Bio-Composites Made From Pine Straw

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ABSTRACT

Pine straw is a renewable natural resource that is under-utilized. The objective of this study was to evaluate the physical and mechanical performances of pine straw composites. Three panel density levels (0.8, 0.9, 1.0 g/cm^3) and two resin content levels (1% pMDI + 4% UF, 2% pMDI + 4% UF) were selected as treatments. For the pine-straw-bamboo-fiber composites, three pine-straw to bamboo-fiber ratios were selected as the treatments. The pine straw fibers were treated with a container blender, which reduced the fiber sizes and increased the roughness of fiber surfaces. The bending and IB strength properties of the pine straw boards meet the requirement of Class 5 of the voluntary product standard PS 60-73. Strength properties of the pine straw boards were positively correlated with the blending time. The waxy cutin on the surface of the needles was a barrier to the bonding quality and blending in the container blender was helpful in the removal of cutin from material surfaces. Linear relationship was found between the internal bond (IB) strength and board density. Higher pMDI resin content led to greater slope values of the IB-density regression lines. The addition of strong bamboo fibers significantly increased the bending and internal bond strength of the pine straw composites.

Keywords: pine straw, pine needles, bio-composites, forest fire, renewable natural resources, bamboo, composites

INTRODUCTION

Pine needles that have fallen from the trees are commonly known as pine straw. They are long and stiff needle-shaped leaves. The needles are usually arranged in bundles containing 2 to 5 needles and stay on the trees for one to two years providing food for the tree through the process of photosynthesis. After needles mature, they turn brown, fall from the tree, and deposit on the forest floor. Thus, a thick and springy carpet of pine needles covers the ground around the trees. The annual yield for 16-year-old loblolly pines varies from 1,700 to 7,400

kg per hectare, depending on the tree density (Moore, et al 1996). The straw has traditionally been harvested for mulch (Jemison 1943, Bateman and Wilson 1961, Sood and Sharma 1985).

Pine needles are chemically composed of cellulous, hemicellulous, lignin, extractives and ash. The outer layer of the adult leaf is the waxy cuticle (cutin) which protects the leaf from moisture loss. After the leaves fall from the tree, the wax remains on the leaves. The cuticle consists of a hard, fibrous outer shell (epidermis). Beneath the epidermis is the hypoderm, which is composed of several compact layers of long, thick-walled, fiber-like cells. The hypoderm gives rigidity to the needle (Koch 1972). The internal structure of the leaf is complex and includes a photosynthesizing parenchyma ("chlorenchyma" or mesophyll) and resin canals. In the center are two parallel vascular bundles, each consists of phloem and xylem (Howard 1973a).

As shown in Table 1, pine needles have a higher cellulose content than sugar cane, which has been used to make bio-based composites (Rowell 1988, Hse and Shupe 2002). Pine straw has been autoclaved with phenol then reduced to powder and pressed into boards (Jain and Gupta 1969). Howard (1974) studied the feasibility of making panels directly from pine straw. The needles were treated with rolling and benzene bath in NaOH solution before they were pressed into 12.7-mm-thick boards. The caustic bath treatment led to desirable modulus of rupture (MOR) and internal bond (IB) properties. Boards from other treatments had poor dimensional and mechanical properties.

Table 1. Chemical components of pinestraw, sugar cane and wood.

	Cellulose (%)	Hemicelluloses (%)	Lignin (%)	Ash-Content (%)	Extractives (%)
Pinestraw[1]	42.6	22.3	37.7	2.4	26.2
Sugar Cane[2]	39.5	28.4	23.4	1.7	2.9
Pine (*Pinus* sp)[1]	51.1	26.8	27.8	0.3	9.1

[1]Howard 1973 (average of the listed data); [2] Ouensanga 1989 (average of the listed data).

Pine straw is a renewable natural resource that is under-utilized. Every year forest fire causes substantial damage to the forest resources. Removal and utilization of pine straw from the forest floor may reduce the fire hazard in the forest and generate profits for forest owners and local communities. Needles that fall from a southern pine tree may equal or exceed the weight of wood added to the stem (Howard 1973). Utilization of pine straw may provide an income that may exceed that from timber (Roise et al. 1991). The development in adhesive technologies and bio-based materials provides a new opportunity for the utilization of pine straw for making composites. The objectives of this study were to make pine straw composites by processing pine straw into fiber materials and evaluate panel density, blending time, and resin content effects on the properties of pine straw and pine-straw-bamboo-fiber composites.

MATERIALS AND METHODS

The pine needles used were from longleaf pine (*Pinus palustris*) stands in Rapides Parish, LA, USA. Pine cones and bark were removed from the pine straw. About 5% dry grass straw residue and understory leaves were mixed with the needles and not removed. The needles were steamed twice in a container for 15 minutes each. This treatment was believed to partially remove cutin wax on the surface of the needles and improve the bonding properties

of pine straw fibers. Before steaming, water was added to one half of the container. After first steaming treatment, the water containing extractives was removed and new water was put in for the second steaming. After the treatment, the wet needles were processed by a rotary grinder without adding additional water. The obtained materials consisted of long needle skins, fiber bundles, fibers, and particles. The materials were then dried in an oven at 80 °C for 16 hours.

Blending was performed in a container blender. On the bottom of the container, a turning blade blew fibers to the air in the container. Resin was sprayed from the top cover of the container to the floating fibers. The adhesives used were commercially prepared pDMI and urea formaldehyde (UF) resins. The two resins were thoroughly mixed before blending and were then applied to fibers by an air-atomizing nozzle.

Since the high speed blade may reduce the fiber size, the effect of the propeller blade on fiber sizes was analyzed. Fibers were blended in the blender for 1, 2,3, 4, and 5 minutes without spraying glue. They were screened after each blending. The 4-minute blending group of fibers was used to analyze fiber size distribution on screens of 20, 35, and 60 mesh. Five hundreds of fibers from each of the screens were randomly chosen for the analysis. Image Pro Plus software and a mounted digital SPOT camera were used to do the measurement.

Two experiments were conducted for the pine straw composites. The first one was to evaluate panel density and blending time effects at 1 percent of pMDI and 4 percent of UF resin solid levels. The treatments were three blending time levels (3, 4, 5 minutes) and three panel density levels (0.8, 0.9, 1.0 g/cm^3). Twenty-seven pine straw composite panels, measuring 25 by 25 by 1.25 cm, were manufactured using a laboratory hot-press with three replications of each treatment combination. The second experiment was to evaluate the performance of panels at a higher resin content. Six pine straw panels were manufactured with two density levels (0.8, 1.0 g/cm^3) at 2 percent pDMI and 4 percent UF. The straw fibers were blended for 5 minutes. The hand-felted mats were pressed for 4 minutes at 177 °C temperature.

For the pine-straw-bamboo-fiber (PSBF) composites, the treatment was three pine-straw-fiber to bamboo-fiber ratios (70 /30, 30 / 70, and 0 / 100 percent by weight). Bamboo culms used in this study were locally obtained at Pineville, LA, USA. The culms were first split and then fed into a chipper. The chips produced were then steamed and wet-grinded into fibers and fiber strands in the rotary grinder. The preparing process of bamboo fibers was similar to that of wet-process hardboard manufacture. The fibers and strands were then dried to 3-6% moisture content in an oven at 80 °C. The dried mixture was dry-separated into fibers with the same grinder. The resin content was 1 percent pMDI and 4 percent UF resin solids based on the oven-dried fiber weight. Target board density was 0.8 g/cm^3. Nine PSBF panels, measuring 25 by 25 by 1.25 cm, were made with three replications for each treatment level.

Physical and mechanical properties of the panels made were evaluated according to the ASTM D1037-98.

RESULTS AND DISCUSSION

Figure 1a shows the fiber-like materials that were obtained from the refinement of pine straw. The materials consist of needle skins, fibrous bundles, and fibers. Figure 1b shows materials that were blended for 5 minutes without the addition of adhesives. As shown in the figure, the materials consist mostly of needle outer skins (epidermis), inner skins (hypoderm), vascular bundles, and small particles. A small amount of fibers was also present in the materials. Pure needle fibers were obtained by refinement of the materials in a wet process and Figure 1c shows the fibers. Solid resin pieces that were 0.1 to 2 mm in diameter were mixed in the materials.

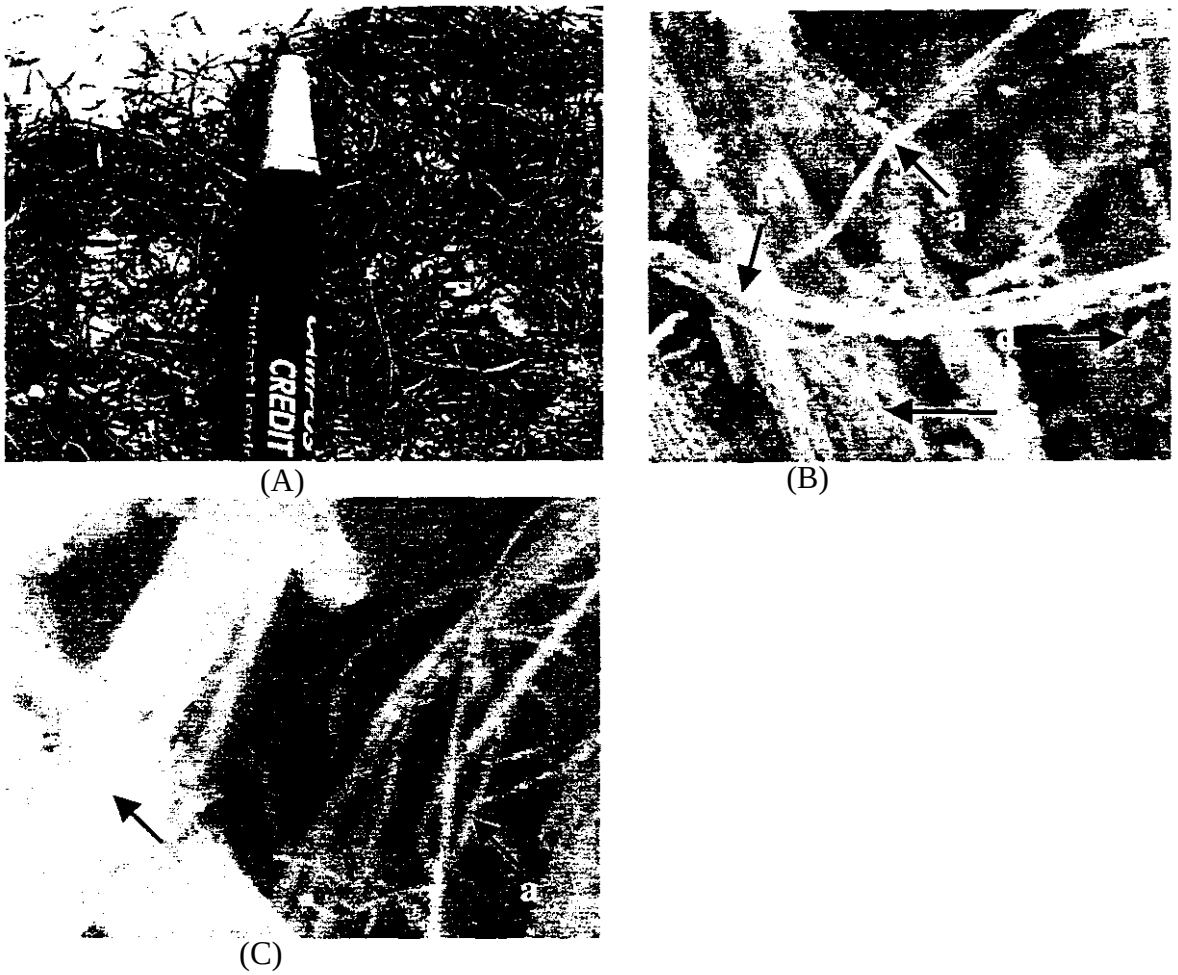

Figure 1. Materials used to make pine straw composites: (A) fiber-like materials obtained from refinement of pine needles, (B) materials obtained after 5-minute blending, (C) pine straw fiber bundles and fibers. On the graphs: a –fiber, b - fiber bundles, c - needle skins, d - particles.

Figure 2 shows fiber length distributions as affected by blending time. The figure illustrates that bigger fiber bundles and needle fractions were greatly reduced to smaller fiber bundles within the first minute of blending. When the blending time was greater than 2 minutes, fiber size distribution changed little and more than 70% of the materials had fiber sizes greater than those on the 60 mesh screen. Figures 3 and 4 show the fiber-length distributions in the 4-minute blending group and fiber frequency distributions within three mesh screens (8, 35 and 60) in this group, respectively. Fibers that stayed on mesh screens 20, 35 and 50 accounted for 73% of the total fiber weight. These indicate that extension of blending time had no

effects on material length after the first minute of blending. Fiber length frequency distributions of these three groups of fibers were skewed to the right, as shown in Figure 4, due to the removal of smaller fibers by the screening.

Modulus of rupture (MOR) and modulus of elasticity (MOE), internal bond (IB) strength, and 24-hour-soaking thickness swell of the pine straw composites are shown in Table 2. These properties of the panels from the second experiment are shown in Table 3. For the needle processing technology used in this study, the bending and IB strength properties of the pine straw boards meet the requirement of Class 5 of the first voluntary product standard PS 60-73. The low IB strength of the boards indicates that high quality bonding between the materials was not achieved. This was possibly due to the wax on the surfaces of the needle outer skins, which were the main component of the materials.

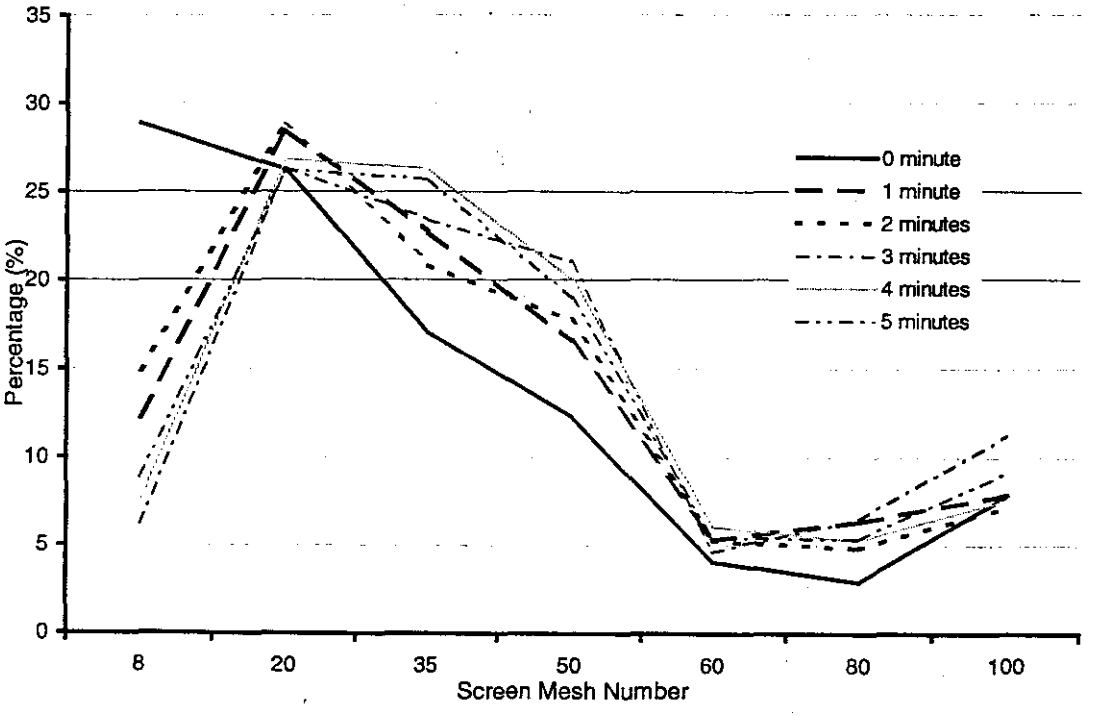

Figure 2. Length distribution of pine straw fiber materials as affected by blending time.

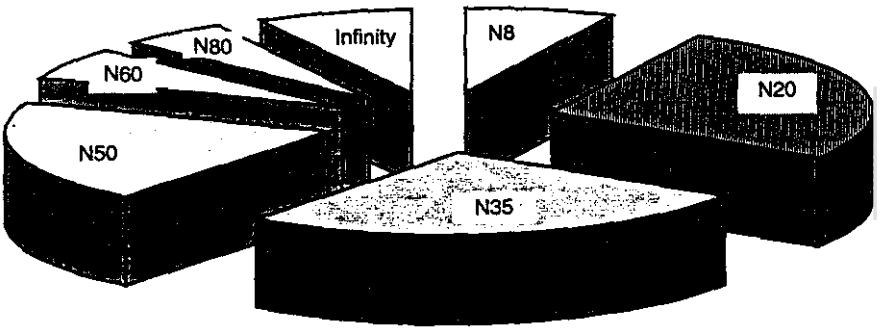

Figure 3. Distribution of pine straw fiber materials blended for 4 minutes.

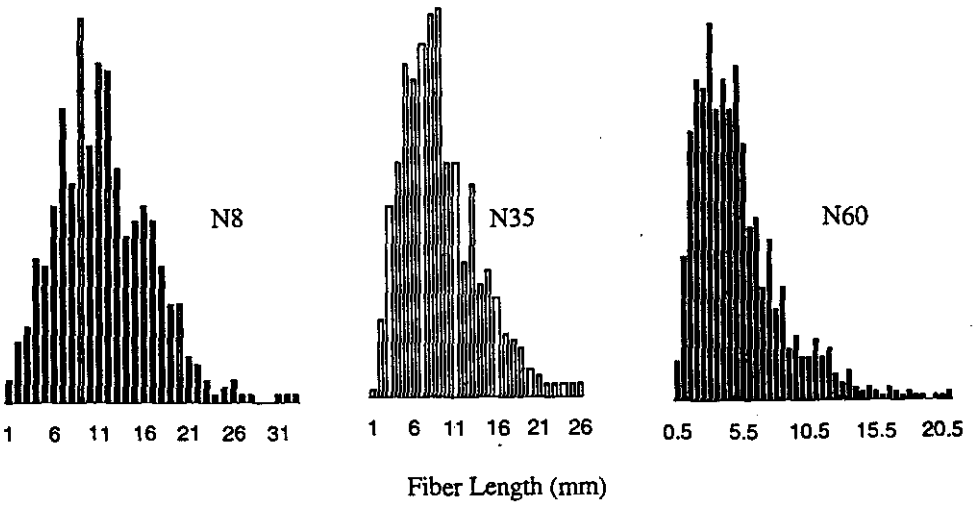

Figure 4. Distribution of pine straw fiber-like materials on the standard screen N8, N35, and N60, after blended for 4 minutes.

Table 2. Properties of pine straw composites with 3 specific gravity and 3 blending time levels at 1% PMDI and 4% UF resin content.

Properties	Blending time (min)	Specific Gravity*		
		0.8	0.9	1.0
MOR (Mpa)	3	7.82	9.28	9.92
	4	13.0	13.89	13.26
	5	15.69	16.65	15.48
MOE (10^3 Mpa)	3	1.04	1.19	1.29
	4	1.82	1.95	1.86
	5	2.28	2.41	2.11
IB (Mpa)	3	0.15	0.17	0.21
	4	0.25	0.26	0.27
	5	0.30	0.29	0.39
TS (%)	3	36.34	35.72	32.47
	4	34.73	36.43	35.92
	5	38.79	39.51	38.33

*** Based on oven-dry conditions.**

It was found that the flexural and bonding strength properties of pine straw composites were affected by panel density, refining time and resin content of the panels. IB strength was very responsive to panel density and resin content. This can be seen through the linear relationship between IB strength and panel density in Figure 5. The effects of resin content were reflected via the slope of the regression lines. Higher resin content led to a greater slope, i.e., IB strength was improved more rapidly with increased panel density at higher resin content levels. Blending time showed a significant effect on all panel properties (Table 2). Bending and IB strength increased with blending time. Since there were little changes in fiber sizes (Figure 2), the blending did not reduce the fiber length. Therefore, the blending effects are attributable to one or both of following two reasons: (1) the improvement in bonding conditions by removal of the waxy cutin on needle skins and (2) better resin distribution through the extended rubbing between fiber surfaces. The blending blade increases the roughness of the waxy surfaces and waxy cutin on needle surfaces was further removed. Furthermore, small particles and fibers generated in the blending produced more bonding sites. These results indicate that more and better refining is needed to improve the bending and IB strength of pine straw composites. One of the methods could be the pressurized grinding, which is widely used in the conventional wet-process hardboard manufacture.

Table 3. Properties of pine straw composites pressed at 2% PMDI and 4% UF resin content.

Specific Gravity*	MOR (Mpa)	MOE (10^3 Mpa)	IB (Mpa)	TS (%)
0.8	8.34	1.10	0.39	32.3
1.0	15.55	2.21	0.46	35.8

*** Based on oven-dry conditions.**

It was expected that pine straw composites would have good dimensional stability due to the hydrophobic waxy cutin and resin in the materials. As shown in Tables 2 and 3, the thickness swell properties of the panels were generally poor. More fresh fiber surfaces were exposed as blending time increased and more thickness swell was expected, even though blending increased the bondability between fibers. Pine straw panels also quickly absorbed surrounding moisture and warped. Panel density shows little effects on thickness swell in the

first experiment (Table 2). As expected, increase of resin content reduced thickness swell (Table 3) of the panels.

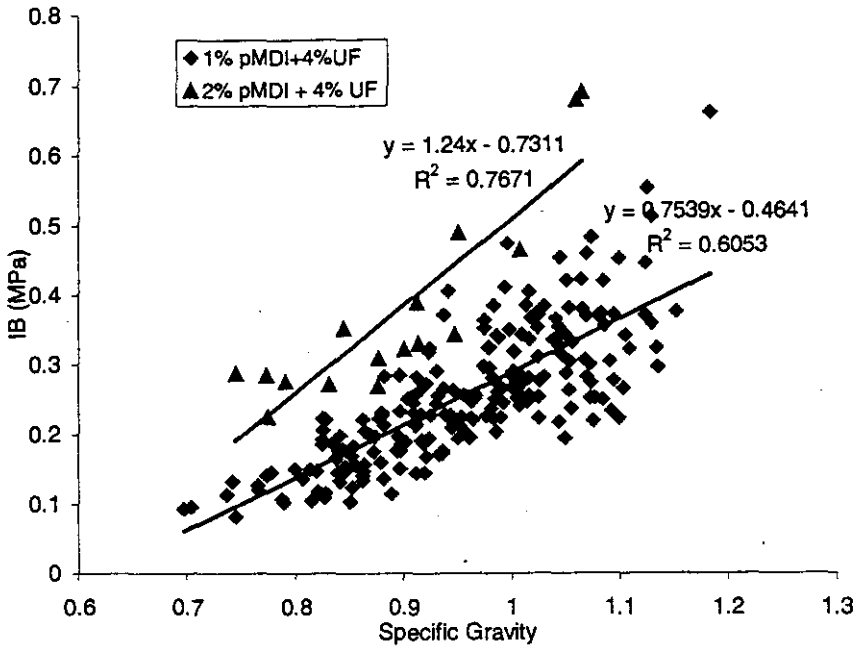

Figure 5. Internal bond strength of pine straw composites as affected by board specific gravity and resin content.

The mechanical properties of PSBF composites are presented in Table 4. The bending and internal bond strength properties of PSBF composites were greater than those made solely from pine straw, especially when the pine-strawhamboo ratio was less than 30 percent. This may be due to the strong fibers and better bondability of the bamboo fibers.

Table 4. Mechanical properties of pine-straw-bamboo-fiber composites.

Pine-straw / Bamboo-fiber (%)	70/30	30/70	0/100 (pure bamboo fibers)
Density (g/cm^3)	0.82	0.82	0.83
IB (Mpa)	0.28	0.62	1.06
MOR (Mpa)	15.9	23.3	22.6
MOE (10^3 Mpa)	2.3	3.6	3.3

SUMMARY AND CONCLUSIONS

Pine straw fibers obtained by steaming and refining pine needles were evaluated. The physical and mechanical properties of pine straw composites were assessed at different panel density, refining time, and resin content levels. Pine-straw-bamboo-fiber composites were made and evaluated. Pine straw materials used in this study mostly consisted of pine needle skins, fibrous bundles, particles, and a small amount of fibers and resin. The wax-coated outer skins and resin were the barriers for good bonding. This requires that more severe fiber processing treatment be applied to enhance the fiber content in the materials. Bending properties, IB strength, and thickness swell were responsive to the changes of refining time, panel density, and resin content The dimensional stability and IB strength of pine straw composites were poor. Pine straw fibers may be used to make composites with other strong and easy-to-bond fibers, such as wood and bamboo fiber. The bending and internal bond strength properties of pine-straw-bamboo-fiber composites were greater than pine straw

composites, especially when the pine-straw-fiber to bamboo-fiber ratio was less than 30 percent.

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