Grinding and classification of pine bark for use as plywood adhesive filler

THOMAS L. EBERHARDT
KAREN G. REED
Research Scientist
and Physical Science Technician
Southern Research Station
USDA Forest Service
2500 Shreveport Highway
Pineville, LA 71360 USA

ABSTRACT

Prior efforts to incorporate bark or bark extracts into composites have met with only limited success because of poor performance relative to existing products and/or economic barriers stemming from high levels of processing. We are currently investigating applications for southern yellow pine (SYP) bark that require intermediate levels of processing, one being the use of carefully ground and classified SYP bark as plywood adhesive fillers. Results can be used to select bark fractions having lower levels of ash. In addition, bark fractions rich in periderm tissue appear to perform better as plywood adhesive fillers relative to that prepared from whole bark.

INTRODUCTION

Since bark contains relatively high amounts of extractives, applications for those extractives have been sought to glean greater value from this biomass resource. For example, condensed tannins from SYP bark have been used to make adhesives for wood composites. Interest in this avenue of bark utilization has waned because of difficulties in competing with entrenched phenolic adhesive systems on both price and performance (1). Promising results have been obtained with substitutes for the more costly resorcinol-based adhesives, however, commercialization still faces barriers (2). An alternative to using bark chemicals for wood composites has been the pressing of bark together to make bark-based composites. Generally, the incorporation of bark results in lower strength relative to that for composite structures made with wood (3-5).

We are investigating applications for bark that require intermediate levels of processing. A few reports have suggested some potential for SYP bark as a filler for plywood adhesives (6). Concerns have been raised about the extractives interfering with the resin cure and high ash contents that would result in higher levels of tool wear (7, 8). The outer bark of SYP is non-living and comprised of obliterated phloem tissue partitioned by periderms (Figure 1). Through specific grinding and classification techniques, we can obtain fractions rich in either of these two tissues. Here we report our results demonstrating the utility of carefully ground and classified SYP bark fractions as plywood adhesive fillers.

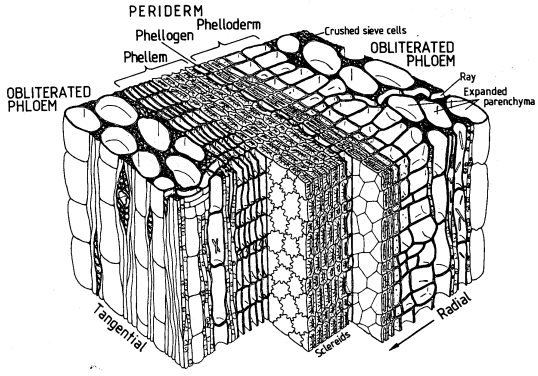

Figure 1. Periderm and obliterated phloem tissues in SYP outer bark (9)

MATERIALS AND METHODS

Bark Preparation

SYP bark (essentially all *Pinus taeda* L.) was collected near the debarking station at a local plywood plant. Samples of bark were ground as received with an electric chipper shredder (Echo, Inc., Model SH-5000) and dried under ambient conditions. Additional samples were sorted by hand to obtain samples of large (mostly 2-8 cm wide by 12-15 cm long) and small (mostly 0.4 cm^2 to 2 cm wide by 2-5 cm long) pieces. Samples of unsorted and sorted bark were shaken with a detergent solution (0.05% Triton X-100), rinsed with water, and dried under ambient conditions. All bark samples were subsequently ground in a Wiley mill (10 mesh grinding screen), sealed in plastic bags, and stored in a freezer. Ash contents for the bark samples were determined in a muffle furnace set to 450 ºC.

Bark Fillers

Previously ground bark was ground further in 100 g batches using a blender (Waring Laboratory, Model 36BL23). Bark was ground at high speed for two 1-minute periods, between which, the canister was removed and briefly shaken by hand. The blender-ground bark was then classified for 30 minutes on a sieve shaker (W.S. Tyler, Ro-Tap, Model RX-29) equipped with 20, 35, 80,

100, 140, and 200 mesh sieves. Materials retained on each screen and in the bottom pan were pooled from repeated operations to obtain enough material at each particle size range for evaluation.

The material passing the 200 mesh sieve was observed by light microscopy and appropriately designated as the obliterated phloem filler. Materials retained on the 35 and 80 mesh screens were rich in periderm tissue; these were ground further in small batches (25 g) using an ultra-centrifugal grinding mill (Retsch, Inc., Model ZM 200) equipped with a 12-tooth rotor and 0.12 mm ring sieve. The finely ground material was then classified as above. Most of the material passed the 200 mesh screen and was designated as the periderm filler. Finally, the whole bark filler was prepared by grinding the coarsely ground bark directly in the ultra-centrifugal grinding mill and classifying it to obtain those materials passing a 200 mesh screen.

Adhesive Preparation

Adhesive mixes were prepared as shown in Table 1 using the 3 bark-derived fillers and a furfural residue control (FuraTex, Bates & Co., Inc.). The filler was mixed with the added water followed by the addition of the extender (HW-200, Bates & Co, Inc.). As necessary, up to 200 g resin (6500B, Borden Chemical) was added to obtain a suitable consistency for working the extender gluten. Additional resin was then added to adjust the mix viscosity; the total amount of resin added before the addition of the caustic (50% NaOH) was 350-400 g. After the caustic addition and mixing (15 min.), the remainder of the resin was added to obtain the final adhesive mix. Further mixing (5 min.) was followed by the determination of the mix viscosity (Brookfield).

Table 1
Adhesive mix for bonding SYP plywood

	Mix Solution (%)	Mix Solids (%)
Filler	6.7	15.0
Extender	7.4	16.6
Sodium Hydroxide Solids	1.6	3.6
Resin	28.9	64.8
Total Mix Solids	44.6	100
Total Mix Water	55.4	
Total Mix	100	

Plywood Assembly and Testing

SYP wood veneers (305 mm x 305 mm x 3.175 mm) were sorted to remove those that had unacceptable defects (e.g., rough veneer, staining). A roll spreader (Black Bros. Inc.) was used to apply the adhesive mix to core veneers at a rate of 366 g/m^2, double glueline basis. Panels were immediately assembled in sets of 4 to give 10, 20, 40 and 60 minute assembly times. After 5 minutes into the assembly process, all panels were pre-pressed (690 kP, 5 min.) with an air pod press (Tyler Manufacturing Co.). The panels with a 10 minute assembly time were transferred as quickly as possible to the hot press (Williams-White Co.) for pressing (157 ºC, 1240 kP, 3 min.). Finished panels were stored in a hot box overnight. For each experiment, panels were prepared in duplicate for each filler and assembly time.

Plywood panels were subsequently cut to afford 10 test specimens each for testing with the lathe checks in the closed and open configurations. Samples were tested using the standard shear test following the standard vacuum/pressure pretreatment (10). After drying in an oven (75 ºC), wood failures were estimated.

RESULTS AND DISCUSSION

Plywood fillers are relatively inert materials added to the adhesive mix to improve its workability. Specifically, these finely ground organic and/or inorganic materials promote good bond formation by facilitating the retention of the adhesive mix on the veneer surface where it is needed for bonding (11). Aside from good performance, desirable features for filler include low cost, consistent quality, and sufficient supplies.

Commonly used plywood adhesive fillers include furfural residue, alder bark, and nutshell flours. Elimination of domestic supplies of furfural residue, and the demand for nutshell flours by other industry sectors, has revived interest in finding alternatives. Issues raised about the use of finely ground SYP bark include high ash contents and potential interference of the extractives with the resin cure. Regarding the latter, research has shown that grinding and classifying bark can afford different cell types (e.g., parenchyma, stone cells) concomitant with different levels of extractives (12, 13). SYP bark from an industrial source was used for our study since it provided a more realistic representation of currently available bark resources.

Reported ash contents for *P. taeda* bark are slightly less than 1 % (14, 15). In addition to this ash, an appreciable amount of grit

typically accompanies industrial supplies of bark and thereby contributes significantly to the value for ash content. Analysis of the SYP bark we obtained gave an ash content that was about 7-fold higher (Table 2). Through a simple washing procedure, we were able to reduce the ash content by one half. A greater reduction in ash content was achieved by simply selecting the larger pieces of bark for grinding. Accordingly, given available supplies of industrial bark, simple onsite screening operations could provide material containing considerably less ash. Washing of the smaller bark pieces might allow greater consumption of the available bark supplies, however, would be impractical given level of grit removal relative to the time and energy needed for both water processing and bark drying.

Table 2
Ash contents for bark samples before and after washing with a detergent solution

	Ash Content (%)[a]	
Bark Sample	Unwashed	Washed
Whole Bark	6.9	3.2
Whole Bark, Large Pieces[b]	1.7	1.6
Whole Bark, Small Pieces[c]	17.6	6.6

a) Percent of oven-dry bark.
b) Fragments were mostly 2-8 cm wide by 12-15 cm long.
c) Fragments were mostly 0.4 cm^2 to 2 cm wide by 2-5 inches long.

Prior research has shown some success with the use of finely ground SYP bark as an adhesive filler for plywood manufacturing (6). In some instances, a lack of success reflected the inability to obtain a material that was sufficiently ground. Unlike reports on other alternative fillers (16, 17), our attempts to obtain sufficiently ground material with a Wiley mill were unsuccessful even with new knives and careful knife adjustment. An acceptable product was subsequently obtained using an ultra-centrifugal mill, albeit in small batches (25 g).

Plywood adhesive mixes were first prepared with the whole bark filler and the furfural residue control. The viscosity for the adhesive mix containing the whole bark filler was 7100 mPa·s after preparation and 13,000 mPa·s after 24 hours at ambient temperature; for the furfural residue control, the viscosity rose from 8800 mPa·s to 20,000 mPa·s. Three-ply plywood panels were then manufactured using 4 assembly times to assess the performance of the bark-based filler. Results from the vacuum/pressure testing of the test specimens showed average wood failures ranging from 62 to 85% for the whole bark filler (Table 3). The highest value for wood failure, coincided with the lowest value for shear strength. This most likely reflects core veneers that are weak relative to the adhesive bonds (8). Despite the fact that most of the wood failures were below the target of 85%, the results demonstrate that under the conditions employed, a whole bark filler with minimal processing performs nearly the same as an established control filler. Detracting from this observation is the acknowledgement that this whole bark filler still contained high levels of ash.

Table 3
Wood failures and shear strengths for plywood made with whole bark and control fillers

	Filler	Assembly Time (Min.)			
		10	20	40	60
Wood Failure (%)	Whole Bark	72	85	75	62
	Control	74	77	82	84
Shear Strength (kP)	Whole Bark	1770	1390	1660	1740
	Control	1840	1720	1880	1990

In our next experiment, we addressed concerns related to the potential interference of bark extractives with the adhesive mix cure. Upon further grinding of the bark with a laboratory blender, we obtained a bimodal distribution of particles with about 40% of the mass in particles smaller than 35 mesh and larger than 80 mesh; about 35% of the mass was in particles that passed through the 200 mesh screen. Another cycle of grinding for fractions coarser than 80 mesh resulted in little additional material passing through this mesh size. Observations of these fractions under a light microscope showed that our coarser material was rich in periderm tissues as evidenced by clumps of spiculate sclereids (Figure 1). In the case of the fine material, observations showed cellular debris with cell wall thicknesses consistent with obliterated phloem tissue. The coarse fractions (i.e., materials retained on 35 and 80 mesh screens) were combined and ground further in an ultra-centrifugal mill. The resultant finely ground material was then classified to obtain a

material suitable for use as a plywood filler. The material that passed the 200 mesh screen after grinding with the blender was suitable for direct use as a filler, and therefore, it received no further processing.

Plywood adhesive mixes were prepared as before with the two bark-based fillers (i.e., periderm, obliterated phloem) and the furfural residue control filler. The viscosities of the mixes prepared with the periderm, obliterated phloem, and control fillers were 16,000, 10,000, and 4,500 mPa·s, respectively. After 24 hours at ambient temperature, the viscosity for the control filler showed the highest relative increase in viscosity to 15,000 mPa·s. The viscosities for the adhesive mixes with the periderm and obliterated phloem fillers were 20,000 and 18,000 mPa·s, respectively.

Three-ply panels were assembled shortly after these adhesive mixes were prepared. Results from the vacuum/pressure testing are shown in Table 4. Again, we found that the wood failure for the control was below the target of 85%. Although, the control adhesive mix viscosity was at least one half that for the other mixes and our target value (10,000 mPa·s), no problems were encountered during adhesive mix application. For the plywood panels assembled up to the point, the veneers were used as received. The average moisture content of the veneers was determined to be 7.8% on an oven-dry basis for selected veneers. The art of making good plywood involves a balance between multiple interrelated processing parameters. We speculated that the low wood failures resulted from being on the high end of the range for acceptable veneer moisture content (3-8%).

Table 4
Wood failures and shear strengths for plywood made with periderm, obliterated phloem, and control fillers

	Filler	Assembly Time (Min.)			
		10	20	40	60
Wood Failure (%)	Periderm	78	89	88	87
	Oblit. Phloem	36	38	58	72
	Control	64	75	64	74
Shear Strength (kP)	Periderm	2070	1550	2080	1590
	Oblit. Phloem	1580	1860	1740	1730
	Control	1660	1800	1990	1720

Of particular interest from this experiment was the stark contrast in the wood failures for the periderm and obliterated phloem fillers. Acceptable wood failures were obtained for the periderm filler at assembly times of 20, 40, and 60 minutes. Very low wood failures were observed for panels prepared with the obliterated phloem filler; these samples showed significant amounts of undercure. Despite obvious bond failure during the shear testing of the panels with the obliterated phloem filler, no trend was apparent in the shear values between any of the fillers or assembly times.

Given concerns about the relatively high veneer moisture content could have contributed to the observed undercure, another experiment was conducted to compare all fillers together with dried veneers. Veneers were placed in racks and dried in an oven at 70 ºC. The moisture content of the veneers was subsequently determined to be 2.3% on the basis of the oven-dry weight. Exposure to ambient conditions prior to plywood assembly resulted in a slight increase in moisture content to 3.9%.

Adhesive mixes were prepared as outlined. The viscosities were close to the target (10,000 mPa·s) and ranged from 8000 to 12,000 mPa·s. The viscosities were also measured after 24 hours under ambient conditions and ranged from 12,000 to 17,000 mPa·s. Plywood panels were prepared with the aged adhesive (24 hours) as before. During panel assembly, it was observed that the dried veneers showed greater rigidity than those used before. Pre-press tack appeared to deteriorate with time as evidenced by partial separations for panels with longer assembly times. To address concerns that the apparently over-dried veneers would impart lower bond quality, an additional control was included with veneers acquired from the plywood mill that same day. Although these veneers also had a low moisture content (2.8% average), they were not nearly as rigid and therefore gave a better pre-press.

Results from the vacuum/pressure testing of these plywood panels are shown in Table 5. By drying the veneers, we were successful in reducing the occurrence of undercure for the panels prepared with the obliterated phloem filler. Marginally higher levels of wood failure for the shorter assembly times indicated that the very poor performance of this filler could be attributed to excessive moisture for the adhesive mix coupled with the conditions employed. Especially interesting was that the periderm filler wood failures were generally higher than those for the obliterated phloem and whole bark fillers. This result validated the differences in the performance we

observed for our bark-based fillers during our initial experiments.

Table 5
Wood failures and shear strengths for plywood made with whole bark, periderm, obliterated phloem, and control fillers

	Filler	Assembly Time (Min.)			
		10	20	40	60
Wood Failure (%)	Whole Bark	50	72	73	67
	Periderm	63	78	72	73
	Oblit. Phloem	58	64	64	66
	Control 1	68	82	76	74
	Control 2	88	91	76	80
Shear Strength (kP)	Whole Bark	1230	1430	1320	1450
	Periderm	1760	1710	1650	1410
	Oblit. Phloem	1370	1380	1210	1290
	Control 1	1360	1340	1460	1320
	Control 2	1340	1240	1670	1460

Wood failure values for the panels prepared with the control filler were marginally higher than those for any of the bark fillers. Our concerns about over drying our veneers were validated by the acceptable wood failure values (>85 %) for our second control. Similar wood failures were observed for the periderm and control fillers for panels prepared under the same conditions. Higher shear strengths for the periderm filler may suggest that the core veneers were stronger and therefore afforded wood failures closer to the controls.

CONCLUSIONS

Experimental results demonstrate that the high levels of ash in commercial supplies of SYP bark can be greatly reduced by simple screening operations to remove high levels of grit accompanying the smaller bark fragments. Grinding and classifying SYP bark affords a filler rich in periderm tissue that performs at least as well as a furfural residue control. Moreover, problems with the poor performance of unclassified SYP bark likely results from components in the obliterated phloem that interfere with the resin cure.

FUTURE WORK

Additional plywood panels will be assembled under conditions that will provide acceptable controls thereby allowing a better assessment of the performance of our bark-based fillers. Characterization of the bark-based fillers will be pursued to determine the cause(s) for the different levels of filler performance.

REFERENCES

1. Kreibich, R.E. – 457-478 **Chemistry and significance of condensed tannins** (Hemingway, R.W., Karchesy, J.J, eds): Plenum Press, NY (1989)
2. Hemingway, R.W. – 80-83 **Wood residues into revenue: residual wood conference proceedings**, Richmond, BC, Canada, Nov. 4 & 5 (1997)
3. Blanchet, P., Cloutier, A., Riedl, B. – *Wood Science and Technology* **34**:11(2000)
4. Chow, S. – *Forest Prod. J.* **25**(11):32(1975)
5. Muszynski, Z., McNatt, J.D. – *Forest Prod. J.* **34**(1):28(1984)
6. Sellers, T., Jr. – *Forest Prod. J.* **44**(6):33(1994)
7. Sellers, T., Jr. – *Forest Prod. J.* **39**(4):39(1989)
8. Sellers, T., Jr., Gardner, D.J. – *Forest Prod. J.* **39**(3):34(1989)
9. Howard, E.T. – *Wood Sci.* **3**(3):134(1971)
10. NIST - 40 pp. **Voluntary product standard PS-1**: National Inst. of Standards and Technology, MD (1996)
11. Sellers, T., Jr. – 471-493 **Plywood and adhesive technology**: Marcel Dekker, NY (1985)
12. Ottone, S.P., Baldwin, R.C. – *Wood and Fiber* **13**(2):74(1981)
13. Ross, W.D., Krahmer, R.L. – *Wood and Fiber* **3**(1):35(1971)
14. Labosky, P. – *Wood Sci.* **12**(2):80(1979)
15. McGinnis, G.D., Parikh, S. – *Wood Sci.* **7**(4):295(1975)
16. Oh, Y., Sellers, T., Jr. – *Forest Prod. J.* **49**(3):61(1999)
17. Oh, Y., Sellers, T., Jr., Kim, M.G., Strickland, R.C. – *Forest Products J.* **47**(10):107(1997)

ACKNOWLEDGEMENTS

This study was made possible through the detailed information and guidance provided by David Perry, Bates & Co., Inc. Dale Huntsberry assisted with plywood preparation and testing. Gary Smith assisted by collecting and processing the bark. The authors especially appreciate the gifts of bark, veneers, and adhesive provided by local plywood producers, Boise Cascade Corporation and Hunt Forest Products, Inc.