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Interfacial and mechanical properties of environment-friendly "green" composites made from pineapple fibers and poly(hydroxybutyrate-co-valerate) resin

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Physical and tensile properties of pineapple fibers were characterized. Tensile properties of pineapple fibers, like most natural fibers, showed a large variation. The average interfacial shear strength between the pineapple fiber and poly(hydroxybutyrate-co-valerate) (PHBV) was 8.23 MPa as measured by the microbond technique. Scanning electron microscopy (SEM) photomicrographs of the microbond specimens revealed an adhesive failure of the interface. Fully degradable and environment-friendly "green" composites were prepared by combining pineapple fibers and PHBV with 20 and 30% weight content of fibers placed in a 0°/90°/0° fiber arrangement. Tensile and flexural properties of these "green" composites were compared with different types of wood specimens. Even though tensile and flexural strength and moduli of these "green" composites were lower than those of some wood specimens tested in grain direction, they were significantly higher than those of wood specimens tested in perpendicular to grain direction. Compared to PHBV virgin resin, both tensile and flexural strength and moduli of these "green" composites were significantly higher. SEM photomicrographs of the fracture surface of the "green" composites, in tensile mode, showed partial fiber pull-out indicating weak bonding between the fiber and the matrix.

1. Introduction

Fiber-reinforced polymeric composites have become popular for a variety of applications because of their high specific strength and modulus. Most composite materials currently available in the market, with long term durability in mind, are made from nondegradable polymeric matrices and fibers. Although disposal of these composites, after their intended use, is not an immediate problem, with double digit growth in their use, their disposal is expected to become critical in the near future. Environment-friendly and fully degradable "green" composites, made from both biodegradable polymeric matrices and fibers, should be advantageous in such situations. These composites could be easily disposed of or composted after their intended use without harming the environment. "Green" composites, in general, may not have high strength as in the case of advanced composites. However, there are many mass volume, noncritical applications at present where composites with moderate strength may be desirable.

Poly(hydroxybutyrate-co-valerates) (PHBVs) are naturally occurring biodegradable polymers produced from a wide range of microorganisms [1, 2]. Mechanical properties of PHBV polymers are comparable to those of traditional thermoplastics such as polyethylene and polypropylene [3, 4]. Although PHBV polymers represent a new generation of biodegradable polymers, their applications have been limited because of their high cost [5]. Incorporation of fillers or fibers could not only make them more affordable but also improve their mechanical properties. There have been some studies on use of fillers, such as clay and calcium carbonate [6] and wood fibers [7] to modify properties of PHBV resins. Biodegradable fillers and fibers, such as strong natural cellulosic fibers, would not only provide reinforcement for PHBVs but also keep the advantage of complete biodegradability. Even though many reports have been published on using natural cellulosic fibers, including jute, sisal etc., as reinforcements for polymers [8–11], much less research has been published on using natural cellulosic fibers as reinforcements for biodegradable polymers such as PHBVs. No continuous or long staple, biodegradable fiber reinforced PHBV composites have been reported up to now.

The mechanical properties of fiber reinforced polymeric composites are of great importance in deciding their end applications. Mechanical properties of composites depend on the properties of constituent fibers, the matrix, and the fiber/matrix interfacial shear strength (IFSS) [12]. Fibers are the main load bearing

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components of a composite material, and the fiber strength generally reflects directly the strength of composite. Matrix is responsible for holding the fibers in place and, more importantly, transferring the load from the broken fibers to their neighboring, intact fibers through the interface. The IFSS is a critical factor that controls the toughness, transverse mechanical properties, and interlaminate shear strength of composite materials [12]. Improvement of the IFSS increases the tensile and flexural strength of a composite, while lowering the impact strength and toughness. While measuring the mechanical properties of fibers and resins is straightforward, measuring IFSS is much more complex. Three major techniques based on micromechanical analysis: the microbond test [13], single fiber composite (SFC) test [14, 15] and single fiber pullout test [16, 17], have been commonly employed to measure the IFSS.

This paper presents mechanical properties of "green" composites made from long staple pineapple fibers, a natural cellulosic fiber, and PHBV resins. These fibers are extracted from the leaves of the plant *Ananas Cosmosus* belonging to Bromeliacese family [18]. Mechanical properties of these "green" composites are compared with those of a variety of woods. Pineapple fiber/PHBV interfacial shear strength (IFSS) values have been measured using the microbond test.

2. Experimental

2.1. Materials

Pineapple fibers were provided by Dr. Villegas of University of Phillipines at Los Banos. These fibers, with an average length of about 1.0 m, were extracted from pineapple leaves. PHBV polymer with a melting temperature, T_m, of 162 °C was provided by Zeneca Bio Products and used as received.

2.2. Fiber characterization

Tensile properties of pineapple single fibers were measured using an Instron tensile testing machine, Model 1122, according to ASTM D3379. Specimen gauge length was 50 mm and testing was performed at a strain rate of 0.04 min^{-1}. One hundred pineapple single fiber specimens were picked out randomly and their diameters were measured using a Leitz polarized light microscope, Model Ortholux, with a calibrated (micrometer) eyepiece. Individual single fiber specimens were then mounted and glued on a paper tab with a drop of ultra super glue. Detailed description of single fiber specimen preparation for tensile test can be found elsewhere [19]. Specimens were tested after being equilibrated at standard ASTM conditions of 21 °C and 65% relative humidity for more than 24 h. Fifty successful tests were conducted to obtain the tensile properties of pineapple fibers. Mass density of fibers was measured using a density gradient column at TRI/Princeton in Princeton, NJ.

2.3. Interface characterization

Pineapple fiber/PHBV interfacial shear strength (IFSS) was obtained using the microbond test [13]. Specimens for the microbond tests were prepared by keeping two small pieces of thin PHBV films, at 2 cm apart, on a single pineapple fiber followed by quickly heating the films to 180 °C in an oven to melt the PHBV and form microdroplets. Specimens were removed from the oven after five minutes at 180 °C and cooled down to room temperature. All specimens were then equilibrated at standard conditions of 21 °C and 65% relative humidity for more than 24 h before performing the microbond test on the Instron testing machine using a special microvise. Schematic of the microbond test is shown in Fig. 1a. The diameter, d, and embedded length, l, of the fiber were measured prior to the microbond test using the optical microscope with a calibrated eyepiece. To conduct a microbond test, fiber was pulled out from the microdroplet at a rate of 0.2 mm/min. Prior to the test the microvise plates, as shown in Fig. 1a, were positioned just above the microdroplet and brought closer until they just touched the fiber surface. At that point, a small frictional resistance was detected. As the fiber was pulled out, shear load at the fiber/microdroplet interface increased. Debonding occurred when the load exceeded the interfacial bond strength. Average IFSS, τ, was calculated using the following Equation 1

$$\tau = \frac{F}{\pi \times d \times l} \tag{1}$$

where F is the load to debond the microdroplet [13]. It was assumed that the shear strength was uniform along the entire fiber/microdroplet interface. A typical load vs. displacement plot of a successful test is shown in Fig. 1b. Fifty successful tests were conducted to obtain the average IFSS value.

2.4. Composite laminate preparation

Composite laminates with a thicknesses of 1 mm were made by sandwiching three layers of fibers between four layers of PHBV films. Laminate thickness was controlled by using a 1 mm stainless steel spacer. Three layers of pineapple fibers were aligned between PHBV films in a parallel array and the whole assembly was carefully placed in a special mold made for making composite laminates. The three fiber layers were arranged in 0°/90°/0° directions with 25% of the fiber weight in the top and bottom layers each and the remaining 50% in the middle layer. The exact fiber content of composite laminates was calculated from their weight and the weight of fibers added. The mold was placed on a Carver laboratory press with a temperature and pressure control. Heating was started immediately and 140 MPa pressure was applied when the temperature reached 180 °C. The mold was removed from the press after equilibrating for 5 min at 180 °C and quickly cooled down (quenched) by a fan. Preliminary studies had indicated that 5 min at 180 °C was sufficient for the resin to completely impregnate the fibers without significant degradation. Both PHBV virgin resin sheets and composite laminates were prepared with the same thermal history.

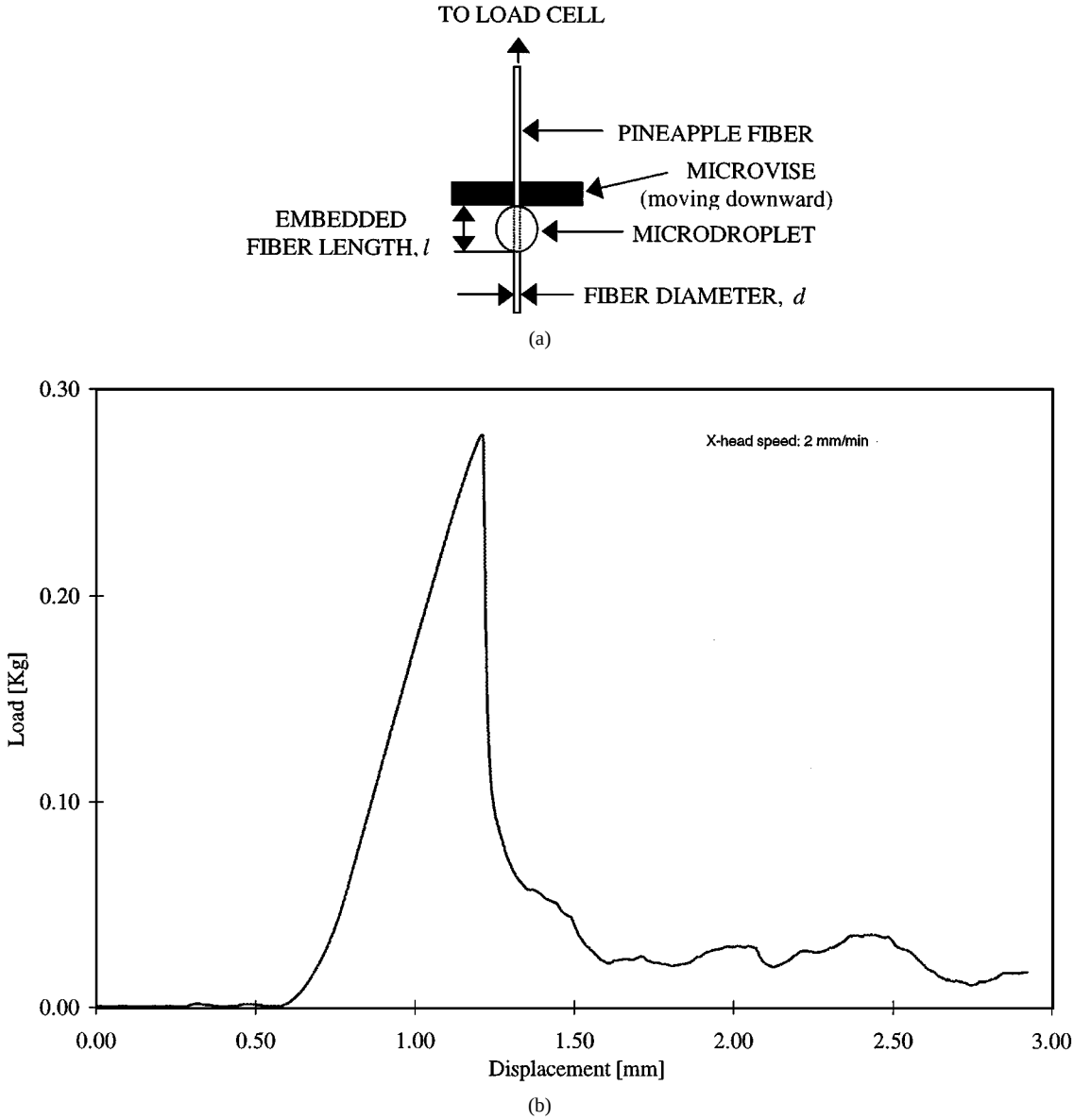

Figure 1 (a) Schematic of the microbond test, (b) the typical load vs. displacement plot of a microbond test.

2.5. Measurement of mechanical properties of composites and woods

Composite specimens of 90 mm × 10 mm × 1 mm and 50 mm × 25 mm × 1 mm dimensions were cut from the laminates to determine their mechanical properties. The specimen length direction was parallel (0°) to fiber axis. Wood tabs were glued to both ends of the 90 mm × 10 mm × 1 mm specimens to obtain a gauge length of 50 mm. Tensile tests were performed using the Instron tensile tester according to ASTM D3039 at a strain rate of 0.04 mm^{-1}. Specimens with dimensions of 50 mm × 25 mm × 1 mm were used for three-point bending tests on the Instron machine with a cross-head speed of 0.43 mm/min according to ASTM D790M. Three different wood specimens; basswood, cherry wood, and walnut wood, with same dimensions were cut from wood sheets, parallel and perpendicular to grain directions. Tensile and flexural tests of wood specimens were performed under the same conditions as those for composite laminates. All tests were performed under standard ASTM conditions of 21 °C and 65% relative humidity. Seven successful tests were conducted to obtain average tensile and flexural properties.

2.6. Surface characterization

Surface topography of fibers, microbond specimens and failed interfacial surface, and fracture surface of the composite laminates after tensile tests were investigated using a scanning electron microscope (SEM), Model LEICA 440.

3. Results and discussions

3.1. Characterization of pineapple fibers

Tensile properties of pineapple fibers are presented in Table I. Although the average strength of pineapple fibers is 445 MPa, and low compared to advanced fibers such as Kevlar and graphite, the strength is sufficient for their use as reinforcement in composites with moderate strength, for noncritical applications. The strength data

TABLE I Tensile properties of pineapple fibers

Density ρ (g/cm^3)	Average strength $\sigma_{average}$ (MPa)	Young's modulus E (GPa)	Fracture strain ε_{break} (%)
1.36	445 (40.93)[a]	13.21 (31.78)	3.37 (20.47)

[a]Numbers in the parenthesis are percent coefficient of variation.

were also fit to a two parameter Weibull distribution of the following formula

$$F(X) = 1 - \exp[-(X/X_0)^m] \quad (2)$$

where X_0 and m are Weibull scale and shape parameters, respectively. The Weibull scale and shape

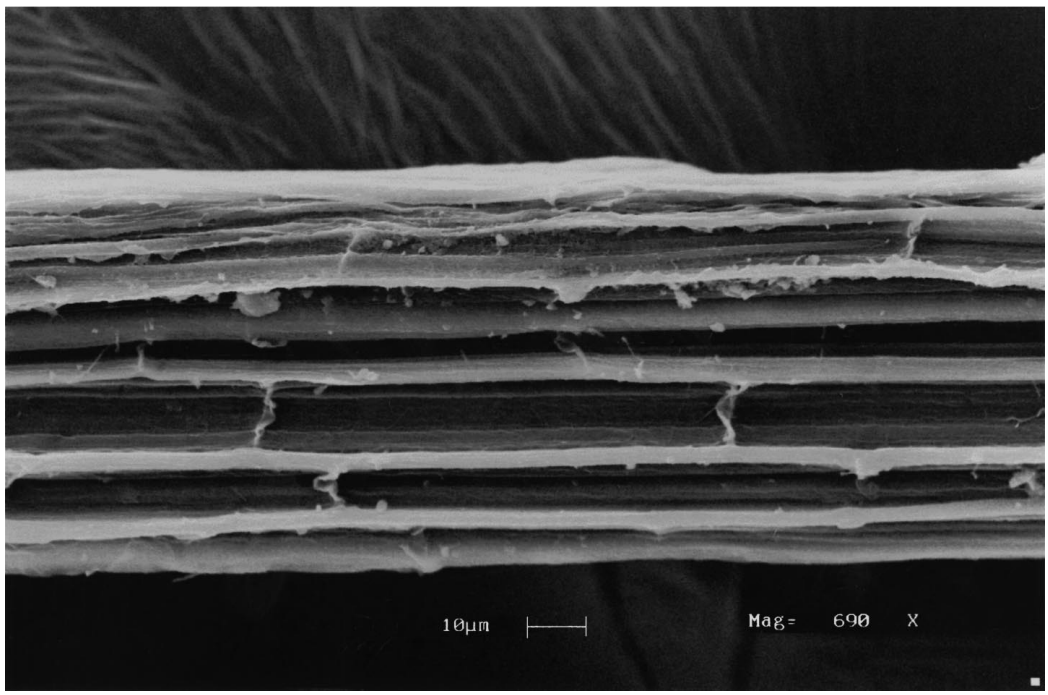

Figure 2 A SEM photomicrograph of a pineapple fiber.

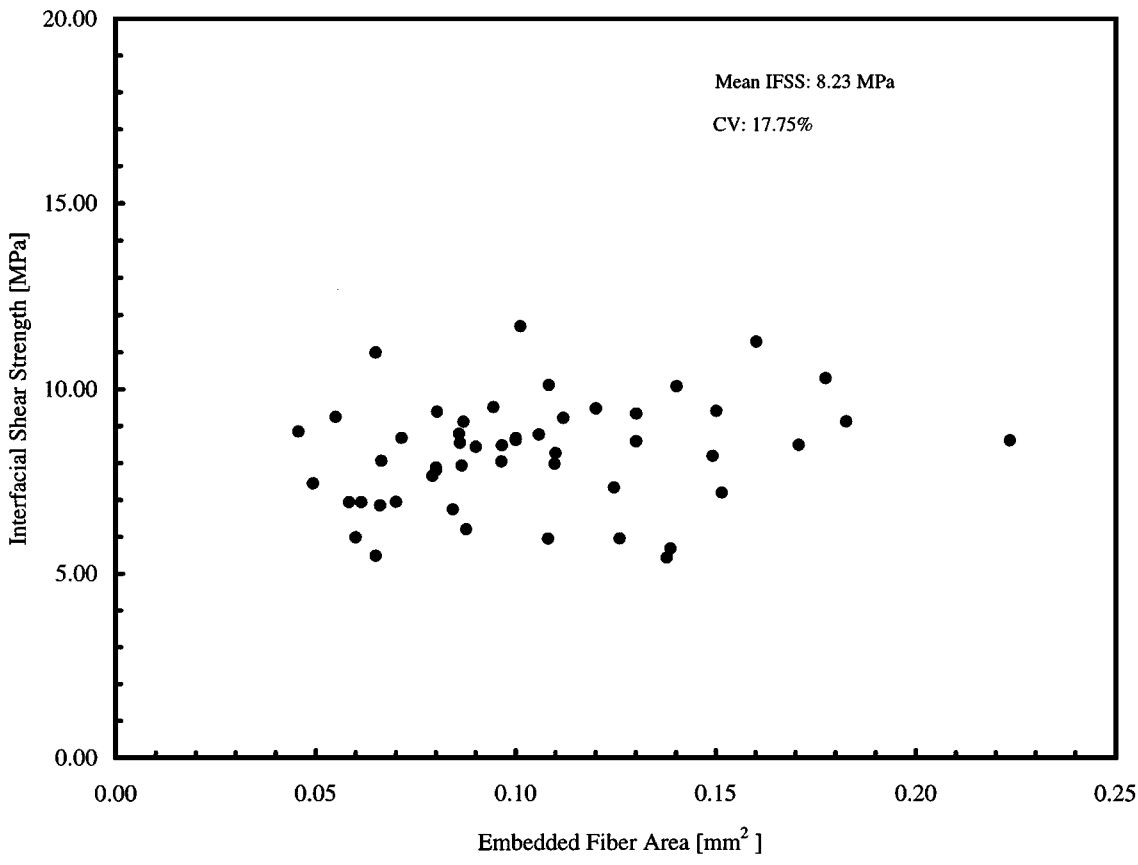

Figure 3 Pineapple fiber/PHBV interfacial shear strength vs. embedded area.

parameters for pineapple fibers were calculated to be 487 MPa and 4.67, respectively. Compared to the strength value of pineapple fibers obtained by Mukherjee and Satyanarayana [20], 362–748 MPa, our value is slightly lower. One possible reason could be different sources from which fibers were obtained. One important characteristic, common for all natural cellulosic fibers, is high variation in their mechanical properties such as fracture stress, strain, and Young's modulus, due to their high inherent irregularity as clearly seen from data presented in Table I. Factors such as geological location of plant, position along the length of the leaf and age contribute to this variation.

Fig. 2 shows a photomicrograph of a pineapple fiber. It is clear that pineapple fibers have a cellular structure. The cells together form fibrils with tissues connected with each other at several locations along the length to form fibers. The fibers are somewhat irregular in the cross section and the diameter varies along its axis, like most other natural cellulosic fibers. Extreme care was taken while separating pineapple fibers. However, some damage still occurred, reducing the fiber strength. This may be one reason for higher strength deviation as well as lower strength compared to the value obtained by Mukherjee and Satyanarayana [20]. However, cellular structure makes these fibers porous giving a higher estimate of the diameter that results in a lower fracture stress value. Without the porosity, the fracture stress would have been significantly higher than the calculated average value of 445 MPa, in this study.

3.2. Interface characterization

The microbond test used in this study provides a simple and effective way to measure pineapple fiber/PHBV interfacial shear strength (IFSS) values. A plot of IFSS vs. embedded fiber area is shown in Fig. 3. The mean IFSS value obtained is 8.23 MPa, with a coefficient of variation of 17.75%. Compared to the 30 to 60 MPa for the IFSS between fibers, such as kevlar, glass and graphite, and epoxy resin [15], this value is much lower. The IFSS depends mainly on two factors: mechanical interlocking and chemical bonding. In the case of pineapple fiber/PHBV resin, hydrogen bonding is possible between the ester group on the resin and -OH group on the fiber. However, because of the hydrophobicity of methyl and ethyl pendant groups in the resin, hydrogen bonding probability is low. As a result, the IFSS is mainly attributed to the high surface irregularity of pineapple fibers, as seen in Fig. 2, and the resulting mechanical interaction. However, high viscosity of the resin seems to preclude much mechanical bonding.

Fig. 4a–c are SEM photomicrographs of a microbond specimen, the bottom end, and the upper end of the microdroplet, respectively, after debonding. Fig. 4b and c, show clean fiber surface with no resin attached to the fiber surface and no fibrils pulled out after debonding indicating adhesive fracture at the interface. An adhesive failure of the interface confirms the low IFSS measured by the microbond technique as well. In cases where the IFSS is larger than the strength of matrix or fiber, matrix or fiber failure is observed before debonding. In pineapple fiber/PHBV resin only interfacial failure was observed.

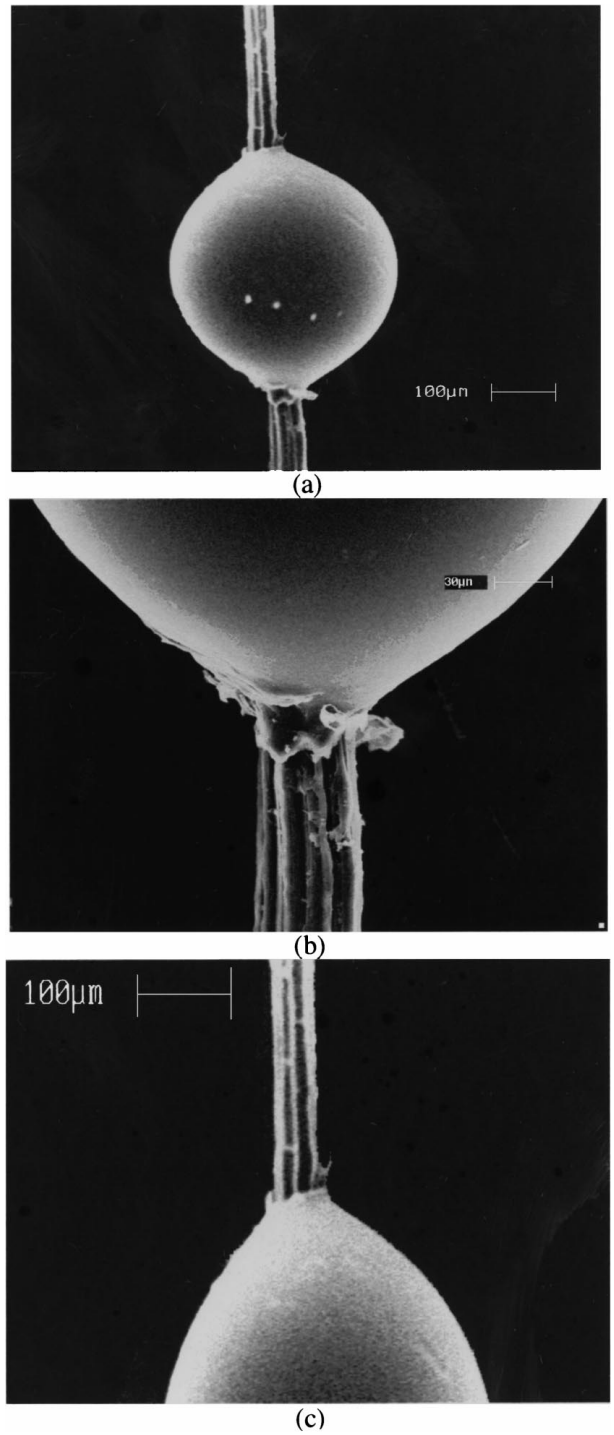

Figure 4 SEM photomicrographs of a microdroplet. (a) After debonding; (b) the bottom end of the debonded microdroplet; (c) the upper end of the debonded microdroplet.

3.3. Tensile and flexural properties of "green" composites and woods

Fully degradable "green" composites were made from pineapple fibers and PHBV resin with total fiber contents of 20 and 30% by weight. Typical tensile and flexural stress vs. strain plots of the "green" composites in fiber axis direction (0°) are shown in Fig. 5. Fig. 6 presents the maximum tensile stress and flexural

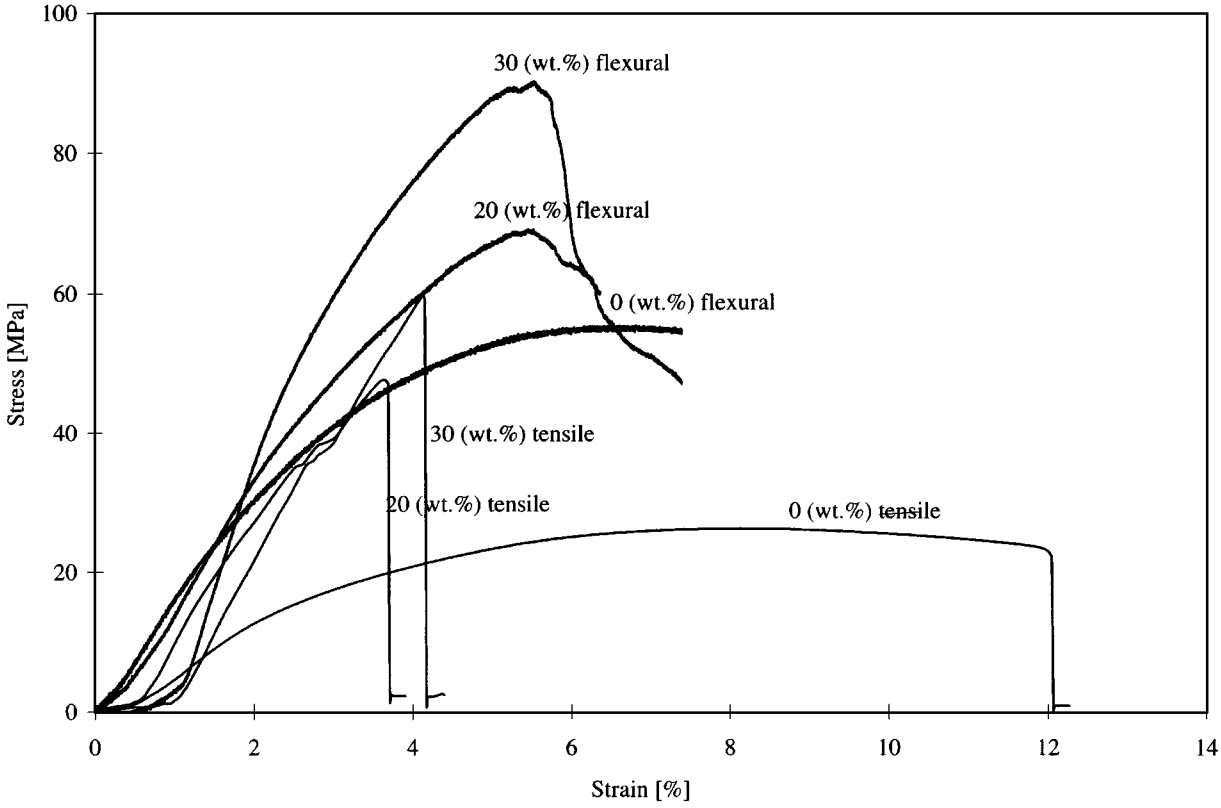

Figure 5 Typical stress vs. strain plots (0° fiber axis direction), in both tensile and flexural modes, of pineapple fiber/PHBV "green" composites (0°/90°/0°).

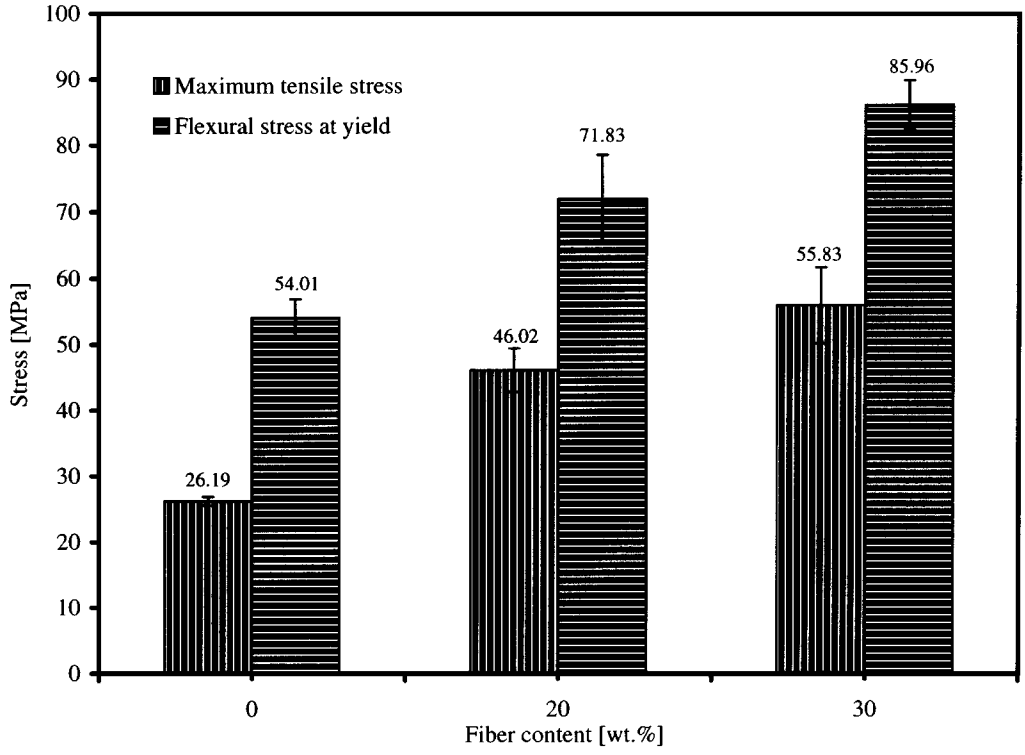

Figure 6 Tensile and flexural stress (0° fiber axis direction) vs. total fiber content of pineapple fiber/PHBV "green" composites (0°/90°/0°).

stress at yield in fiber axis direction (0°) as a function of fiber content. It can be seen, as expected, that the incorporation of pineapple fibers improves the strength of PHBV, both in tensile and flexural mode, while decreasing the strain to failure in tensile mode and flexural strain at yield. Composites with 30% fiber weight content show an increase of the tensile strength by about 100% and flexural strength by about 60%. With 20% fiber weight content the changes in tensile strength and flexural strength are over 75 and 32% respectively. It

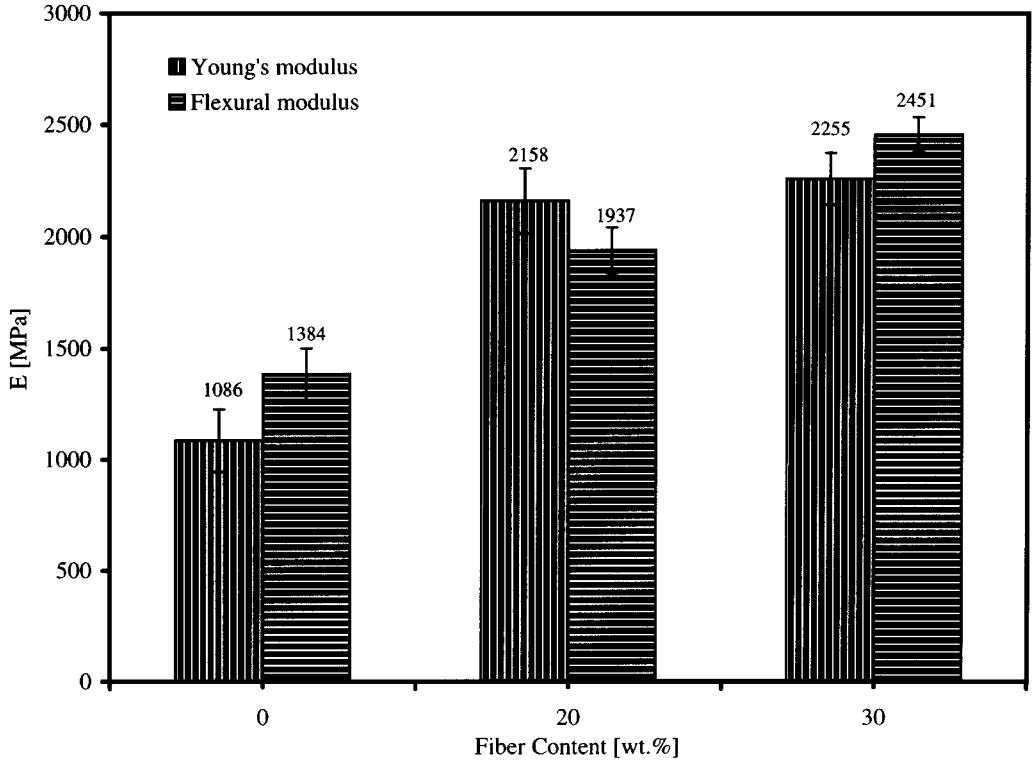

Figure 7 Young's and flexural modulus (0° fiber axis direction) vs. total fiber content of pineapple fiber/PHBV "green" composites (0°/90°/0°).

should be noted that for composites with 30% fibers, only 15% fibers are aligned in 0° direction. Mean values of Young's and flexural moduli of PHBV and pineapple fiber reinforced composites in (0°) fiber axis direction are shown, as histograms, in Fig. 7. Compared to PHBV virgin resin, composites with 30% fiber weight content show an increase of Young's modulus by about 107% and flexural modulus by about 77%. With 20% fiber weight content the increases in Young's and flexural moduli are 99 and 40% respectively.

According to the rule of mixture [12], average tensile strength and modulus of unidirectional continuous fiber reinforced composites can be given by

$$\sigma_c = \sigma_f V_f + \sigma_m V_m \quad (3)$$

$$E_c = E_f V_f + E_m V_m \quad (4)$$

where σ_c, σ_f and σ_m are the stress of composites, fiber and matrix; E_c, E_f and E_m are the modulus of composites, fiber and matrix; V_f and V_m are the volume fraction of fiber and matrix. Since fiber content in fiber axis direction (0°) is the same as that in perpendicular direction (90°), we assume these composites to have same properties in both directions. The calculated values of tensile strength of composites in 0° direction of 52 and 72 MPa with total fiber weight contents of 20 and 30%, respectively, are higher than the experimentally obtained values of 46 and 56 MPa. This is attributed to somewhat less than unidirectional nature of the composites obtained in this study and lower IFSS values. Even though the fibers were aligned in a parallel array prior to placing in the mold, they moved slightly away from the center during the molding process because of the high viscosity of the PHBV melt and 140 MPa pressure, resulting in a bow-like non-unidirectional fiber orientation. Also, for composites with 30% fiber content, it was found that some fibers were unimpregnated by the resin, which resulted in higher deviation of experimental value from the theoretical value than composites with 20% fiber content. Another factor contributing to lower experimental strength values could be the very small amounts of voids present in the composite specimens. With further improvements in the processing of composites, the tensile strength could increase and be comparable to theoretical values. Composites with a 20% fiber content have higher experimental value of Young's modulus, 2.2 GPa, than theoretical value of 1.7 GPa. However, composites with a 30% fiber content have the same experimental value of Young's modulus, 2.3 GPa, as the theoretical value. Higher deviation from unidirectional orientation was noticed for fibers in the center layer for composites with a 20% fiber content than for composites with a 30% fiber content, which results in the higher experimental value in 0° direction.

Both virgin resin and composite specimens showed yielding in the three-point bending test. The quick cooling down process during virgin PHBV resin and composite processing gives low crystallinity of PHBV, which results in increased flexibility of both virgin resin sheets and composite laminates.

Fig. 8 presents the fracture strain in tensile mode and flexural strain at yield, of virgin PHBV resin and pineapple fiber reinforced composites in histogram format. The composites, as expected, have lower tensile fracture strain and flexural strain at yield than the virgin resin. The fracture strain in tensile mode of composites with total fiber weight contents of 20 and 30% is about 3%, which is close to the fracture strain of pineapple fibers and significantly lower than over 10% for PHBV resin. Although much more brittle than the resin, it can

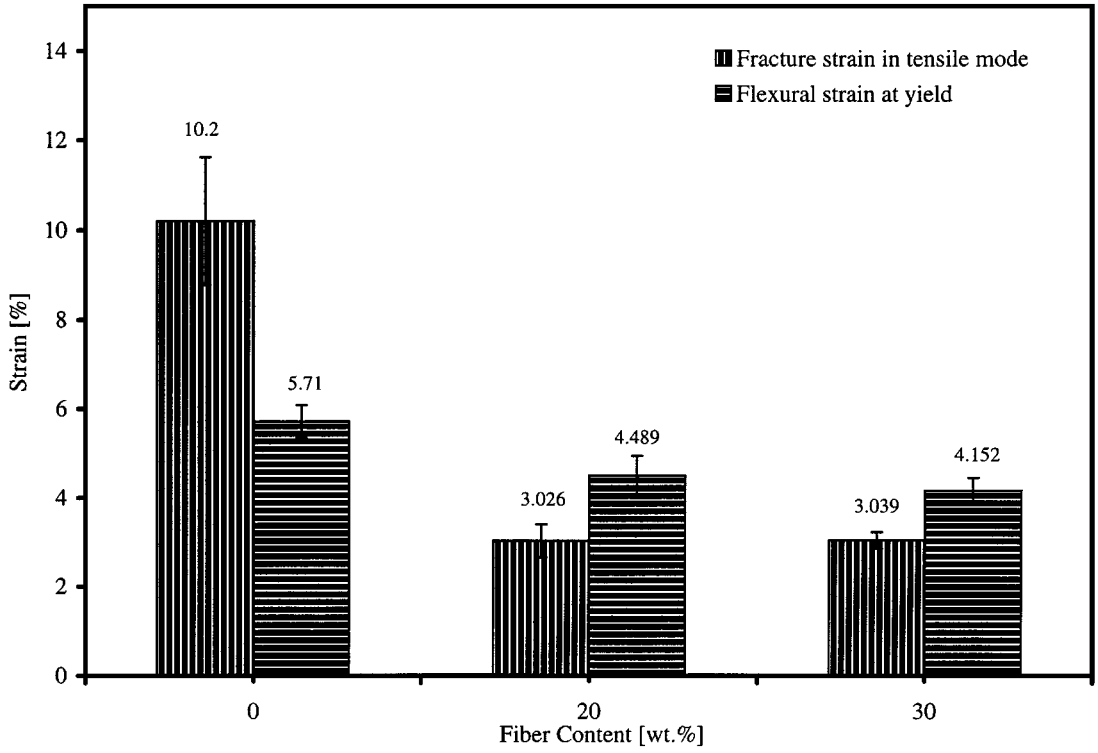

Figure 8 Tensile and flexural strain (0° fiber axis direction) vs. total fiber content of pineapple fiber/PHBV "green" composites (0°/90°/0°).

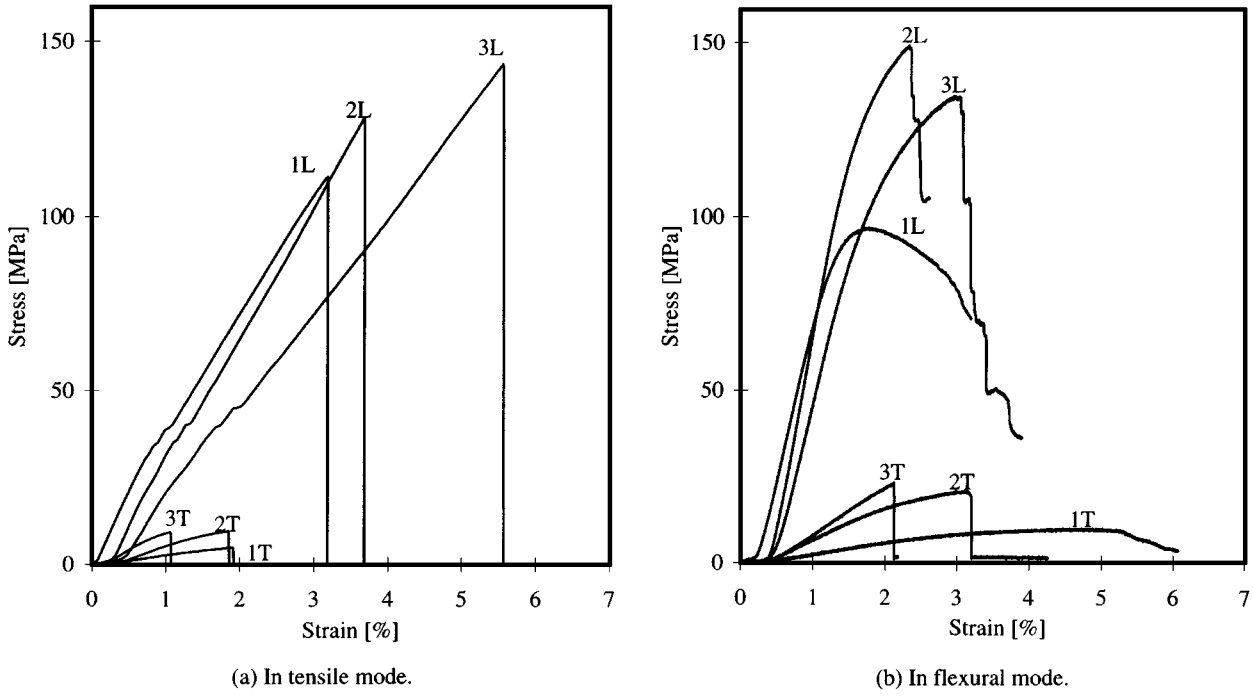

Figure 9 Typical stress vs. strain plots of woods in (a) tensile mode and (b) flexural mode.

be seen from Fig. 5 that the composites show some yielding. The flexural strain at yield of composites, however, decreases slightly with the increase of fiber content.

Typical stress vs. strain plots of three different wood specimens tested in grain and perpendicular to grain directions are shown in Fig. 9a in tensile mode and (b) in flexural mode. Fig. 9a shows that bass, cherry, and walnut woods exhibit nonisotropic behavior with high tensile strength, modulus, and strain to failure in grain direction and low values in perpendicular to grain direction. Fig. 9b shows that flexural strength and moduli of these woods in perpendicular to grain direction are much lower than those in the grain direction.

Strength and modulus of wood specimens, in both grain direction and perpendicular to grain direction, are presented in Figs 10 and 11. Even though the woods have high strength and modulus in grain direction (0°), their strength and modulus in perpendicular to grain direction (90°) are very low. Pineapple fiber/PHBV composites in this study exhibit somewhat lower strength and modulus than these woods in grain direction,

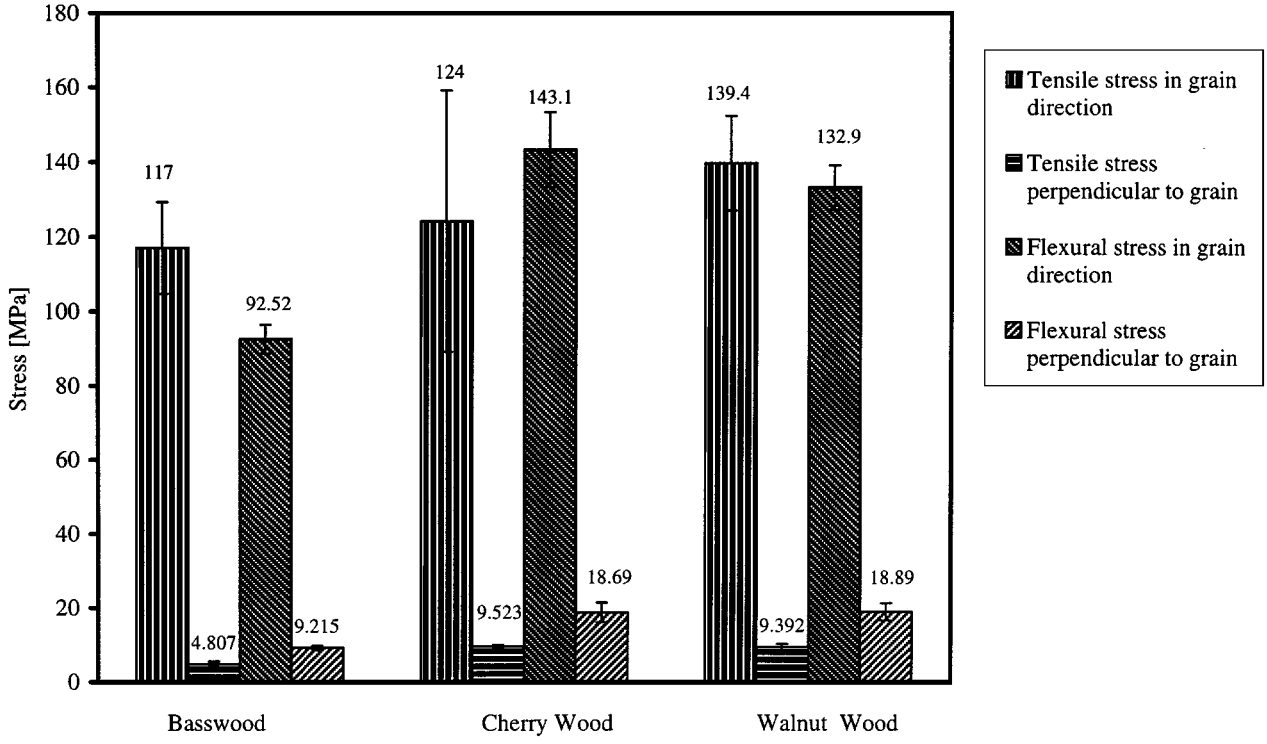

Figure 10 Tensile and flexural stress of three different wood.

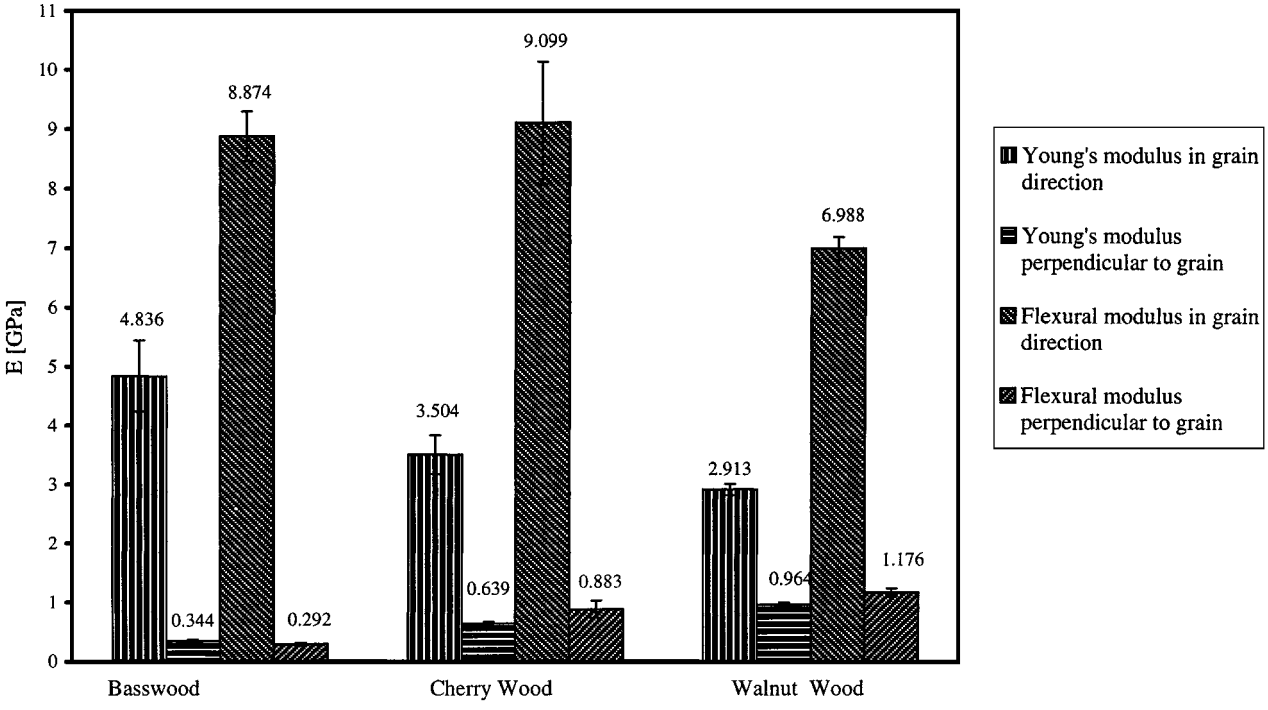

Figure 11 Tensile and flexural modulus of three different wood.

however, their strength and modulus values are much higher than those of woods in perpendicular to grain direction. A significant advantage of "green" composites, as in the case of all composites, is that their mechanical properties could be easily engineered for specific applications. The hydrophobicity of the matrix could also be an advantage of these "green" composites in the application of somewhat humid environments. However, they should degrade fast in a compost environment.

Fig. 12 shows, in histogram format, the tensile fracture strains and flexural failure strains of bass, cherry, and walnut woods. It can be seen that fracture strain in tensile mode of these woods is higher in grain direction than in the perpendicular direction. The fracture strains in flexural mode of bass and cherry woods are higher in perpendicular to grain direction than in grain direction while walnut wood shows opposite behavior. Even though the strain to failure of these woods can be

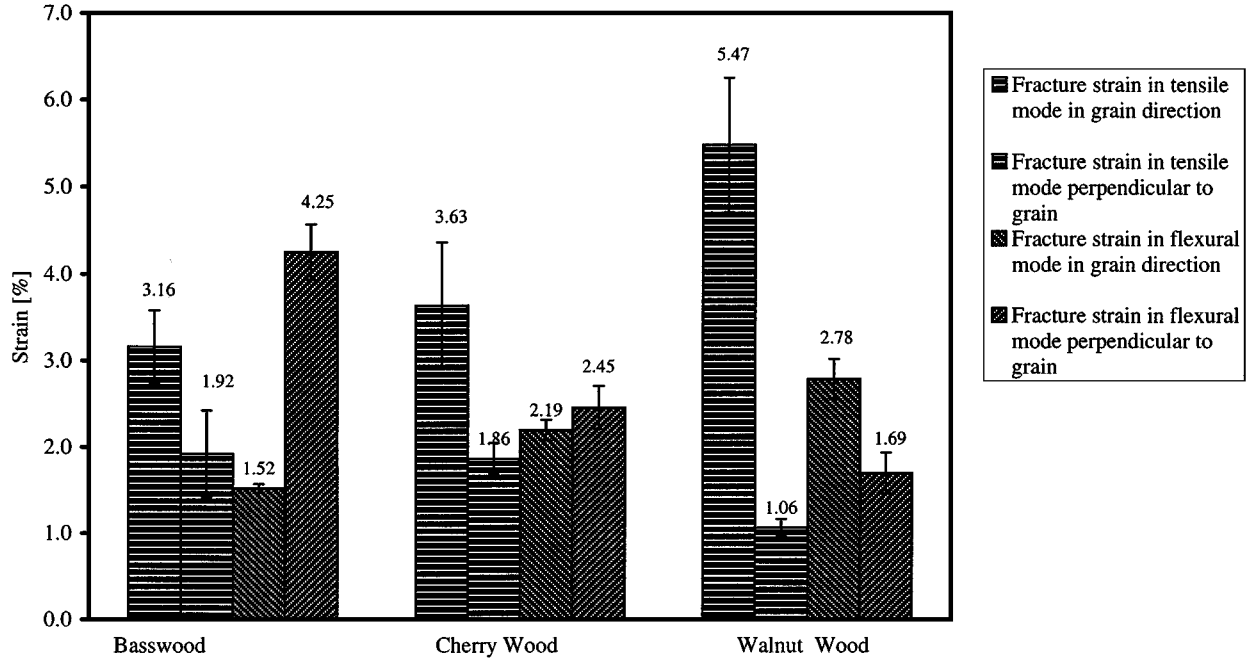

Figure 12 Tensile and flexural strain of three different wood.

higher than those of "green" composites, the flexural strains at failure in most cases are lower than flexural strains at yield of "green" composites. In other words, these "green" composites are much more tougher than the wood varieties tested in this study. Catastrophical failure of these woods occurred in the three-point bending test while "green" composites only yielded without failure.

3.4. Fracture surface of "green" composites

Fig. 13 shows the fracture surfaces of pineapple fiber/PHBV resin "green" composites, in tensile mode, with 30% fiber content. Although many fibers broke at the fracture surface, several other fibers were pulled out from the matrix with no resin adhering to the fibers. This is due to the weak fiber/matrix bonding as indicated by the low IFSS values from microbond tests. In case of good bonding between fiber and matrix, continuous fibers will break just at the fracture surface of composites with no broken fibers protruding from the fracture surface or resin sticking to fibers. In general, the longer the length of the fiber pulled out from the matrix, the weaker the bonding between the fiber and the matrix. These results suggest that surface treatment of pineapple fibers to increase the IFSS would also improve the strength of "green" composites.

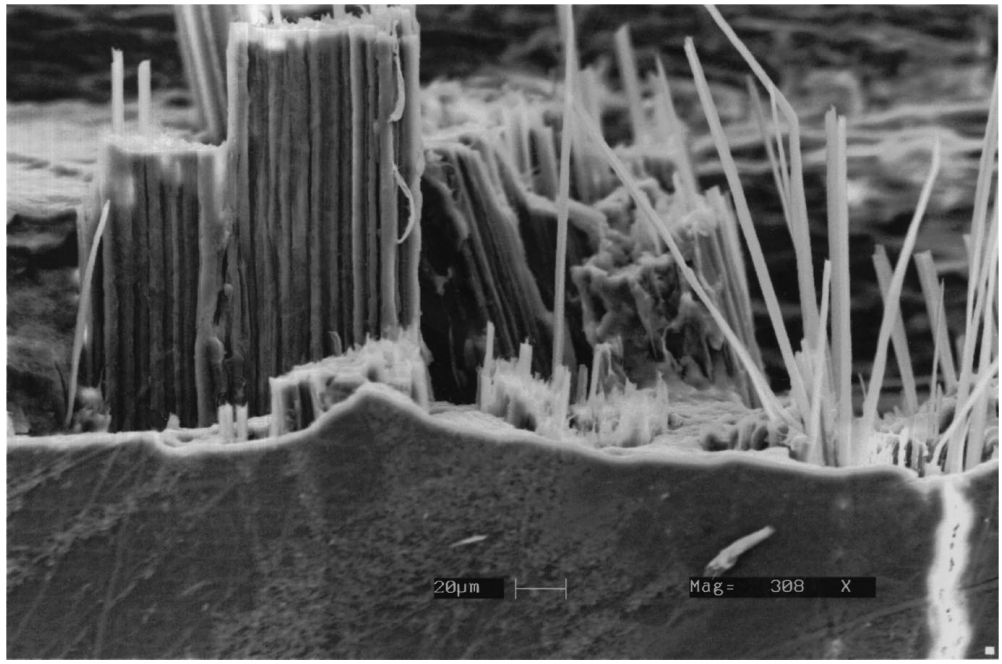

Figure 13 A SEM photomicrograph of the fracture surface in tensile mode of pineapple fiber/PHBV "green" composites (0°/90°/0°) with 30% fiber content.

4. Conclusions

A new class of totally biodegradable, environment-friendly "green" composites were prepared using pineapple fibers and PHBV resin. Pineapple fibers have an average tensile strength of 445 MPa and Young's modulus of 13.21 GPa. As is the case with all natural cellulosic fibers, pineapple fibers showed high variability in their mechanical properties. Pineapple fiber/PHBV resin interfacial shear strength measured using microbond technique was found to be 8.23 MPa. SEM photomicrographs revealed that the failure was adhesive. This suggests that fiber surface may be treated to improve fiber/PHBV IFSS.

Tensile and flexural strength of composites are significantly higher than those of the virgin resin, while their strain to failure in tensile mode and flexural strain at yield decrease. The fracture strain of both composites with 20 and 30% fiber weight content is nearly the same as that of pineapple fibers.

Some woods have higher tensile and flexural strength in grain direction, while lower strength in perpendicular to grain direction than those of pineapple fiber reinforced PHBV "green" composites. These composites are much more tougher than the three varieties of wood specimen tested in this study.

Pineapple/PHBV "green" composites show promising properties to be applied in mass volume noncritical applications. As in the case of most composites, their mechanical properties can be designed with specific application in mind.

References

1. A. A. CHOWDHURY, *Arch. Mikrobiol.* **47** (1963) 167.
2. P. A. HOLMES, S. H. COLLINS and L. F. WRIGHT, US Patent no. 4.477.654 (1984).
3. P. H. BARHAM and A. LELLER, *J. Polym. Sci.: Polym. Phys. Ed.* **24** (1986) 69.
4. P. H. BARHAM, *J. Mater. Sci.* **19** (1984) 392.
5. A. J. HUANG, A. S. SHETTY and M. S. WANG, *Adv. Polym. Tech.* **10**(1) (1990) 23.
6. P. A. HOLMES, in "Development in Crystalline Polymers II," edited by by D. C. Bassett (Elsevier, London, 1988) pp. 1–65.
7. P. GATENHOLM, J. KUBAT and A. MATHIASSON, *J. Appl. Polym. Sci.* **45** (1992) 1667.
8. A. C. KARMAKER and G. HINRICHSEN, *Polym. Plast. Eng.* **30** (1991) 609.
9. S. VARGHESE, B. KURIAKOSE, S. THOMAS, C. K. PREMALATHA and A. T. KOSHY, Platics, *Rubber Compos. Process. Applic.* **20** (1993) 93.
10. R. G. RAJ, B. V. KOKTA, G. GROULEAU and C. DANEAULT, *Polym. Plast. Tech. Eng.* **29**(4) (1990) 339.
11. P. R. HORNSBY, E. HINRICHSEN and K. TARVERDI, *J. Mater. Sci.* **32** (1997) 443.
12. S. W. TSAI and H. T. HAHN, in "Introduction to Composite Materials" (Technomic Publishing, Lancaster, PA, 1980).
13. P. M. MILLER and L. REBENFELD, *Compos. Sci. Technol.* **28** (1987) 17.
14. A. N. NETRAVALI, L. T. T. TOPOLESK, W. H. SACHSE and S. L. PHOENIX, *ibid.* **36** (1989) 13.
15. A. N. NETRAVALI, Z. F. LI, W. H. SACHSE and H. F. WU, *J. Mater. Sci.* **26** (1991) 6631.
16. Z. F. LI and A. N. NETRAVALI, *J. Appl. Polym. Sci.* **44** (1992) 333.
17. S. CHUA and M. R. PIGGOTT, *Compos. Sci. Technol.* **22** (1985) 33.
18. J. GEORGE, S. S. BHAGAVAN, N. PRABHAKARAN and S. THOMAS, *J. Appl. Polym. Sci.* **57** (1995) 843.
19. A. N. NETRAVALI, R. B. HENSTENBURG, S. L. PHOENIX and P. SCHWARTZ, *Polym. Compos.* **10** (1989) 226.
20. P. S. MUKHERJEE and K. G. SATYANARAYANA, *J. Mater. Sci.* **21** (1986) 51.

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