. Then for 15% wood only significant tensile strength was obtained. Above 60% total filler content, the tensile strength of HDPE/Cassava/Wood composite was not quite reasonable for load bearing applications because it was too low as shown in Fig. 3.0.

The addition of wood flour without cassava flour significantly reduced the tensile strength of the composites due to the poor interfacial adhesion between wood fibre and the HDPE resin matrix.

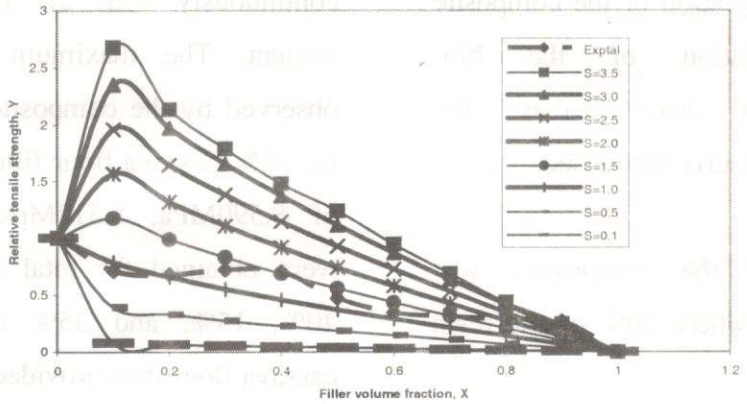


Fig. 5.0: Stress concentration, S, effect on the relative tensile strength vs filler volume fraction curves

This higher stress concentration at the interface is also confirmed by the value of $\alpha = 2.8$. As a consequence, the matrix deformation (and hence the composite deformation) decreased in a pseudo-logarithm manner with increasing total filler content.

The addition of the filler reduced the ductility of the polymer as evident from the decrease in elongation at break. Thus increasing filler content resulted in an increase in the brittleness of the composite.

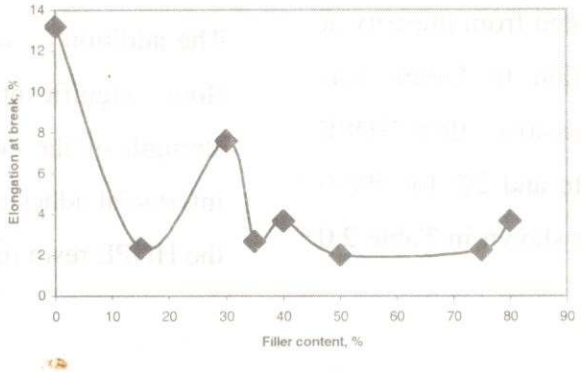


Fig. 6.0: Elongation at break of HDPE/Cassava/Wood composites versus filler content

The highest elongation at break of the composites was observed with 30% cassava flour content. 50% wood flour filler content exhibited the least break elongation (Fig.6.0). It is important to note that sample with 80% total filler content showed a dramatically higher break elongation than those with 15% wood flour only. This phenomenon can be attributed to the higher contribution of the cassava flour in 80% total filler content. For the system under study, the elongation at break is inversely proportional to the filler content. Typically, the elongation at break for HDPE/Cassava/Wood composite decreased as filler content was increased as shown in Fig. 6.0.

The general tensile elongation behaviour of polymeric composites with particulate fillers can be predicted by two mathematical equations (Ghosh and Maiti, 1996, Bigg, 1987) viz in equation 7 and 8.

$$\frac{\varepsilon_c}{\varepsilon_p} = (1 - \Phi^{1/3}) \quad (7)$$

$$\frac{\varepsilon_c}{\varepsilon_p} = (1 - K.\Phi^{2/3}) \quad (8)$$

where:

ε_c = Relative elongation of the composite

ε_p = relative elongation of the polymer

K = parameter depending on filler particle size and the applied treatment.

Table 4.0 Experimental results (elongation at break) applied to mathematical model equations (7-8)

% Filler content	Eqn 7	Eqn 8	Exptal
0	1	1	1
15	0.469	0.718	0.177
30	0.331	0.552	0.577
35	0.295	0.503	0.202
40	0.263	0.457	0.278
50	0.206	0.370	0.152
75	0.091	0.175	0.169
80	0.072	0.138	0.277

Table 4.0 shows the comparison of the experimental results of relative elongation at break with the mathematical model equations (7-8). From the equations the trend decreased in elongation at break as the total filler content increased. There was downward reduction of elongation at break but occasional jump as a result of synergetic effects of the two fillers as in the previous analysis, hence the experimental results did not correlate with either of the equations.

Fig. 7.0 presents the relative elongation at break of the HDPE/Cassava/Wood composites as a function of total filler volume fraction. The elongation at break decreased with increasing wood filler to 15%. Reinforcing with 30% cassava flour increased the elongation at break until a combination of the fillers resulted to further decrease. Above this value no significant influence was observed. A quantitative analysis indicated that influence of cassava flour content was higher than that of wood flour.

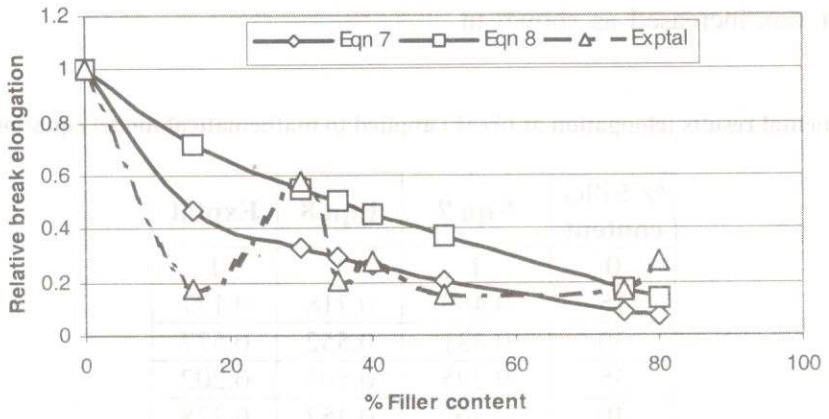


Fig. 7.0: Relative break elongation for HDPE/Cassava/wood composites as a function of % filler content. Experimental curves are plotted as dashed lines and the corresponding theoretical models [described by eq (7): eq(8)] are plotted as full lines

Table 5.0 and Fig 8.0 clearly showed that the modulus of elasticity for HDPE/Cassava/Wood composite decreased as filler content increased. The highest modulus of elasticity was observed in 40% total filler content (see Table 2.0). Also one can see that the modulus of elasticity for HDPE/Cassava/Wood composites was significant only for total filler contents between 0 and 40%. Above 40% total filler content, the Young's Modulus decreased swiftly to 0. Again the modulus of elasticity of this system was dependent on the amount of individual filler. At wood filler content above 5%, there was no significant result.

Thus, to improve modulus of elasticity of this composite system, a simple and practicable way is to increase the cassava flour content of the batch. However, in 2% Secant modulus the results showed that 15% wood flour only and 40% total filler content improved significantly the modulus.

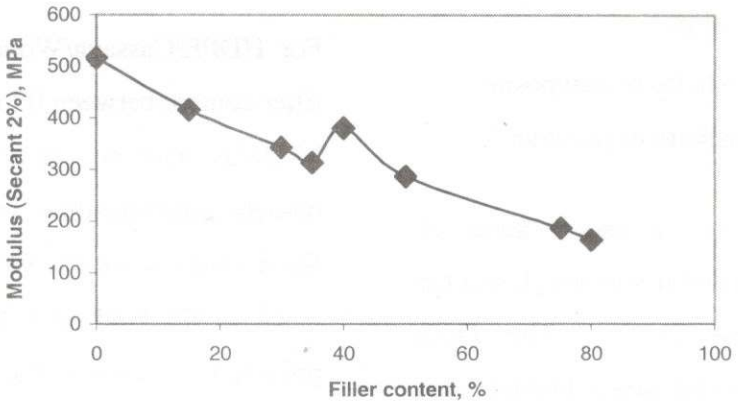


Fig. 8.0: Modulus (Secant 2%) of HDPE/Cassava/Wood composites versus filler content

Significant equations that predict the modulus of elasticity of polymeric composites were developed by Einstein (equation 9), Guth (equation 10), Thomas (equation 11) and Quemada (equation 12)

$$\frac{E_c}{E_p} = (1 + 2.5\,\Phi) \quad (9)$$

$$\frac{E_c}{E_p} = (1 + 2.5\Phi + 14.1\Phi^2) \quad (10)$$

$$\frac{E_c}{E_p} = [1 + 2.5\Phi + 10.05\Phi^2 + 0.00273\exp(16.6\Phi)] \quad (11)$$

$$\frac{E_c}{E_p} = \frac{1}{(1 - \tfrac{1}{2}K\Phi)^2} \quad (12)$$

E_c = Modulus of elasticity of composite

E_p = Modulus of elasticity of polymer.

Guth's equation was a modification of Einstein's equation and it was developed for high filler volume fraction. The Guth equation is very useful since Einstein's is valid only for polymers filled with non-interactive spherical particles. The Thomas' equation is an empirical relationship based on data generated by a mono-disperse spherical particle system whereas the Quemada's equation introduces a variable coefficient highlighting particle interactions and geometrical differences.

Other background equations of importance are equations 13 and 14 (Ghosh and Maiti, 1996, Bigg, 1987).

$$\frac{E_c}{E_p} = (1 - \Phi^{2/3}) \quad (13)$$

$$\frac{E_c}{E_p} = \frac{1 - \Phi^{2/3}}{1 - \Phi^{2/3} + \Phi} \quad (14)$$

For HDPE/Cassava/Wood composites with filler content between 0 to 40%, the Young's Modulus may be predicted by theoretical models corresponding to polymers highly filled with non-interactive spherical particles (such as the Guth and Thomas equations), provided the wood content does not exceed 5%. Above this, none of the theoretical models described above could predict the modulus of elasticity of HDPE/Cassava/Wood composites.

Table 5.0 Experimental results Modulus (Secant 2%) applied to mathematical model equations (9-14)

% Filler content	Exptal	Eqn 9	Eqn 10	Eqn 11	Eqn 12	Eqn 13	Eqn 14
0	1.000	1.000	1.000	1.003	1.000	1.000	1.000
15	0.806	1.375	1.692	1.634	1,169	0.718	0.827
30	0.664	1.750	3.019	3.052	1.384	0.552	0.648
35	0.607	1.875	3.602	4.017	1.469	0.503	0.590
40	0.737	2.000	4.256	5.697	1.563	0.457	0.533
50	0.555	2.25	5.775	15.748	1.778	0.370	0.425
75	0.362	2.875	10.806	705.362	2.560	0.175	0.189
80	0.317	3.000	12.02	1607.493	2.778	0.138	0.147

From Table 5.0 the experimental results correlated with the mathematical model equations (13-14) only. In equations (9-12) the trend was increase in modulus (Secant 2%) as the total filler content increased. In equations (13-14) including the experimental results the trend decreased in modulus (Secant 2%) as the total filler content increased.

Fig. 9.0 shows the experimental relative modulus (Secant 2%) curve and theoretical predictions, corresponding to equations (9-14) of the HDPE/Cassava/Wood composites as a function of total filler volume fraction.

Then experimental curves were relatively in agreement with equation (13 and14). In equations (9 to12) the Modulus (Secant 2%)increased with increase in volume fractions of the fillers.

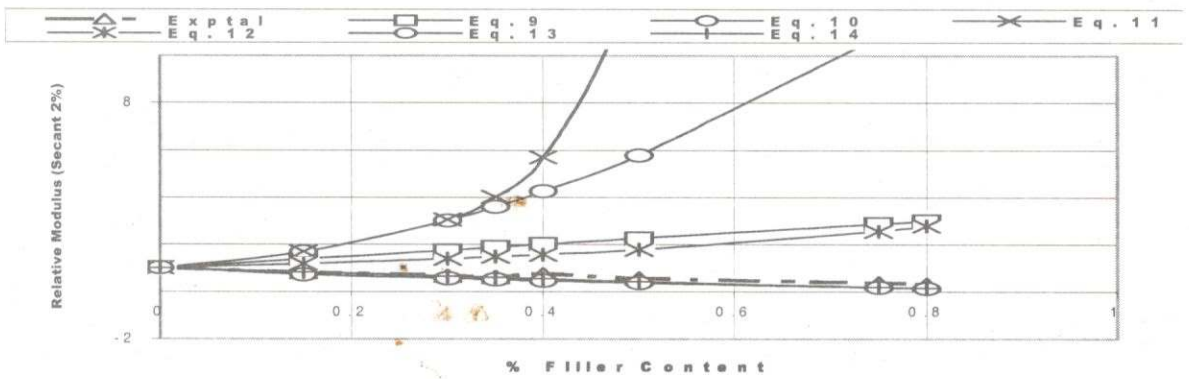


Fig. 9.0: Relative Modulus (Secant 2%) for HDPE / Cassava / Wood Composites as a Function of % Filler Content

Conclusions

The tensile strength for samples with 15%, 40%, 35%, 30% and 50% total filler were high enough for load bearing applications such as furniture.

The highest elongation at break of the composite was observed in 30% cassava flour content. 50% wood flour filler content exhibited the least break elongation.

The same trend was also true for the 2% secant modulus and the modulus of elasticity (stiffness) of some of the composites.

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