Mechanical and Physical Properties of Wood Fiber-Reinforced, Sulfur-Based Wood Composites

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ABSTRACT

Sulfur-based composite was made from sulfur impregnated, oven dried, wet-formed fiber mats. The fiber mats consisted of a 50/50 mixture of recycled newsprint pulp and mechanical hardwood pulp from several species made from chips in a laboratory refiner. The thickness of the composites was 0.125 inch and the specific gravity of the unimpregnated fiber mat was 0.2. The average MOR of the sulfur-based composites is in the order of the MOR of particleboard that contains more than three times as much fiber per unit volume (i.e., specific gravity of 0.65). The high MOE of the sulfur-based composites, nearly four times greater than that of particleboard, is most impressive. After the 24-hour soak test, the composites retained 64 percent of MOR and 28 percent of MOE. They swelled significantly less in thickness as compared to most wood composites and expanded significantly more in length. The only species used to make the composite that appeared to be consistently superior in mechanical properties was sweetgum. The lack of permeability of white oak might also have accounted for its good show in its wet MOR and MOE. A conceptual understanding of the high strength properties of the sulfur-based composites is presented.

INTRODUCTION

Over the years the abundant supplies of by-product sulfur available to world markets has stimulated considerable interest in the use of elemental sulfur as a component of wood composite or wood bonding systems. Some 80 years ago element sulphur impregnated wood was used for telegraph poles, railroad ties, and the like (3). It was shown that wood can absorb up to 100 percent of its dry weight in sulphur by immersing wood for several hours into liquid sulphur at 140 C - 1.50 C, yielding materials which are strong, durable, and resistant to fungi (5). However, sulphur impregnated wood was not widely accepted because the treated materials were brittle and it was cumbersome to prepare them. Likewise, the use of sulphur as a wood bonding agent (4,8) has not been widely established as the bond tends to be dense and brittle. Nevertheless, elemental sulphur was shown to be highly compatible with UF resin, and substantial quantities of UF resin can be replaced with elemental sulfur with little or no drop in mechanical strength of the panel products (7). The high mechanical strength properties of sulphur bonded wood products were attractive and led to the development of sulphur bonded composites which included numerous fiber species. For instance, kraft corrugating fiber mat dipped in molten sulfur until its weight doubled and the resulting impregnated paper was corrugated while hot (6); Corrugated roofing panels were made with elemental sulphur and agricultural residues (1); and structural sulphur-based building materials were made from kraft pulp screenings (2).

The purpose of this paper is to present experimental data to demonstrate that recycled fibers and fiber recovered from field-chipped, low-grade hardwoods can be made into exterior, structural, sulphur based composites with remarkable properties.

EXPERIMENTAL PROCEDURE

Design of the Experiment

The variables for the experiments were:

1. Species
   a. white oak
   b. red oak
   c. hickory
   d. sweetgum
   e. mixed species

2. Sulfur Impregnation Time
   a. 2 minutes
   b. 4 minutes
   c. 6 minutes

3. Coarseness of Pulp
   a. fine
   b. coarse

4. Three Replications

5. Fixed Conditions
   a. mat density--O.2 gm/cc
   b. mat thickness--O. 125 inches

Preparation of Whole Tree Chips

For each species, three trees were selected, felled and field chopped without debarking with a Swathe-Felling mobile chipper in Auburn, Alabama. The chips for each species were mixed thoroughly. Approximately 300 pounds were collected randomly from the mixed pile and shipped to Mississippi State University for defiberization.

Defiberization

After steaming at atmospheric pressure for approximately 40 minutes, the wood chips were converted to fiber in a laboratory refiner at the Mississippi State University, Forest Products Laboratory. Pulp was made at each of two plate settings and was labeled as "fine" and "coarse" pulp. In both cases, however, the pulps contain a fair amount of unseparated fiber bundles. The average freeness measured by using a Canadian Freeness Tester was 850 ml.

Mat Forming and Panel Fabrication

Because the pulp quality was not the type that would be used to make commercial insulation board or hardboard, recycled newsprint pulp was added to the slurry in order to make a fiber mat of the required density. A 50/50 mixture of repulped newsprint and hardwood pulp was used.

The recycled pulp was prepared by soaking shredded newspaper in water and then pulping this paper in about 30 seconds in a bench-top, Waring-type blender. This pulp was added to the hardwood mechanical pulp on an equal weight basis and adjusted to about one percent consistency before being poured into the sheet mold (i.e. decklebox) for the formation of the mats. The weight of the fiber in the slurry was calculated for the target thickness of 0.125 inches and air-dry specific gravity of 0.2 of the mats to be formed.

Fiber mats were made in a one-foot square decklebox equipped with a vacuum surge tank to speed water drainage. While still in the decklebox, the mats were pressed to remove as much water as possible by mechanical means with a vacuum assist by placing a board over the wet mat and then drawing a vacuum in the chamber beneath the mat.

The wet mats were formed on a removable screen which facilitated their transfer to a hydraulic press for removal of additional water. they were then placed in trays and put on a rack for air drying. With provision for good air circulation in the drying racks, an equilibrium moisture content of about 10 percent was reached in 24 hours under room temperature conditions.

Commercial flaked sulfur was heated in a steel pan on a thermostatically controlled hot plate set at 135" C (At that temperature sulfur has a watery viscosity). The dried mats were immersed in the molten sulfur for periods of 2, 4, and 6 minutes. They were turned over halfway through the immersion cycle to equalize the exposure of both surfaces to the radiant head differential at the bottom and the top of the pan and to permit the escape of air bubbles on the lower surfaces of the mats during impregnation.

To ensure uniform thickness and density and the removal of excess sulfur, the mats were placed in a cold, hydraulic platen press immediately after removal from the molten sulfur. They were pressed to stops at pressures of less than 50 psi. Press time was approximately one minute during which the sulfur solidified.

Sampling and Testing

All composite panels were conditioned at room temperature for 24 hours before being cut into 8 static bending specimens of 2 c 5 inches and one dimensional stability specimen of 2 x 11 1/2 inches. Specimens were conditioned for at least one month before testing.

For static bending tests, three specimens were tested in the dry state and three in the water-soaked condition. ASTM standards i.e. (D 1037-64) were used. To attain the water-soaked condition and to study the dimensional stability, specimens were subjected to a vacuum-pressure cycle which involved 30 minutes in vacuum when immersed in water followed by application of pressure of 60 psi for 24 hours. Weight, thickness, and length measurements were taken where appropriate in both tests.

RESULTS AND DISCUSSION

Average physical and mechanical properties of the sulfur-based composites are summarized in Table 1. Effects of wood species, pulp quality, and sulfur impregnation time were evaluated by analysis of variance at the 0.05 level of probability, and all differences discussed were significant at that level.

Bending Strength (MOR)

The average MOR values for the dry specimens tested ranged from 2050 psi for red oak board with fine pulp at 4 minutes sulfur impregnation to 3270 psi for sweetgum board with coarse pulp at 6 minutes impregnation (Table 1, column 4). The overall average MOR was 2680 psi. This is in the order of the MOR of particleboard that contains more than three times as much fiber per unit volume (i.e.

Table 1. Physical and Mechanical Properties of Sulfur-Based Composites.

Species	Sulfur Im-pregna-tion Time	MOR	MOE	LE	TS	MOR	MOE	Wet/Dry MOE	Wet/Dry MOE
	Min.	psi	1000 psi	-percent-		psi	1000 psi	-percent-	
Whiie oak 0.2-fine	2	2700	1415	0.98	7.2	2190	431	81	30
	4	2600	1423	1.07	8.1	2340	553	90	39
	6	2550	1362	1.08	8.9	2150	548	84	40
0.2 coarse	2	2300	1282	1.08	9.0	1540	347	67	27
	4	2480	1315	1.03	14.9	1630	332	66	25
	6	2540	1321	1.05	13.1	1680	330	66	25
Red Oak 0.2-fine	2	2190	1151	1.04	7.4	1440	380	66	33
	4	2050	1224	1.04	9.3	1500	402	73	33
	6	2230	1340	1.06	8.4	1600	427	72	32
0.2 coarse	2	2390	1234	0.88	10.5	1630	389	68	32
	4	2640	1290	0.98	11.2	1720	414	65	32
	6	2600	1395	0.95	11.3	1660	406	64	29
Hickory 0.2-fine	2	2500	1273	1.22	16.2	1570	380	68	32
	4	2490	1237	1.19	14.0	1740	432	54	29
	6	2630	1397	1.24	11.9	1630	410	61	32
0.2-coarse	2	2530	1251	1.02	15.8	1650	345	62	30
	4	2530	1215	1.14	14.0	1780	349	65	32
	6	2860	1283	1.16	12.4	1710	354	64	32
Sweet-gum 0.2-fine	2	2850	1348	0.95	11.2	1930	426	63	30
	4	2890	1393	0.97	14.3	1570	388	70	35
	6	3040	1459	1.03	12.0	1850	463	62	30
0.2-coarse	2	3050	1439	0.90	14.2	1890	431	65	28
	4	3090	1400	0.98	11.3	2010	450	70	29
	6	3270	1457	0.98	13.0	2090	460	60	28
Mixed Species 0.2-fine	2	2780	1424	1.13	13.8	1610	364	59	26
	4	2940	1309	1.19	13.2	1680	348	57	27
	6	2790	1361	1.14	13.7	1570	370	56	27
0.2-coarse	2	3100	1398	1.00	16.7	1660	311	54	22
	4	3040	1414	1.14	14.2	1720	353	57	25
	6	2870	1409	1.03	13.4	1660	386	59	27

Each value is the average ofthreetest specimens.

specific gravity of 0.65, ref.9). In the case of modulus of rupture, the major contributor to the strength value is the fiber fraction. As in all randomly oriented composition boards, the failure in bending is initiated on the tension face. Sulfur, a soft crystalline material with little or no tensile strength, contributes very little to the bending strength.

The dry MOR values differed significantly with species. The dry MOR values were highest for sweetgum, followed in decreasing order by mixed species, hickory, white oak and red oak.

The average MOR values for the water-soaked specimens ranged from 1440 psi for red oak board with fine pulp at 2 minutes sulfur impregnation to 2340 psi for white oak board with fine pulp at 4 minutes sulfur impregnation (Table 1, column 8).

Fiber quality interacted with species to affect bending strength. For fine fiber, the bending strength was highest for white oak, followed in decreasing order by sweetgum, hickory, mixed species, and red oak; while for coarse fiber, the bending strength was highest for sweetgum, followed in decreasing order by hickory, mixed species, red oak, and white oak. It was noted that the white oak panels resulted in highest bending strength with fine fiber and lowest bending strength with coarse fiber.

The wet MOR values are shown as a percentage of the dry MOR values of matched specimens from the same panel (Table 1, column 10). It was noted that the overall average wet MOR of 1750 psi was 64 percent of the average dry value. Under the best combination of species and impregnation time, the wet MOR was as high as 90 percent of the dry MOR. Such remarkably high wet values may be better understood if one remembers that the properties of wood-based fiberboards in flexure are generally determined by their tension properties, as mentioned earlier. Sulfur can contribute little or nothing to the tensile strength of the composite except to essentially eliminate the spring constant effects. Also, by filling the capillary spaces with sulfur, the fibers and fiber bundles are stressed more equally than in fiber composition board. In the latter, some bonded fibers are stressed to the point of failure, while others are simply being unkinked and are far short of failure.

Stiffness (MOE)

Average MOE values for dry specimens ranged from 1,151,000 psi for red oak board with fine pulp at 2 minutes sulfur impregnation to 1,457,000 psi for sweetgum boards with coarse pulp at 6 minutes sulfur impregnation (Table 1, column 5).

The high MOE of the sulfur-based composites is impressive in light of the fact that the specific gravity of the fiber mat was low (0.2) compared with the specific gravity of the wet process hardboard (about 1.0). The MOE of hardboard is in the order of 500,000 psi with nearly five times that amount of fiber content (9). This discrepancy can be explained on the basis that in composition boards, such as hardboard, the deflections (i.e. strain measurements) used to calculate MOE do not represent the true strain components of the fibers in the axial direction. Instead the measured strain reflects a type of spring constant rather than the MOE of the material from which the spring is made. The spring constant effect in fiber composition board is the result of the twisting under stress of the fibers at their bonded crossings and the unkinking or uncoiling of the twisted or curled fibers which must be stretched out in tension (or further compressed in compression) before pure strain can be developed in tension or compression along the axis of the randomly oriented fibers.

In the sulfur-impregnated composite the randomly-oriented fibers are rigidly fixed by the surrounding sulfur which has an inherent MOE of about 500,000 psi (2). Therefore, when stress is applied to the composite the fibers cannot twist, unkink, or buckle as long columns. And the strain that is measured in testing does, indeed, measure the weighted average strain components of the wood fiber axis in the stress direction as well as the strain in the sulfur itself as suggested by the Rule of Mixtures (2). In the property of modulus of elasticity, then, the difference between a fiber-reinforced composite and a fiber-based composition board is dramatically illustrated.

By analysis of variance, the dry MOE differed significantly with species. The dry MOE values were highest for sweetgum, followed in decreasing order by white oak, mixed species, hickory, and red oak.

There was little difference in MOE between 2- and 4- minutes sulfur impregnation, but the MOE increased slightly as impregnation time increased to 6 minutes. Average MOE values were:

Impregnation Time (min.)	MOE (1000 psi)
2	1321
4	1322
6	1381

On average, fine fiber resulted in slightly higher MOE as compared to that of coarse fiber.

Fiber quality interacted with species to affect the panel stiffness. For fine fiber, both white oak and sweetgum had same high MOE values, followed in decreasing order by mixed species, hickory, and red oak; while the MOE values were highest for sweetgum, followed in decreasing order by mixed species, red oak, white oak, and hickory, for coarse fiber.

Modulus of elasticity suffered more dramatically than MOR in the water-soaked specimens. The average value, of all conditions, was 399,000 psi which represents only 28 percent of the average dry value for matched specimens. However, under the best combination of conditions the wet MOE exceeded 500,000 psi which is within the range of the average MOE (dry) of untempered hardboard with four to five times more fiber per unit volume (9).

Fiber quality interacted also with species to affect the wet MOE. For the fine fiber, the wet MOE values were highest for white oak, followed in decreasing order by sweetgum, hickory, red oak, and mixed species. For the coarse fiber, the wet

MOE values were highest of sweetgum, followed in decreasing order by red oak, mixed species, hickory, and white oak. It is noted that the white oaks yield highest average wet MOE with fine fiber while lowest wet MOE with coarse fiber.

The only species that appeared to be consistently superior in the mechanical properties of the composite was sweetgum (Table 1). Since this was the only moderately dense, diffuse-porus hardwood included in the study, it is suggested that the reason for this behavior is the fact that it produced a more uniform mechanical pulp. This species contains fewer short-fibered parenchyma cells which cannot contribute to the MOR and the MOE of the composite and fewer large-diameter vessel segments which also detract from these two strength properties.

The well-known anatomical characteristics of white oak, which account for its lack of permeability to liquids, might also have accounted for its good showing, compared to sweetgum, in its wet MOR and MOE averages. Otherwise, it is not surprising that the major factor which accounts for the differences in the mechanical properties between species with similar anatomical characteristics (e.g. ring porosity), namely specific gravity, is no longer a factor in reconstituted wood fiberboards when these same species are compared at the same specific gravity.

Thickness Swelling

The average thickness swelling ranged from 7.2 to 16.7 percent. The coarse fiber panels swelled significantly more that those of fine fiber panels due to the stresses produced by the cross grain expansion of the large, densed fiber bundles.

The analysis of variance showed that the fiber quality interacted with species to affect thickness swelling. For fine fiber, the thickness swell was least with white oak, followed in increasing order by red oak, sweetgum, mixed species, and hickory; while the thickness swell for coarse fiber was least for red oak, followed in increasing order by white oak, sweetgum hickory, and mixed species.

Overall thickness swelling was 12.1 percent which is strikingly lower than most of the wood composites. Since the sulfur-based composite was made with the mat specific gravity of 0.2 and no hot pressing, there is no compression set to be released upon soaking. Hence, the low thickness swelling may reflect primarily the swelling of the cell wall rather than a combination of swelling plus relief of compression set.

Linear Expansion

The average linear expansion ranged from 0.88 to 1.24 percent. The coarse fiber panels were slightly more stable than those made with fine fiber.

The fiber quality interacted with species to affect the linear expansion. For fine fiber, the sweetgum panels were most stable; followed in decreasing order by white oak, red oak, mixed species, and hickory. For coarse fiber, on the other hand, red oak panels were most stable; followed in decreasing order by sweetgum, white oak, mixed species, and hickory.

CONCLUSIONS

The fiber fraction is the major contributor to the strength value of the sulfur-based composite because sulfur, a soft crystalline material with little or no tensile strength and low elastic modulus, contributes very little to their mechanical properties. However, by filling the capillary spaces with sulfur between fibers in a wet-formed fiber mat, the fiber bundles are stressed more equally, and the spring constant effect that detractes from the bending strength and stiffness of wood-based fiberboards is essentially eliminated. Accordingly, when low-density fiberboards are saturated with sulfur, the resulting fiber-reinforced composites reflect the very high tensile and elastic properties inherent in the cell wall of wood fibers. Thus, fiber-reinforced sulfur-based composites yield very high MOR and MOE values.

Since the sulfur-based composite are made with the mat specific gravity of about 0.2 with no hot pressing, there is no compression set to be released, and they show very low thickness swelling in the water soak test.

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