Lignocellulosic Polymer Composite IV

OLFAT Y. MANSOUR, SAMIR KAMEL, MONA A. NASSAR

Cellulose and Paper Department, National Research Center, Dokki, Cairo, Egypt

Received 14 March 1997; accepted 7 December 1997

ABSTRACT: Palm leaves as a woody lignocellulose, together with polystyrene, were used to produce composites. Chemithermal mechanical pulps (CTMP) were obtained from palm leaves under alkaline or acidic conditions. Appropriate bending strength was obtained from palm leaves and their CTMP pulps prepared under neutral or acidic conditions. The bulky fibers resulted from the alkaline pulps lead to composites of low bending strength. Thus, the cooking conditions of the palm leaves to obtain CTMP pulp play an important role on the properties of the composites. The nonbulky fibers lead to the formation of trapped pockets air as the number of the hydrogen bond are few. The presence of these air pockets allows the polystyrene solution to enter forming bonding between the interfaces. It is also found that the lower the density of the composites, the lower the internal bond strengths. The chemical constituents of the CTMP pulps, as well as the yields of the pulps, may influence the properties of the composites. Increasing the percentage of polystyrene in the composites, the mechanical properties increased. The water uptake and the swellability decreased until 20% polystyrene concentration and then levelled off. The thickness and density behaved the same. However, the type of substrate of the composite and the weight fraction are the important factors in determining the properties of the composites. © 1998 John Wiley & Sons, Inc. J Appl Polym Sci 69: 845-855, 1998

Key words: lignocellulosics; composites; polymer composites; lignocellulosics plastics; plastic composites

INTRODUCTION

The word "composite" means the material that consists of two or more distinct parts. Thus, a material having two or more distinct constituent materials or phases may be considered, as composites may consist of one or more discontinuous phases. The discontinuous phase is usually harder and stronger than the continuous phase and is called reinforcement or reinforcing material, whereas the continuous phase is termed the matrix. Cellulose, in the form of timber, wood pulp or cotton, is cheap and relatively abundant and provides a suitable substrate, to which improved properties can be given by the use of chemicals and, hence, can be compressed into a composite. 1

One of the most important factors determining the properties of the composites is the relative proportions of the matrix and reinforcing materials. The relative proportions can be given as the weight fractions or the volume fractions. The void content of a composite may significantly affect some of its mechanical properties. Higher void contents usually mean lower fatigue resistance and greater susceptibility to water penetration, weathering, and increased variation in strength properties.² Many varieties of mechanical and physical properties can be developed through appropriate compounding of polymers with fibers and fillers. Ellwood³ has noted that maximum property improvements are usually achieved with maximum polymer content. It has also been mentioned by some investigators⁴ that composites are

Correspondence to: O. Y. Mansour.

Journal of Applied Polymer Science, Vol. 69, 845–855 (1998) © 1998 John Wiley & Sons, Inc. CCC 0021-8995/050845-11

more compact than untreated wood, and their specific gravities usually fall below 0.85 and 1.15. The hardness is increased, as well as the bending, compression, and shear strengths. The dimensional stability increased as well, and the antistatic efficiencies up to 70% have been recorded due to their reduced water uptake. Composites also resist the decay and the attack by fungi and insects. Composites or mechanically oriented sheet materials of tensile strengths approaching the strengths of metal alloys are theoretically possible.⁵ It has been found that composite had increased mechanical flexural strength when the wood particles were 0.5-0.1 mm, and the filler content was 50%. The tensile strength of composites reached its maximum when the wood content was 40-50%.

Bamboo-plastic composite (BPC) was prepared under co-60 gamma radiation using methyl methacrylate as (MMA) as the bulk monomer combined with methanol at different proportions. Water absorption was low with BPC compared to the untreated bamboo.⁶

Due to the vast potential for use for cellulosic fillers in thermoplasic and thernosets, Varma and Chavan⁷ used chemically modified cellulosic fillers by attacking pendant primary amine groups (diamino propane). The modified cellulosic fillers were used for curing on epoxy resin diglycidyl ether of bisphenol. Gel times thus obtained are much less than systems containing unmodified cellulose.

Moldas and Kokta⁸ compared the performance of composites of maleic anhydride modified cellulosic materials, for example, chemithermo mechanical pulp (CTMP) of hardwood flour as hardwood aspen and nutshell flour in polyethylene and polystyrene composites. Experimental results indicate that the performance of the three different cellulosic materials are not the same, and it also varies with the concentration of fillers and the nature of thermoplastics. The same authors found that fibers of CTMP are more separated and they are more flexible than the other two cellulosic. They stated that CTMP is, without doubt, the best pulp as far as mechanical properties of composites are concerned.

In this work, different alkaline, acidic, or neutral pulping methods were considered to produce CTMP pulps. The difference in bending strength and water penetration for the composites prepared there from were investigated as the first aim.

The increased polymer contents in the compos-

ite has its effect on the strength properties; therefore, different contents of polystyrene were used and the bending strength and the water penetration were investigated as the second aim of this work.

MATERIAL AND METHOD

Preparation of Composite Sample

Palm leaves or their CTMP pulps of moisture content ~ 25% were fluffed using a Pfeiffer shredder for 20–25 min. The samples were then dried at 45°C for 24 h. The samples were ground and passed through mesh 20. The fine particles were excluded. The composite samples were prepared from the ground samples by impregnation into polymer solutions as follows.

X number of grams of polystyrene (depending on the needed polymer percent) was placed in 150 mL of benzyne in a 500-mL beaker, then 50 g of each crude palm leaves or CTMP pulp samples were impregnated with the polymer solution in the beaker and kept covered for 24 h at room temperature. The solvent was evaporated and dried, and the homogeneous mixture was then kept to be pressed as follows: 50- or 100-g dry samples were placed in a disc form of a diameter of 15.0 cm, heated to 160°C, and then pressed under 50, 75, 100, or 125 KN disc for 10 min.

Measurements of the Composite Sample

The composite samples were subjected to the following measurements.

Thickness

Thickness was measured using a dial micrometer.

Density

The density was calculated using the following equation:

density =
$$\frac{\text{weight of the composite sample}}{\text{volume of the composite sample}}$$

Water Uptake

Small pieces having the same dimensions of composites under investigation were impregnated in water for different periods, namely, 24, 48, and 72 h, and also for 7 days, at room temperature $(\sim 25^{\circ}C)$. After each period, a piece was centrifuged at 5000 rpm for 10 min, then weighed and dried at 105°C for 24 h. The percentage of water uptake was calculated as follows:

water uptake =
$$\frac{B-A}{A} \times 100$$

where *B* is the weight of the centrifuged sample, and *A* is the weight of the sample dried at 150° C.

Swellability

The volume of small pieces of composites under investigation were measured and then impregenated in water for different periods, as in the case of water uptake. After each period, the volume of the pieces was measured accurately. The percentage of swellability was calculated as follows:

swellability =
$$\frac{X - Y}{Y} \times 100$$

where X is the volume of sample after the impregnation, and Y is the volume of the sample before the impregenation.

Bending Strength

Bending strength was determined as the weight at which the composite sample broke and is distinguished as kg/cm^2 . The apparatus used was an Instron 1128.

RESULTS AND DISCUSSION

Properties of Palm Leaves and the Prepared CTMP Pulp

25-mesh ground palm leaves and the CTMP pulp prepared from them using different pulping conditions were investigated for holocellulose, α -cellulose, hemicellulos, lignin, and ash content. The results are found in Table I. It is clear that the yields of the pulps produced by neutral or acid cooking conditions are high compared with those of alkaline pulping. Regarding the chemical analysis, the holocellulose, as well as the α -cellulose of the CTMP pulps cooked with water or acid sulfite, are higher than those cooked under alkaline conditions. The hemicellulose contents are high for CTMP numbers 1 and 2 cooked with water compared with those cooked with the other reagents. Lignin contents of CTMP pulps are lower than the untreated palm leaves (Table I). It is also clear that the alkaline cookings lead to CTMP pulps of lower liginin contents than the water, or sulfite, cookings (Table I). On the other hand, it is evident from Table I that CTMP pulps obtained by water or sulfite cooking have high ash contents compared with those CTMP pulps prepared under alkaline conditions.

Composites from Palm Leaves and the CTMP Pulps

Palm leaves and the CTMP pulps of mesh size 25 were impregenated into polymer solution (20% polystyrene, based on oven dry weight) and thoroughly mixed for 24 h at room temperature. Then the solvent was evaporated. The mixture was left to dry and then was pressed into composite under 50 KN for 10 min at 160°C.

From Tables IIA and IIB, it is clear that the density of the all composites were inversely proportional to the thickness of the samples, and the density ranged between $0.74-1.14 \text{ g/cm}^3$. Maximum density was obtained from CTMP pulps (no. 1 in Table IIA), which were prepared by cooking the palm leaves with water at 170° C for 1.5 h, while minimum density was attained from soda CTMP pulps (no. 3 in Table IIA) cooked at 155° C (Table IIA).

Bending strength of the composite from untreated palm leaves and polystyrene was higher than that of obtained from CTMP pulps cooked with sodium hydroxide or with sodium hydroxide and sodium sulfide (kraft) or with sodium sulfite, (CTMP pulps from nos. 3-8), together with polystyrene. The bending strength of the composite prepared from CTMP cooked by water at 170°C for 1.5 h (CTMP no. 1) was higher than that of composite obtained from untreated palm leaves (Tables IIA and IIB). It is also clear from Tables IIA and IIB that the bending strength of the composite from CTMP cooked with sulfite at temperature 155°C (CTMP no. 7) was higher than that of composite obtained from water cooked one for 3 h (CTMP no. 2). So, appropriate bending strengths were obtained on using untreated palm leaves and water-cooked or sulfite-cooked palm leaves (CTMP nos. 1, 2, 7 and 8, respectively) while other cooking conditions lead to composites of very low bending strengths, which could not be detected on the bending strength apparatus, (Table IIA).

Regarding the water uptake and the swellability of the composites, the maximum values were reached after 24 h and the morphology deteriorated of the composite samples obtained from un-

Pulping Condition				Chemical Analysis								
Reagent Used	Temperature (C)	Time (h)	Yield (%)	Holocellulose (%)	$\substack{\alpha\text{-Cellulose}\\(\%)}$	Hemicellulose (%)	Lignin (%)	Ash (%)				
_	_	_	100.6	71.85	40.22	31.63	19.51	7.57				
1. Water	170	1.5	60.6	82.29	63.94	18.35	12.50	6.21				
2. Water	170	3.0	58.0	80.23	61.90	19.53	10.44	8.33				
3. NaOH	155	1.5	42.4	82.29	58.84	23.45	7.91	9.40				
4. NaOH	170	1.5	45.1	79.01	59.7	19.31	6.44	12.45				
5. NaOH + Na ₂ S	155	1.5	44.3	82.49	57.76	24.73	7.68	10.04				
6. NaOH + Na ₂ S	170	1.5	40.0	78.81	55.89	22.92	5.34	12.35				
7. Na_2So_3	155	1.5	61.5	86.66	69.55	28.11	8.50	8.14				
8. Na_2So_3	170	1.5	58.4	88.00	68.00	20.11	7.90	8.21				

Table I Chemical Analysis of Raw Material and Different Pulps

treated palm leaves and that cooked with sodium hydroxide-sodium sulfide mixture at $170^{\circ}C$ (CTMP no. 6 in Table IIA). The temperature was kept constant for 48 h, then increased and reached a maximum value after 7 days. The swelling percent also remained constant for 48 h, then increased and reached a maximum value after 7 days. The water uptake and the swelling percent of the composites obtained from other cooked palm leave samples increased with the increasing time of impregenation of the samples into water (Table IIA).

It is also seen from Tables IIA and IIB, that the water uptake and swelling percent are generally increased in the following order: composites [that obtained from water-cooked palm leaves (CTMP nos. 1 and 2)] < sulfite-CTMP nos. 7 and 8 < soda-CTMP nos. 3 and 4 < that obtained from kraft (CTMP nos. 5 and 6).

On conclusion, maximum bending strength and low water uptake and swellability percent were obtained for composites prepared from CTMP nos. 1 and 2.

If the materials used in the disc to be pressed into composite were doubled and the pressure was 50 KN for 10 min at 160°C, the properties of the composites changed (Table IIB). From Table IIB, it is evident that the thickness of these composites either doubled or significantly increased to be either less than the double or more. The density of the composites decreased for the composites made from untreated or water-cooked CTMP or kraft (sodium hydroxide-sodium sulfide) treated ones, (Table IIB). The other treated palm leaves lead to composites of the same or more densities compared with the corresponding ones of Table IIA. The bending strengths of the first two composites decreased, while for the other composites, they increased, except for the three ones whose values are still undetectable Table IIB. However, the bending strength of the composites of Table IIB decreased in the following order: sulfite-cooked palm leaves at 155°C (CTMP no. 7) > water cooked one for 1.5 h (CTMP no. 1) > water cooked for 3 h (CTMP no. 2) > sodium sulfite cooked at 170°C (CTMP no. 8) > untreated palm leaves. The cooking with sodium hydroxide showed no change of the bending strength for composites of Tables IIA or IIB.

It is seen from Tables IIA and IIB that the water uptake and the swelling percent of the composites obtained from untreated palm leaves and the kraft-cooked palm leaves at 170°C (CTMP no. 6) reached maximum values, and the composite samples are deformed after 24 h from the impregnation in water.

On conclusion, minimum water uptake, and, also, minimum swelling percent, were obtained for composites made from water-cooked palm leaves for 1.5 h (CTMP no. 1), while maximum water uptake, and, also, the swelling percent, were obtained for composites obtained from CTMP nos. 5 and 6 (palm leaves cooked with a sodium hydroxide-sodium sulfide mixture); thus, it is evident from Tables IIA and IIB that the water uptake and the swelling percent of the composites (Table IIB) increased in the following order: composites obtained from CTMP nos. 1 and 2 < from CTMP nos. 3 and 4 < from CTMP nos. 7 and 8 < those obtained from CTMP nos. 5 and 6.

In principle, composites can be constructed of any of two or more materials. For the purpose of obtaining specific characteristics and purposes,

					Water	Uptake		Swellability (%)					
Pretreatment	Thickness (cm)	Density (g/cm ³)	Bending (kg/cm ²)	After 24 h	After 48 h	After 72 h	After 7 days	After 24 h	After 48 h	After 72 h	After 7 days		
— 1. H₂O at 170°C	0.34	0.96	49.27	121.76	—	—	—	125.01	—	—	—		
for 1.5 h 2. H ₂ O at 170°C	H_2O at 170 C0.271.1473.66 H_2O at 170°C610.3310.9535.17Soda treatment510.3310.9510.17	73.66	27.19	28.01	28.74	30.40	21.39	21.87	26.68	27.15			
for 3 h 3. Soda treatment		35.17	33.36	34.97	38.23	54.50	33.46	33.87	38.19	56.51			
at 155°C 4 Soda pulping	0.42	0.74	undetected	78.77	81.36	83.03	101.39	101.57	118.83	131.31	178.74		
at 170°C	0.35	0.89	undetected	86.60	89.54	93.82	96.99	118.12	126.30	129.09	137.99		
at 155°C	0.35 0.89 undetecte	undetected	111.58	115.45	119.69	136.29	120.72	120.93	124.42	137.94			
at 170°C	0.40	0.80	undetected	89.97	—	—	—	107.21	—	—	—		
at 155°C	0.30	1.02	44.27	43.34	51.99	56.65	68.14	93.12	120.78	126.71	136.97		
at 170°C	0.30	1.06	24.25	54.18	60.83	66.25	67.97	70.62	84.87	92.87	93.44		

Table IIA Effect of Pretreatment of the Palm Leaves on the Composite Properties Prepared Therefrom (Series I) (Series I)

Table IIB Effect of Pretreatment of the Palm Leaves on the Composite Properties Prepared Therefrom (Series II)

					Water	Uptake		Swellability (%)					
Pretreatment	Thickness (cm)	Density (g/cm ³)	Bending (kg/cm ²)	After 24 h	After 48 h	After 72 h	After 7 days	After 24 h	After 48 h	After 72 h	After 7 days		
 1 H_O at 170°C	0.59	0.91	58.32	8	_	_	_	198.01	_	_			
for 1.5 h 2. H ₂ O at 170°C	0.67	0.90	80.22	46.02	50.97	53.33	54.79	18.29	19.25	21.69	21.41		
for 3 h 3 Soda treatment	n_2 or u 110 °C 0.63 0.98 55.67 Soda treatment 0.72 0.86 22.64 Soda pulping 0.75 0.85 undetecte Soda pulping 0.75 0.85 undetecte	55.67	54.81	65.55	81.42	85.83	22.54	33.88	37.88	49.94			
at 155°C 4 Soda pulping		22.64	78.34	79.48	85.91	103.06	52.27	52.70	100.41	106.81			
at 170°C 5 Soda pulping		undetected	54.06	53.51	57.64	75.01	23.82	32.20	42.58	97.86			
at 155°C 6 Kraft pulping	0.74	0.88	undetected	110.6	117.86	119.43	130.94	121.15	123.45	129.84	138.29		
at 170°C 7 Sulfite pulping	0.75	0.85	undetected	5	106.66	—	—	98.10	—	—	—		
at 155°C 8 Sulfite pulping	0.60	1.03	66.67	78.92	95.42	94.49	110.85	169.95	180.29	182.02	196.27		
at 170°C	0.55	1.15	37.42	55.02	57.59	63.41	66.63	58.48	71.48	79.51	82.53		

the constituents retain their identity such that they exhibit an interface between one another. In some cases, the continuous region is a distinct added phase, called an interphase. When such an interphase is present, there are two interfaces, one between each surface on the interphase and its adjoining constituent. The interphases of composites (the bonding agent) is the region where loads are transmitted between the reinforcement and the matrix. The extent of interaction between

						Water	Uptak	e	Swellability (%)			
Pulping Condition	Polystyrene (%)	Thickness (cm)	Density (g/cm ³)	Bending (kg/cm ²)	After 24 h	After 48 h	After 72 h	After 7 days	After 24 h	After 48 h	After 72 h	After 7 days
_	10	0.34	0.96	49.27	121.76				125.01	_		
	15	0.35	1.02	56.81	79.59	81.69	85.15	113.28	154.56	157.26	180.94	196.65
	20	0.37	1.05	86.54	74.46	75.31	83.42	89.32	99.75	110.25	118.34	141.84
	25	0.37	0.89	92.53	63.64	73.59	81.12	82.60	78.65	77.58	92.07	93.38
H_2O at												
170°C for												
1.5 h	10	0.27	1.14	73.66	27.19	28.01	28.74	30.40	21.87	21.39	26.88	27.15
	15	0.30	1.16	82.76	20.02	20.93	24.23	27.89	11.82	11.82	16.78	19.98
	20	0.31	1.17	92.37	18.49	21.26	25.84	24.54	10.06	11.34	13.09	13.72
	25	0.31	1.11	110.22	17.97	20.64	21.02	24.16	9.96	11.04	13.90	14.41
$\begin{array}{c} 10\% \ Na_2SO_3\\ at \ 155^\circ\!C \end{array}$												
for 1.5 h	10	0.30	1.02	44.27	43.34	51.99	56.65	68.14	93.12	120.78	126.71	136.97
	15	0.31	1.04	58.16	36.46	38.45	39.06	66.0	90.20	117.23	119.45	130.97
	20	0.33	1.06	59.30	30.60	32.40	36.40	62.10	80.42	110.22	110.24	120.98
	25	0.33	1.02	106.87	28.25	30.82	32.63	60.81	68.98	74.42	75.58	81.56

Table IIIB Effect of Polystyrene Percent on Composites Properties (Series II)

					Water Uptake			Swellability (%)				
Pulping Condition	Polystyrene (%)	Thickness (cm)	Density (g/cm ³)	Bending (kg/cm ²)	After 24 h	After 48 h	After 72 h	After 7 days	After 24 h	After 48 h	After 72 h	After 7 days
_	10	0.69	0.91	58.32	141.58	_	_	_	198.01		_	
	15	0.69	0.93	62.61	91.93	93.27	97.90	99.40	142.78	142.45	154.89	153.03
	20	0.68	0.95	92.41	72.44	81.25	93.02	93.33	100.67	102.43	108.57	108.25
	25	0.73	0.94	108.16	87.48	88.87	98.89	98.95	122.34	130.37	129.72	130.55
H ₂ O at 170°C for												
1.5 h	10	0.67	0.90	80.22	46.02	50.97	53.33	54.79	18.29	19.25	21.69	21.41
	15	0.65	1.06	95.36	21.99	22.25	22.46	24.62	10.69	10.69	12.67	14.12
	20	0.60	1.11	120.72	19.09	22.58	25.61	29.20	10.33	12.47	13.19	14.21
	25	0.75	0.92	122.43	18.52	20.67	21.53	22.29	4.88	7.66	8.42	8.38
$10\% Na_2So_3$												
at 155° C												
for 1.5 h	10	0.60	1.03	66.67	78.92	95.42	94.49	110.85	169.55	182.02	182.29	196.27
	15	0.58	1.05	74.84	68.24	75.53	83.52	88.40	150.23	155.94	172.44	182.51
	20	0.56	1.07	105.70	62.67	66.32	70.34	72.64	110.96	108.77	112.83	116.10
	25	0.55	0.98	111.52	74.29	76.25	82.13	98.19	132.85	132.35	135.00	137.63

the reinforcement and the matrix is a design variable and may vary from strong chemical bonding to weak frictional forces. The bond is critical to the long-term stability of the composite, playing a key role in fatigue properties, environmental behavior, and resistance to hot and wet conditions.⁹

Woody plants, such as palm leaves, can offer the

attractive combination of low cost, good strengthto-weight ratio, and easy processing. The finer size of wood-fiber-reinforced plastics, which are similar to particleboard compositions, ensure better distribution and wetting of the reinforcement and, consequently, better strength. However, cellulose fibers have been evaluated for reinforcing thermoplastics, such as polystyrene. The constit-



Figure 1 Effect of polystyrene percent on composite properties (series I).

uent of the wood or the cellulosic fibers has its effect on the properties of the composites. The weight of the wood or cellulosic fibers in the composite disc also affects the composite properties. It is worthy to mention that Moldas and Kokta⁸ found that CTMP is the best pulp as far as mechanical properties of composite are concerned.

Applying the aforementioned principle, it is clear that the cooking conditions of the palm leaves to obtain CTMP pulp play an important role on the properties of the composites. From Tables IIA and IIB, the water-cooking and the sulfite-cooking CTMP nos. 1 and 2 and 7 and 8, respectively, gave a composite of detectable bending strengths (73.66, 35.17, 60.22, and 55.67 and 44.27, 24.25, 66.67, and 37.42, respectively). Untreated palm leaves led to composites having bending strengths higher than composites prepared from CTMP nos. 3-6. The alkaline cooking of palm leaves caused swelling of the cellulosic fibers. The number of hydroxyl groups available for hydrogen bonding increased. As a consequence, the density of the composite decreased,

as is evident from Tables IIA and IIB. On the other hand, the alkaline pulping led to bulky fibers without air pockets between them. The other neutral or acid-cooking lead to fibers which may form air pockets, as the number of hydrogen bonding between them are few. The present of air pockets between these fibers allows the polyester solution to enter, forming bonding between the interfaces. In this case, the plastic layer or the interphase forms a protrusion between the different interfaces. Therefore, the bending strength increased. Thus, the bulky fibers resulting from the alkaline pulping led to composites of low bending strength. It is obvious from the results that the density of the composites prepared from untreated palm leaves, or the CTMP pulps nos. 1, 2, 7, and 8, are higher than the other composites. The bending strengths of these composites are high as well. Since the internal bond (IB) strength is very dependent on the density: the lower the density, the lower the internal bond strength; thus, it is clear that internal bond strengths of composites prepared from CTMP pulps obtain by



Figure 2 Effect of polystyrene percent on composite properties (series II).

neutral or acid-cooking are higher than those obtained by alkaline cooking or alkaline CTMP pulps. The increased internal bond strengths may be attributed on one hand to the protrusions formed in between the fibers instead of the air pockets. On the other hand, they may be due to the flow or film formation of the polystyrene in the composite structure.

The decreased water uptake and the swellability of the composites prepared from CTMP pulps nos. 1, 2, 7, and 8 than those obtained from CTMP pulps nos. 3–6 may be due to the increased swelling of the CTMP pulps obtained by alkaline conditions. Thus, it is reliable that the fiber of the alkaline CTMP pulps are more swollen than those of acid or neutral CTMP pulps.

As regards the effect of the chemical constituents of the cellulosic fibers on the properties of the composites prepared, it is clear from Tables IIA and IIB that the composites prepared from CTMP pulps nos. 1-3 and 8 have relatively high bending strengths and low water uptake and swellability. It is shown from Table I that the CTMPS nos. 1, 2, 7, and 8 contain high α -cellulose and lignin contents. On the other hand, for the composites prepared from alkaline CTMP, the yield obtained of these pulps are low, and the properties also are low (Tables I, IIA, and IIB).

Thus, the chemical constituents of the pulps as well as the yield of the pulps may play certain role on the properties of the composites. The role of the cellulosic fibers is concerned in the crystallinity, which led to increased bending strength. The role of the increased lignin content on the increase of the properties of the composites is related to the increase mobulity of the matrix polymer, that is lignin, especially near its transition temperature, which reached more than 160°C due to the pressing of the composites at this temperature for 10 min. Under these conditions, the lignin will be in a rubbery phase, which can permit more orientation and, thus, more crystallinity of the cellulosic microfibrils and, hence, increased bending strength.¹⁰ It is of great interest to say that the CTMP pulps obtained by using the neutral or acid-pulping conditions are the best pulps as far (=Bending kg/cm blank #Bending kg/cm H O at 170°C for 1.5 hr =Bending kg/cm 10% Ng SQ at 155°C for 1.5 hr



Figure 3 Effect of polystyrene percent on composite properties (series I).

as the mechanical and water properties of the composites are concerned.

Effect of Polystyrene Percent

In this part of the work, the composites were obtained from untreated palm leaves, water-cooked CTMP no. 1 pulp (pulping time 1.5 h), and sulfite pulp CTMP no. 7 (pulping temperature 155° C) using different concentrations of polystyrene solutions, namely, 10, 15, 20, and 25%, based on the oven dry weight of the pulp. The ground celluloses of mesh size 25 were impregnated in the solutions and thoroughly mixed for 24 h, then the mixtures were dried, then pressed under 50 KN at 160°C for 10 min. The results for all composite samples were gathered in Tables IIIA and IIIB and Figures 1–4.

Two series of experiments were considered: Series I (Table IIIA), in which only 50 g of the mixture was pressed, series II (Table IIIB), in which 100 g of the mixture was pressed. From Table IIIA and Figure 1 (series I), it is seen that the thickness obtained for different pulp increased with increasing polymer concentrations until 20% then levelled off.

On the other hand, the density of the composites slightly increased with the increase of the polymer concentration and reached maximum values at 20% polystyrene concentration, then slightly decreased with the increase of the polymer concentration (Tables IIIA and IIIB and Fig. 1).

From series II (Table IIIB and Fig. 2), the increased amount of CTMP-polystyrene mixture, namely, 100 g to be pressed into composite, the thickness of the composite of this series doubled than those of series I, while the densities slightly increased or decreased (Table IIIB and Fig. 2).

For the same cellulosic sample, the increase of the polystyrene concentration from 10 to 15 to 20 to 25, and the thickness decreased with the increase of the polymer concentration until minimum at 20% polystyrene concentration, then increased (Table IIIB and Fig. 2). On the other hand, the density increased as the concentration of the polymer increased until maximum at 20%



Figure 4 Effect of polystyrene percent on composite properties (series II).

polystyrene concentration, then decreased (Table IIIB and Fig. 2).

Regarding the bending strength of the composites of series I and II it is clear from Table IIIA and IIIB (Figs. 3 and 4) that the values increased as the polystyrene concentration increased. Also, the bending strength of the composites of series II are higher than those of series I (Table IIIA and Figs. 3 and 4). Maximum bending strength was obtained with composite from CTMP no. 7, while with other pulps, the difference in the bending strength depends on the polymer percent. At 10 and 20% polymer concentrations, the bending strength of the composites from crude palm leaves is higher than that of the composites from sulfite pulp (CTMP no. 7); but at 15 and 25% polymer, the bending strength of the composites from untreated palm leaves is less than that of composites from sulfite pulp CTMP no. 7 (Tables IIIA and IIIB and Figs. 3 and 4).

CONCLUSION

The maximum bending strength was obtained with composites from water pulp at different poly-

styrene percentages. The water uptake and the swellability of the composites both of series I and II decreased as the polystyrene concentration increased (Tables IIIA and IIIB). In the meantime, the water uptake and the swelling percent of the composites of series I are slightly low than those of composites of series II (Tables IIIA and IIIB).

The effect of the polymer concentration on the properties of the composites can be explained as follows. The increase in density and strength of the composites as the concentration of the polymer increased were attributed to the flow and film formation of the polystyrene in the composite structure. In spite of the slight decrease in the density at 25% polystyrene concentration, the bending strength increased. It is worthy to mention that the internal bond strength (IB) is very dependent on the density.¹¹ However, the increased bending strength as a result of the increased internal bond (IB) with the increase of the density until maximum then the forward increase of the bending strength and the slight decrease of the density at polymer concentration 20% represent the levelling off of the density, at

which the greater increase of the internal bond affects only the bending strength and not the density.

The decreased water uptake as the polymer concentration increased revealed that the void content of the composites decreased because it has been mentioned before¹ that higher void contents usually mean greater susceptibility to water penetration. It is worthy to mention that the CTMP no. 1 (the palm leaves treated with H_2O at 170°C for 1 h) possess the lower water uptake, and, hence, the lower void contents. These results are in agreement with the increased bending strengths of the composites prepared from palm leaves. Also, the composites prepared from CTMP no. 7 possess low water uptake compared with the palm leaves, and, consequently, the latter showed increased bending strength than the former.

Therefore, the treatments of the woody plant or the use of CTMP pulp is very important to obtain composites with low void contents and high bending strength.

However, the increase of the amount of lignocellulose-polymer composite led to increased thickness and bending strength, but the density and the water uptake and swellability percent showed an insignificant increase or a slight increase (Tables IIIA and IIIB). These results showed that the void contents of the composites are independent of the amount or the weight of the composite. The void contents are mainly dependent on the type of pulps as well as the polymer content in the composite. These results are in agreement with the statement mentioned before (1) that the relative properties, that is, the weight fractions or the volume fractions have a great effect on the properties of the composites.

Thus, the type of the substrate of the composites and the weight fraction or volume are the important factors in determining the properties of the composites.

REFERENCES

- Bhagw, D. Agarwal, and L. J. Broutman, Analysis and Performance of Fiber Composites, New York, 1979, pp. 1–10.
- G. Lubin, Handbook of Fiber Glass and Advanced Plastic Composite, Van Nostrand Reinhold, New York, 1959, p. 3.
- R. Ellwood, An Investigation of Certain Physical and Mechanical Properties of Wood-Plastic Combinations, ORO, p. 368.
- 4. P. R. Hillo and D. T. McGahan, Atom, 1, 76 (1971).
- 5. J. C. Halpin, Chem. Abstr., 86, 17760w (1977).
- A. S. Bashar, A. Mubarak, Khan, and K. M. Idriss Ali, *Polym. Plast. Technol. Eng.*, **35**, 581 (1996).
- A. J. Varma and V. B. Chavan, Cellul. J., 215, (1994).
- D. Moldas and V. Kokta, *Cellulosics: Chemical, Biochemical, and Material Aspects, J. F. Kennesy, G. O. Phillips, and P. A. Williams, Eds., Ellis Horwood, New York, 1993, pp. 519–524.*
- 9. M. M. Schwartz, *Composite Materials Handbook*, 2nd ed., McGraw-Hill, New York, 1992, pp. 3-11.
- T. G. Rials and W. G. Glasser, Wood Fiber Sci., 21, 80 (1989).
- 11. H. Hall and J. Haygreen, Forest Prod. J., 29 (1983).
- 12. H. Hall and J. Haygreen, Forest Prod. J., 34 (1984).