Iranian Polymer Journal
16 (4), 2007, 271-278

Available online at: http://journal.ippi.ac.ir

Prediction of the Elastic Modulus of Wood Flour/Kenaf Fibre/Polypropylene Hybrid Composites

Jamal Mirbagheri[1], Mehdi Tajvidi[1], Ismaeil Ghasemi[2*] and John C. Hermanson[3]

(1) Department of Wood and Paper Science and Technology, College of Natural Resources University of Tehran, Iran
(2) Department of Plastic, Processing Faculty, Iran Polymer and Petrochemical Institute P.O.Box: 14195/115, Tehran, Iran
(3) USDS Forest Laboratory, Advance Housing Research Centre, One Gifford Pinchot Drive Madison, 53726-2398, WI, USA

Received 10 January 2007; accepted 30 April 2007

ABSTRACT

The prediction of the elastic modulus of short natural fibre hybrid composites has been investigated by using the properties of the pure composites through the rule of hybrid mixtures (RoHM) equation. In this equation, a hybrid natural fibre composite assumed as a system consisting of two separate single systems, namely particle/polymer and short-fibre/polymer systems. However, there is no interaction between particles and short fibres. Polypropylene was used as the polymer matrix and 40-80 mesh kenaf fibre and 60-100 mesh wood flour were used as the fibre and the particulate reinforcements, respectively. Hybrid composites were produced by kenaf fibre/wood flour ratios of 40:0, 30:10, 20:20, 10:30, and 0:40. Maleic anhydride and DCP have been also used as the coupling agent and initiator, respectively. Mixing process carried out in an internal mixer at 180°C and 60 rpm. The rule of hybrid mixtures (RoHM) equation has been employed using the weight and volume fractions of the reinforcements. The relationship between experimental and predicted values was evaluated and the accuracy of the estimation of the model was controlled. The results indicated that RoHM equation is able to predict the elastic modulus of the composites. The comparison between experimental and predicted values showed that they are in good agreement.

Key Words:

rule of hybrid mixtures;
kenaf fibre;
wood flour;
polypropylene;
hybrid composites;
modulus of elasticity.

(*) To whom correspondence to be addressed:
E-mail: i.ghasemi@ippi.ac.ir

INTRODUCTION

The growing interest in using natural fibres as a reinforcement of polymer-based composites is mainly due to their advantages such as lower cost and density, ease of preparation, lower energy requirements for processing, biodegradability, and wide availability over traditional reinforcing fibres such as glass and carbon. However, some limitations in using natural fibres in composites are the lower allowable processing temperatures, incompatibility between the hydrophilic natural fibres and hydrophobic polymers, high moisture absorption of the fibres, and the resulting swelling of the manufactured composite [1-3].

Thermoplastics, which are used

in such composites, consist of polyethylene (high and low density), polypropylene, polyvinyl chloride and polystyrene. On the other hand, kenaf, jute, sisal, coir, flax, banana, wood flour, rice hulls, newsprint, pulp, and cellulose fibres are the main natural fibres used as reinforcement [4]. Among the various non-wood natural fibres, kenaf fibres provide high stiffness and strength values. They also have higher aspect ratios making them suitable to be used as the fibrous phase [5].

Hybrid composites are materials made by combining two or more different types of fibres in a common matrix [6-9]. Hybridization of two types of short fibres having different lengths and diameters offers some advantages over the use of either of the fibres alone in a single polymer matrix [10-12]. Hybrid composites have long taken the attention of many researchers as a way to enhance the mechanical properties of composites. However, hybrid composites using natural fibres have been studied rarely and the most published reports limited to the hybrid composite which are consists of one kind of natural fibre and one kind of non-natural fibre. For example using of natural fibre/glass fibre, talc, and carbon fibre has been reported [13-17].

For interpreting the mechanical properties of composite by several variable parameters additional measurements are required when changes such as relative volumes of the constituents, constituent properties, and fabrication process occur in the composite system variables. Thus, experiments may be time consuming and cost prohibitive [18]. Therefore, theoretical and semi-empirical methods of determining composite properties should use to predict the effects of a large number of system variables [19].

The mechanical properties of hybrid short fibre composites can be evaluated using the rule of hybrid mixtures (RoHM) equation, which is widely used to predict the strength and modulus of hybrid composites [20]. It is assumed that the RoHM works best for modulus prediction than the strength values. Since, strength values in such composites are not primarily functions of the strengths of the components they are, however dependent on fibre/matrix interaction and interface quality. In tensile tests, any minor (microscopic) imperfection on the specimen surfaces may lead to stress build-up and fracture that could not be predicted directly by using the RoHM equation. Therefore, the present work only considers its application for modulus values, as they are uniform and obtain well below the maximum fracture load.

One of the most important factors determining the properties of composites is the relative proportions of the matrix and reinforcing materials to each other. The relative proportions can be expressed as the weight fractions or the volume fractions. Natural fibres (such as kenaf) are very bulky and the bulk densities as low as 0.2 g/cm^3 are even expected. The present processing equipment normally uses a gravimetric feeding system which has proved to be more efficient than the volumetric ones in this case. Therefore, in the industry, weight fractions are used instead of volume fraction. For this reason, we have considered the possibility of using weight fraction in the well-established RoHM to introduce a much simpler analysis.

This is the first attempt to introduce a simple practical method to determine the modulus of the all-natural-fibre hybrid composites. Other publications have mainly focused on hybrid composites of a natural fibre with a synthetic one. Having the results confirmed, we can predict the improvements in modulus values when longer fibres are added to the wood flour/PP composite system.

The objective of our work was to predict the elastic modulus of kenaf fibre/wood flour/polypropylene hybrid composites by using the properties of the pure composites by employing the RoHM equation. Comparison of the predicted results was also made by using the weight and volume fractions of the fibres.

The Rule of Mixtures (RoM) Equation

Elastic Modulus

Most fibre-reinforced composites are anisotropic, i.e., these materials have at least 5 or 6 independent elastic moduli. Thus, their properties are differet at various directions. Often all the fibres are aligned in one direction to give a uniaxial oriented material. Among the five or six moduli, two are considered as the most important moduli in most situations:

- The longitudinal Young modulus of composite E_{cL} in which the load is applied parallel to the direction of the fibres
- The transverse Young modulus of composite E_{cT} in which the load is applied perpendicular to the direction of the fibres.

In the longitudinal direction, a tensile force tends to stretch the fibre and the matrix to the same extent. Therefore, on the assumption that fibre, matrix, and composite experience equal strains, the longitudinal Young modulus in tension is given:

$$E_{cL} = E_m V_m + E_f V_f \quad (1)$$

where E_m and E_f are the modulus of matrix and fibres, and V_m and V_f are the corresponding volume fractions of the two phases, respectively. In the transverse direction on the assumption that fibre, matrix, and composite experience equal stresses, the Young' modulus in tension can be estimated as follows:

$$1/E_{cT} = V_m / E_f + V_f / E_m \quad (2)$$

Tsai and Pagano have developed a similar method of calculating of the Young' modulus of composites containing short fibres which are oriented randomly in the composite [21].

$$E\ (\text{random}) = 3/8\ E_L + 5/8\ E_T \quad (3)$$

where, E_L and E_T are the longitudinal and transverse muduli, respectively.

Density

The density of the composite material can be easily obtained using the eqn (4).

$$p_c = p_f V_f + p_m V_m \quad (4)$$

where V_f and V_m are the volume fraction of fibre and matrix, and P_c, P_f and P_m are the densities of composite, fibre, and matrix, respectively.

The Rule of Hybrid Mixtures (RoHM) Equation

By considering a hybrid composite as a system consisting of two single composite systems and assuming that there is no interaction between the two single systems, we can apply the iso-strain condition to the two single systems, i.e.,

$$\varepsilon_c = \varepsilon_{c1} = \varepsilon_{c2} \quad (5)$$

where ε_c, ε_{c1}, and ε_{c2} are the strains of the hybrid composite, the first system, and the second system, respectively [19]. Force equilibrium requires that

$$E_c\varepsilon_c = E_{c1}\varepsilon_{c1}V_{c1} + E_{c2}\,\varepsilon_{c2}\,V_{c2} \quad (6)$$

Then, the modulus of the hybrid composite can be evaluated from the RoHM equation by neglecting the interaction between two systems as follows:

$$E_c = E_{c1}V_{c1} + E_{c2}V_{c2} \quad (7)$$

where E_c, V_{c1}, and V_{c2} are the elastic modulus of the hybrid composite, the relative hybrid volume fraction of the first and the second system, respectively. It should be considered that the expressions listed below are valid for the assumed system

$$V_{c1} + V_{c2} = 1 \quad (8)$$

$$V_{c1} = V_{f1} / V_t \quad (9)$$

$$V_{c2} = V_{f2} / V_t \quad (10)$$

$$V_t = V_{f1} + V_{f2} \quad (11)$$

where V_t is the total reinforcement volume fraction. In addition $V_{f1} + V_{f2}$ should be used as reinforcement volume fraction for calculation of the elastic modulus (E_{c1} and E_{c2}) of both of the single composites. A positive or negative hybrid effect is defined as a positive or negative deviation of the elastic modulus of the hybrid composites from the RoHM equation, which has been indicated in the presented results.

EXPERIMENTAL

Materials

Polypropylene, as homopolymer, with a melt flow index of 8 g/10 min and a density of 0.9 g/cm^2, supplied by Bandar Imam Petrochemical Industries Co., Iran, with PI0800 trade name was used as the polymer matrix. The 40-80 mesh kenaf fibres and 60-100 mesh beech wood flour were obtained from the Research Forest of the Natural Resources Faculty of the University of Tehran, Iran. Maleic anhydride (MA) (the coupling agent) and DCP (as initiator) were supplied by Merck Co. (Germany) and Hercules Co. (U.S.A), respectively.

Sample Preparation

After grinding and passing kenaf fibre and wood flour through determined sieves, they were dried in an oven at 65±2ºC for 24 h before being blended with PP.

Polypropylene, kenaf fibre, and wood flour were

Table 1. Composition of the studied formulations (wt%).

Code	Polypropylene content	Kenaf fibre content	Wood flour content
PP	100	0	0
KF40	60	40	0
KF30-WF10	60	30	10
KF20-WF20	60	20	20
KF10-WF30	60	10	30
WF40	60	0	40

PP: Polypropylene; KF: kenaf fiber; WF: wood flour

formulated according to the various fibre contents (Table 1). MA and DCP were also added at 1 and 0.1 wt% of the batch weight after melting of PP, respectively. The total time of mixing was 8 min at 180ºC and 60 rpm using a Hakke internal mixer (SYS 9000 model). The compounded materials were grinded using a pilot scale grinder (Wieser, WG- LS 200/200 model).

Then, the mixed blends were dried at 105ºC for 4 h. Test specimens were injection moulded at 190ºC to produce standard ASTM tensile specimens. The specimens were stored under controlled conditions (50% relative humidity and 23ºC) for at least 40 h before testing.

It may be noted that the extraction of the natural fibres' extractives is not economically feasible and percticable in industry. In addition, it would not contribute to our study, as it does not affect the analysis.

Fibre and Composite Characterization

After the tensile test was performed, the specimens were cut into geometrically regular shapes samples. Then, the density of the composites was measured, experimentally. Kenaf fibre and wood flour densities were then calculated using the RoM equation.

The elastic modulus of composites was determined using tensile tests. Tensile tests were performed according to ASTM D 638 specification. Tensile tests were carried out using an MTS testing machine with a load cell capacity of 10 kN (model 10/M_Instron, UK) at a cross-head speed of 5 mm/min. Tensile elastic moduli were determined from the slopes of the stress-strain curves.

The volume and weight fractions results were evaluated and the elastic modulus of constituents (kenaf fibre and wood flour) was calculated by back-calculation through RoM equation. However, because of the anisotropy of the fibres and their random distributions, these are spatially taken as averaged. The RoHM has the advantage of not relying on the moduli of the constituents rather it uses the modulus of pure composites. These values are presented only to give an idea of the differences in the elastic moduli.

At least five specimens of each formulation were tested for density and tensile properties determination. The data reported in our work are the mean values of five measurements. Then, values predicted by the RoHM were compared with experimental results for the elastic modulus of the hybrid composites using regression equations and t test was performed for comparing means values. Sigma Plot statistical software was used for data analysis.

RESULTS AND DISCUSSION

Figure 1 shows the tensile moduli of the six studied formulations. The composite containing only kenaf fibre has the highest modulus whereas the composite containing wood flour exhibits the lowest value. As mentioned earlier, kenaf fibres have the high aspect ratio in comparison with wood flour. Thus, by increasing the amount of the kenaf fibres in the composites, their elastic modulus improves. It can be expected

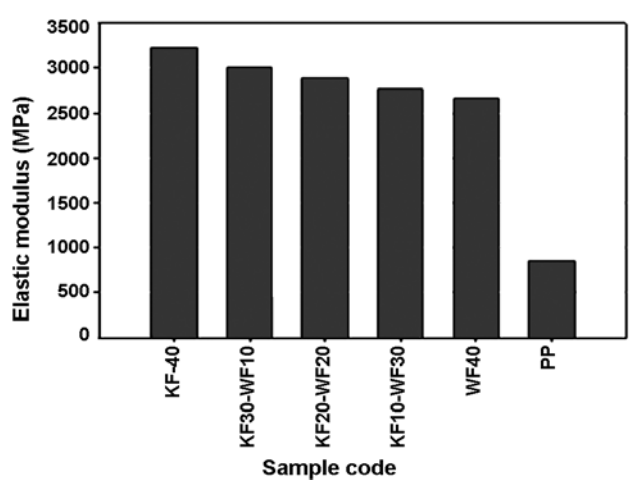

Figure 1. Tensile moduli of various composite formulations.

Table 2. Elastic modulus of the constituents of the composites.

Constituent	Elastic modulus (MPa)
Polypropylene	846.771
Kenaf fib re	17986.5
Wood flour	13543.93

that all hybrid composites present elastic modulus values between the elastic modulus value of pure kenaf fibre and that of the wood flour composites.

Table 2 presents the modulus values of the constituents of the composites determined by RoM equation. As shown in Table 2, kenaf fibres have higher modulus than the wood flour. Hence, it would be expected that hybrid composites containing greater proportions of kenaf fibres have higher elastic modulus. It is evident as the polymer matrix shows the lowest modulus.

Results Based on Weight Fraction

Table 3 illustrates the experimental and predicted (by RoHM equation) elastic moduli values of each hybrid composite employing different weight fractions of the fibres. The difference between the predicted and experimental values is also presented. The double letters A in the forth column (significance level) indicates no significant difference between the predicted and experimental moduli for any of the hybrid formulations as determined by t test. Moreover, the differences between two moduli are quite small. It is also noted that this difference is higher when the kenaf fibre fraction is higher. The RoHM works better in case of particulate (wood flour) than fibrous (kenaf fibres) systems. That is the reason why at higher kenaf fibre concentrations the deviation increases.

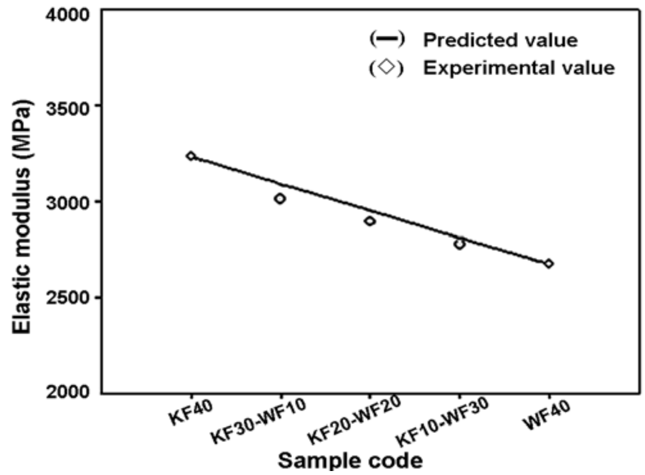

Figure 2. Experimental and RoHM- predicted elastic moduli based on weight fractions.

Figure 2 shows the experimental and predicted values of the elastic modulus of each formulation using weight fraction of the constituents. Due to the nature of the RoHM equation, a linear trend is observed in the predicted values. However, the experimental values exhibit more or less a non-linear trend. As shown, the highest difference between experimental and predicted elastic modulus is observed for KF30 -WF10 formulation (consisting 30% kenaf fibre and 10% wood flour) whereas the lowest difference is seen for KF10 -WF30 (consisting 10% kenaf fibre and 30% wood flour). As mentioned earlier, the reason for

Table 3. Comparison of the predicted and experimental tensile modulus means by t test based on weight fraction.

Sample code	Variable	Mean (MPa)	Significance P = 0.05	Difference (%)
KF40	Experimental Predicted	3229.55 3229.55	-	-
KF30 -WF10	Experimental Predicted	3008.06 3089.90	AA	2.72
KF20 -WF20	Experimental Predicted	2891.01 2950.25	AA	2.04
KF10 -WF30	Experimental Predicted	2771.89 2810.60	AA	1.39
WF40	Experimental Predicted	2670.94 2670.94	-	-

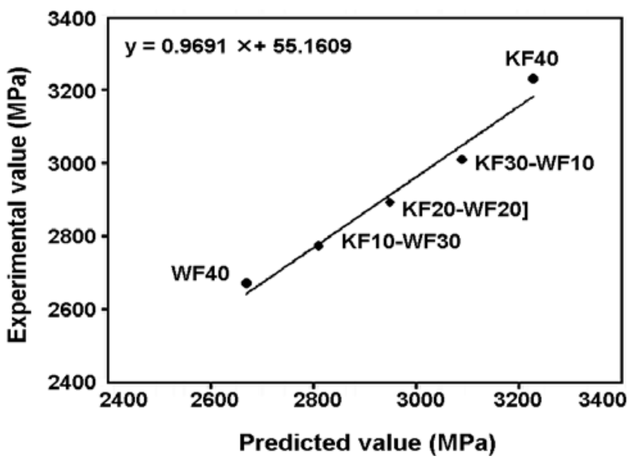

Figure 3. Regression between experimental and RoHM-predicted elastic moduli based on the weight fractions.

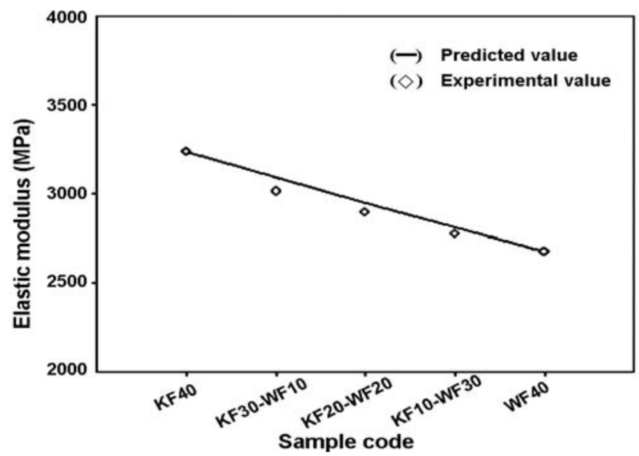

Figure 4. Experimental and RoHM-predicted elastic moduli based on the volume fractions.

the rate of these deviations is the greater compatibility of the RoHM equation with particulate systems than with the fibrous systems. No significant difference is found between the predicted and experimental values at 95% confidence level.

Figure 3 shows the regression between experimental modulus and predicted modulus (calculated by RoHM equation by weight fraction). Analysis of variance of the regression between experimental and predicted values has shown that the regression is significant at 95% confidence level. The regression equation between these values is also presented in Figure 3. The coefficient of determination (R-squared) between experimental and predicted moduli was determined to be 0.986 indicating a strong linear correlation.

Results Based on Volume Fraction

Table 4 shows the difference of mean values between experimental and predicted elastic modulus values of each hybrid composite based on the volume fraction of the constituents. Again, the double letters A indicates no significant difference between experimental and predicted elastic modulus values for all hybrid composites. It is also noted that the difference between experimental and predicted values becomes higher at higher kenaf fibre fractions.

A comparison of the experimental and predicted values of the elastic modulus of each formulation using volume fraction of the constituents is presented in Figure 4. It is clear that the predicted results are very close to the experimental ones. However, as shown, the highest difference between experimental and predicted elastic moduli is observed for KF30 -WF10 formulation (consisting 30% kenaf fibre and 10% wood flour) whereas the lowest difference is seen for KF10 -WF30 (consisting 10% kenaf fibre and

Table 4. Comparison of the predicted and experimental tensile modulus means by t test based on volume fraction.

Sample code	Variable	Mean (MPa)	Significance P = 0.05	Difference (%)
KF40	Experimental Predicted	3229.55 3229.55	-	-
KF30 -WF10	Experimental Predicted	3008.06 3085.95	AA	2.58
KF20 -WF20	Experimental Predicted	2891.01 2945.03	AA	1.86
KF10 -WF30	Experimental Predicted	2771.88 2806.72	AA	1.25
WF40	Experimental Predicted	2670.94 2670.94	-	-

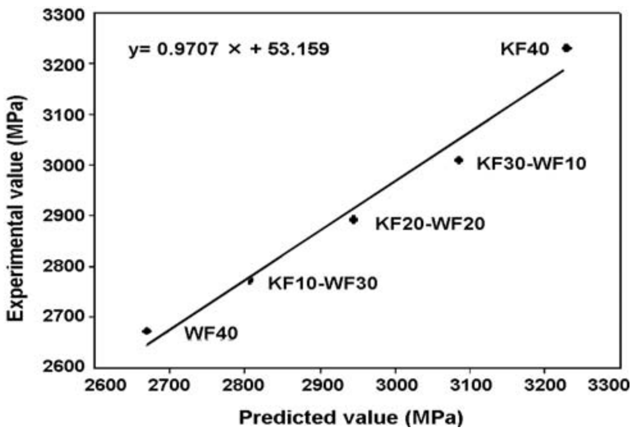

Figure 5. Regression between experimental and RoHM-predicted elastic moduli based on the volume fractions.

30% wood flour). The reason of this deviation has been explained before. As mentioned earlier, no significant difference found between the predicted and experimental values at 95% confidence level.

Figure 5 presents the regression between experimental modulus and predicted modulus (calculated by RoHM equation by weight fraction). Analysis of variance of the regression between experimental and predicted values has proved that the regression is significant at 95% confidence level. The regression equation between these values is also presented. The coefficient of determination (R-squared) between experimental and predicted moduli was determined to be 0.988 indicating a strong linear correlation.

Table 5 presents a summary of the values of deviation from the model for both weight and volume fractions. Both approaches have resulted in very small differences between predicted and experimental values. However, volume fraction seems to be more accurate as can be seen by its lower error values for all three hybrid composite formulations. In all cases, the difference between predicted and experimental values decreases at higher wood flour contents.

Table 5. Percent deviations from the RoHM model based on weight and volume fractions for each hybrid formulation.

Formulation	Deviation from the model (%)	
	Weight fraction	Volume fraction
KF30-WF10	2.72	2.58
KF20-WF20	2.04	1.86
KF10-WF30	1.39	1.25

CONCLUSION

In this study, the elastic modulus of random short discontinuous natural fibre reinforced hybrid composites was predicted using the rule of hybrid mixtures (RoHM) equation. This equation was employed using weight and volume fractions of the reinforcements. It was observed that the RoHM equation predicted the elastic modulus of hybrid composites a little higher than experimental elastic modulus values. However, comparison between experimental and predicted modulus values indicated that the best prediction of modulus by RoHM equation was achieved when volume fraction of the constituents was employed in the equation. Furthermore, in all cases the relationship between experimental and predicted values was determined to be strong by the high R^2 values obtained. This means that a good linear relationship can be expected.

Finally, this suggests that RoHM equation can be suitably applicable to the prediction of the elastic modulus of the short natural fibre hybrid composites. It was also concluded from the experimental values that the linear trend inherently present in the RoHM equation did not seem to be very compatible with the almost non-linear experimental results. This may make necessary the employment of some weighting coefficients for different fibres and/or fillers in order to compensate the higher and/or lower reinforcing efficiencies. The application of the simple RoHM equation for other mechanical characteristics of natural fibre hybrid composites such as tensile strength will also be interesting.

REFERENCES

1- Wu J., Yu D., Chan C., Kim J., Mai Y., Effect of fiber pretreatment condition on the interfacial strength and mechanical properties of wood fiber/PP composites, *J .Appl. Polym. Sci.*, **76**, 1000-1010, 2000.

2- Coutinho F.M.B., Costa T.H.S., Carvalho D.L., Polypropylene-wood fiber composites: Effect of treatment and mixing conditions on mechanical properties, *J .Appl. Polym. Sci.*, **65**, 1227-1235, 1997.

3- Rowell R.M., Sanadi A.R., Jacobson R.E., Caulfield D.F., Kenaf Properties, Processing and Products, In: *Properties of Kenaf/Polypropylene composites*, Jackson M.S. *(Ed.)*, Mississippi State University, Ag & Bio Engineering, Ch. **32**, 381-392, 1999.

4- Rowell R.M., Sanadi A.R., Caulfield D.F., Jacobson R.E., Utilization of natural fibers in plastic composites: Problems and opportunities. In: *Lignocellulosic-plastics Composites*, Leao A.L., Carvalho F.X., Frollini E. (Eds.), University of Rio de Janeiro, 23-51,1997.

5- Sanadi A.R., Caulfield D.F., Jacobson R.E., Rowell R.M., Renewable agricultural fibers as reinforcing fillers in plastics: Mechanical properties of kenaf fiber-polypropylene composites, *Ind. Eng. Chem. Res.*, **34**, 1889-1896, 1995.

6- Sreekalaa M.S., George J., Kumaran M.G., Thomasa S., The mechanical performance of hybrid phenol-formaldehyde-based composites reinforced with glass and oil palm fibres, *Comp. Sci. Technol.*, **62,** 339-353, 2002.

7- Jiang H., Pascal Kamdem D., Bezubic B., Ruede P., Mechanical properties of poly(vinyl choloride)/wood flour/glass fiber hybrid composites, J. *Vinyl Addit. Techn.*, **9**, 138-145, 2003.

8- Jacob M., Thomas S., Varughese K.T., Natural rubber composites reinforced with sisal/oil palm hybrid fibers: Tensile and cure characteristics, *J. Appl. Polym. Sci.*, **93**, 2305-2312, 2004.

9- Kalapasad G., Joseph K., Thomas S., Influence of short glass fiber addition on the mechanical properties of sisal reinforced low density polyethylene composites, *J. Comp. Mater.*, **31**, 509-527, 1997.

10-Rozman H.D, Tay G.S., Kumar R.N., Abubakar A., Polypropylene hybrid composites: A preliminary study on the use of glass and coconut fiber as rienforcements in polypropylene composites, *Polym. Plast. Technol. Eng.*, **38**, 5, 997-1011, 1999.

11-Marcovich N.E., Ostrovsky A.N., Aranguren M.I., Reboredo M.M., Wood flour/sisal fibers as hybrid reinforcement of thermoset polymers, *Nat. Polym. Comp. Conf. Proc.*, 14th-17th June, 419-21, 2000.

12-Thwea M.M., Liaob. K, Durability of bamboo-glass fiber reinforced polymer matrix hybrid composites, *Compos. Sci. Technol*, **63**, 375-387, 2003.

13-Mishra S., Mohanty A.K., Drzal L.T., Mishra M., Parija S., Nayak S.K., Tripathy S.S., Studies on mechanical performance of biofiber/glass reinforced polyester hybrid composites. *Compos. Sci. Technol*, **63**, 1377-1385, 2003.

14-Jacobson R.E., Rodney E., Daniel F. Caulfield, Hybrid composites: Combining cellulose fibers and wollastonite mineralfibers into a nylon 6 matrix, *Proc. 7th Int. Conf. Woodfiber-Plastic Compos.*, 9th-12th June, Madison, WI, USA, 271-276, 2003.

15-Paiva L., Junior C.Z., de Carvalho L.H., Fonseca, V.M. Monteiro S.N., d'Almeida J.R.M., Analysis of the tensile strength of polyester/hybrid ramie–cotton fabric composites, *Polym. Test.*, **23**, 131-135, 2004.

16-Rozman H.D., Hazlan A., Abubakar A., Preliminary study on mechanical stability of rice husk-glass fiber hybrid polyester composites, *Polym. Plast. Technol. Eng.*, **43**, 1129-1140, 2004.

17-Tajvidi M., Falk R.H., Hermanson J.C., Felton C., Static and dynamic mechanical properties of a kenaf fiber-wood flour/ polypropylene hybrid composite, *J. Appl. Polym. Sci.*, **98**, 665-672, 2005.

18-Nielsen L.E., Landel R.F., *Mechanical Properties of Polymers and Composites*, 2nd ed., Marcel Dekker, London, 557, 1994.

19-Agarwal B.D., Broutman L.J., *Analysis and Performance of Fiber Composites*, John Wiley, New York, Ch.1, 1980.

20-Fu S.-Y., Xu G., Mai Y.-W., On the elastic modulus of hybrid particle/short-fiber polymer composites, *Compos. Part B- Eng.*, **33**, 291- 299, 2002.

21-Gibson R.F., *Principles of Composite Material Mechanics*, McGraw-Hill, New York, 421, 1993.