BINDERLESS PANELS MADE WITH BLACK SPRUCE BARK

Zhenhua Gao, a,* Xiang-Ming Wang, b Hui Wan, b and Gilles Brunette b

The bark of black spruce was thermo-mechanically refined and used to manufacture binderless bark-based fiberboard with various pressing temperatures, times, and panel structures in order to utilize an abundant bark resource for a better value-added application. The test results indicated that it is technically feasible to manufacture binderless fiberboard with refined black spruce bark through self-bonding under elevated temperatures over a reasonable period of pressing time. Binderless bark-based fiberboards with a homogeneous structure had very poor flexural properties due to the poor strength of bark itself; however, by using a sandwich structure with 30wt% wood fiber in the surface layers and 70wt% bark in the core layer it was possible to sufficiently improve panel flexural properties so that the manufactured binderless bark-based fiberboards was able to meet the mechanical property requirements of 115-grade fiberboard according to ANSI A208.2 (2009). Refining conditions had a great impact on the mechanical properties of binderless bark-based fiberboard.

Keywords: Bark; Binderless fiberboard; Refining conditions; Mechanical properties

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INTRODUCTION

Bark is an abundant and renewable bio-based material and is estimated to make up 15 to 20% of a tree by volume and 10 to 15% by mass (Wang and Xu 2003; Kofujita et al. 1999). However, most bark is underutilized because of its heterogeneous structure, diverse chemical composition, and very low strength compared to wood. The properties of bark are highly dependant on tree species, tree age, climatic growth conditions, and growth site (Harkin and Rowe 1971; Blanchet et al. 2000). Therefore, effective utilizations of bark with high value added approaches are conducted case by case, and more and more attention has been paid to the value-added utilizations of bark.

Some bark that contains abundant tannins has been utilized in the past to prepare metal ion removers and biopolymers. Metal ion removers with good adsorption ability to different metal ions can be prepared from Pinus radiata bark and tannins by chemically modifying them with an acidified formaldehyde solution (Palmaa et al. 2003). High-quality wood adhesives without or with very low formaldehyde emission can be prepared from tannin that is extracted from various species of bark such as pine, spruce, fir, wattle, chestnut, and so on (Kim 2009; Lee and Lan 2006; Vázquez et al. 2009; von Leyser and Pizzi 1991). Bark also can be converted by liquefying it in phenol at elevated temperatures in the presence of acidic catalysts into fusible and soluble chemical products for preparing some bio-based polymers such as adhesives, polyurethane (PU) foams, and

With declining forest resources and the rising cost of wood materials, researchers have been studying various ways of using different kinds of lignocellulosic residual materials for wood composites production since the 1970s (Velasquez et al. 2003). Medium density fiberboard (MDF) panels with acceptable bond strength and flexural strength can be made by refining black spruce bark (Xing et al. 2007); particleboards meeting ANSI indoor requirements can be also made from hammer-milled bark residues (Blanchet et al. 2000). However, UF resin as a binder is necessary for such bark panels, which leads to the emission of hazardous formaldehyde.

Technology of binderless panel is able to not only eliminate the emission of hazardous formaldehyde from panels but also to reduce the dependence of composites boards on petroleum-based synthetic resin. The work by Wellons and Krahmer (1973) and Chow (1975) has demonstrated the possibility of manufacturing binderless composite panels from ground bark residues. Chow (1975) obtained a high density self-bonding bark particleboard after hot pressing at 300°C with 1.23 MPa of internal bond strength and 10.80 MPa of modulus of rupture; however the pressing temperature at 300°C was too high for commercial uses due to the hazards of self-combustion of bark. Refining under thermo-mechanical conditions with the combination of high moisture content is believed to be able to improve the surface morphology and chemistry of refined lignocellulosic materials, and therefore increase reaction activity and adhesion of products refined. It was confirmed that the lignin content on the surfaces of refined wood increased (McDonald et al. 1999; Koljonen et al. 2003). Thermo-mechanical pretreatment was carried out to hydrolyze most of the hemicelluloses and to plasticize the lignin in Miscanthus sinensis, and the resulting fibers were able to be hot pressed to produce fiberboard without synthetic resin (Velasquez et al. 2003). Similarly, bark after refining is also believed to have improved surface properties, contributing to the self-bonding in manufacturing binderless bark panels. The main goal of this research is to evaluate the possibility of manufacturing binderless bark panels from refined black spruce bark with lowered pressing temperature and shortened cycle, and to optimize the manufacturing parameters for binderless bark panels.

**EXPERIMENTAL**

**Materials and Preparations**

Fresh black spruce (*Picea mariana*) bark just peeled off from flesh logs was supplied by a local sawmill in Quebec, Canada. After sand and stones were removed from the bark by a sieving machine equipped with a 10-mesh screen, the bark was oven-dried at 70°C for 24 hours, resulting in a moisture content of 2.7%.

Oven-dried bark was ground using a Wiley mill and then separated by 5-, 10-, 18-, and 48-mesh Tyler screens with various factions (5-10, 10-18, and 18-48 mesh, respectively, corresponding to particle sizes of 4.0-2.0, 2.0-1.0, and 1.0-0.356 mm). Water was added into oven-dried bark to increase its moisture content to around 47%. The wet bark was then refined with an Andritz disk refiner at FPInnovations’ MDF pilot
plant with various steam pressures (Pressure), plate distances (Distance), retention time (Time) and plate speed (Speed), as summarized in Table 1. After refining, the fiber was discharged through a blow-line and dried by a flash tube dryer to final moisture content about 5 to 7%. No wax and resin were injected during refining.

Table 1. Refining Parameters for Bark

<table>
<thead>
<tr>
<th>Refining ID</th>
<th>Distance (mm)</th>
<th>Pressure (bar)</th>
<th>Time (min)</th>
<th>Speed (rpm)</th>
</tr>
</thead>
<tbody>
<tr>
<td>I</td>
<td>1.0</td>
<td>12</td>
<td>5</td>
<td>2200</td>
</tr>
<tr>
<td>II</td>
<td>0.5</td>
<td>12</td>
<td>5</td>
<td>2200</td>
</tr>
<tr>
<td>III</td>
<td>0.1</td>
<td>12</td>
<td>5</td>
<td>2200</td>
</tr>
<tr>
<td>IV</td>
<td>0.1</td>
<td>7</td>
<td>5</td>
<td>2200</td>
</tr>
<tr>
<td>V</td>
<td>0.1</td>
<td>3</td>
<td>5</td>
<td>2200</td>
</tr>
<tr>
<td>VI</td>
<td>0.1</td>
<td>12</td>
<td>3.5</td>
<td>2200</td>
</tr>
<tr>
<td>VII</td>
<td>0.1</td>
<td>12</td>
<td>2</td>
<td>2200</td>
</tr>
</tbody>
</table>

Black spruce wood chips were refined using the Andritz disk refiner with the following refining parameters: steam pressure 12 bar, plate distances 0.1 mm, retention time 5 min, and plate rotational speed 2200 rpm. The moisture content of refined wood fibers was about 5.7%.

Panel Manufacturing

About 260 to 300 g of ground bark particles, refined bark fibers, or the mixture of wood fibers and refined bark fibers were used to form a mat with a dimension of 250 mm×250 mm, and the initial mat weight \(M_o\) and moisture content \(MC_o\) were measured. The mat was then immediately hot-pressed at temperatures ranging from 200 to 260°C for various period of times (6 to 20 min) under a pressure of 2.8 MPa for 1 min first and then 1.2 MPa for 5 to 19 min. Before trimming, the panel weight \(M_i\) and moisture content \(MC_i\) were measured again for determining the weight loss of panels during hot pressing. Two replicate panels were manufactured with each condition.

Some binderless bark particleboards were prepared from ground bark with various particle sizes as controls. The ground black spruce bark with moisture content of 5.02% was hot pressed at 260°C for 20 min, during which the pressure was held at 2.8 MPa for 1 min and then kept at 1.2 MPa for 19 min, resulting in binderless bark particleboard with a target thickness of about 6.5 mm. The stop bars were once used in an attempt to control the target density of binderless bark particleboards at 1000 or 1100kg/m³.

In this study, three panel structures were designed as follows: A - homogeneous structure with refined bark only as control; B - homogeneous structure with hybrid fibers by evenly mixing 30wt% wood fiber with 70wt% bark fiber; and C - sandwich structure by using 30wt% of black spruce wood fiber on two face layers and 70wt% of refined bark fibers in the core layer.

For homogeneous binderless bark panels prepared with various hot-press temperatures (200, 220, 240 and 260 °C) and time (20, 15, 12, 9 and 6 min), the bark fibers of refining batch VII in Table 1 with moisture contents ranging from 6.4 to 7.0%
were used. The forming and hot-press processes were similar to those for binderless bark particleboards as mentioned above.

For sandwich-structure (three-layer) panels, wood fibers were used for the two face layers of a panel (15wt% + 15wt% on a basis of total mass) and refined bark fibers for the core layer (70wt% on a basis of total mass). The binderless fiberboard panels were hot pressed at 260°C for 6 to 8 min without stop bars.

Panel Evaluation

Panel properties including internal bond (IB) strength, modulus of rupture (MOR), modulus of elasticity (MOE), and thickness swelling (TS) were measured in accordance with standard test methods of ANSI A208.1-2009 (ANSI 2009 for particleboard), ANSI A208.2-2009 (ANSI 2009 for fiberboard), and EN 317-1993 (for determining TS). Three specimens were used for determining panel density and IB, and 2 specimens for MOR and MOE for each panel. Weight loss (WL) of panel, referring to the percentage of solid mass lost during the hot pressing due to thermal degradation, was calculated by WL = \((M_2 - M_3)/M_2 \times 100\), where \(M_2\) was the solid mass of bark before hot pressing calculated by \(M_2 = M_0/(1+MC_0)\), and \(M_3\) was solid mass of bark panel after hot pressing calculated by \(M_3 = M_1/(1+MC_1)\).

Thermogravimetric Analysis (TGA) of Refined Bark Fibers

A Netzsch TG 209 F3 (Netzsch Co., Germany) thermogravimetric analyzer was employed to stimulate the thermal-degradation of refined bark fiber occurring during the hot pressing. After about 5 μg of refined powder bark was added into an Al₂O₃ crucible for the TGA test, the sample was quickly heated from 25°C to a target temperature (200, 220, 240 or 260°C) at a heating rate of 80°C/min, and then it was kept at the target temperature in the presence of air. The total period of heating time (TGA scanning time) was 20 min, the same as the longest hot-press circle used in panel manufacturing.

RESULTS AND DISCUSSION

Binderless Panels with Ground Bark Particles

Binderless bark panel can be prepared from bark without additional synthetic adhesive, but it needs a high hot-press temperature and a long hot-press time for bark self-bonding via polymerization of the phenolic extractives and lignin in bark that exhibit strong adhesive characteristics (Chow 1975). Shen (1991) believed that the self-bonding of lignocellulosic materials during hot pressing or molding operation included in-situ transformation, crosslinking, polymerization, and thermosetting processes of the free sugar, sugar polymers, dehydrated carbohydrates, furfural products, organic acids, lower-molecular water soluble compounds, and other decompositions in lignocellulosic materials.

In order to evaluate the effect of refining pretreatment on manufacturing binderless bark panels, some binderless panels were prepared from ground bark with various particle sizes as controls based on the studies of Chow (1975), Shen (1991), and Troughton and coworkers (1998). Effects of bark particle sizes and using stop bars on
some properties of bark particleboard panels are shown in Table 2. Obviously, these results confirmed that the ground bark could be hot-pressed into binderless particleboard panels with quite high bond strength. However, high board density, high pressing temperature, and long pressing time were all indispensable.

<table>
<thead>
<tr>
<th>Particle Size (mm)</th>
<th>Stop Bar</th>
<th>Board Density (kg/m³)</th>
<th>IB (MPa)</th>
<th>MOR (MPa)</th>
<th>MOE (MPa)</th>
<th>TS (%)</th>
<th>Weight Loss (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>2-4</td>
<td>No</td>
<td>996(12)</td>
<td>0.77(0.09)</td>
<td>7.15(1.56)</td>
<td>1924(155)</td>
<td>3.48(0.25)</td>
<td>20.5</td>
</tr>
<tr>
<td>1-2</td>
<td>No</td>
<td>1009(15)</td>
<td>1.11(0.24)</td>
<td>7.74(1.40)</td>
<td>1616(27)</td>
<td>3.31(0.09)</td>
<td>21.0</td>
</tr>
<tr>
<td>0.356-1</td>
<td>No</td>
<td>1029(17)</td>
<td>1.53(0.08)</td>
<td>6.16(0.77)</td>
<td>1202(150)</td>
<td>2.70(0.39)</td>
<td>19.0</td>
</tr>
<tr>
<td>0.356-1#</td>
<td>Yes</td>
<td>852(28)</td>
<td>0.13(0.02)</td>
<td>1.56(0.39)</td>
<td>231(130)</td>
<td>4.37(0.76)</td>
<td>19.3</td>
</tr>
<tr>
<td>0.356-1##</td>
<td>Yes</td>
<td>886(46)</td>
<td>0.42(0.15)</td>
<td>3.85(0.68)</td>
<td>754(205)</td>
<td>3.46(0.64)</td>
<td>18.9</td>
</tr>
</tbody>
</table>

Values in parentheses are standard deviations; Panel a and b were prepared with stop bars and target densities of 1000kg/m³ and 1100kg/m³, respectively.

When stop bars were used in the preparation of the first two panels with attempt to obtain panels with target densities of 1000 kg/m³ and 1100 kg/m³, the actual panel densities were 852 kg/m³ and 886 kg/m³, respectively, which were much lower than the target values due to both elastic and visco-elastic rebounds of compressed bark particles generated during the hot pressing. Consequently, these panels had much lower mechanical properties than those pressed without stop bars. In other words, a density higher than 950 kg/m³ was necessary for manufacturing binderless bark particleboard with sound mechanical strengths in terms of high IB, MOR, and MOE.

All these results indicated that intimate contact between bark particles under a high compression ratio was necessary to achieve self-bonding during hot pressing. The boards using bark particles sizes ranging from 0.356 to 1 mm without stop bars had IBs ranging from 1.11 to 1.53 MPa, more than the required IB for H3-grade particleboard (>0.90MPa) according to ANSI A208.1 (2009). However, the MOR and MOE were lower than the requirements for the H1-grade particleboard according to the ANSI (2009) (>14.9 MPa and >2160 MPa, respectively) due to the low strength of bark. Therefore, improving flexural property was another key point for this study.

The thickness swells of binderless bark particleboards ranged from 2.70% to 4.37%, which was much lower than the required values specified in EN 312-2003 for load-bearing boards for use in dry conditions (<16%) and in humid conditions (<11%). The lower thickness swelling should be attributed to the polymerization, crosslinking and/or other transformations of hydrophilic components of bark into hydrophobic products during hot pressing under elevated temperatures for a sufficient period of time.

### Homogeneous Binderless Bark Panels Using Refined Spruce Bark

The mechanical properties of binderless fiberboard panels prepared from refined black spruced bark fibers alone are shown in Table 3. It was found that the hot-press conditions used in this study resulted in fiberboard panels with density ranging from 985 to 1029 kg/m³. It was also found that higher pressing temperature showed more advantages for in-situ transformation, degradation, crosslinking, polymerization, and thermosetting processes of bark; consequently, more and more of the bark mass was
degraded, as shown by the weight loss of panel after hot pressing. The weight loss (WL%) of panels showed a linear relation to pressing temperature ($T$) as $WL\% = 0.2495T - 48.76$ ($R^2 = 0.9948$). As hot-press temperature increased from 200°C to 260°C, for 20-min pressing cycle, the IB of binderless bark fiberboards increased from 0.34 MPa to 1.42 MPa gradually. However, the flexural properties (MOR and MOE) started to increase when the temperature reached 240°C and then to decrease as hot-press temperature increased further, due to the strong degradation of bark, especially the bark on surface layers of panel that were directly contacted with hot platens, as confirmed by the TGA results shown in Fig. 1. When the refined bark was subjected to the hot-press temperature for 20 min, the TGA curve showed two apparently different zones: Zone I (or heating zone) during which the platens quickly brought the temperature of the bark from 25°C to the target levels (200-260°C), and Zone II (or the isothermal zone) in which the bark was kept isothermally at the target temperature. This TGA testing model could ideally stimulate the thermal-degradation of surface bark contacted with hot platens during the hot pressing because the temperature of refined bark sample in the TGA test quickly reached the temperature employed during hot-press and then was kept for the duration at the same temperature as the hot-press cycle. After being kept isothermally for 20 min, TGA curves in Fig. 1 clearly indicate that the higher hot-press temperature resulted in the more mass loss of the bark. The mass losses obtained by TGA (ranging from 76.7% to 90.3% corresponding the scanning temperature from 260°C to 200°C) were much higher than those given in Table 3, which were obtained by determining the mass losses of panel before and after hot pressing. This was likely attributable to much lower temperature of panel core than the panel faces (directly contacted with the hot plates), resulting in much less thermal degradation in the panel having bark in the interior. At hot-pressing temperatures of 240°C or higher, the IB of binderless bark fiberboard panels could meet the required value for the 155-grade fiberboard panels according to ANSI A208.2 (2009) (>0.81 MPa). Hot pressing at 220°C for 20 min achieved the specifications for 130-grade fiberboard, for which IB >0.54 MPa.

**Table 3. Properties of Homogeneous Binderless Bark Panels using Refined Bark**

<table>
<thead>
<tr>
<th>Press Temperature (°C)</th>
<th>Press Time (min)</th>
<th>Board Density (kg/m³)</th>
<th>IB (MPa)</th>
<th>MOR (MPa)</th>
<th>MOE (MPa)</th>
<th>TS (%)</th>
<th>Weight Loss (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>200</td>
<td>20</td>
<td>942(31)</td>
<td>0.34(0.06)</td>
<td>5.52(1.01)</td>
<td>471(115)</td>
<td>21.04(2.06)</td>
<td>0.7</td>
</tr>
<tr>
<td>220</td>
<td>20</td>
<td>1003(65)</td>
<td>0.81(0.10)</td>
<td>7.63(1.96)</td>
<td>762(180)</td>
<td>10.46(0.47)</td>
<td>6.7</td>
</tr>
<tr>
<td>240</td>
<td>20</td>
<td>1019(81)</td>
<td>1.02(0.38)</td>
<td>10.87(1.62)</td>
<td>1244(225)</td>
<td>4.88(0.64)</td>
<td>11.3</td>
</tr>
<tr>
<td>260</td>
<td>20</td>
<td>996(44)</td>
<td>1.42(0.15)</td>
<td>9.80(1.17)</td>
<td>1077(91)</td>
<td>3.88(0.09)</td>
<td>15.8</td>
</tr>
<tr>
<td>260</td>
<td>15</td>
<td>1009(58)</td>
<td>1.49(0.23)</td>
<td>9.52(1.87)</td>
<td>1160(159)</td>
<td>5.53(0.51)</td>
<td>13.8</td>
</tr>
<tr>
<td>260</td>
<td>12</td>
<td>1029(70)</td>
<td>1.43(0.35)</td>
<td>10.63(2.58)</td>
<td>1082(237)</td>
<td>5.70(0.67)</td>
<td>13.1</td>
</tr>
<tr>
<td>260</td>
<td>9</td>
<td>951(21)</td>
<td>1.00(0.13)</td>
<td>8.59(1.54)</td>
<td>883(119)</td>
<td>5.73(0.24)</td>
<td>11.9</td>
</tr>
<tr>
<td>260</td>
<td>6</td>
<td>985(66)</td>
<td>0.99(0.18)</td>
<td>8.38(0.72)</td>
<td>946(128)</td>
<td>6.38(0.66)</td>
<td>9.0</td>
</tr>
</tbody>
</table>

Values in parentheses are standard deviations.
Though hot-pressing at 220°C showed potential to produce binderless bark fiberboard, with the bond strength reaching the 130-grade requirement, and at 240°C reaching the 155-grade requirement for fiberboard, it took a long pressing time (20 min) to develop adequate bond strength, which is not practical for the commercial production of bark-based composite boards. However, the knowledge of modern polymer science states that the increase in temperature accelerates molecular and segmental motion of polymers, bringing the system more rapidly to equilibrium or apparent equilibrium and accelerating all types of visco-elastic behaviors, which is called time-temperature correspondence (Akinay et al. 2002; Strobl 1997). With this principle in mind, some more panels were manufactured at a higher pressing temperature (260°C) with shortened pressing time (6 to 15 min). The test results of these panels are also shown in Table 3. The data show that both IB and MOR properties of bark panels were improved when pressed at 260°C and 12 min, as compared to 240°C and 20 min. Chow and Pickles (1971) concluded that bark at temperatures above 180°C underwent chemical degradation and polymerization reactions; they attributed the strong adhesive characteristics of binderless bark panel at higher temperature to the polymerizations of phenolic extractives and lignin in bark. Therefore, the improved bond quality of bark at higher temperature was due to the more chemical degradation of bark at 260°C (in terms of more weight loss during hot pressing) that produced more components that were beneficial to self-bonding. However, further decreasing of the pressing time from 12 min to 6 min resulted in decreased IB strength and flexural properties due to the insufficient time for bark to develop self-bonding. This indicated that an increase in pressing time at 260°C resulted in increased level of bark degradation during hot pressing, and the weight losses of panels, WL, as a function of pressing time (t, from 6 to 20 min) can be expressed by the following equation: WL% =
5.3713Ln(t) - 0.3619 ($R^2 = 0.9824$). The above test results showed that the 6-min pressing time at 260°C was able to produce the 155-grade binderless bark fiberboard that achieved IB>0.81MPa according to ANSI A208.2 (2009). Though this hot-press time was longer than that used for producing wood composites with the addition of synthetic adhesive as binder, it was sharply reduced as compared to the pressing time used for the binderless bark panels made with the ground bark, which required 20 min to achieve adequate bonding strength, as shown in Table 2. This result was attributed to the improved surface reactivity and chemical properties of refined bark after the thermo-mechanical pretreatment via refining. However, all bark panels with various pressing temperatures or times had MOR and MOE values that were lower than those required for either 155-grade (>27.9MPa and >2792MPa) or 115-grade (>12.4MPa and >1241MPa) fiberboard panels according to ANSI A208.2 (2009) due to the low strength of bark fiber itself. Thus, improving flexural properties of bark fiberboards became a key issue for producing binderless bark panel with respect to potential commercial applications.

The thickness swellings of binderless bark fiberboards decreased gradually from 21.04% to 3.88% as the hot-pressing temperature increased from 200°C to 260°C, indicating that higher hot-pressing temperature benefited to the transformations of hydrophilic components of bark into hydrophobic products. As the hot-pressing time decreased from 20 min to 6 min, the TS just showed a slight increase from 3.88% to 6.38%. All binderless bark fiberboards except the one prepared at 200°C for 20 min had TS lower than the required values specified in EN 622-3-2004 for general purpose high-density fiberboards for use in dry conditions (<15%) and in humid conditions (12%), showing good moisture resistance.

The results in Table 2 also indicate that the particle size had a great impact on the mechanical properties of the resulting binderless bark boards. As particle size decreased gradually, the bond strength (IB) increased obviously from 0.77 to 1.53 MPa. These facts gave clear evidence that the grinding treatment improved the crosslinking or self-bonding ability of bark, which technically showed the possibility to reduce hot-pressing temperature and time. Likewise, the bark underwent more severe treatment during the thermo-mechanical refining in the presence of high moisture, which would lead to much better self-bonding ability of bark and technical possibility to reduce hot-pressing temperature and time. In addition, all bark panels lost 18.9 to 21.0% of their mass during the hot pressing due to the chemical degradation of bark at a high temperature (260°C) over a long pressing time (20 min). Therefore, reducing hot pressing temperature or time is also necessary to avoid mass loss of resultant panels.

**Effects of Panel Structure on Properties of Binderless Fiberboards**

By hot pressing at high temperatures (200-260°C) for a proper cycle, the binderless bark panels showed good internal bond strength for the self-bonding. However, these panels had very poor flexural strength due to the poor mechanical properties of bark itself. Wood fiber is usually stronger than bark fiber because of higher cellulose content and much less of other components such as free sugar, sugar polymers, dehydrated carbohydrates, furfural compounds, organic acids, and other lower-molecular water soluble materials. With this concern, some wood fibers were added, in order to improve flexural properties of bark panels, by forming 3 panel structures.
When 30wt% wood fiber was mixed even ly with 70wt% bark fiber and hot-pressed at 260°C for 6 min, it produced a homogeneous binderless hybrid fiberboard (Panel structure B) with an IB of 1.16 MPa that was 17.2% higher than that of a control panel made from bark fiber only (Panel structure A). This IB was more than the required values for 155-grade fiberboard panels according to ANSI A208.2 (2009) (>0.81MPa). Due to the introduction of 30% wood fiber with better strength properties, the MOR and MOE of binderless hybrid fiberboard were also improved (by 15.8% and 7.3%, respectively); however, they were still much lower than those required for either 155-grade (>27.9MPa and >2792MPa) or 115-grade (>12.4MPa and >1241MPa) fiberboard panels according to ANSI A208.2 (2009). These results indicated that there was still a limitation for improving the flexural properties of bark panels through mixing wood fiber with bark fiber. Though the TS (11.56%) was much higher than control (0.38%), it was still lower than required values specified in EN 622-3-2004 for general purpose high-density fiberboards for use in dry conditions (<15%) and in humid conditions (12%).

Table 4. Properties of Binderless Bark Panels with Various Panel Structures

<table>
<thead>
<tr>
<th>Panel Structure</th>
<th>Press Time (min)</th>
<th>Board Density (kg/m³)</th>
<th>IB (MPa)</th>
<th>MOR (MPa)</th>
<th>MOE (MPa)</th>
<th>TS (%)</th>
<th>Weight Loss (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>A</td>
<td>6</td>
<td>985(66)</td>
<td>0.99(0.18)</td>
<td>8.38(0.72)</td>
<td>946(128)</td>
<td>0.38(0.66)</td>
<td>9.0</td>
</tr>
<tr>
<td>B</td>
<td>6</td>
<td>967(47)</td>
<td>1.16(0.04)</td>
<td>9.70(1.61)</td>
<td>1105(129)</td>
<td>11.56(0.47)</td>
<td>7.8</td>
</tr>
<tr>
<td>C</td>
<td>6</td>
<td>943(24)</td>
<td>0.62(0.12)</td>
<td>14.17(1.62)</td>
<td>1858(157)</td>
<td>11.91(0.57)</td>
<td>10.4</td>
</tr>
<tr>
<td>C</td>
<td>8</td>
<td>954(12)</td>
<td>1.09(0.29)</td>
<td>15.70(0.07)</td>
<td>1973(114)</td>
<td>11.67(0.42)</td>
<td>10.9</td>
</tr>
</tbody>
</table>

Values in parentheses are standard deviations.

In terms of flexural properties (MOR and MOE), which are dependent on the material strength and bond quality of surface layers of test panel, a sandwich structure of panel (Panel structure C) was proposed by placing a total of 30wt% wood fiber on both faces of panel with bark fiber in the core. The test results are given in Table 4. It was observed that the flexural properties of sandwich bark panels were apparently improved (panel coded C). The MOR and MOE of these panels were 14.17 MPa and 1858 MPa, respectively, corresponding to 69.1% and 96.4% of improvement as compared with those of controlled homogeneous panels with bark only. The binderless bark fiberboard panels with the sandwich structure met the requirements for the 115-grade (>0.47 MPa for IB, >12.4 MPa for MOR, and >1241MPa for MOE) fiberboard panels according to ANSI A208.2 (2009). However, the sandwich structure also led to decreasing IB strength from 0.99 MPa to 0.62 MPa under the press cycle of 6 min. This result might be attributed to the heat-transfer hindrance of the wood fiber on the face layers of panel, resulting in insufficient conduction of energy to bring about the well self-bonding of bark. To verify this speculation, another sandwich fiberboard was prepared with a longer pressing time (8 min). The test result of this panel showed that the IB was apparently increased from 0.62MPa to 1.09MPa or a 76% improvement compared to that hot-pressed for 6 min, while the TSs remained almost the same.
Effects of Refining Parameters on Properties of Binderless Fiberboards

Many previous studies have shown that thermal–mechanical refining conditions have significant effects on final MDF panel properties. Krug and Kehr (2001) suggested that increasing the steam pressure resulted in shorter fiber lengths, lower strength, and weaker elastic properties. Xing et al. (2006) reported that both preheating retention time and steam pressure or refining temperature directly affected the properties of MDF panels with bark bonded by UF resin. Based on these concerns, the effects of bark refining parameters on the mechanical properties of binderless fiberboards were investigated in this work. The test results of binderless panels made various bark fibers are shown in Table 5. All these panels had a sandwich structure, i.e., wood fibers on both face layers (15wt%/15wt%) and bark fiber in the core layer (70wt%) and were hot pressed at 260°C for 8 min without stop bars.

Table 5. Bark Refining Parameters and Properties of Resultant Binderless Bark Fiberboards

<table>
<thead>
<tr>
<th>Refining ID</th>
<th>Refining Parameters</th>
<th>Panel Properties</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Distance (mm)</td>
<td>Pressure (bar)</td>
</tr>
<tr>
<td>I</td>
<td>1.0</td>
<td>12</td>
</tr>
<tr>
<td>II</td>
<td>0.5</td>
<td>12</td>
</tr>
<tr>
<td>III</td>
<td>0.1</td>
<td>12</td>
</tr>
<tr>
<td>IV</td>
<td>0.1</td>
<td>7</td>
</tr>
<tr>
<td>V</td>
<td>0.1</td>
<td>3</td>
</tr>
<tr>
<td>VI</td>
<td>0.1</td>
<td>12</td>
</tr>
<tr>
<td>VII</td>
<td>0.1</td>
<td>12</td>
</tr>
</tbody>
</table>

Values in parentheses are standard deviations.

Figure 2. Refined bark fibers (Batch I, Batch II, and Batch III, as indicated in Table 5, from left to right)

When the plate distance decreased from 1 mm to 0.1 mm, the refined bark fibers became less coarse, as shown in Fig. 2. This result indicated that the bark fibers refined with a smaller plate distance were finer due to the smaller plate distance or larger mechanical force imposed on the bark, resulting in more friction between bark materials.
during refining and more activated fibers for the self-bonding of bark. Consequently, the binderless fiberboard made from the refined bark with 0.1 mm plate distance had much higher IB and lower TS, as compared with other plate distances (0.5 and 1.0 mm). However, the plate distance had a slight effect on MOR and MOE of the resulting fiberboards. Therefore, the plate distance was set at 0.1 mm for further investigating the effects of other refining parameters.

Steam pressure correlates to the temperature of steam for heating bark during refining. Steam pressures at 12, 7, and 3 bars are equivalent to the steam temperatures of 188, 165, and 133°C, respectively. The results in Table 5 indicate that the binderless fiberboards prepared from the bark fibers refined at steam pressures of 12 and 7 bars had much higher IBs (1.82 MPa and 1.85 MPa, respectively) and lower TSs (7.65% and 8.47%, respectively) than those from the bark fibers refined at 3 bars. This was probably attributable to the improved surface reactivity due to some physical and chemical transitions of some components of bark at elevated refining temperatures. These transitions also led to some effects on the MOR and MOE. The binderless bark fiberboards prepared from the bark fibers refined at 7 and 12 bars of steam pressure had the highest and lowest flexural properties, respectively.

Generally, the bark refined with a long preheating retention time corresponds to the bark being fed slowly and allows more time for it to be preheated and softened in a digester; such bark has a better chance to be activated during refining, leading to better self-bonding ability. The data in Table 5 confirm that binderless bark fiberboards had improved IB strength as the preheating retention time of bark increased from 2 min to 5 min. With respect to the MOR property, the 3.5-min retention time resulted in a strong binderless bark panel (20.25 MPa).

The above results confirmed that bark refining parameters had great impact on the mechanical properties of binderless bark fiberboards. Though all binderless bark fiberboards achieved IB, MOR, and MOE values that met the requirements of 115-grade fiberboard panels (>0.47MPa, >12.4MPa and >1241MPa, respectively) according to ANSI A208.2 (2009), the best refining parameters were: plate distance of 0.1 mm, steam pressure of 12 bars, and preheating retention time of 3.5 min.

CONCLUSIONS

1. It is technically feasible to manufacture binderless fiberboard from refined black spruce bark with the mechanical properties meeting the requirements of 115-grade fiberboard according to ANSI A208.2 (2009) by hot pressing refined bark at 260°C for 6 min.
2. In comparison with the grinding treatment, the thermo-mechanical treatment of bark via refining could more effectively lower the required pressing temperature and shorten the pressing time needed for manufacturing binderless bark fiberboard.
3. A sandwich structure with wood fiber at each surface layer (15wt% of the panel weight) and refined bark in the core layer (70% of the panel weight) sufficiently improved flexural properties of binderless fiberboard.
4. Refining parameters had great effect on the mechanical properties of resultant binderless bark fiberboard, and optimized refining parameters were: plate distance of 0.1 mm, steam pressure of 12 bars, and preheating retention time of 3.5 min.

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