

A study of wood-plastic combinations based on low-density woods

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Six low-density tropical woods were impregnated with various vinyl monomers and polymerized by irradiation with a ^{60}Co source. The wood-plastic combinations were subjected to standard tests of mechanical properties, and their fracture surfaces were studied by scanning electron microscopy. It was found that, even though most mechanical properties are enhanced by addition of plastics, the properties of wood-plastic combinations fall below those of high-density natural woods on a per unit weight basis. The direct observation of fracture surfaces gave indications of non-uniform penetration of the plastic and little bonding between the polymer and cellulose fibres. Although the wood-plastic combinations produced by the present methods may not be recommendable for applications where increased strength is desired on the basis of cost/quality considerations, they may be suitable for uses where increased abrasion resistance, dimensional stability and lower anisotropy of compressional properties are primary considerations.

1. Introduction

The advent of composite materials as a solution to the relentless need for ever stronger and better materials has led to the study of multitudinous combinations of different fibres and matrix materials. One such combination that has drawn the attention of numerous researchers over the past decade is plastic impregnated wood, where it is hoped to combine the tensile strength of cellulose fibres with the hardness, abrasion resistance, and incompressibility of the polymeric materials used. These materials also offer the particular advantage of greater dimensional stability (i.e. resistance to swelling).

A great deal of the accumulated research effort has gone into the study of permeability and conditioning of wood and the formulation of optimum irradiation conditions for the monomers used in impregnation. Although a large number of wood-plastic materials have been tested for their mechanical and physical properties, such data have generally not been analysed past the stage of estab-

lishing an improvement or deterioration of properties. Since wood-plastic combinations are proposed not only as "erstaz" type materials for natural woods but also as improved materials, a more careful cost-quality relationship seems necessary. As has been pointed out by Tarkow [1], cost considerations have severely impaired the development of modified (plasticized, plastic-impregnated, and compressed) woods.

An interesting comparison of the effect of plastics on the properties of wood is the comparison of resin-treated and compressed wood (*staypack*) [1, 2]. This comparison shows a superior quality, at the same density, for the compressed material, indicating that "wood substance" is a superior quality filler. Unfortunately, compressed woods are as costly to produce as wood-polymer composites.

Tropical woods, because of their broad range of densities (0.26 to 1.18 g cm⁻³ among 144 species of the Venezuelan Guayana [3]) offer a particularly interesting base for comparison of natural

and plastic-impregnated woods. For this reason it was deemed interesting to compare, on a per unit incremental density basis, the increment in mechanical properties produced by filling density woods with various polymeric materials, and to compare the properties of these wood-polymer combinations to high-density woods. Also, direct examination of fracture surfaces by SEM was made in an attempt to correlate the strength of the materials tested to the morphology of the fracture surfaces.

2. Experimental

The following six woods, based on their availability and low density, were selected for impregnation studies (common name in parentheses):

<i>Erismia uncinatum</i> (muriello)	$\rho = 0.59 \text{ g cm}^{-3}$
<i>Hieronyma laxiflora</i> (sangrón)	$\rho = 0.63 \text{ g cm}^{-3}$
<i>Jacaranda superba</i> (girasol)	$\rho = 0.35 \text{ g cm}^{-3}$
<i>Pithecellobium jupunba</i> (saman)	$\rho = 0.62 \text{ g cm}^{-3}$
<i>Pterocarpus vernalis</i> (sangre de drago)	$\rho = 0.71 \text{ g cm}^{-3}$
<i>Tabebuia rosea</i> (apamate)	$\rho = 0.63 \text{ g cm}^{-3}$

The densities quoted above refer to air-dried specimens.

The following monomers were used for impregnation of samples of each of the above woods: methylmethacrylate (MMA) commercial grade of the Aldrich Chemical Co.

methylmethacrylate (80%) and unsaturated polyester (20%) (MMA-AR). The unsaturated polyester used was AROPOL 2731 of Ashland Chemical Co.

styrene (60%) and acrylonitrile (40%) (ST-AN) commercial grade of Aldrich Chemical Co.

styrene (48%), acrylonitrile (32%) and polyester (20%) (ST-AN-AR).

The impregnation was carried out in a steel tank of 50 cm \times 52 cm \times 13.5 cm. Sixteen samples of 6 cm \times 6 cm \times 50 cm were placed simultaneously in the tank, which was then evacuated to a pressure of 2 to 3 mm Hg and held at this pressure for 2 to 3 h. Monomer was then introduced maintaining the vacuum during this time. Subsequently a pressure of 2 atm was introduced on the monomer solution holding the immersed wood samples. This pressure, supplied from a nitrogen tank, was maintained for 14 h after which time the tank was depressurized and the impregnated samples removed. A schematic drawing of the impregnation apparatus is shown in Fig. 1.

The samples were subsequently irradiated from a ^{60}Co source of 7000 Ci activity. The samples were placed in an aluminium irradiation tank in a position where the average radiation intensity was of $0.045 \text{ Mrad h}^{-1}$. The total radiation doses used for the four monomers were the following: MMA-0.46 Mrad, MMA-AR-0.60 Mrad, ST-AN-1.10 Mrad and ST-AN-AR-0.35 Mrad.

Using the following values for the densities (ρ_M) of the polymerized plastic materials: MMA- 0.95 g cm^{-3} , MMA-AR- 1.01 g cm^{-3} , ST-AN- 0.87 g cm^{-3} and ST-AN-AR- 0.94 g cm^{-3} and a value of the density of the wood substance (ρ_w) of about 1.5 g cm^{-3} , the impregnation efficiency (ratio between the absorbed volume of monomer and the free pore volume of the wood, in percent) was calculated as $\text{imp. eff.} = 100.M/(\rho_M.V_f)\%$, where

IMPREGNATION FACILITY

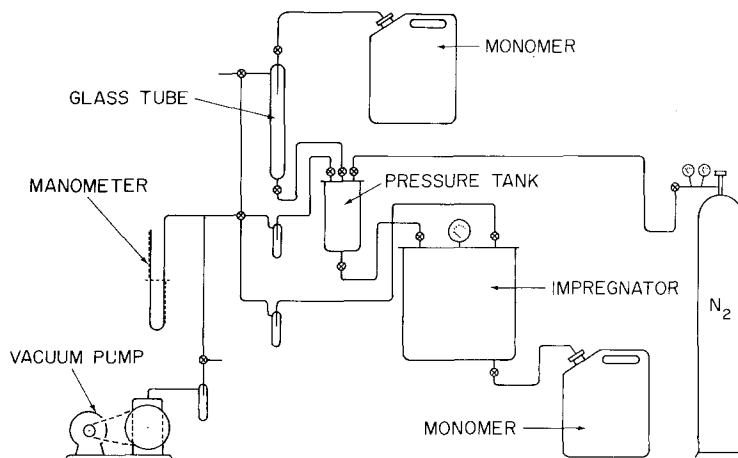


Figure 1 Schematic drawing of impregnation apparatus.

TABLE I Calculated mean impregnation efficiencies

Base wood	Polymer	Amount impregnated (g cm ⁻³)	Impregnation efficiency (%)
Mureillo	MMA	0.56	97
	MMA-AR	0.60	98
	ST-AN	0.53	10
	ST-AN-AR	0.57	99
Sangrón	MMA	0.58	105
	MMA-AR	0.24	41
	ST-AN	0.39	77
	ST-AN-AR	0.50	91
Girasol	MMA	0.71	98
	MMA-AR	0.63	82
	ST-AN	0.55	83
	ST-AN-AR	0.63	87
Saman	MMA	0.32	57
	MMA-AR	0.02	3.4
	ST-AN	0.27	53
	ST-AN-AR	0.14	25
Sangre de drago	MMA	0.41	82
	MMA-AR	0.42	78
	ST-AN	0.39	84
	ST-AN-AR	0.35	70
Apamate	MMA	0.50	91
	MMA-AR	0.33	56
	ST-AN	0.31	61
	ST-AN-AR	0.28	51

V_f , the free volume of the samples, is given by $V(1-\rho/\rho_M)$, where V is the total volume and ρ the density of the unimpregnated wood sample. The impregnation efficiencies so calculated are listed in Table I.

The polymer-containing samples, as well as control samples of natural wood, were then cut according to the specifications of standardized tests for the determination of their mechanical properties. All the tests used were ASTM (D143/45-92) [5], except for toughness, which was evaluated according to the specifications of the U.S. Forest Products Laboratory [4]. These tests were conducted at the facilities of the Laboratorio Nacional de Productos Forestales in Mérida, Venezuela.

During the cutting of the test samples considerable difficulties were encountered due to the poor machinability of the plastic containing materials. Cutting wheels reinforced with stellite had to be used for cutting and a high degree of tool wear was observed during the subsequent machining operations.

Aside from the standardized tests, a number of samples of natural and ST-AN containing samples of girasol and sangre de drago were prepared for tensile testing in an Instron machine, and subsequent observation of the fracture surfaces. The

test specimens had a cylindrical reduced section 2.5 cm long and 0.75 cm diameter, and samples were cut with the fibre at 0°, 45° and 90° to the direction of the tensile load. The results of these tests are discussed below.

3. Results

3.1. Mechanical properties

The results of the standardized tests performed on the composite materials and the corresponding control samples are listed in Tables II to IV. It is evident from these results that the effect of the presence of plastics varies widely from one property to another, as well as between samples and little, if any, correlation can be established between the types of woods and plastics and the resulting properties.

Hardness, shear strength parallel to the grain, and compression strength perpendicular to the grain are the most consistently improved properties. On the other hand, the effect of the plastics on toughness was found to be generally negative with a decrease of more than 50% in a number of cases. These results are essentially in agreement with those obtained for plastic-wood combinations of other researchers working with temperate climate woods such as birch, alder, pine, aspen, etc. [7, 8].

TABLE II Results of static bending tests on natural woods and wood-plastic combinations.

Wood	Impregnation (density)*	Proportional limit (% change)† (kg cm ⁻²)	Static bending tests ultimate strength (% change) (kg cm ⁻²)	Modulus of elasticity (% change) (10 ³ kg cm ⁻²)
Mureillo (<i>Erismia uncinatum</i>)	NI (0.59)	670 ± 145 ⁶²	960 ± 195 ⁶²	125 ± 25 ⁶²
	MMA (1.15)	880 ± 150 ⁵ (+31%)	1430 ± 210 ⁵ (+49%)	165 ± 30 ⁵ (+36%)
	MMA-AR (1.19)	890 ± 15 ² (+33%)	1380 ± 30 ² (+44%)	165 ± 5 ² (+36%)
	ST-AN (1.12)	850 ± 60 ² (+27%)	1320 ± 120 ² (+38%)	165 ± 20 ² (+36%)
	ST-AN-AR (1.16)	790 ± 60 ³ (+18%)	1170 ± 50 ³ (+22%)	160 ± 10 ² (+28%)
Sangron (<i>Hieronyma laxiflora</i>)	NI (0.63)	600 ± 155 ⁵	980 ± 70 ⁵	115 ± 10 ⁵
	MMA (1.21)	700 ± 120 ⁴ (+17%)	1180 ± 190 ⁴ (+20%)	135 ± 25 ⁴ (+17%)
	MMA-AR (0.87)	910 ± 120 ² (+52%)	1240 ± 200 ² (+27%)	155 ± 30 ² (+17%)
	STAN (1.02)	960 ± 120 ² (+60%)	1380 ± 200 ² (+41%)	175 ± 30 ² (+52%)
	STAN-AR (1.13)	700 ± 120 ² (+17%)	1010 ± 200 ² (+3%)	135 ± 30 ² (+17%)
Girasol (<i>Jacaranda supereba</i>)	NI (0.35)	280 ± 50 ⁸	510 ± 90 ⁸	75 ± 15 ⁸
	MMA (1.06)	790 ± 80 ² (+182%)	1160 ± 130 ² (+127%)	150 ± 10 ² (+100%)
	MMA-AR (0.98)	850 ± 80 ² (+204%)	1150 ± 65 ² (+125%)	155 ± 10 ² (+107%)
	ST-AN (0.90)	780 ± 105 ² (+179%)	1140 ± 250 ² (+124%)	160 ± 5 ² (+113%)
	ST-AN-AR (0.98)	810 ± 40 ² (+189%)	1140 ± 70 ² (+124%)	165 ± 10 ² (+120%)
Saman (<i>Pithecellobium Jupunba</i>)	NI (0.62)	650 ± 155 ¹⁶	1040 ± 265 ¹⁶	140 ± 30 ¹⁶
	MMA (0.94)	610 ± 150 ⁴ (-6%)	1010 ± 335 ⁴ (-3%)	120 ± 50 ⁴ (-14%)
	MMA-AR (0.64)	380 ± 30 ² (-42%)	670 ± 50 ² (-36%)	70 ± 20 ² (-50%)
	STAN (0.89)	460 ± 90 ² (-29%)	600 ± 95 ² (-42%)	85 ± 15 ² (-39%)
	STCAN-AR (0.76)	850 ± 155 ² (+31%)	1290 ± 170 ² (+24%)	150 ± 10 ² (+7%)
Sangre de drago (<i>Pterocardus vernalis</i>)	NI (0.71)	710 ± 210 ¹⁰	1210 ± 195 ¹⁰	130 ± 15 ¹⁰
	MMA (1.12)	900 ± 75 ² (+27%)	1310 ± 120 ³ (+8%)	145 ± 10 ² (+12%)
	MMA-AR (1.13)	870 ± 60 ⁴ (+22%)	1370 ± 130 ⁴ (+13%)	160 ± 20 ⁴ (+23%)
	ST-AN-AR (1.10)	930 ± 20 ⁴ (+31%)	1320 ± 160 ⁴ (+9%)	160 ± 10 ⁴ (+23%)
	ST-AN-AR (1.06)	840 ± 20 ³ (+18%)	1410 ± 125 ³ (+17%)	145 ± 10 ³ (+12%)
Apamate (<i>Tabebuia Rosea</i>)	NI (0.63)	660 ± 40 ⁸	1040 ± 125 ³	105 ± 5 ⁸
	MMA (1.03)	940 ± 80 ⁵ (+42%)	1530 ± 80 ⁵ (+46%)	160 ± 10 ⁵ (+52%)
	MMA-AR (0.96)	980 ± 70 ² (+48%)	1460 ± 175 ² (+40%)	155 ± 5 ² (+48%)
	ST-AN (0.94)	900 ± 90 ³ (+36%)	1470 ± 140 ³ (+41%)	160 ± 15 ³ (+52%)
	ST-AN-AR (0.91)	950 ± 100 ³ (+44%)	1370 ± 110 ³ (+32%)	150 ± 5 ³ (+43%)

*Density in g cm⁻³ at 12% humidity.

†Based on values for non-impregnated wood.

Superscript numbers indicate the number of samples on which the r.m.s. deviations are calculated.

One of the wood specimens (saman) showed rather irregular behaviour with regard to both the impregnation efficiency and the properties of the resulting wood-polymer combination, appearing to suffer severe damage from the introduction of plastics.

Excluding the saman-based materials, 13 of the 20 wood-polymer combinations tested exhibited a decrease in toughness on introduction of plastics, while only 2 of the 20 samples showed a decrease in some of the other mechanical properties.

Since the combination with polymer entails the absorption of a considerable amount of material with a corresponding change in density, a fair comparison of the properties of natural and combined materials should be made on a per unit

weight basis, i.e. on specific properties. For this same reason it is of particular interest to compare wood-plastic composites with high-density natural woods.

The nearly linear correlation between most mechanical properties of woods and their specific weight (density) is generally recognized [3, 6]. As an illustration, Fig. 2 and 3 show the ultimate strength in bending and side hardness, respectively of 24 tropical woods and an equal number of temperate climate woods, as a function of their density. The data shown on these diagrams were selected at random from extensive compilations of data on Venezuelan woods [3] and temperate climate American woods [6]. These figures also show that climatic conditions do not affect the

TABLE III Results of compression tests on natural woods and wood-plastic combinations.

Wood	Impregnation (Density)*	Compression tests			
		Parallel to grain (% change)†		Perpendicular to grain	
		Proportional limit (kg cm ⁻²)	Ultimate strength (kg cm ⁻²)	Modulus of Elasticity (10 ³ kg cm ⁻²)	Proportional limit
Mureillo (<i>Erisma uncinatum</i>)	NI (0.59)	380 ± 120 ⁶⁶	530 ± 95 ⁶⁶	105 ± 25 ⁶⁶	45 ± 10 ⁶⁹
	MMA (1.15)	480 ± 60 ⁵	660 ± 30 ⁵	120 ± 20 ⁵	330 ± 10 ³
	MMA-AR (1.19)	530 ± 65 ⁵	690 ± 20 ⁵	115 ± 10 ⁵	180 ± 60 ⁵
	ST-AN (1.12)	480 ± 55 ⁵	740 ± 55 ⁵	135 ± 20 ⁵	320 ± 105 ⁵
	ST-AN-AR (1.16)	490 ± 30 ⁵	610 ± 40 ⁵	115 ± 5 ⁵	200 ± 75 ⁴
Sangron (<i>Theironyma laxiflora</i>)	NI (0.63)	330 ± 45 ⁵	480 ± 45 ⁵	80 ± 10 ⁵	60 ± 6 ¹⁰
	MMA (1.21)	340 ± 30 ²	670 ± 60 ²	165 ± 20 ²	100 ± 25 ²
	MMA-AR (0.87)	440 ± 30 ⁵	680 ± 30 ⁵	115 ± 15 ⁵	145 ± 15 ⁵
	ST-AN (1.02)	570 ± 30 ⁵	730 ± 85 ⁵	130 ± 25 ⁵	205 ± 30 ⁵
	ST-AN-AR (1.13)	470 ± 30 ²	820 ± 60 ²	225 ± 20 ²	125 ± 20 ²
Girasol (<i>Jacaranda superba</i>)	NI (0.35)	165 ± 50 ⁹	260 ± 75 ⁹	35 ± 10 ⁹	20 ± 10 ⁵
	MMA (1.06)	445 ± 50 ²	790 ± 55 ²	195 ± 15 ²	75 ± 20 ²
	MMA-AR (0.90)	455 ± 20 ⁵	640 ± 25 ⁵	120 ± 20 ⁵	50 ± 5 ⁵
	ST-AN (0.90)	455 ± 55 ⁵	540 ± 55 ⁵	125 ± 5 ⁵	80 ± 40 ⁵
	ST-AN-AN (0.98)	455 ± 80 ⁵	560 ± 85 ⁵	110 ± 15 ⁵	65 ± 5 ⁵
Saman (<i>Pithecellobium Jupuneca</i>)	NI (0.62)	440 ± 120 ¹²	520 ± 115 ¹²	115 ± 40 ¹²	70 ± 25 ¹²
	MMA (0.94)	310 ± 55 ⁵	500 ± 55 ⁵	65 ± 75 ⁵	115 ± 25 ⁵
	MMA-AR (0.64)	290 ± 50 ⁵	430 ± 70 ⁵	55 ± 15 ⁵	65 ± 5 ⁵
	ST-AN (0.89)	350 ± 35 ⁵	630 ± 95 ⁵	75 ± 10 ⁵	120 ± 5 ⁵
	ST-AN-AR (0.76)	270 ± 40 ⁵	360 ± 30 ⁵	45 ± 5 ⁵	80 ± 25 ⁵
Sangre de drago (<i>Pterocarpus vernalis</i>)	NI (0.71)	400 ± 55 ¹⁰	530 ± 45 ¹⁰	100 ± 15 ¹⁰	60 ± 5 ⁸
	MMA (1.12)	460 ± 75 ⁵	580 ± 65 ⁵	115 ± 15 ⁵	165 ± 80 ⁵
	MMA-AR (1.13)	290 ± 60 ⁵	430 ± 45 ⁵	100 ± 20 ⁵	175 ± 15 ⁵
	ST-AN (1.10)	500 ± 25 ⁴	630 ± 35 ⁴	125 ± 10 ⁴	325 ± 80 ⁵
	ST-AN-AR (1.06)	460 ± 40 ⁵	680 ± 25 ⁵	100 ± 10 ⁵	210 ± 25 ⁵
Apamate (<i>Tabebuia rosea</i>)	NI (0.63)	400 ± 70 ⁵	500 ± 40 ⁵	95 ± 20 ⁵	50 ± 5 ⁸
	MMA (1.03)	470 ± 130 ⁵	600 ± 65 ⁵	125 ± 20 ⁵	185 ± 45 ⁵
	MMA-AR (0.96)	360 ± 45 ⁵	510 ± 35 ⁵	100 ± 10 ⁵	140 ± 15 ⁵
	ST-AN (0.94)	440 ± 85 ³	540 ± 130 ³	105 ± 5 ³	170 ± 15 ⁵
	ST-AN-AR (0.91)	460 ± 85 ⁴	600 ± 80 ⁴	105 ± 10 ⁴	150 ± 10 ⁵

* Density in g cm⁻³ at 12% humidity.

† Based on values for non-impregnated wood.

Superscript numbers indicate the number of samples on which the r.m.s. deviations are calculated.

TABLE IV Hardness, shear strength and toughness of natural woods and wood-plastic combinations.

Wood	Impregnation (density)	Hardness (kg)		End (% change) [†]	Shear strength parallel to grain (kg cm ⁻²)		Toughness (m kg (% change)) [†]	
		Side (% change) [†]						
Mureillo (<i>erisma uncinatum</i>)	NI (0.59)	360 ± 90 ⁷¹		520 ± 110 ⁷¹	80 ± 15 ¹¹⁷	1.20 ± 0.35 ⁷⁶		
	MMA (1.15)	1480 ± 240 ²	(+310%)	N.A.	200 ± 35 ¹⁰	1.35 ± 0.60	(+13%)	
	MMA-AR (1.19)	1140 ± 270 ¹⁰	(215%)	850 ± 165 ⁵	(+63%)	180 ± 25 ¹⁰	1.95 ± 1.40 ⁵	(+54%)
	ST-AN (1.12)	1270 ± 200 ²	(+255%)	860 ± 30 ²	(+255%)	145 ± 35 ¹⁰	0.60 ± 0.10 ⁵	(-50%)
	ST-AN-AR (1.16)	1120 ± 250 ⁹	(+210%)	700 ± 55 ²	(+35%)	155 ± 55 ¹⁰	0.65 ± 0.10 ⁵	(-46%)
Sangron (<i>Hieronyma laxiflora</i>)	NI (0.63)	510 ± 95 ⁸		690 ± 95 ⁸	105 ± 15 ¹⁰	1.25 ± 0.55 ⁶		
	MMA (1.21)	2040 ± 280 ²	(+300%)	1890 ± 230 ²	(+175%)	160 ± 20 ²	0.95 ± 0.15 ²	(-24%)
	MMA-AR (0.87)	2080 ± 240 ¹⁰	(+310%)	2180 ± 195 ⁵	(+215%)	185 ± 25 ¹⁰	0.95 ± 0.10 ⁵	(-24%)
	ST-AN (1.02)	1520 ± 320 ¹⁰	(+200%)	1580 ± 265 ⁴	(+130%)	200 ± 20 ¹⁰	0.80 ± 0.20 ⁵	(-36%)
	ST-AN-AR (1.13)	1630 ± 280 ²	(+230%)	1600 ± 230 ²	(+130%)	200 ± 20 ²	0.50 ± 0.15 ²	(-60%)
Girasol (<i>Jacaranda superba</i>)	NI (0.35)	190 ± 100 ⁶		300 ± 120 ⁶	60 ± 15 ¹⁰	0.55 ± 0.15 ⁵		
	MMA (1.06)	1470 ± 95 ²	(+670%)	1040 ± 100 ²	(+250%)	115 ± 15 ²	1.25 ± 0.90 ²	(+125%)
	MMA-AR (0.98)	1230 ± 130 ¹⁰	(+550%)	1220 ± 90 ³	(+310%)	105 ± 10 ⁹	1.15 ± 0.20 ⁵	(+110%)
	ST-AN (0.90)	1190 ± 100 ⁸	(+530%)	760 ± 100 ²	(+150%)	125 ± 15 ¹⁰	0.80 ± 0.10 ⁵	(+45%)
	ST-AN-AR (0.98)	1220 ± 60 ¹⁰	(+540%)	1130 ± 110 ³	(+280%)	120 ± 10 ¹⁰	0.95 ± 0.24 ⁵	(+73%)
Saman (<i>Pithecellobium jupunba</i>)	NI (0.62)	495 ± 140 ²⁰		460 ± 110 ²⁰	105 ± 20 ²⁸	2.10 ± 0.80 ¹⁷		
	MMA (0.94)	680 ± 200	(+37%)	650 ± 270 ⁵	(+41%)	120 ± 20 ¹⁰	1.20 ± 0.15 ⁵	(-42%)
	MMA-AR (0.64)	660 ± 140 ¹⁰	(+33%)	550 ± 125 ⁵	(+20%)	100 ± 10 ¹⁰	0.55 ± 0.05 ⁵	(-74%)
	ST-AN (0.89)	995 ± 200 ¹⁰	(+100%)	785 ± 25 ²	(+71%)	125 ± 10 ¹⁰	0.90 ± 0.10 ⁵	(-57%)
	ST-AN-AR (0.76)	630 ± 115 ¹⁰	(+25%)	625 ± 120 ⁵	(+36%)	115 ± 15 ¹⁰	0.75 ± 0.10 ⁵	(-64%)
Sangre de drago (<i>Pterocarpus vernalis</i>)	NI (0.71)	630 ± 120 ⁸		740 ± 130 ⁸	115 ± 20 ¹⁰	2.60 ± 0.80 ¹⁰		
	MMA (1.12)	2030 ± 125 ¹⁰	(+180%)	1550 ± 380 ²	(+110%)	185 ± 20 ¹⁰	1.15 ± 0.20 ⁵	(-56%)
	MMA-AR (1.13)	1750 ± 235 ⁸	(+91%)	1410 ± 310 ²	(+91%)	160 ± 20 ¹⁰	2.50 ± 0.35 ⁵	(-4%)
	ST-AN (1.10)	2060 ± 135 ⁷	(+230%)	1290 ± 360 ²	(+74%)	180 ± 45 ⁸	0.95 ± 0.15 ⁵	(-79%)
	ST-AN-AR (1.06)	1650 ± 375 ⁷	(+160%)	1200 ± 390 ²	(+62%)	170 ± 25 ¹⁰	1.80 ± 0.50 ⁵	(-31%)
Apamate (<i>Tabebuia</i>)	NI (0.63)	440 ± 50 ⁸		630 ± 80 ⁸	110 ± 15 ⁸	1.35 ± 0.70 ⁵		
	MMA (1.03)	1490 ± 155 ¹⁰	(+240%)	1520 ± 110 ⁴	(+140%)	165 ± 10 ⁹	1.60 ± 0.15 ⁵	(+19%)
	MMA-AR (0.96)	1130 ± 220 ¹⁰	(+160%)	1320 ± 100 ⁵	(+110%)	145 ± 20 ⁵	1.15 ± 0.15 ⁵	(-15%)
	ST-AN (0.94)	1550 ± 120 ⁵	(+250%)	1040 ± 90 ²	(+65%)	140 ± 30 ⁶	0.90 ± 0.05 ⁸	(-33%)
	ST-AN-AR (0.91)	1420 ± 95 ⁸	(=220%)	1630 ± 60 ⁴	(+160%)	145 ± 20 ⁸	0.95 ± 0.05 ⁴	(-30%)

* Density in g cm⁻³ at 12% humidity.

[†] Based on values for non impregnated wood.

Superscript numbers indicate the number of samples on which the r.m.s. deviations are calculated.

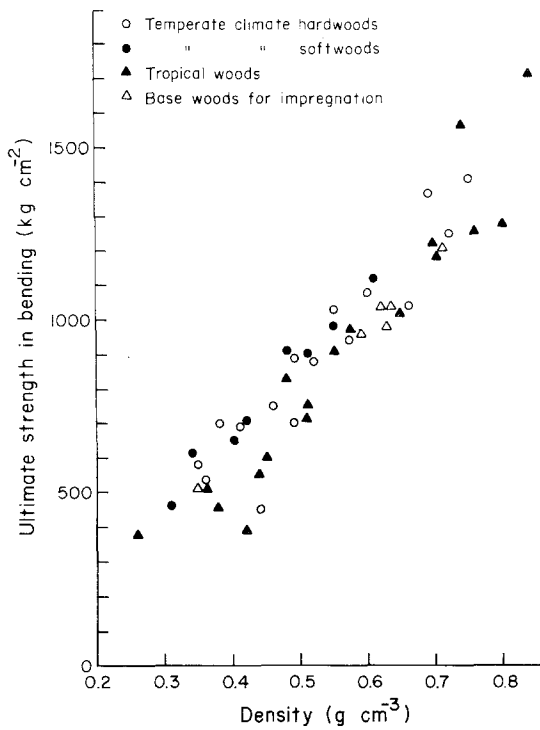


Figure 2 Ultimate strength in bending of tropical and temperate climate woods.

distribution of mechanical properties in function of density. It is interesting to note, however, that while most woods of temperate climate have densities (at 12% humidity) in the range of 0.4 to 0.8 g cm^{-3} , there are a large number of tropical woods with densities in the 0.9 to 1.2 g cm^{-3} range, which corresponds to the densities obtainable in wood-plastic combinations. These higher density woods allow an interesting comparison to be made between the increment in mechanical properties by increased density through addition of plastic resins on the one hand, and natural cellulose on the other.

Four of the mechanical properties of wood-plastic combinations over an equivalent density range are shown in Fig. 4 to 7. It is evident from these diagrams that the side hardness and the strength in compression are essentially independent of the exact nature of the material and are determined primarily by the density.

As regards strength in bending and toughness, however, it appears that natural woods are superior to plastic-containing woods of equivalent density, and thus natural cellulose is a filler superior to the resins used.

A more quantitative interpretation of these

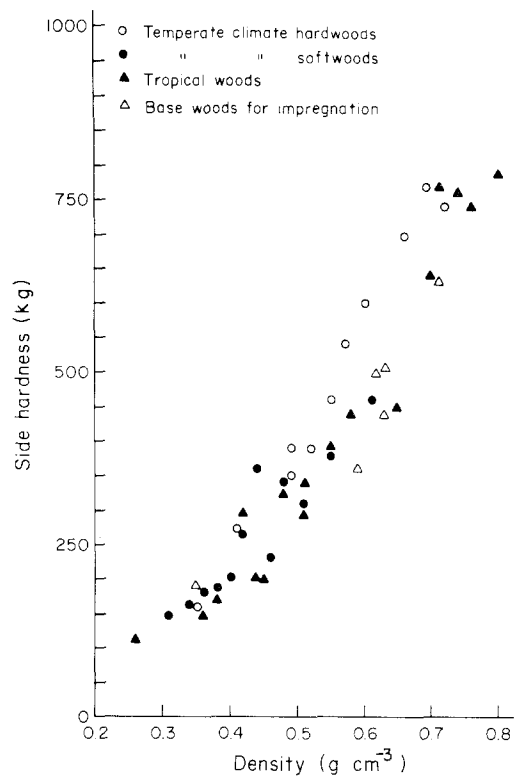


Figure 3 Side hardness of tropical and temperate climate woods.

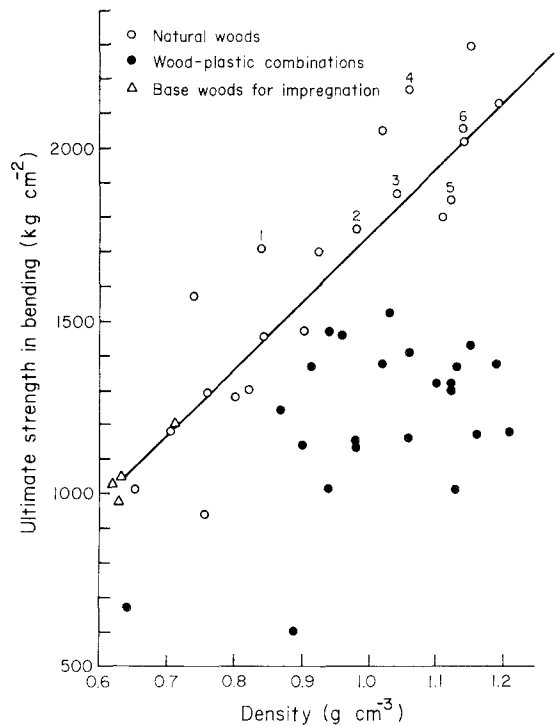


Figure 4 Ultimate strength in bending of natural woods and wood-plastic composites as a function of density.

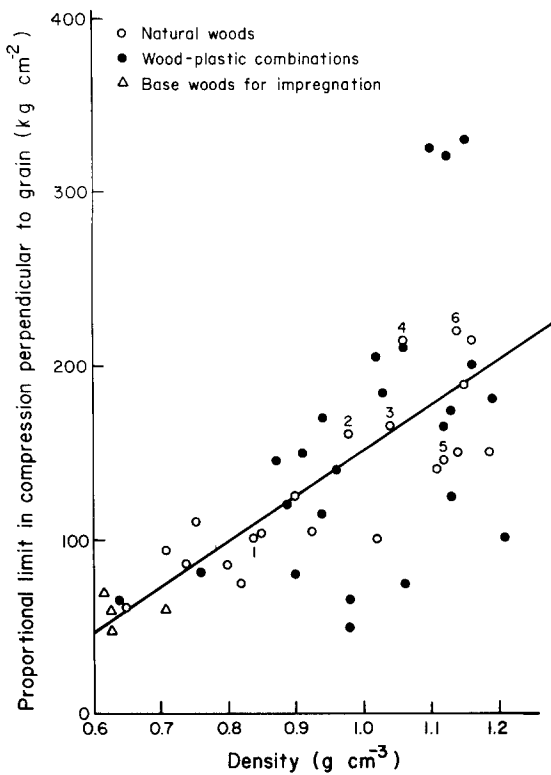


Figure 5 Proportional limit in compression perpendicular to grain of natural woods and wood-plastic composites as a function of density.

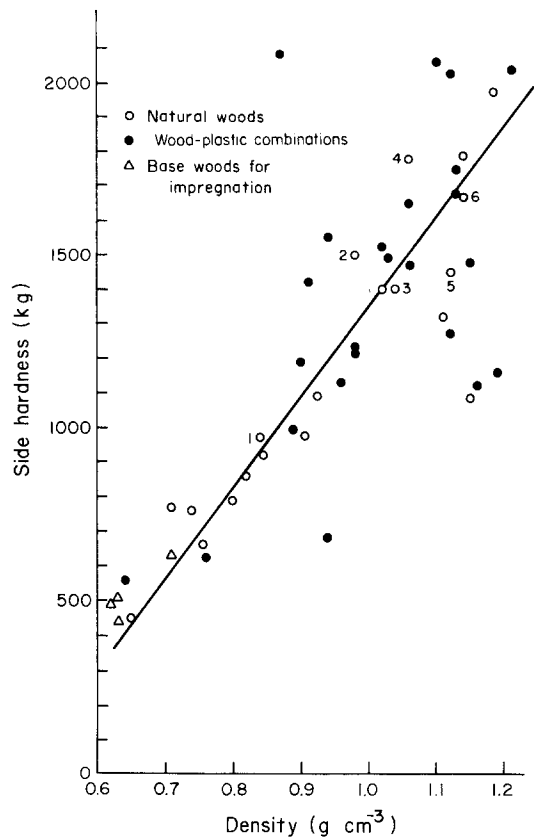


Figure 6 Side hardness of natural woods and wood plastic composites as a function of density.

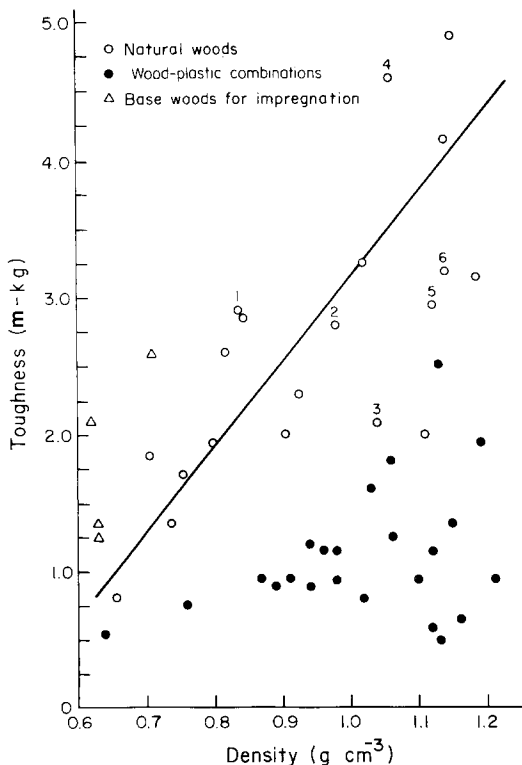


Figure 7 Toughness of natural woods and wood-plastic composites as a function of density.

effects can be established by the relationship:

$$Y_c = d_w \bar{Y}_w + (d_c - d_w) \bar{Y}_p \quad (1)$$

where Y denotes any given mechanical property, d concentration in g cm^{-3} , and the subscripts c , w and p refer to the composite material, natural wood, and plastic, respectively. The quantities \bar{Y} are the specific values of Y , i.e. Y/d for each of the constituents of the composite.

Since Y_c , \bar{Y}_w , d_c and d_w are experimentally measured parameters, Equation 1 may be solved for \bar{Y}_p which represents the increment in the property Y per unit density change produced by addition of plastic material.

The same type of incremental effect can be calculated on the basis of a comparison of a low and high-density wood (w_l and w_h):

$$Y_{wh} = d_{w1} \bar{Y}_{w1} + (d_{wh} - d_{w1}) \bar{Y}_{c1} \quad (2)$$

where \bar{Y}_{c1} is the equivalent of \bar{Y}_p but refers to filling by natural cellulose. Since the costs of plastics and production of wood-plastic combinations can be compared directly to the cost of

TABLE V Proportional limit in compression perpendicular to grain of natural woods and wood-plastic composites as a function of density.

Y =	USB	PLC1	PLC2	HS	HE	SS	T
NI (\bar{Y}_w, \bar{Y}_{w1})	1700	560	85	890	1040	165	3.60
MMA (\bar{Y}_p)	240	150	260	3410	1980	160	-3.50
MMA/AR (\bar{Y}_p)	370	- 270	270	2650	1600	105	-0.25
ST/AN (\bar{Y}_p)	570	270	680	3670	1410	165	-4.20
ST/AN/AR (\bar{Y}_p)	560	175	430	2920	1320	150	-2.30
Tocorito (\bar{Y}_{c1})	3860	1190	290	2580	3030	220	2.60
Algarrobo (\bar{Y}_{c1})	2060	1440	360	3200	3160	260	0.75
Mora (\bar{Y}_{c1})	2000	1140	320	2300	2300	135	-1.45
Zapatero (\bar{Y}_{c1})	2760	1070	430	3290	2440	155	5.75
Cacaito (\bar{Y}_{c1})	1560	705	205	2000	1590	35	0.80
Hierro (\bar{Y}_{c1})	1970	930	370	2400	1120	35	1.45

Units: USB, PLC1, PLC2 and SS in $\text{kg cm}^{-2} (\text{g cm}^{-3})^{-1}$, HS and HE in $\text{kg} (\text{g cm}^{-3})^{-1}$, T in $\text{kg-m} (\text{g cm}^{-3})^{-1}$.

higher density woods, \bar{Y}_p and \bar{Y}_{c1} may be used as the base for establishing a quality/cost ratio for these materials.

For a comparison on this basis, we selected seven properties that span the range of the different mechanical tests to which the samples were subjected, and six common high-density woods, as given below. Basic mechanical properties for comparison of natural and plastic containing woods were: (1) ultimate strength (USB); (2) fibre stress at proportional limit in compressions parallel to grain (PLC1); (3) fibre stress at proportional limit in compression perpendicular to grain (PCL2); (4) side hardness (HS); (5) end hardness (HE); (6) maximum shearing strength parallel to grain (SS); (7) toughness (T).

The high-density natural woods used for comparison (densities, at 12% humidity, in parenthesis) were: (1) *Lonchocarpus latifolius* (tocorito) ($\rho = 0.84 \text{ g cm}^{-3}$); (2) *Hymenaea courbaril* (algarrobo) ($\rho = 0.98 \text{ g cm}^{-3}$); (3) *Mora gongrijpii* (mora) ($\rho = 1.04 \text{ g cm}^{-3}$); (4) *Peltogyne*

porphyrocardia (zapatero) ($\rho = 1.06 \text{ g cm}^{-3}$); (5) *Eschweilera grata* (cacaito) ($\rho = 1.12 \text{ g cm}^{-3}$); (6) *Licania alta* (hierro) ($\rho = 1.14 \text{ g cm}^{-3}$).

The high-density woods were selected to span the density range of the wood-plastic combinations and to be representative of average properties - the points corresponding to these high-density woods are marked 1 to 6 in Figs. 4 to 7.

Tables V and VI show the values of \bar{Y}_p and \bar{Y}_{c1} of the various plastics and the six high-density natural woods calculated relative to the highest and lowest density base woods used for impregnation (sangre de drago $\rho = 0.71$, and girasol $\rho = 0.35$). The result, which is typical of the other natural woods as well, shows that $\bar{Y}_p \approx \bar{Y}_{c1}$ for properties such as compression perpendicular to the grain, hardness, and maximum shearing strength, hence these properties appear to be functions of, primarily, the density of the material. On the other hand, for compression parallel to the grain, static bending, and toughness, natural cellu-

TABLE VI Incremental specific properties (\bar{Y}_p and \bar{Y}_{c1}) relative to girasol.

Y =	USB	PLC1	PLC2	HS	HE	SS	T
NI (\bar{Y}_w, \bar{Y}_{w1})	1460	480	55	535	860	170	1.55
MMA (\bar{Y}_p)	920	390	80	1810	1040	75	1.00
MMA/AR (\bar{Y}_p)	1020	460	45	1650	1460	65	0.95
ST/AN (\bar{Y}_p)	1140	520	110	1830	830	110	0.45
ST/AN/AR (\bar{Y}_p)	1000	440	70	1640	1310	90	0.60
Tocorito (\bar{Y}_{c1})	2450	790	160	1580	1700	170	4.85
Algarrobo (\bar{Y}_{c1})	1990	990	220	2080	2050	200	3.55
Mora (\bar{Y}_{c1})	1970	885	210	1740	1740	145	2.25
Zapatero (\bar{Y}_{c1})	2350	860	270	2250	1820	115	5.70
Cacaito (\bar{Y}_{c1})	1740	680	160	1640	1420	90	3.05
Hierro (\bar{Y}_{c1})	1960	800	255	1870	1170	90	3.35

Units: as in Table V.

TABLE VII Average specific (Y_{w1} and Y_{wh}) and incremental specific properties (\bar{Y}_p and \bar{Y}_{c1}) of plastics and high-density woods relative to six low-density woods.

Y	USB	PLC1	PLC2	HS	HE	SS	T
Low-density woods (\bar{Y}_w, \bar{Y}_{w1})							
MMA (\bar{Y}_p)	1600 ± 80	570 ± 60	80 ± 15	710 ± 130	975 ± 90	160 ± 15	
MMA/AR (\bar{Y}_p)	710 ± 360	185 ± 120	250 ± 165	2500 ± 630	1830 ± 465	135 ± 50	-0.40 ± 1.60
ST/AN (\bar{Y}_p)	890 ± 320	155 ± 290	235 ± 105	2840 ± 1890	2380 ± 1970	155 ± 95	0.05 ± 0.90
ST/AN/AR (\bar{Y}_p)	960 ± 300	345 ± 180	410 ± 185	2680 ± 830	1300 ± 570	150 ± 55	-1.50 ± 1.50
ST/AN/AR (\bar{Y}_p)	640 ± 420	265 ± 95	250 ± 135	2350 ± 800	1670 ± 1060	140 ± 30	-1.10 ± 0.95
High density woods							
Tocorito (\bar{Y}_{c1})	3210 ± 480	900 ± 190	215 ± 45	2250 ± 350	2340 ± 440	205 ± 40	5.95 ± 2.15
Algarrobo (\bar{Y}_{c1})	2090 ± 50	1180 ± 70	290 ± 45	2810 ± 390	2660 ± 360	240 ± 30	3.35 ± 1.35
Mora (\bar{Y}_{c1})	2040 ± 70	980 ± 105	265 ± 35	2160 ± 220	2070 ± 190	140 ± 20	1.35 ± 1.35
Zapatero (\bar{Y}_{c1})	2630 ± 160	940 ± 95	360 ± 55	2930 ± 360	2180 ± 210	155 ± 30	6.80 ± 0.90
Cacaito (\bar{Y}_{c1})	1680 ± 80	660 ± 55	185 ± 15	1940 ± 160	1530 ± 90	60 ± 25	2.70 ± 0.95
Hierro (\bar{Y}_{c1})	2010 ± 50	840 ± 65	320 ± 35	2260 ± 200	1150 ± 80	65 ± 25	3.20 ± 0.90

Units: as in Table V.

lose is superior to the plastic fillers tested ($\bar{Y}_{cl} > \bar{Y}_p$).

Table VII summarizes the values of \bar{Y}_p and \bar{Y}_{c1} averaged over all pairs of low-density wood-wood plastic combinations and low density-high density woods considered, (excluding all data based on saman, because of the highly irregular impregnation efficiencies and properties of the wood-plastic composites based on this wood) together with the mean square deviation from these average values. It is evident from this table that the decrease in toughness constitutes the major difficulty for the use of wood-plastic combinations.

These findings are in general agreement with similar studies on other types of woods. Miettinen [9], for instance, has found that among temperate climate woods some are more impregnable than others and that the type of monomer used has little effect on the properties of the impregnated material. Miettinen also reports possible physical damage to samples during the radiation treatment.

Singer *et al.* [8] have conducted a study similar to the present one using beech and pine as the base woods and methylmethacrylate, vinyl acetate, and a styrene-acrylonitrile mixture as the impregnation polymers, finding that polymethylmethacrylate leads to improved bending strength and hardness while vinyl acetate and styrene-acrylonitrile give rather irregular (frequently negative) effects on the mechanical properties. For comparison with the present results, the average increment of the static bending strength (\bar{Y}_p) for methylmethacrylate in beech, calculated from

Singer's data (8 samples) has a value of $735 \pm 390 \text{ kg cm}^{-2}$, which is similar to the $710 \pm 360 \text{ kg cm}^{-2}$ average obtained on the low-density tropical wood samples.

3.2. Effect of plastic impregnation on elastic properties of wood

In an attempt to obtain a better understanding of the properties of the wood-plastic combinations under study, a number of tensile specimens were tested in an Instron machine. It was observed that samples cut at 0° , 45° and 90° to the grain direction, regardless of the presence or absence of plastic, failed in shear along the fibres, confirming that the cellulose fibres are more highly resistant to tension than either the natural interfibrillar material or the plastic filling material.

Fig. 8 shows typical stress-strain diagrams obtained from these experiments. These diagrams show that the presence of plastic in wood increases its strength and decreases its ductility. The toughness, as measured by the area under the $\sigma - \epsilon$ curves, shows a decrease of the order of 30% for the composite material, which is in agreement with the toughness measured by impact tests (see Table IV).

As can be seen from the data shown in Table II, there is generally a marked increase (up to 100%) in the elastic modulus on introducing plastic material, into the wood structure. These values compare with the 30 to 40% increase in modulus of elasticity of several temperate climate woods reported by Miettinen *et al.* [7].

It is possible to explain the increase in elastic

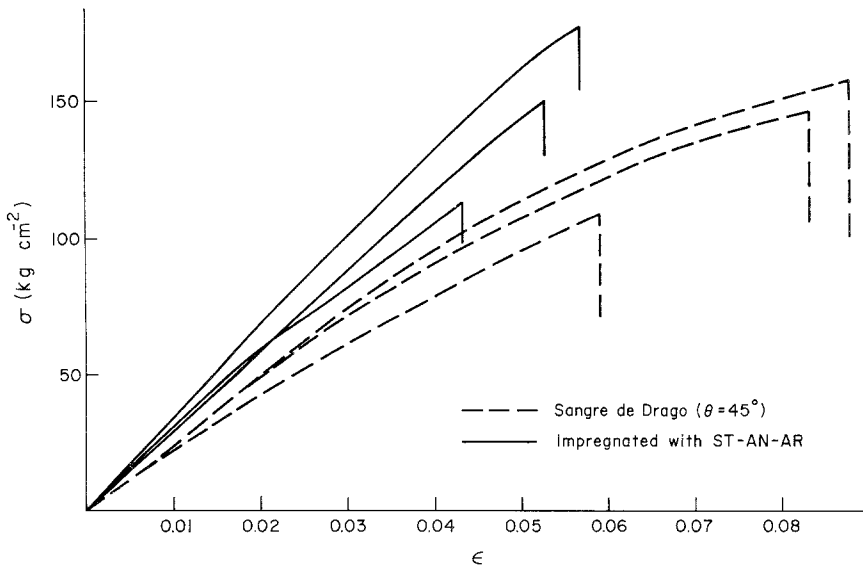


Figure 8 Typical stress-strain diagrams for natural wood and wood-plastic composites. Tensile stress at 45° to grain.

modulus on the basis of a model of natural fibres proposed by Hearle [10]. According to this model, each cellulose fibre consists of smaller, crystalline filaments wound in a spiral and interconnected by amorphous material. The overall elasticity of the material results from the combined effect of the spring-like extension of the spirals, stretching of the crystalline material, and changes in volume (i.e. lateral compression) of the amorphous, interconnecting material. This latter effect is dominant in low-density materials, such as the natural woods under study, since the void fraction forms part of the amorphous fraction of the material. The elastic modulus may then be written as [10]:

$$K = K_0(1 - \cot^2 \theta)^2 \quad (3)$$

where θ is the spiral angle of the cellulose fibres

$$\text{and} \quad K_0 = \frac{(1 - \alpha)^2}{(1 - \gamma)} k_a + \frac{\alpha^2}{\gamma} k_c. \quad (4)$$

In Equation 4 $\alpha = \Delta V_c / \Delta V$ is the fraction of the total volume change occurring in the crystalline material, γ the fraction of crystalline material, and k_a and k_c constants that depend of the properties of the amorphous and crystalline materials. Generally, the crystalline part of the material is much less compressible than the non-crystalline part, hence $\alpha \ll 1$ and

$$K_0 \approx \frac{k_a}{1 - \gamma} + \frac{\alpha^2}{\gamma} k_c. \quad (5)$$

Since the introduction of plastics reduces the void fraction within the non-crystalline part of the wood, it can be expected to increase the value of k_a and, hence, the elastic modulus. Correspondingly, it would be expected that the increase in the elastic modulus in going to higher density natural woods would be considerably larger, since in this case crystalline cellulose would partly act as the "filler" in place of the synthetic materials and $k_c \gg k_a$. The experimental results confirm this inference: for the six high-density woods used previously for comparisons, the elastic moduli in static bending and compression range from 1.92×10^5 to $2.87 \cdot 10^5$ kg cm^{-2} , and $1.54 \cdot 10^5$ to $2.88 \cdot 10^5$ kg cm^{-2} , respectively [3]; meanwhile the maximum values of the corresponding elastic moduli for the wood-plastic composites are $1.75 \cdot 10^5$ and $2.25 \cdot 10^5$ kg cm^{-2} , being generally much lower than these particular values (See Tables II and III). It is also interesting to examine the experimental data in terms of the conventional representation of deformation of composite materials in terms of the equal strain and equal stress limits.

If it is assumed that the strain in the cellulose and polymer phases is the same, as would be the case if the cellulose fibres and polymer filler were perfectly bonded along the direction of the applied force, the stress in the composite material would be:

$$\sigma_T = \sigma_w v_w + \sigma_p v_p \quad (6)$$

TABLE VIII Experimental and calculated elastic moduli in compression for wood-MMA combinations.

Wood	E_w (10^3 kg cm^{-2})	v_w	E_p (10^3 kg cm^{-2})	v_p	E' (10^3 kg cm^{-2})	E'' (10^3 kg cm^{-2})	$E(\text{exp})$ (10^3 kg cm^{-2})
Mureillo	318	0.393	31.6	0.588	144	50	165 ± 30
Sangron	274	0.420		0.580	133	50	135 ± 25
Girasol	322	0.233		0.761	98	40	75 ± 15
Saman	339	0.413		0.252	148	108	140 ± 30
S. de drago	275	0.473		0.432	143	65	130 ± 15
Apamate	250	0.420		0.530	120	55	105 ± 5

TABLE IX Experimental and calculated elastic moduli in compression for wood-MMA combinations.

Wood	E_w (10^3 kg cm^{-2})	v_w	E_p (10^3 kg cm^{-2})	v_p	E' (10^3 kg cm^{-2})	E'' (10^3 kg cm^{-2})	$E(\text{exp})$ (10^3 kg cm^{-2})
Mureillo	267	0.393	35.6	0.588	126	56	120 ± 20
Sangron	190	0.420		0.580	101	54	165 ± 20
Girasol	150	0.233		0.761	62	43	195 ± 15
Saman	278	0.413		0.252	124	116	65 ± 15
S. de drago	211	0.473		0.432	115	70	100 ± 15
Apamate	226	0.420		0.530	114	60	95 ± 20

where σ and v denote the stress in the individual phases and their volume fractions respectively. Since $\sigma_w = E_w e$ and $\sigma_p = E_p e$, it is readily seen that the elastic modulus of the combined material is given by

$$E' = v_w E_w + v_p E_p. \quad (7)$$

The other extreme assumption, i.e. that of equal stress in both phases, which would be applicable if both phases were perfectly bonded along the direction normal to the applied stress, implies that the total strain is given by

$$e = e_w v_w + e_p v_p = \sigma \left(\frac{v_w}{E_w} + \frac{v_p}{E_p} \right). \quad (8)$$

Consequently, the elastic modulus of the composite material may be expressed in terms of those of the constituents as

$$E'' = \left(\frac{v_w}{E_w} + \frac{v_p}{E_p} \right)^{-1}. \quad (9)$$

Since the mechanical properties of polymethylmethacrylate are well known, the experimentally measured moduli of wood-MMA combinations can be readily compared to those calculated by Equations 7 and 9. The values of E_w for the various woods were calculated by dividing the experimentally measured moduli of the wood samples by the volume fraction of "wood material", assuming a density of 1.5 g cm^{-3} for this material. For polymethylmethacrylate the elastic modulus in bending is $31.6 \times 10^3 \text{ kg cm}^{-2}$

and the elastic modulus in compression is $35.6 \times 10^3 \text{ kg cm}^{-2}$ [11].

The results obtained from bending and compression tests are compared to calculated moduli in Tables VIII and IX. As can be seen from these results, the elastic moduli in bending conform, fairly closely, with the results predicted by the equal strain hypothesis. This is in agreement with previously reported on basswood-MMA combinations prepared by catalyst-heat polymerization [12].

Of the measured elastic moduli in compression, on the other hand, three fall outside the range of the values predicted by the equal strain and equal stress hypotheses. In the case of saman, which shows a general deterioration of mechanical properties upon the introduction of plastic filler, the decrease of the elastic modulus to values below those of the unimpregnated wood may be attributable to some form of disruption of the wood structure. On the other hand, some positive synergistic effect appears to be at the root of the larger than expected increase in the elastic modulus in compression of polymethylmethacrylate-filled sangron and apamate. This result is indicative of the possibility that particular wood-polymer combinations may behave, in regard to at least some properties, as true composite materials.

4. Morphology of fracture surfaces

Detailed investigations of fracture of polymer-filled woods reported by other investigators [13]

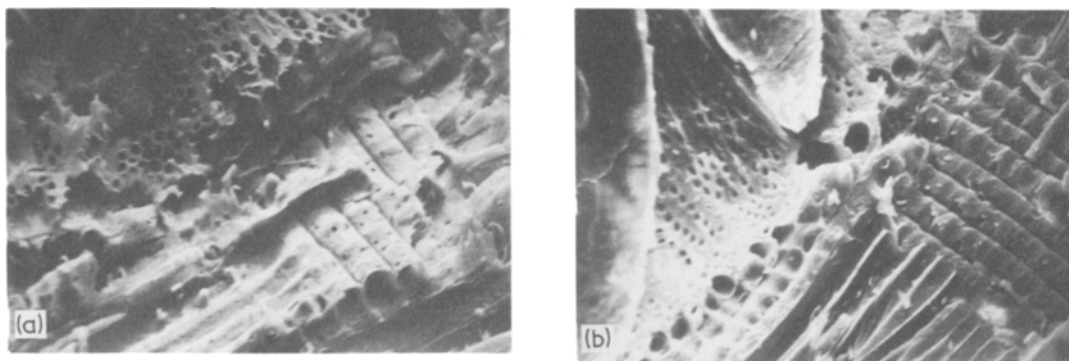


Figure 9 Areas of the fracture surface of sangre de drago: (a) natural wood, and (b) after introductions of ST-AN, $\times 55$.

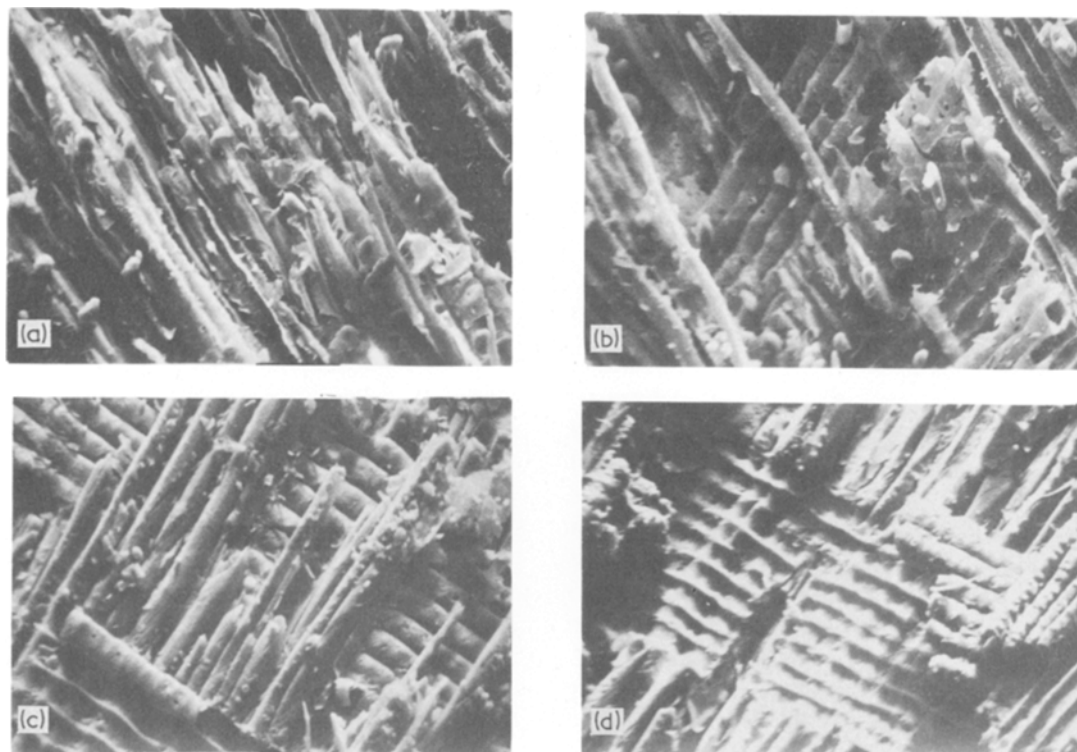


Figure 10 Typical surface morphologies of fracture of sangre de drago: (a, b) natural wood and (c, d) after introductions of ST-AN, $\times 55$.

show that under certain conditions the presence of polymer can alter dramatically the failure mode of the base material. The change in the fracture mode appears to be related to the degree of penetration of the cell walls and the properties (e.g. the glass transition temperature) of the polymer itself.

A qualitative examination of the fracture surfaces of the polymer-wood combinations and the corresponding control samples of untreated woods showed that portions of the wood-plastic material appear to be virtually unaffected by the

impregnation and polymerization process, as evidence by identical morphologies of the fracture surfaces, as is illustrated, for example, in the micrographs of Fig. 9. There are, of course, other regions in the wood-plastic material that show extensive penetration of polymer in the wood structure, but mechanical properties that depend on crack nucleation and propagation may be practically unaffected by this partial filling. The micrographs of Fig. 10 show the most typical morphologies of the fracture surfaces of the materials

studied. Although these observations are, of course, insufficient to draw any conclusions on the nature and extent of grafting between the polymer and the cell walls, at least there appears to be no significant change in the prevalent fracture mechanism of the treated and untreated materials.

5. Conclusions

The results obtained in this study show that the plastic impregnated woods obtained by present day techniques are not truly composites, in the sense that composite materials should have properties that are equal or superior to the sum of the properties of the constituent materials. The reasons for this, in the case of the materials studied here, appear to be the non-uniformity of penetration of plastic into the wood structure and, possibly, a weak bonding between the polymer the cellulose cell walls. Consequently, it would appear that methods leading to increasing the permeability of the woods to be treated, and to grafting of polymers onto cellulose might be the crucial improvements needed for making true wood-plastic composites.

Since the loss of toughness appears to be the main drawback in a comparison with higher density woods, it may also serve as the crucial parameter for the selection of particular plastic combinations. The reduced toughness of wood-plastic combinations is also a serious limitation as regards some conventional construction methods (i.e. nailing [14]). It is also evident that a detailed investigation of the fracture mechanisms in these materials might be useful for the formulation of better wood-plastic combinations.

The purpose of the study reported here was to evaluate the cost effectiveness of polymer impregnation of wood for obtaining general purpose materials of improved quality. Our results, combined with other previous studies, appear to indicate that such result may only be attainable on a very selective basis of wood-plastic combinations and processing methods. The main utility of wood combinations, as has been proposed by previous researchers, appears to be in special applications where lesser anisotropy, increased abrasion resistance and better dimensional stability justify the production costs of wood plastic combinations.

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