

Tensile Properties of Wood Flour/Kenaf Fiber Polypropylene Hybrid Composites

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ABSTRACT: Hybrid composites of wood flour/kenaf fiber and polypropylene were prepared at a fixed fiber to plastic ratio of 40 : 60 and variable ratios of the two reinforcements namely 40 : 0, 30 : 10, 20 : 20, 10 : 30, and 0 : 40 by weight. Polypropylene was used as the polymer matrix, and 40–80 mesh kenaf fiber and 60–100 mesh wood flour were used as the fiber and the particulate reinforcement, respectively. Maleic anhydride and dicumyl peroxide were also used as the coupling agent and initiator, respectively. Mixing process was carried out in an internal mixer at 180°C at 60 rpm. ASTM D 638 Type I tensile specimens of the composites were produced by injection molding. Static tensile tests were performed to study the mechanical behavior of the hybrid composites. The hybrid effect on the elastic modulus of the composites was also investigated using the rule of

hybrid mixtures and Halpin–Tsai equations. The relationship between experimental and predicted values was evaluated and accuracy estimation of the models was performed. The results indicated that while nonhybrid composites of kenaf fiber and wood flour exhibited the highest and lowest modulus values respectively, the moduli of hybrid composites were closely related to the fiber to particle ratio of the reinforcements. Rule of hybrid mixtures equation was able to predict the elastic modulus of the composites better than Halpin–Tsai equation. © 2007 Wiley Periodicals, Inc. *J Appl Polym Sci* 105: 3054–3059, 2007

Key words: hybrid composites; kenaf fiber; wood flour; polypropylene; rule of hybrid mixtures; Halpin–Tsai equation; elastic modulus

INTRODUCTION

The growing interest in using natural fibers as a reinforcement of polymer-based composites is mainly due to their advantages such as lower cost, renewability, acceptable specific properties, lower density, ease of preparation, lower energy requirements for processing, biodegradability, wide availability, and relative nonabrasiveness over traditional reinforcing fibers such as glass and carbon. However, some limitations in using natural fibers in composites are the lower allowable processing temperatures, incompatibility between the hydrophilic natural fibers and hydrophobic polymers, and high moisture absorption of the fibers and the resulting swelling of the manufactured composite.^{1–3} Thermoplastics 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 fibers are the main natural fibers used as reinforcement.⁴

Hybrid composites are materials made by combining two or more different types of fibers in a common matrix. They offer a range of properties that cannot be obtained with a single kind of reinforcement. Hybridization of two types of short fibers having different length and diameter can offer some advantages over using each of the fibers alone in a single polymer matrix. Hybrid composites have long taken the attention of many researchers as a way to enhance mechanical properties of composites. However, hybrid composites using natural fibers are less studied. And in such studies, the hybrid composite often consists of one natural fiber and one non-natural fiber.^{5–15} Studies on hybrid composites with two natural fiber reinforcement phases are extremely rare.¹⁶

Generally, mechanical properties of a composite are dependent upon the properties of the matrix and reinforcement, the interaction between the matrix and reinforcement, amount, type, arrangement of the fibers within the composite, and fabrication process.¹⁷ Properties of composites are determined through experimental measurements. Experimental methods may be simple and direct. However, one set of experimental measurements determines the properties of a fixed fiber–matrix system produced by a single fabrication process. Additional measurements are required when any change in a system variable occurs such as

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TABLE I
Composition of the Studied Formulations (wt %)

No.	Code	Polypropylene content	Kenaf fiber content	Wood flour content
1	PP	100	0	0
2	KF40	60	40	0
3	KF30	60	30	10
4	KF20	60	20	20
5	KF10	60	10	30
6	WF40	60	0	40

PP, polypropylene; KF, kenaf fiber; WF, wood flour.

relative volumes of the constituents, constituent properties, and fabrication process. Experiments may become time consuming and cost prohibitive. Mechanics based models and semiempirical methods of determining composite properties can therefore be useful to predict the effects of a large number of system variables.¹⁸

The rule of hybrid mixtures equation

The elastic moduli of hybrid short fiber composites can be evaluated using the rule of hybrid mixtures (RoHM) equation, which has been widely used to predict the strength and modulus, etc. of hybrid composites.¹⁹ The modulus of the hybrid composite can be evaluated from the RoHM equation by neglecting the interaction between two systems as:

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

where E_c is the elastic modulus of the hybrid composite. V_{c1} and V_{c2} are the relative hybrid volume fraction of the first system and second system, respectively, and $V_{c1} + V_{c2} = 1$. Also, $V_{c1} = V_{f1}/V_t$ and $V_{c2} = V_{f2}/V_t$, where V_t is the total reinforcement volume fraction and equals $V_{f1} + V_{f2}$.²⁰

The basic rule of mixtures can also be used to determine the “average” stiffness of the reinforcement using back-calculation as indicated in the following equation:

$$E_f = \frac{E_c - E_m V_m}{V_f} \quad (2)$$

where E_f , E_c , and E_m are the elastic moduli of the reinforcement, the nonhybrid composite and the matrix, respectively. V_f is the volume fraction of the reinforcement and V_m is that of the matrix.¹⁹

Halpin–Tsai equation

A modified Halpin–Tsai equation can be used to calculate the elastic moduli of hybrid composites. Therefore, the values of the composites moduli E_{11} and E_{22}

can be derived by using the modified Halpin–Tsai model as follows:

$$E_{11} = \left\{ \frac{1 + 2(l_{f1}/d_{f1})\eta_{L1}V_{f1}}{1 - \eta_{L1}V_{f1}} E_m \right\} + \left\{ \frac{1 + 2(l_{f2}/d_{f2})\eta_{L2}V_{f2}}{1 - \eta_{L2}V_{f2}} E_m \right\} \quad (3)$$

$$E_{22} = \left\{ \frac{1 + 2\eta_{T1}V_{f1}}{1 - \eta_{T1}V_{f1}} E_m \right\} + \left\{ \frac{1 + 2\eta_{T2}V_{f2}}{1 - \eta_{T2}V_{f2}} E_m \right\} \quad (4)$$

$$\eta_{L1} = \frac{(E_{f1}/E_m) - 1}{(E_{f1}/E_m) + 2(l_{f1}/d_{f1})} \quad (5)$$

$$\eta_{T1} = \frac{(E_{f1}/E_m) - 1}{(E_{f1}/E_m) + 2} \quad (6)$$

$$\eta_{L2} = \frac{(E_{f2}/E_m) - 1}{(E_{f2}/E_m) + 2(l_{f2}/d_{f2})} \quad (7)$$

$$\eta_{T2} = \frac{(E_{f2}/E_m) - 1}{(E_{f2}/E_m) + 2} \quad (8)$$

and in the case of random distribution:

$$E_{\text{random}} = \frac{3}{8}E_{11} + \frac{5}{8}E_{22} \quad (9)$$

where E is the elastic modulus; V is the volume fraction; l and d are length and diameter of the fiber, respectively; and the subscripts m , f_1 , and f_2 designate matrix, first fiber, and second fiber, respectively.²¹

The objective of the present study was to evaluate tensile properties of hybrid composites consisting kenaf fiber, wood flour, and polypropylene. The elastic modulus of such composites has also been predicted by employing the RoHM and Halpin–Tsai equations.

EXPERIMENTAL

Materials

Polypropylene, homopolymer, with a melt flow index of 8 g/10min and a density of 0.9 g/cm³, supplied by Arak Petrochemical Industries, Iran, was used in this work as the polymer matrix.

40–80 mesh kenaf fibers 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) was supplied by Merk, Frankfurt, Darmstadt, Germany, and dicumyl peroxide (DCP) (the initiator) was supplied by Hercules, Wilmington, DE.

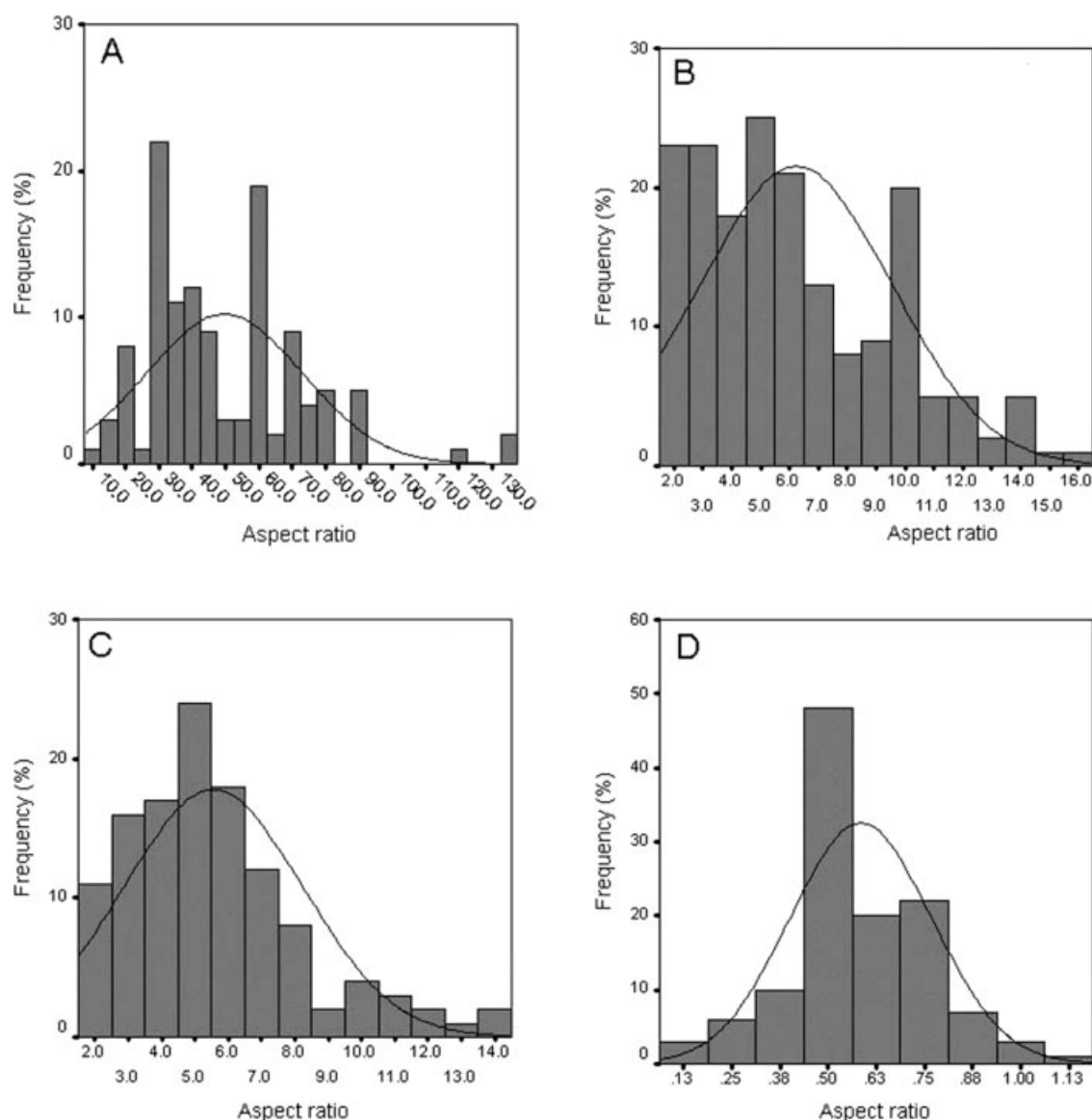


Figure 1 Frequency distributions of the aspect ratios of kenaf fibers and wood flour: (A) kenaf fiber before processing; (B) kenaf fiber after processing; (C) wood flour before processing; and (D) wood flour after processing.

Methods

Fiber preparation

After grinding and passing kenaf fiber and wood flour through predetermined sieves, they were dried in an oven at $(65 \pm 2)^{\circ}\text{C}$ for 24 h before being blended with PP.

Composite preparation

Polypropylene, kenaf fiber and wood flour, were weighed and bagged according to the various fiber contents indicated in Table I. MA and DCP were also added at 1 and 0.1% of the batch weight, respectively. They were then blended at 180°C for 8 min at 60 rpm using a HAAKE internal mixer (SYS 9000 model). The

compounded materials were then ground to prepare the granules using a pilot scale grinder (WIESER, WG-LS 200/200 model).

The mixed blends were then dried at 105°C for 4 h. Test specimens were injection molded at 190°C and an injection pressure of 7 MPa to produce standard ASTM D638 Type I tensile specimens. The specimens were stored under controlled conditions (50% relative humidity and 23°C) for at least 40 h prior to testing.

Fiber and composite characterization

Tensile tests were performed according to ASTM D 638 specification. They were carried out using an MTS testing machine with a load cell capacity of 10 kN (model 10/M) at a cross-head speed of 5 mm/min. An

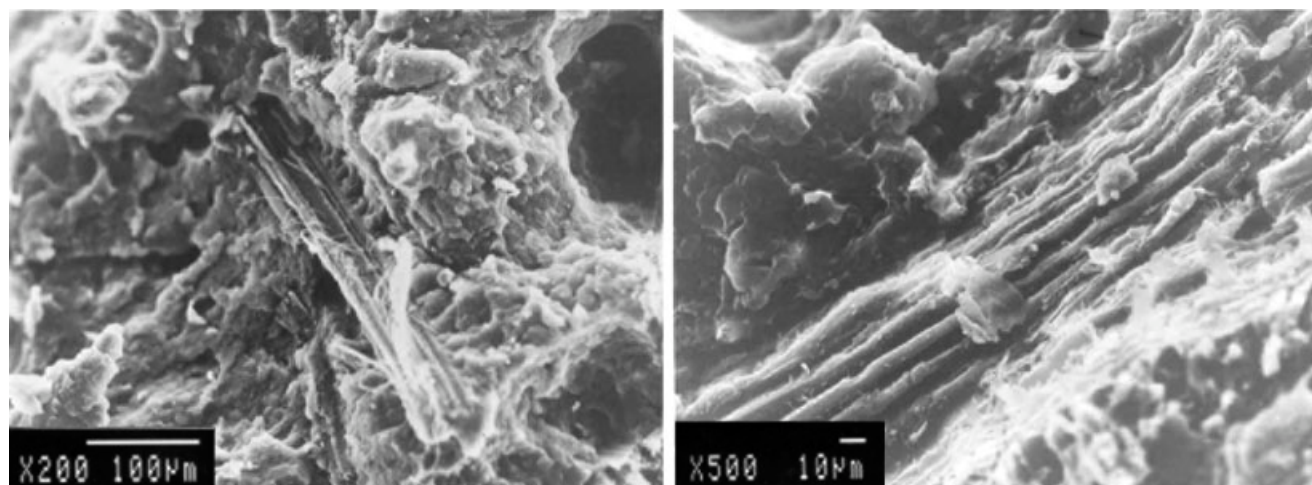


Figure 2 SEM micrographs of fractured surfaces of the KF-20 formulation.

MTS extensometer was mounted on dog-bone specimens and strain data were recorded over a gauge length of 50 mm. Tensile elastic moduli were determined from the slopes of the stress–strain curves.

To study the effect of processing on the fiber characteristics, fiber length, and diameter analyses were performed using light microscopy before and after mixing with polymer matrix. To study the microstructure of the hybrid composites, scanning electron microscopy (SEM) was performed on fractured surfaces of typical samples.

Data analysis

At least five specimens of each formulation were tested. Hence, data reported in the present article are the mean values of five measurements. Values predicted by the RoHM or Halpin–Tsai equations were compared with experimental results for the elastic modulus of the hybrid composites using independent sample *t* tests. All comparisons have been made at 95% confidence level. For fiber length/diameter analysis, at least 120 individual fibers were measured.

RESULTS AND DISCUSSION

Figure 1 exhibits the aspect ratio analysis of both natural fibers before and after processing into composites. Normal distribution curves are also presented for comparison. Dramatic reductions in the aspect ratios of both fibers are easily seen. For kenaf fibers, the mean aspect ratio dropped from 49.6 to 6.2 because of shearing in the mixer and the consequent fiber length reduction during injection molding. These values were 5.6 and 0.59 for wood flour, respectively. In both conditions however, kenaf fibers presented considerably higher aspect ratios than wood flour indicating that in a hybrid system they can be considered as the

fibrous reinforcement while wood flour is mostly particulate. The less than unity value of wood flour aspect ratio after processing is due to the fact that wood fibers are in fact fiber bundles having higher thicknesses than individual fibers. After processing, the fiber lengths are reduced whereas their diameters are almost intact.

SEM micrographs of typical fractured surfaces of tensile specimens of KF20 formulation are presented in Figure 2. A long fractured kenaf fiber in the center of the image clearly indicates both the higher aspect ratio of kenaf fibers as compared with wood flour and the effectiveness of the compatibilizer.⁴ The image also shows the random distribution of fibers in the matrix.

The stress–strain curves of various composite formulations are presented in Figure 3. The curve for pure PP is also presented for comparison. These curves are the average of five specimens. The lowest curve in composite formulations corresponds to the nonhybrid composite of 40% wood flour whereas the

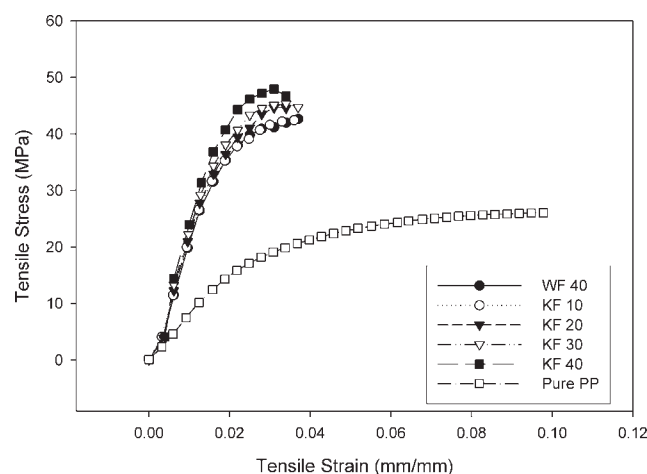


Figure 3 Stress–strain curves of various hybrid and nonhybrid composites.

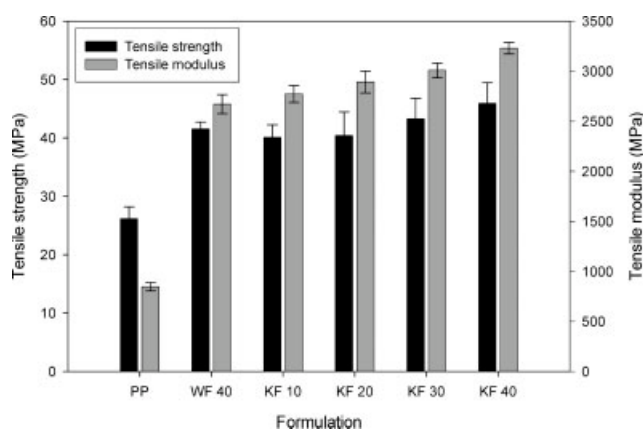


Figure 4 Variations of tensile strength and modulus in various composite formulations.

highest one belongs to the nonhybrid composite formulation containing 40% kenaf fibers. Improvements in both maximum tensile stress and slope of the curves because of the presence of more kenaf fibers are easily detectable. This confirms higher reinforcement of kenaf fibers as compared with that of wood flour.

The values of the tensile strengths and moduli of different nonhybrid and hybrid composites are plotted in Figure 4 where the bar for pure PP is also given for comparison. Following a slight drop (not significant at 95% confidence level) in tensile strength when 10% kenaf fibers are added to the system, the values of tensile strength regularly increased when the kenaf fiber ratio increased. Therefore, hybridization has improved the tensile strength of the composites. A constant increase in the stiffness values is also observed when the kenaf fiber fraction is increased.

Table II presents the modulus values of the constituents of the composites determined by back-calculating a rule of mixtures equation.¹⁹ As shown in Table II, kenaf fibers have higher modulus than that of wood flour. Hence, it would be expected that hybrid composites containing greater proportions of kenaf fibers have higher elastic modulus. The polymer matrix shows the lowest modulus. These values have been used in the Halpin–Tsai equation. It should be however mentioned that natural fibers are very anisotropic, so their moduli in Table II are spatially average. A comparison of these values with those reported in the literature reveals that the calculated values are close to reported values.^{3,4}

TABLE II
Elastic Modulus of the Constituents of the Composites

Constituent	Elastic modulus (MPa)
Polypropylene	847
Kenaf fiber	17,987
Wood flour	13,544

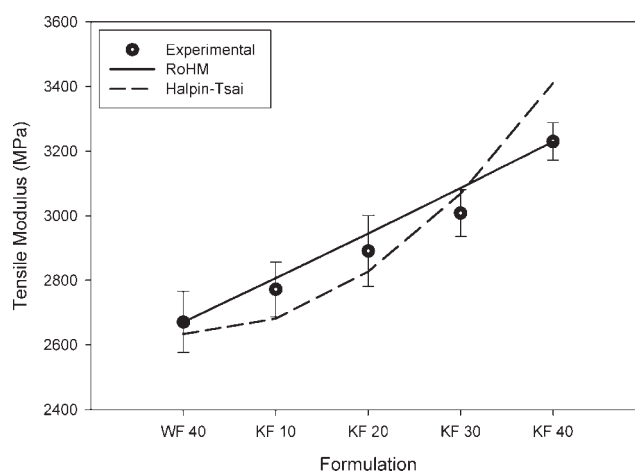


Figure 5 Comparison of modulus experimental results with RoHM and Halpin–Tsai equations.

A comparison of experimental modulus values with predicted values using RoHM and Halpin–Tsai equations is presented in Figure 5. The black circles indicate experimental values. Because of the nature of RoHM equation, a linear trend is observed for its predicted values (white squares). All predicted values are also higher than experimental ones. However, in the case of the Halpin–Tsai equation, a different nonlinear trend is observed. Furthermore, the Halpin–Tsai equation underestimated the modulus values for WF40, KF10, and KF20 formulations whereas it overestimated this property at higher kenaf fiber contents.

A detailed statistical analysis of the accuracy of the two equations in predicting the elastic modulus of hybrid composites is presented in Table III. A double A letter indicates no significant difference between experimental and predicted elastic modulus values, whereas the letters AB indicate a statistically significant difference. Because the RoHM predicts the properties of the hybrid composites using the properties of nonhybrid composites, no data are presented for nonhybrid composites of KF40 and WF40. It is also noted that the difference between experimental and predicted values becomes higher at higher kenaf fiber fractions. RoHM predicted values are very close to those obtained experimentally as seen by small difference values. It is also observed that the difference between predicted and experimental values becomes smaller at higher wood flour contents. Fu et al. reported that for a hybrid system of short glass fiber/calcite ABS composite, the rule of hybrid mixtures performed well in the estimation of the elastic modulus.¹⁹ The results of the present study also confirm that the simple rule of hybrid mixtures can provide a good means for rapid estimation of the elastic properties as seen by small deviations from the model. Halpin–Tsai equation on the other hand, is able to predict the modulus of nonhybrid composites (WF40 and KF40) as well. All predicted values are significantly

TABLE III
Comparison of the Predicted and Experimental Tensile Modulus Means by *t* test

Equation	Formulation	Variable	Mean (MPa)	Significance <i>P</i> = 0.05	Difference (%)
RoHM	KF40	Experimental	3,229	–	–
		Experimental	3,008	AA	2.58
	KF20	Predicted	3,086		
		Experimental	2,891	AA	1.86
	KF10	Predicted	2,945		
		Experimental	2,771	AA	1.25
	WF40	Predicted	2,807		
		Experimental	2,671	–	–
	KF40	Experimental	3,229	AB	5.61
		Predicted	3,411		
Halpin–Tsai	KF30	Experimental	3,008	AA	2.02
		Predicted	3,069		
	KF20	Experimental	2,891	AA	2.24
		Predicted	2,827		
	KF10	Experimental	2,771	AA	3.23
		Predicted	2,682		
	WF40	Experimental	2,671	AA	1.37
		Predicted	2,634		

similar to the experimental values with the exception of the KF40 formulation. However, even in that case, the difference is quite small (5.61%). The results of using the modified Halpin–Tsai equation are better than those obtained by Biagiotti et al. for the composite system of glass fiber/natural fiber polypropylene composites.²¹

CONCLUSIONS

Hybridization can improve the mechanical properties of natural fiber plastic composites. The results of the present study confirm that it is possible to enhance such properties by adding longer fibers (such as kenaf) to wood flour plastic composites. Both tensile modulus and tensile strength were improved when kenaf fibers were added to the wood flour/PP system. In this study, the elastic modulus of random short discontinuous natural fiber reinforced hybrid composites was also predicted using the rule of hybrid mixtures and a modified Halpin–Tsai equation. Both equations satisfactorily predicted the elastic moduli of the hybrid composites. However, the RoHM proved better by providing smaller error values. The RoHM is also considerably easier to use because it only requires data on the properties of nonhybrid composites and there is no need to have information on the individual fibers mechanical properties or aspect ratios. It is also concluded from the experimental that the linear trend inherently present in the RoHM equation seems not to be very compatible with the almost nonlinear experimental results. This may make necessary the employment of some weighting coefficients for different fibers and/or fillers to compensate for higher and/or lower reinforcing efficiencies. The application of the simple RoHM equation for other mechanical characteristics of natural fiber hybrid composites will also be interesting.

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