Evaluation of coupling agents to manufacture hybrid hardboard made from industrial waste fiberglass and wood fiber

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Abstract

Every day, tons of fibrous material are landfilled that could otherwise be used for structural panel products. In this study, we looked at combining fibers from industrial fiberglass insulation trim waste with commercial hardboard fibers and with recycled corrugated container fibers to improve the properties of a structural hardboard-like panel. This study also investigated the effectiveness of two coupling agents at enhancing fiber-to-fiber bonding between fiberglass and wood fibers. Processing characteristics, physical properties, and mechanical properties of the wet-formed mats and finished panels were measured to determine the effects of the following three factors: 1) fiberglass loading level (0%, 7.5%, and 15%); 2) coupling agent; and 3) wood fiber type (steam-exploded hardboard or recycled corrugated). Also, panel strength and linear expansion properties of the experimental boards were compared with commercial hardboard. As applied, the coupling agents did not improve the fiberglass-to-wood-fiber bond. All panels exceeded minimum strength and dimensional requirements for hardboard; however, mechanical properties decreased as fiberglass loading level increased. The addition of fiberglass improved dimensional stability for all panels and improved drainage rate for panels made with recycled corrugated containers. Panels made from recycled corrugated container fibers were significantly stronger than those made from commercial hardboard fibers. Industrial waste fiberglass can be incorporated with either wood fiber (commercial hardboard or recycled corrugated container) and still exceed minimum dimension and strength requirements for hardboard.

In the United States, millions of tons of useful fibrous materials are landfilled that could otherwise be used for structural panel products. One of these materials is fiberglass (FG) fiber derived from insulation manufacturing facilities where thousands of tons of trim waste are generated per plant per year (Hart 1995). Insulation FG is bonded and coated with phenolic resin, and in most cases, this trim material has been processed, making it too costly to recycle, so it is landfilled. Paper and paperboard, another fibrous material, represent a large portion of the recycling waste stream. In 1995, the U.S. Environmental Protection Agency (EPA 1997) estimated there were 74 million metric tons classified as paper and paperboard in the municipal solid waste stream. Approximately 40 percent of this material was recycled, yet nearly 44 million metric tons were still landfilled. There may be an opportunity to use these waste fibrous materials for industrial structural panel products. Panel products do not have the same strict requirements for fiber cleanliness as paper products, but there are stringent requirements for structural performance and dimensional stability. We need to

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find more uses for both of these waste fibers because doing so would save some of our natural resources while at the same time reducing landfill pressures.

Background

This report is the second of two that investigated the use of recycled materials in panel products such as hardboard (HB). In the first report (Hunt and Vick 1999), comparisons were made between properties of boards made with commercial HB fibers and boards made with old corrugated container (OCC) fibers. The results showed that panels made from OCC fibers were three times stronger and two times stiffer than those made from HB fibers. The OCC panels were five times stronger than the minimum American Hardboard Association (AHA) standard (AHA 1995) for standard HB. Panels from both fiber types had linear expansion values less than the AHA siding standard. The effects of fiber forming on drainage rate and mat thickness were also presented. This second report looks at the effects of FG addition to boards made with one of two fiber types and the effectiveness of coupling agents at enhancing the bond between FG and the HB or OCC fibers. We expect that if the bond between FG and wood fiber could be improved, the panels produced would be stronger, stiffer, and more dimensionally stable. Engineered fiberboard products or structures could then be made for more structurally and environmentally demanding applications.

Fiberglass addition

Research efforts to use FG in combination with wood fibers in structural panel products are not new. Other researchers have found that adding FG improved performance in some areas and reduced performance in others. Paper made with increasing percentages (up to 25%) of two types of waste FG showed only marginal strength improvements up to 5 percent FG and decreasing strength with additional FG (Hart 1995). The main benefit was a 42 percent increase in drainage rate with 25 percent FG. Other research (Cavlin and Back 1968) investigated the effects of chopped FG on the dimensional stability of fiberboard with densities ranging from 200 to 1,000 kg/m^3. They found dimensional change was reduced by 40 percent in low-density boards (200 to 600 kg/m^3, with 10% FG). Two other studies (Nishikawa et al. 1974, 1975) investigated the effects of chopped FG on fiberboard strength properties, dimensional stability, and drainage time for panels at densities of 650 and 1,000 kg/m^3. In the first study, they found FG decreased drainage time by 30 and 41 percent at 10 and 50 percent FG loadings, respectively. In the second study, linear expansion decreased from 0.28 to 0.17 percent when 20 percent FG was added to the high-density panel. Bending modulus of rupture (MOR) and modulus of elasticity (MOE) also decreased from 59.8 to 47.0 MPa and from 5.1 to 4.6 GPa, respectively, when FG was added. However, in these studies, no attempts were made to enhance the bond between the FG and wood fibers and all the FG fibers were from virgin fiber sources without phenol-formaldehyde (PF) resin coating.

Coupling agents

Good fiber-to-fiber adhesion is key to improved mechanical performance of a composite. For conventional wet-formed HB, adhesion between the wood fibers occurs primarily through hydrogen bonding, hemicellulose, and lignin flow, supplemented by low concentrations of resin precipitated onto the fiber surface. Fiberglass, on the other hand, has low surface reactivity and requires an adhesive to bond fibers together. Phenolic adhesives do not adhere well to FG, so a coupling agent is used to provide an effective chemical link between the adhesive and the FG. Applying a coupling agent is normal practice when bonding virgin FG in nonwood composites. With post-cured FG trim waste, however, the coupling agent is already coupled with a PF resin. The outside surface, which is now the cured PF resin surface, also has low surface energy, and a coupling agent is needed to enhance fiber-to-fiber bonding with the PF resin. A coupling agent then needs to be applied to provide a reactive surface for bonding to wood fibers. Aminosilane coupling agents are capable of reacting with FG surfaces and PF resins. Hydroxymethylated resorcinol (HMR) coupling agent is capable of reacting with wood fiber, PF resin, and perhaps FG.

Organofunctional silane. — The silane coupling agent, gama-aminopropyl triethoxysilane, has proven itself in many industrial applications for its capability to couple phenolic resins to FG (Marsden 1990). Silquest A-1100 (Osi Specialties, Inc.–Crompton Corp., Philadelphia, Pennsylvania) was recommended for this study.

The surface of the cured phenolic coating on the waste FG is physicochemically different from the original FG surface. These coated surfaces also differ in wettability. Some areas of the phenolic-coated fibers could be restored to their original mineral surfaces by abrasion, causing the aminosilane and resins to more effectively wet and perhaps bond to the waste FG surfaces. Aminosilane coupling agents are generally bonded to mineral surfaces at elevated temperatures. In this study, the coupling agent was applied at room temperature in the wet-slurry process water.

Hydroxymethylated resorcinol. — The HMR coupling agent, patented by the USDA Forest Service, Forest Products Laboratory (FPL) (Vick et al. 1996), is used to enhance adhesion of all thermosetting wood adhesives with wood. HMR, similar to silane, is normally pre-dried after application for maximum reactivity on the surface.

Hardboard panels made from recycled corrugated containers

Wet-formed HB made from recycled paper waste is not new. Steinmetz (1974) investigated using wax-coated OCC as a partial or total replacement for HB fibers. He formed a series of panels with increasing percentages of OCC without phenolic resin. He found that adding 15 percent OCC did not significantly change properties, and subsequent addition up to 100 percent OCC only increased the bending strength by 10 percent, stiffness by 4 percent, and dimensional stability by 63 percent, although it decreased drainage rate by 109 percent. Kruse (1995) reported similar results from a commercial trial where 20 percent OCC was added to HB fibers. He found that the MOR was the same as for HB panels. This led Kruse to conclude that although adding recycled fibers to the manufacturing system did require some adjustments to the process equipment, these were minor and should cause no problems if implemented. Both Steinmetz and Kruse noted that when recycled paper fibers were used, the drainage rates were slow compared with that for conventional HB fibers.

Yao (1978) made HB panels from 100 percent recycled shredded paper from

Table 1. — A full 2^3 factorial experimental design with one midpoint was used in this study.

Fiberglass type	Experimental design factors		
	Coupling agent	Wood fiber furnish	Fiberglass addition (%)
Pink	Water and silane	HB and OCC	0 and 15, midpoint 7.5
Yellow	Water and HMR	HB and OCC	0 and 15, midpoint 7.5

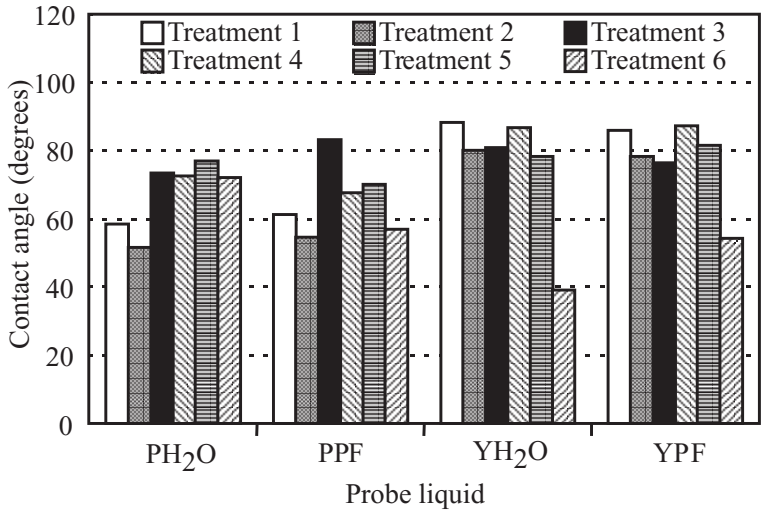

Figure 1. — Wilhelmy fiber contact angle measurements for pink (P) and yellow (Y) fiberglass in water (H_2O) and PF probe liquids using six treatment methods: 1) dry, as is; 2) 3 days in water; 3) 30 minutes in silane solution, then fibers air-dried; 4) 30 minutes in silane solution; 5) 30 minutes in silane solution, then 3-day soak; and 6) 30 minutes in HMR solution.

municipal waste. He investigated the effects of binder type, heat treatment, wax treatment, and one or two screens on the mechanical properties of HB panels. He found that HB panels from recycled household trash could meet or exceed the minimal commercial requirements for tensile strength, MOR, internal bond, and thickness swell. He also found that properties significantly improved if two screens on either side of the panel were used during drying.

Experimental

There were two specific objectives for this study. The first was to investigate the benefits of using a coupling agent on waste FG to enhance tensile properties and dimensional stability of a high-density FG-reinforced HB composite panel product. The second was to investigate the use of fibers derived from OCC in composite panels with FG incorporated to enhance drainage rates. A full 2^3 factorial experimental design for each of two FG types, pink and yellow, was used for this study (**Table 1**). There were 32 panels for each FG type, 3 replicates, and 8 midpoints for 3 factors. The factors were coupling agents (silane or water with pink FG and HMR or water with yellow FG), wood fiber furnish (HB or OCC fibers), and FG addition (0% or 15% by weight and with a midpoint of 7.5%).

Fiber furnishes

Hardboard. — Commercial wet-processed HB fiber was obtained from Georgia Pacific (Duluth, Minnesota). The fiber mix was 90 percent hardwood and 10 percent softwood fiber. No additional processing was done to the HB fiber for this study.

Old corrugated cardboard. — Shredded OCC material was hydropulped 20 minutes at 10 percent consistency in 40°C water. The material was then passed through an atmospheric refiner with a gap set at 0.26 mm (0.010 in.) to break up small fiber bundles. The Canadian Standard Freeness (CSF) was 623, and the length-weighted average fiber length was 2.09 mm (0.082 in.).

Fiberglass. — Two types of trim waste FG were provided from a commercial FG insulation manufacturer. Pink FG fibers were 5 to 25 mm long (0.2 to 1.0 in.), loosely matted together in bundles, and coated with approximately 6 percent phenolic resin. Yellow FG fibers were only 2 to 12 mm (0.08 to 0.50 in.) long, in loosely matted bundles, and coated with 12 percent phenolic resin. Preliminary HB panels made with the pink and yellow FG as received had small FG balls on the panel surface. These were visually and structurally unacceptable. Our desire was to have a uniform dispersion of FG throughout the panel. For both FG types, bundles were broken up by passing a water-slurry of FG through a 20.3-cm- (8-in.-) diameter disk-refiner with the gap set at 0.38 mm (0.015 in.). Running the FG through the refiner also shortened the FG fiber length to approximately 5 mm for both FG types.

Wettability of phenolic-coated fiberglass surfaces

Wilhelmy dynamic contact angle analysis is an accepted measure of wettability of individual fibers (Young 1986; Gardner et al. 1991,1996; Saver 1992). We used the same method where individual fibers were prepared as probes and immersed in either deionized water or a 0.1 percent aqueous solution of PF resin. Contact angles were determined for both types of FG fibers with six surface treatments in two probe liquids. A lower contact angle indicates a higher surface energy and generally a greater potential for bonding. The FG treatments were as follows: 1) dry (as received from the manufacturer); 2) 3 days in water; 3) 30 minutes in silane solution, then air-dried; 4) 30 minutes in silane solution; 5) 30 minutes in silane solution, then 3-day water soak; and 6) 30 minutes in HMR solution. Treatment 4 was selected for the pink FG fibers and treatment 6 for the yellow FG fibers (**Fig. 1**). All of the above wettability measurements were conducted on FG that had not passed through the wet-refiner. Abrading and removing some portion of the cured PF resin coating may have significantly changed the surface energy and increased the wettability of

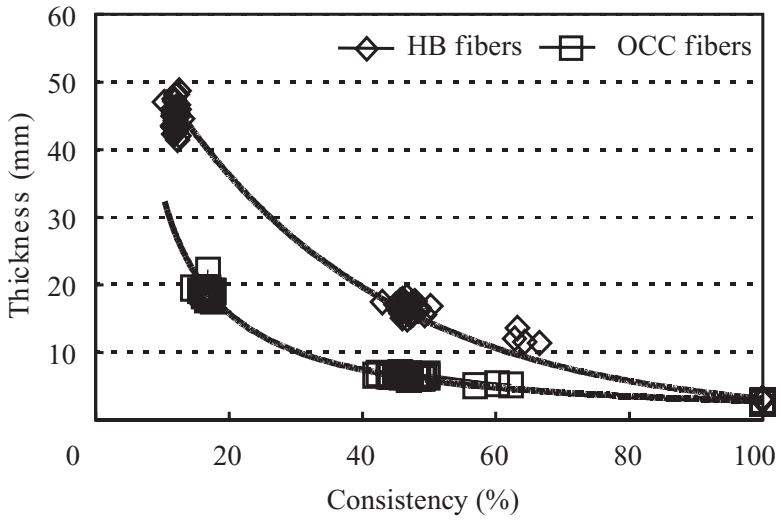

Figure 2. — Panel thickness compared with panel consistency for HB and OCC fiber.

fibers by water and the coupling agent treatments.

Treatment of fiberglass with coupling agents

The following procedures were used to treat the FG with the coupling agents. For silane treatment, a 0.1 percent solution was prepared and added to the FG fibers 30 minutes prior to forming the panels. For panels made without FG but with silane, the silane was added to the HB or OCC fibers instead.

For HMR treatment, FG fibers were placed in 325 mL of 2.3 percent HMR solution for 30 minutes. For those panels formed without FG but with HMR, 325 mL of solution was added into the mixing tank. We assumed complete retention of HMR on the fibers and no discharge with the process water. We did not measure the exact amount of HMR retained in the panels.

Panel processing

For each panel, either HB or OCC fibers were dispersed in water to a consistency of 0.83 percent. The prescribed amount of FG was added. PF resin, on a 1.5 percent dry-weight basis, was added and mixed in with the slurry. The mixed fiber slurry was poured into a 51- by 51-cm (20- by 20-in.) forming box with water added to bring the consistency to 0.72 percent. The drain time, formed wet-thickness, and weight were recorded for each mat. The target weight was 714 g and target thickness was 3 mm (0.125 in.).

After forming, the mats were wet-pressed between screens at 1.72 MPa (250 psi) for 1 minute. After pressing, thickness and weight data were recorded again.

The wet-pressed mats were placed between screens and hot-pressed at 1.72 MPa (250 psi), 170°C (340°F) for 10 minutes to ensure ovendry panel conditions. The dry panel thickness and weight were measured.

Testing procedures

All panels were conditioned at 22°C (72°F) and 50 percent RH for 4 days before testing. Stress wave times across the diagonal of each panel were measured using a stress wave timer, Model 239A (Metriguard, Pullman, Washington). Specimens for tensile and linear expansion tests were then cut from each panel and evaluated according to the ASTM D 1037 test method (ASTM 1996).

Results and discussion

Before and after refining, we observed that the pink fibers were clearly more hydrophilic than the yellow, but refining greatly improved the wetting and dispersion of both FG fibers when they were added to the wood fiber slurries prior to forming. We also observed that forming consistency and agitation kept the FG in suspension and prevented it from settling to the bottom of the mixing tank or forming box. Uniform distribution of all the fibers is essential for uniform property development within HB.

Drainage rate

Wood fiber type and FG addition had a significant effect on drainage. The HB fiber had a faster drainage rate at 2.2 cm/sec. compared with the OCC fiber at 0.64 cm/sec. The faster drainage rate of the HB was caused by the stiffer HB fibers, which resulted in a more porous mat structure. This is evident from differences in mat thickness (**Fig. 2**). The HB fibers also have less fibrillated surfaces, so they are less restrictive to water flow than the OCC fibers.

The effects of FG addition on drainage rates were statistically significant with OCC fibers but not with HB fibers. (References to significance indicate statistically significant at $\alpha < 0.05$ level.) With 15 percent addition of FG fibers, drainage rate for OCC fibers increased to 1.0 cm/sec., a 58 percent rate increase. This was one of the desired outcomes and reasons for using FG. Although this was a significant rate increase, it was still only about half the rate of HB fibers. We believe that the stiffer FG fibers provided larger voids for increased water flow. The HB fibers are already stiff and produce an open, porous mat, and the addition of stiff FG fibers did not significantly improve drainage rates. The FG fibers are also proportionately longer than OCC fibers, which reduces the overall fiber fines, a major contributor to slower drain times.

Panel thickness, consistency, and final density

Mat or panel thickness, consistency, and density were measured after wet-forming (~15% consistency), wet-pressing (~45% to 55% consistency), and dry-pressing (100% consistency) (**Fig. 2**). An unexpected result from this study was the significant thickness difference between the two wood fiber types, HB and OCC, after forming and pressing (**Fig. 2**). This effect is an important consideration when designing molds for three-dimensional engineered fiberboard structures from different fiber types (Hunt and Winandy 2002). We believe the main reason for the thickness difference is that OCC fibers are chemically pulped to remove up to 45 percent of the lignin, making them more flexible and conformable than the stiffer semimechanical pulped HB fiber. The HB fibers also have considerably more fiber bundles or shives that are stiff and less conforming than individual fibers.

The FG fibers had a significant effect on final density for all panels: the density decreased as the FG percentage increased (**Table 2**). This indicates that the stiffer FG fibers caused additional separation

Table 2. — Mechanical and physical properties of the panels made with HB and OCC fibers.

Fiberglass type	Coupling agent treatment	Fiberglass loading	Panel properties				
			Specific gravity	Tensile strength	MOE	Linear expansion (30% to 90%)	Sonic MOE
		(%)		(MPa)	(GPa)	(%)	(GPa)
HB pink	Water	0.0	0.944	25.37	4.10	0.205	4.29
		7.5	0.925	22.69	3.99	0.183	4.11
		15.0	0.903	19.03	3.68	0.178	3.86
OCC pink	Water	0.0	1.082	75.53	8.63	0.275	8.55
		7.5	1.048	65.67	7.56	0.264	7.72
		15.0	1.010	52.63	7.04	0.232	6.85
HB pink	Silane	0.0	0.944	25.97	4.10	0.215	4.29
		7.5	0.917	21.11	3.82	0.188	4.00
		15.0	0.902	18.44	3.62	0.173	3.82
OCC pink	Silane	0.0	1.082	73.33	8.62	0.268	8.66
		7.5	1.079	64.40	7.62	0.251	7.87
		15.0	1.005	52.52	6.98	0.221	6.87
HB yellow	Water	0.0	0.928	27.66	4.50	0.182	4.50
		7.5	0.906	24.06	3.94	0.178	4.12
		15.0	0.880	18.60	3.45	0.177	3.80
OCC yellow	Water	0.0	1.079	74.51	9.76	0.271	8.75
		7.5	1.040	64.55	7.82	0.218	7.84
		15.0	0.998	54.05	6.70	0.200	6.91
HB yellow	HMR	0.0	0.902	32.51	5.39	0.194	4.72
		7.5	0.881	26.91	4.56	0.175	4.26
		15.0	0.865	22.20	3.93	0.192	4.05
OCC yellow	HMR	0.0	1.082	80.76	9.33	0.217	8.86
		7.5	1.034	71.17	9.11	0.189	7.17
		15.0	0.995	57.24	8.04	0.193	6.98

Table 3. — Properties of commercial hardboard for comparison.

Commercial hardboard type[a]	Panel properties			
	Specific gravity	Tensile strength	MOE	Linear expansion (30% to 90%)
		(MPa)	(GPa)	(%)
6-mm-thick tempered				
A	0.95	28.54	5.48	0.270
E	0.99	30.54	5.72	0.320
3-mm-thick standard				
A	0.88	28.06	4.55	--
E	0.93	25.44	4.69	--
3-mm-thick tempered				
A	0.95	38.89	6.27	--
E	0.96	32.34	5.86	--

[a]Hardboard types A and E are wet-formed commercial hardboard. Data taken from Suchsland and Woodson (1987).

between fibers, preventing some fiber-to-fiber contact. If the FG fibers were as flexible as the wood fibers, then adding FG fibers would have produced a noticeable decrease in panel thickness, but the opposite was true. The FG addition was based on weight not volume. Thus, adding 15 percent FG should have produced an increase in density by 6 percent. However, density actually decreased by 6 percent for OCC panels (**Table 2**).

Mechanical and physical properties

Stress wave modulus of elasticity (SMOE), tensile modulus and strength, and dimensional stability were measured to determine the main effects and interactions for the three factors: coupling agent, wood fiber type, and FG addition. Results from all the tests are presented in **Table 2** and compared with commercial data and AHA minimum HB standards on **Table 3**.

The HMR coupling agent had a significant effect on tensile properties and linear expansion while silane did not have a significant effect. The HMR-treated panels consistently showed an increase in tensile modulus and strength compared with those treated with silane or water alone (**Figs. 3** and **4**). This may be due to the higher HMR addition rate of 2.3 percent compared with only 0.1 percent for silane. The addition of FG fibers with coupling agents did not overcome the sheet bulking effect (increase in bulk or decrease in density and decrease in fiber-to-fiber contact), which may have been responsible for the loss of panel stiffness and strength. Tensile strengths for panels made from HB fibers were similar to or slightly higher than minimum AHA requirements for standard and tempered HB (AHA 1995). However, panels made from

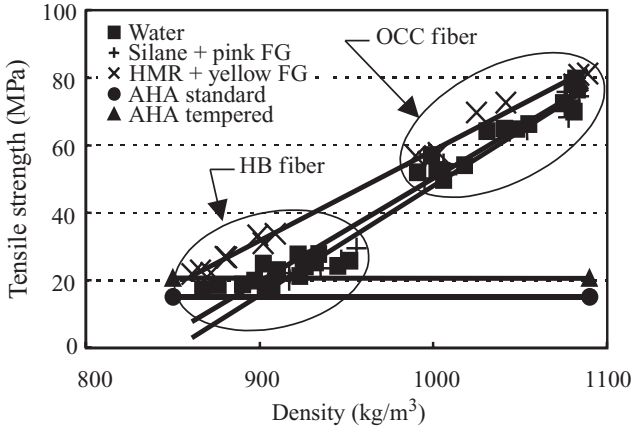

Figure 3. — Tensile strength for OCC and HB fibers with and without coupling agents (silane and HMR) and 0, 7.5, and 15 percent FG fibers. For comparison, minimum strength requirements for AHA standard and tempered HB are shown.

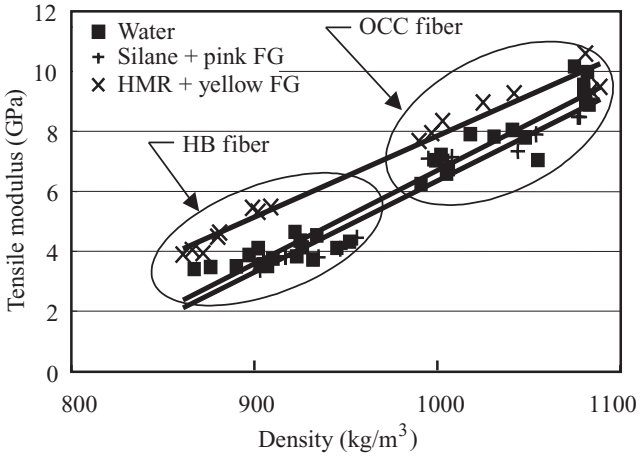

Figure 4. — Tensile modulus for OCC and HB fibers with and without coupling agents (silane and HMR) and 0, 7.5, and 15 percent FG fibers.

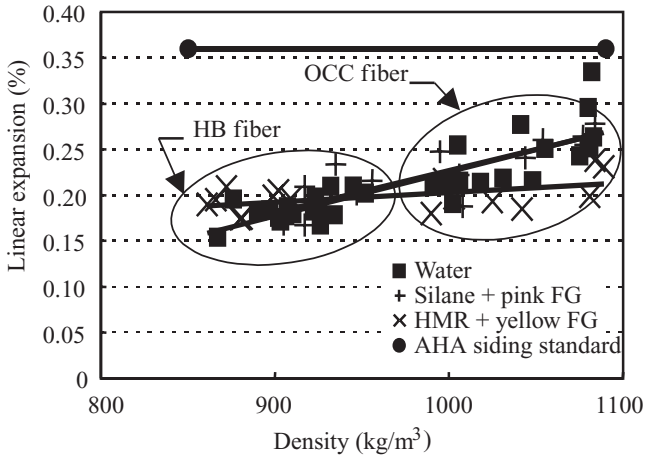

Figure 5. — Linear expansion for all specimens with and without coupling agents (silane and HMR) and 0, 7.5, and 15 percent FG addition. For comparison, AHA linear expansion minimum standards for siding materials are shown.

OCC fibers, with or without FG, had 1.5 to 5 times higher strength than minimum AHA standards. The OCC panels were 1 to 3 times stronger than the HB panels. The OCC panels also exhibited significantly greater stiffness (almost double) compared with HB panels. We believe the OCC fiber morphology and the drying process were the main reasons for this significant increase. Yao (1978) found that hot-pressing recycled fibers between two screens, as was done in this study, significantly improved strength properties compared with those achieved using only one screen. Further research in this area is necessary to fully determine the cause of this increase in strength and stiffness.

The HMR had an interesting effect on linear expansion (**Fig. 5**). It was expected that FG would decrease linear expansion; however, HMR-treated panels had essentially the same linear expansion with or without FG addition. For all other panels, linear expansion decreased with increasing amounts of FG fibers. Linear expansion results for all panels were acceptable because they fell well below the maximum allowable AHA siding standard of 0.36 percent.

The SMOE was compared with the tensile MOE values for all panels (**Fig. 6**). The nondestructive test method successfully predicted the tensile modulus across all factors. The SMOE as shown in **Figure 6** could be used even with varying factors as an effective nondestructive evaluation test to predict tensile MOE. **Figure 6** also shows the significant effects of fiber type (OCC or HB) and FG addition (0%, 7.5%, and 15%) on panel modulus.

Concluding remarks

It is possible to form HB panels using FG fibers from insulation trim waste material. Refining the FG is a required additional processing step essential for uniform distribution and elimination of FG fiber bundles within the wood fiber mat. Adding FG increased drainage rates for OCC fiber mats, which would boost production rates closer to commercial HB rates.

All the panels with FG fully exceeded minimum HB standard requirements. However, we did not fully achieve the potential strength and stiffness available in the FG fibers. All data clearly indicate reduced strength properties with increased FG loadings. Thus, it seems that neither the silane nor HMR coupling agents, as applied in this study, improved adhesion of FG to the wood fibers sufficiently to produce improved tensile properties. The coupling agents probably washed from the fibers during the wet-slurry processing method used

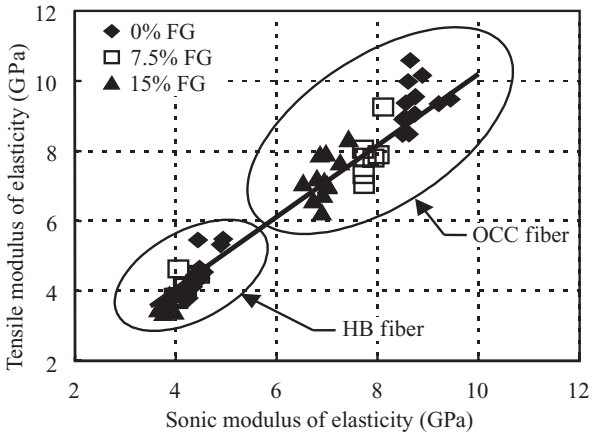

Figure 6. — SMOE compared with measured tensile modulus for both fiber types (OCC and HB) with increasing levels of FG fibers. Regression line has slope of 1.03, and $r^2 = 0.95$.

in this study. More research is needed to develop an effective coupling agent or coupling application process to enhance the wood-fiber-to-FG fiber bond in wet-formed panels.

Panels made from OCC fibers were significantly stronger and stiffer than those made from HB fibers. HB and OCC fibers are morphologically different and therefore respond differently to processing conditions. The effects of processing methods on panel properties when using these high- and low-yield fibers still need to be fully explored.

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