EFFECT OF MICROCRYSTALLINE CELLULOSE, SPECIES, AND PARTICLE SIZE ON MECHANICAL AND PHYSICAL PROPERTIES OF PARTICLEBOARD

Emmanuel Atta-Obeng

MS Student
E-mail: eattaobe@mix.wvu.edu

Brian K. Via†*

Assistant Professor
School of Forestry and Wildlife Sciences
Auburn University
Auburn, AL 36849
E-mail: bkv0003@auburn.edu

Oladiran Fasina

Associate Professor
Biosystems Engineering
Auburn University
Auburn, AL 36849
E-mail: fasinoo@auburn.edu

(Received August 2010)

Abstract. Particleboards made from both sweetgum (*Liquidambar styraciflua*) and southern pine (*Pinus* spp.) were made at a small and large particle size and at 0 and 10% microcrystalline cellulose loading. Modulus of rupture, modulus of elasticity, work to maximum force, and thickness swell (after 2 and 24 h) were measured for all treatment combinations. An increase in particle size had a positive influence on mechanical properties but also allowed for more thickness swell, particularly for the southern pine furnish. Conversely, adding cellulose actually decreased mechanical properties, increased thickness swell, and decreased springback. In the field, the ability to manipulate particle size to control particleboard mechanical properties is perhaps more cost-effective and practical than cellulose addition. Replacing southern pine with sweetgum was viable with equal or better mechanical and physical properties. This suggests that the hardwood species could be a feasible substitute for pine as the demand for woody resources in the southern US continues to grow.

Keywords: Particleboard, sweetgum, southern pine, microcrystalline cellulose, particle size, mechanical properties, physical properties.

INTRODUCTION

Wood-based composites allow for complete use of the tree resulting in better use of timber resources. Worldwide, there has been a growing demand for composite wood products such as plywood, oriented strandboard, hardboard, particleboard, medium-density fiberboard, and veneer board products (Sellers 2000). Consumption of particleboard has grown from 9 Mm^3 in 1965 to 70 Mm^3 in 1997, and worldwide competition for the raw material has also increased, resulting in increased product costs (Buongiorno 2003; Sackey et al 2011). Furthermore, global production is projected to reach 84 Mm^3 by 2013 (Buongiorno 2003). In North America, 76 particleboard mills produced approximately 11 Mm^3 in 1998, accounting for 19% of the

* Corresponding author
† SWST member

total wood composite products produced (Sellers 2000, 2001).

Particleboard panels can be categorized as either appearance- or structural-based. Appearance-based products include end uses such as cabinets, furniture, and core material, whereas structural-based particleboard is used in floor underlayment, stair tread shelving, home construction, and structural sheathing (Maloney 1993; Nemli and Çolakog et al 2005). Urea formaldehyde adhesives are commonly used for appearance-based panels, whereas phenol formaldehyde (PF) is more appropriate for outdoor (structural) applications because of its resistance to liquid water exposure (Gürü et al 2006) and because it improves composite mechanical properties. For structural-based products, problems associated with particleboard manufacture include moderate strength properties caused by small particle size and greater swell potential caused by high board density. Controlling both of these properties may be important for products such as floor underlayment in which high mechanical properties and swelling capacity are important. Factors that can influence particleboard mechanical properties are adhesive type and loading, wood species, vertical density profile, adhesive cure properties, press time, and particle size (Adeyemi and Adeyemi 2002; Papadopoulos et al 2002; Zheng et al 2006; Nemli and Demirel 2007; Ashori and Nourbakhsh 2008; Hassan et al 2009; Mendes et al 2010). During particleboard manufacturing, separation of particles by size is already performed at the screening stage and thus may be a feasible way to control mechanical properties. Increasing particle size is known to improve mechanical properties of other composite products and could be advantageous for particleboard. Conversely, increased particle size results in a rougher surface, and light sanding may be necessary if appearance is important for the structural application.

A possible alternative to increasing wood particle size is through microcrystalline cellulose (MCC) reinforcement, which can increase strength while helping to maintain a smooth surface. Cellulose materials will become increasingly available as pulp mills begin to look for new product opportunities and as fractionation of biomass into cellulose streams becomes more prominent to accommodate the biofuels, bioenergy, and bioproducts industry (Ragauskas et al 2006). Research has already found that inexpensive fillers such as wood flour and wheat flour have a positive reinforcing effect on plywood adhesive systems (So and Rudin 1990). However, microscale cellulose fibers may have an advantage compared with wood particles because of their higher strength-to-weight ratio. The smaller size and larger aspect ratio of cellulose (10-50 μm) might be advantageous during integration of MCC into the adhesive system and could result in better reinforcement efficiency (Seydibeyoğlu and Oksman 2008).

Finally, there is growing concern that conventional wood species such as southern pine will be depleted as new biofuels and other biobased manufacturers emerge. Furthermore, the quality of the southern pine resource has decreased because of higher juvenile wood content in the harvested wood. Juvenile wood can sometimes be undesirable because of the combination of lower density, higher microfibril angle, higher lignin, and lower cellulose content, all of which can have a negative impact on composite mechanical properties (Via et al 2009). In the southeastern US, the volume of hardwood available for processing has increased because of an industry-wide culture of using mostly loblolly pine and other southern pine species. Sweetgum is conventionally a nuisance species that develops on southern pine and other pine-based sites from Connecticut to Florida, ranging west to Texas, Iowa, and Oklahoma. Consumption of sweetgum for structural particleboard may be one way to use an otherwise underutilized raw material. Successful use of sweetgum may be important if consumption of southern pine begins to exceed regeneration or planting.

The objective of this study was to control the species, particle size, and MCC content of particleboard to determine which combination of treatments and levels would yield superior mechanical and physical properties.

MATERIALS AND METHODS

Chemicals and Materials

MCC was purchased from VWR Chemical Co. (St. Louis, MO). Commercial liquid resol PF adhesive (55% solids content, Model #13BO33) for binding wood particles was provided by Louisiana-Pacific Corporation (Hanceville, AL). Two different types of woods were used in this study: sweetgum and southern pine. Southern pine wood particles were provided by Temple Inland (Monroeville, AL). A sweetgum tree with an average diameter at breast height of 210 mm was harvested from a woodland area near Auburn University (Auburn, AL). After foliage was trimmed and bark was removed with a hand shear, the tree was broken down into wood chips using a ring-type flaker. Chips were conditioned at 20°C and 65% RH for 2 wk before they were decreased to particles using a laboratory hammer mill with a 6.35-mm screen. Particles were classified into two size categories, small and large, with oversized and undersized particles removed on a screening machine with 3-, 1.5-, and 0.8-mm openings. Distribution of both southern pine and sweetgum wood particles was evaluated using a digital image processing system (Camsizer; Retsch Technologies, Haan, Germany). The Camsizer works on the principle that bulk material (wood particles) falls between a light source and camera, which allows for rapid measurement of each particle size and consequent distribution. Particles were optically recorded, digitized, and processed by the host computer. Results of the particle distribution test are shown in Fig 1. Figure 1 shows that the distribution of large-sized wood particles was wider and tailed to the right compared with the distribution of small-sized particles. These particles were stored at room temperature (25°C) and 50% RH resulting in 8% MC prior to composite manufacturing.

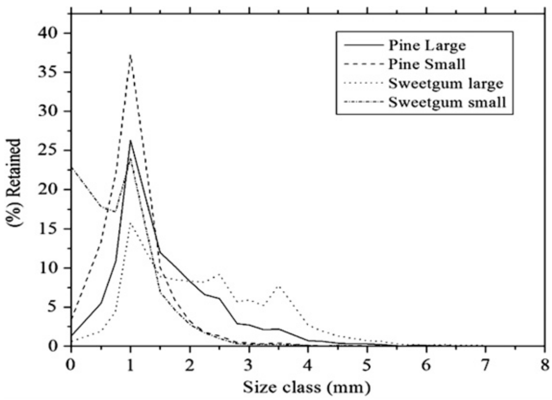

Figure 1. Size distributions of sweetgum and southern pine wood particles.

Panel Manufacturing

Dried wood particles were blended with MCC and PF adhesive. MCC loading rates were 0 and 10% of total furnish weight. As a result, a 15% adhesive loading (weight basis) was applied using an atomizing spray gun, which resulted in complete bonding of all treatment combinations. After blending, materials were placed in a molding box (431.7 × 431.8 mm) and manually formed into a homogeneous single-layer board. The adhesive-coated mats were then compressed on aluminum cauls in a hot press at 200°C and a pressure of 2.94 MPa. Total press time was 5 min for all boards. Press time was considered to be the time from initial contact of the press with the mat until release of press pressure. Stops were placed in position to produce boards of 12.7-mm target thickness. Three panels were made for each treatment at a target density of 660 kg/m^3 for a total of 24 boards. Treatment SYS0 (Table 1) was considered to be the control treatment based on 0% MCC, smaller/conventional-sized particles, and southern pine species. After pressing, panels were trimmed to 431.8 × 431.8 × 12.7 mm and kept in a conditioning room at 65% RH and 23°C until their moisture contents equilibrated. These panels were then cut into test samples following protocols from ASTM (1993).

Water Resistance

Thickness swell and linear expansion were determined using protocols from ASTM (1993). Sample particleboards were cut into 304.8- ×

Table 1. Factorial experimental design for manufacture of particleboard panels.[a]

Panel type	Species	Particle size (mm)	Adhesive loading (%)	Microcrystalline cellulose loading (%)	Avg. board density[b] (kg/m^3)	Avg. board thickness[b] (mm)	Panel replications
SGS0	Sweetgum	0.83	15	0	640 (5)	12.8 (0.0046)	3
SGS10	Sweetgum	0.83	15	10	640 (11)	12.5 (0.0016)	3
SGB0	Sweetgum	2.30	15	0	660 (9)	12.9 (0.0054)	3
SGB10	Sweetgum	2.30	15	10	640 (14)	12.8 (0.0013)	3
SYS0	Southern pine	1.13	15	0	670 (17)	12.8 (0.0052)	3
SYS10	Southern pine	1.13	15	10	650 (4)	12.5 (0.0031)	3
SYB0	Southern pine	1.62	15	0	680 (7)	12.8 (0.0037)	3
SYB10	Southern pine	1.62	15	10	670 (6)	12.8 (0.0042)	3

[a] Panel codes were developed (shown in panel type column) for use in Figs 2-5.
[b] Numbers in parentheses are standard errors.

304.8-mm squares. Three specimens of each treatment were soaked in tap water for 2 and 24 h. To prevent floating, weights were used to fully submerge samples in water. Thickness in the middle of the test sample was measured with a digital thickness gauge (C1050EB; Mitutoyo Co., Kanagawa, Japan). Thickness and length measurements were taken before and after soaking. From these, thickness swell and linear expansion were calculated. Thickness swell percentage was determined from the following equations:

$$\mathrm{TS}_{2\mathrm{h}}(\%) = {}^{(t_2 - t_1)}/_{t_1} \times 100 \qquad (1)$$

$$\mathrm{TS}_{24}(\%) = {}^{(t_{24} - t_1)}/_{t_1} \times 100 \qquad (2)$$

where t_1 is thickness at specimen middle before soaking and t_2 and t_{24} are thicknesses of test specimens after soaking for 2 and 24 h, respectively.

Density Determination

Density is an important variable that influences particleboard mechanical properties such as modulus of elasticity (MOE) and modulus of rupture (MOR). Board density can also be controlled to meet specific application requirements. American National Standard Institute (ANSI 1999) has designated boards with target densities greater than 800 kg/m^3 as high density, between 640 and 800 kg/m^3 as medium density, and less than 640 kg/m^3 as low density. Density of each test sample was measured before testing. Volume of the samples was calculated from length, width, and thickness measurements taken from a digital caliper (CD-800 C; Mitutoyo Co.). Bulk density was determined by dividing particleboard mass (kg) by its volume (m^3).

Mechanical Properties

All test samples were conditioned at 65% RH to a constant temperature of 23°C for at least 1 week before testing. ASTM (1993) was followed for tests of mechanical properties. MOE and MOR measurements were obtained from three-point bending tests. MOE is a ratio of the stress to strain curve that describes potential ability of the board to sustain deformation. MOR, conversely, describes stress to failure. Work to maximum force was also calculated as area under the stress to strain curve from 0 to maximum force.

Samples for three-point bending (evaluation of MOE and MOR) were 228.6 × 76.2 × 12.7 mm. A span of 177.8 mm for bending at loading rate of 10 kN/min was used. Tests were carried out on a Zwick/Roell Universal Testing machine (Z010; Zwick Roell Testing Systems, Ulm, Germany). Six replicates were evaluated for each treatment. MOE and MOR of the sample specimen were calculated based on the following equations:

$$\mathrm{MOR(MPa)} = {}^{3PL}/_{2bd^2} \qquad (3)$$

$$\mathrm{MOE(GPa)} = {}^{P_1L^3}/_{4bd^2Y_1} \qquad (4)$$

where P is maximum load (N), P_1 is load at proportional limit (N), b is specimen width (mm), d is specimen thickness (mm), Y_1 is deflection corresponding to P_1 (mm), and L is span (mm).

Experimental Design and Data Analysis

To study effects of cellulose loading and particle size on physical and mechanical properties of particleboards, a preliminary 3 × 2 factorial experiment was conducted. MCC loading was set at three levels: 0, 5, and 10%. Two levels of particle size were used. A 10% adhesive content was used for all treatments. Results of the preliminary experiment showed that mechanical properties decreased with addition of MCC. Based on results of the preliminary experiment, a 2 × 2 × 2 factorial design was conducted for the main experiment (Table 1). Adhesive content was increased to 15% after it was found that a 10% loading was not enough to support the high surface area MCC as indicated by visual inspection. Triplicates for each treatment were analyzed. Because boards were hand-formed, achieving uniform density along the length of the boards was quite difficult. Therefore, an adjustment using analysis of covariance (ANCOVA) was performed to correct for any variation in density that may have occurred among samples. For all graphs and statistical analysis, density was chosen as a covariate and adjusted means were calculated. Statistical analysis system software (SAS Institute Inc., Cary, NC) was used for all computations.

RESULTS AND DISCUSSION

ANCOVA results of effect of MCC loading (0 and 10%) and other factors (particle size and species type) on mechanical and physical properties of particleboard are summarized in Table 2. Detailed analysis of effect MCC loading, wood

Table 2. Summary of analysis of variance of panel properties within a 2 × 2 × 2 factorial design.

Panel properties	Source of variation	F-ratio	Significance level
Modulus of elasticity	Particle size	18.82	0.0001
	MCC loading	55.12	<0.0001
	Species	19.16	0.0001
	Species*particle size	4.13	0.0497
	Species*MCC	3.59	0.0665
	Particle size*MCC	0.87	0.3561
	Species*particle size*MCC	0.21	0.6506
Modulus of rupture	Particle size	0.30	0.5848
	MCC loading	34.68	<0.0001
	Species	16.93	0.0002
	Species*particle size	22.80	<0.0001
	Species*MCC	0.40	0.5328
	Particle size*MCC	2.45	0.1266
	Species*particle size*MCC	1.07	0.3086
Work to maximum force	Particle size	23.36	<0.0001
	MCC loading	8.45	0.0063
	Species	61.50	<0.0001
	Species*particle size	52.36	<0.0001
	Species*MCC	0.68	0.4153
	Particle size*MCC	2.86	0.0996
	Species*particle size*MCC	1.18	0.2845
24-h thickness swelling	Particle size	134.61	<0.0001
	MCC loading	88.53	<0.0001
	Species	0.87	0.3657
	Species*particle size	12.78	0.0028
	Species*MCC	33.23	<0.0001
	Particle size*MCC	22.92	0.002
	Species*particle size*MCC	30.25	<0.0001

MCC = microcrystalline cellulose.

particle size, and species type on mechanical and physical properties of particleboards are presented in subsequent sections.

A 12% PF loading is more common in the industry and was originally tried in this study. However, for some treatment combinations, the 12% loading was not enough to cover the high surface area of MCC. We therefore used a 15% PF adhesive loading to handle the addition of cellulose. This probably resulted in better adhesive distribution and higher strength properties than conventional particleboard. A uniform adhesive distribution results in more efficient bonding of all flakes in the composite. Bonding strength also increases with adhesive content (Ashori and Nourbakhsh 2008) because of the removal of weak spots.

Mechanical Properties

Table 2 shows a significant effect of MCC loading and species type on bending properties (both MOE and MOR) of particleboard ($\alpha = 0.05$). Also, there was a significant interaction between species type and particle size on MOE and MOR. This supports the theory that effect of particle size on mechanical properties depended on species type. Figures 2 and 3 show that static bending properties (MOE and MOR) of boards manufactured from 0% MCC loadings were higher than those made from 10% MCC loading. MOE of all boards varied from 1.3-1.9 GPa, whereas MOR values ranged from 5.8-11 MPa. MOE of boards made from small- and large-sized sweetgum wood particles decreased by 31 and 26%, respectively, with addition of 10% MCC. Addition of 10% MCC to small- and large-sized southern pine particles also decreased board MOE by 21 and 11%, respectively.

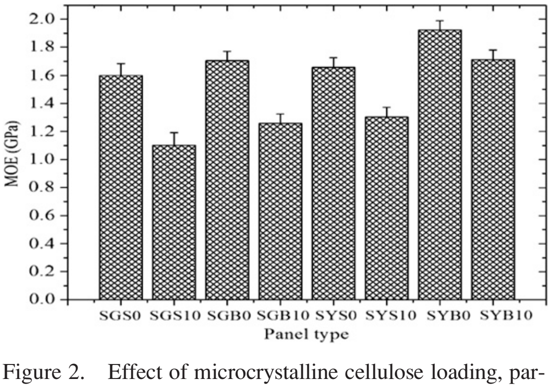

Figure 2. Effect of microcrystalline cellulose loading, particle size, and species on particleboard modulus of elasticity.

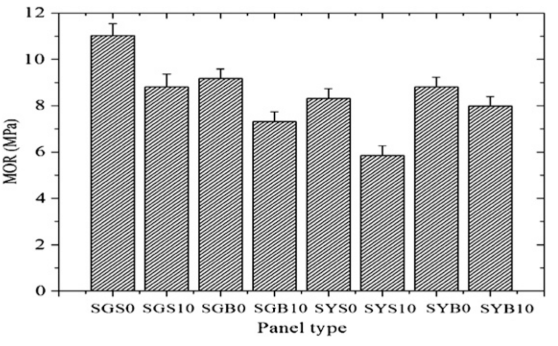

Figure 3. Effect of microcrystalline cellulose loading, particle size, and species on particleboard modulus of rupture.

For breaking strength, addition of 10% MCC decreased MOR by 20% for both small and large particle treatments (sweetgum), whereas the same treatment decreased MOR by 30 and 9% for southern pine. ANSI (1999) states that minimum values of MOE and MOR required for M-1 grade particleboard are 1.725 GPa and 11 MPa, respectively. Boards produced from large-sized sweetgum and southern pine particles met the MOE requirement. However, almost all boards produced had an MOR close to the ANSI requirement but slightly lower, including the control. When data were normalized such that the control met ANSI standards, changing species or particle length resulted in acceptable or improved mechanical properties.

Mechanical properties (MOE and MOR) of boards made at 0% MCC loading were better than at 10% MCC loading. Irrespective of the wood species or particle size used, addition of MCC to particleboard panels resulted in decreased mechanical properties (Table 2). Analysis of board thickness out of the press, with a *t* test, revealed a statistical difference between treatments. Panels with no MCC exhibited a negligible to 2.3% higher springback based on thickness measurements out of press. Higher springback in panels with no cellulose added was initially unexpected because it was anticipated that cellulose elasticity would result in a higher springback. However, after careful assessment of the data, it was found that addition of cellulose resulted in thinner mats during forming to reach the same density. It was hypothesized that addition of cellulose to the mat resulted in decreased strain during pressing and resulted in less springback on press release. This finding is quite important and suggests that any variation in cellulose loading within the manufacturing process could result in unusual thickness variation compared with a conventional particleboard product.

It is important to understand that springback is an irreversible thickness swell caused by release of compression stress imparted to the board when the furnish is pressed in the hot press (Palardy et al 1989; Mohebby et al 2009). During springback, if the forces are great enough, the adhesive can debond from the wood resulting in a weak spot within the composite matrix and an interruption in the mechanical interlocking that once existed within the compressed particles of the composite (Nemli and Demirel 2007). These results were similar to those found in acetylated wood-based particleboard in which a 2% increase in springback resulted in significant and similar decreases in MOR and MOE (Mohebby et al 2009). They attributed this decrease in mechanical properties to a debonding of the fiber and adhesive matrix.

An increase in particle size is known to sometimes improve bending strength properties of composites (Kelemwork et al 2009; Hashim et al 2010). Figures 2 and 3 show that increasing particle size in both species resulted in an increase in board MOE. A similar trend was observed for MOR values of southern pine boards. An increase in board strength with larger particle size may be explained by the hypothesis that larger particles have less surface area compared with smaller particles for a given volume of wood particles. When larger particles are used, adhesive coverage is more effective, resulting in a greater percentage of particle coverage because of lower surface area. This results in better bond and subsequently higher strength values of the large particle-based composite.

Research has shown that an increase in horizontal and vertical density profile has a positive influence on bonding strength of wood-based composites (He et al 2007; Nemli and Demirel 2007; Nirdosha et al 2009). Table 1 describes average density for each panel type. Panel density varied between 640 and 680 kg/m^3. This density range was slightly lower than the 700 kg/m^3 recommended by ANSI (1999) for construction-grade particleboard. If panels had been manufactured to a higher density, a linear adjustment of the data to 700 kg/m^3 shows that all treatments would have exceeded the mechanical properties recommended by the standard.

Thickness Swell

Table 2 shows a significant effect of MCC addition and particle size on thickness swell properties. Also, there was a three-way interaction among species type, MCC loading, and particle size on thickness swell. This could be important to the manufacturer and suggests that multiple changes to the process could have complex effects on thickness swell performance. Conversely, addition of sweetgum resulted in better dimensional stability compared with addition of

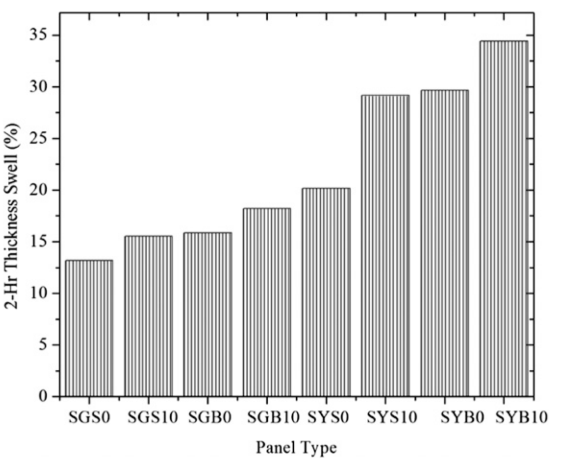

Figure 4. Effect of microcrystalline cellulose loading, particle size, and species on particleboard 2-h thickness swell.

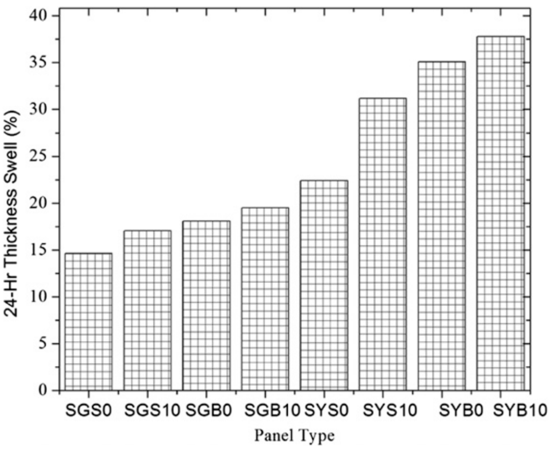

Figure 5. Effect of microcrystalline cellulose loading, particle size, and species on particleboard 24-h thickness swell.

pine. Any complex interaction effect is likely to have a minimal impact when switching from pine to sweetgum because thickness swell properties improved relative to industry standards.

Figures 4 and 5 show that using a 10% MCC loading increased thickness swell properties (both 2 and 24 h) of the boards irrespective of particle size or species used. Once again, this was hypothesized to be attributable to debonding between the adhesive and woody matrix resulting in microdefects that allowed entry of liquid water. Conversely, it may also be possible that addition of cellulose provided more available hydroxyl groups for moisture sorption and consequent thickness swelling. It was also found that thickness swell increased with increasing particle size. Increased particle size resulted in more void space during consolidation resulting in higher porosity and easier diffusion of water into boards during soaking.

CONCLUSIONS

Results obtained from this study led to the following conclusions:

1. Adding microcrystalline cellulose to particleboard resulted in a decline in board mechanical properties and higher thickness swell.
2. Particle size had a significant effect on mechanical properties. MOE and MOR increased with increasing particle size.
3. Sweetgum performed favorably compared with southern pine and may be a reasonable substitute.

ACKNOWLEDGMENTS

This research was in alignment with 5-year goals set forth in Hatch-Project Number ALA-031-1-09020. We thank the Center for Bioenergy and Bioproducts for allowing us to use their facilities.

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