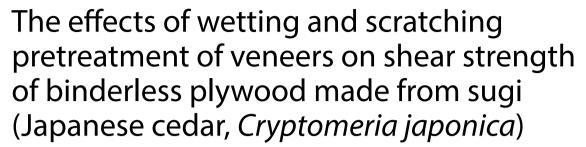


ORIGINAL ARTICLE

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Abstract

To produce practicable binderless laminated veneer lumber (LVL) and plywood, their water resistance needs to be improved. The objective of this study was to manufacture highly water-resistant binderless plywood from sugi (Japanese cedar, *Cryptomeria japonica*), basing on a self-bonding mechanism, without any resins or powders. To achieve this, a pretreatment of wetting and scratching veneers was applied. Two types of wetting conditions, soaking for 12 h, and wetting for 1 min, were examined. Sandpaper or a wire brush was used for scratching. Three veneers were laminated in perpendicular directions and hot pressed at 200–220 °C for 20–45 min using two types of pressing schedule: simple 1-step pressing, or 4-step pressing while gradually increasing the pressure. As a result, whichever wetting time was employed, and regardless of the scratching tool used, plywood could be manufactured with both pretreatments. As the wet shear strength showed a comparable value to dry shear strength, the plywood had high water resistance. The highest wet shear strength was 0.6 MPa, for the plywood made by pretreatment wetting of the surface for 1 min and scratching with a wire brush followed by 1-step pressing at 220 °C for 20 min. This was fairly close to the Japanese Agricultural Standard (JAS) requirement. This study indicated that easy pretreatment and mild pressing conditions achieved a binderless plywood with high water resistance.

Keywords: Plywood, Binderless, Self-bonding, Water resistance, Sugi

Introduction

Bonding without adding any adhesives is called self-bonding. Many kinds of self-bonded binderless particle boards or fiber boards have been manufactured from various biomass, making them environmentally friendly. The raw materials are reduced to a small size and then formed into a mat in a frame, then hot pressed under high temperature and pressure. The moisture in the raw materials and high pressing temperature and pressure are considered to provide the self-bonding property.

This method of self-bonding has been applied for laminated veneer lumber (LVL) and plywood recently. LVL, in which veneers are laminated with parallel grain

directions, was successfully manufactured without adding any adhesives [1, 2]. It was observed that the bonding line was curved, showing that during compression the cells at the surface of one of the veneer layers tried to find room to match into the other layer. In contrast, plywood, in which veneers are laminated with perpendicular grain directions, has not been thoroughly examined. Although it was reported to be possible to manufacture plywood from beech veneer, the shear strength was three times lower than that of the corresponding LVL product [1]. The alternating orientation of the grain seemed to be a disadvantage, because incapacity of the cells to entangle at the bonding line due to the orthotropic structure of wood was observed [1]. One method to improve the bonding strength of binderless plywood is to use botanical powders. The powder plays the role of binder, filling the space among veneers [3, 4]. Scratching the surface

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of veneers to increase the roughness of the surface and entangling the veneers with each other might be another method to improve the bonding of plywood.

Weak water resistance is a disadvantage of self-bonding. Dry shear strength has been reported, but wet shear strength has not despite both European Norm (EN-314) [5] and Japanese Agricultural Standard (JAS) [6] requiring the measurement of wet shear strength as an accelerated deterioration test [2, 7]. It was reported that the binderless LVL specimens manufactured by pressing at 200 °C were delaminated during soaking in water and their wet shear strengths could not be measured. With the increasing temperature, an increase in the dry shear strength was also observed. However, binderless LVL could not be formed at high temperatures such as 260 °C [2]. To achieve high water resistance and wet shear strength, alternative methods that allow shape forming are necessary, even if pressed at lower temperature. It was found that two methods were effective in improving water resistance: wetting veneers before hot pressing, and heat treatment after hot pressing [8]. It was suggested that the bond integrity increased because the ratio of delamination occurrence decreased when samples were soaked in water, and that moisture improves the formability of lignocellulose materials, most likely by improving the mobility of the lignin-hemicellulose matrix and by increasing the contact area [8].

The objective of this study was to manufacture highly water-resistant binderless plywood without adding any resins or powders. It was required that the wet shear strength satisfy the 0.7 MPa JAS requirement for softwood [6]. To achieve high water resistance of binderless

plywood, wetting pretreatment before hot pressing and scratching of veneers was applied. The efficiency of wetting and scratching of veneers before hot pressing was examined.

Materials and methods

Raw materials

Rotary-cut commercial sugi (Japanese cedar, *Cryptomeria japonica*) veneers were obtained from Seihoku Company (Ishinomaki, Japan). Only heartwood was used for manufacturing plywood, because it was reported that the internal bonding of binderless particleboard made from sugi heartwood was higher than that of sapwood [4]. The thicknesses of veneers were 3.0-3.2 mm. Veneers were cut to a size of 200×200 mm, and stored in an airdried state to keep the moisture content at 6-8%.

Pretreatment

Table 1 shows the pretreatment and hot pressing conditions. Wetting pretreatments were carried out to increase moisture content throughout or only on the surface of veneers. To increase the moisture content of whole veneers, they were soaked in tap water at 20 °C for 12 h. The excess water was removed by pressing under a weight of 3 kg to minimalize the variation of moisture content of samples. The final moisture content was adjusted to approximately 50–60%. The other veneers were wet with running water for a minute to increase the moisture content only at their surfaces. The moisture contents of resultant veneers were approximately 12%.

After the wetting treatment, the veneers were scratched with sandpaper (#40) or a steel wire brush (SUN UP

Table 1 Pretreatment and pressing conditions of plywood

Sample	Pretreatment conditions				Pressing conditions		
	Time for soaking in water	Tool for scratching	Scratching direction	Scratching times	Temperature, °C	Pressure schedule	Time, min
A	12 h	Sandpaper (#40)	Open and close	50–60 sets	220	1-step	30
В							45
C						4-step	
D					200		
E	12 h	Wire brush	Open	20-40 times	220	4-step	45
F				40-60 times			
G				60-80 times			
Н			Close	40-60 times			
1	1 min	Wire brush	Open	40-60 times	220	1-step	30
J			Circular	50 sets			
K				100 sets			
L				50 sets			20
М							45

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Fig. 1 Steel wire brush for scratching veneers. **a** The side of the brush. **b** The bottom of the brush

Channel brush No. 29457, Taiyo shokai Co. Ltd., Osaka,

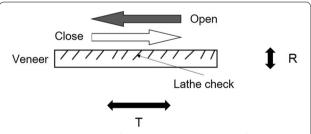


Fig. 2 The cross-section of a veneer and the direction of scratching to open or close lathe checks

Japan) shown in Fig. 1, to compare the coarseness of scratching tools. With sandpaper, the scratching direction was perpendicular to the longitudinal direction to open and close lathe checks as shown in Fig. 2. Hereby we call "1 set" as scratching once in each direction. For the wire brush, two patterns were applied. The first pattern was scratching linearly to open or close lathe checks as shown in Fig. 2. To scratch all the surfaces of a veneer equally, a range of the number of scratching times was

necessary. The second pattern was scratching in a circular pattern as shown in Fig. 3, where "1 set" consists of four small circular scratchings.

Consequently, three pretreatment patterns were used in this study, by combining wetting and scratching treatments: combination of 12-h wetting and sandpaper scratching (Table 1A–D), combination of 12-h wetting and wire brush scratching (Table 1E–H), and combination of 1-min wetting and wire brush scratching (Table 1I–M).

Manufacture of binderless plywood

Three veneers were laminated in perpendicular directions to each other. Distance bars were set on both sides of the veneers to keep the thickness of resultant plywood constant, and to avoid explosion caused by excessive compression of the veneers. The compression rate was determined by the ratio of the target thickness of the plywood to the sum of the three veneers before hot pressing and was kept constant at 40–50% for all samples.

Two different types of pressing schedules were applied: 1-step pressing at 5 MPa constant pressure, and 4-step pressing. For the latter, the pressure was 0 MPa for the first 5 min, raised to 3 MPa for the next 5 min, reduced to 0 MPa again for another 5 min, and finally increased to 5 MPa for 30 min. The pressing temperature was constant at 200 or 220 °C for both types of pressing schedule. Two plywoods were manufactured under each manufacturing condition.

Shear strength test

Shear strength testing was carried out according to JAS for plywood [6]. The test specimens were cut to a size of 75×25 mm. Ten specimens from each manufacturing condition were tested. Shear strength was measured in dry conditions, and in wet conditions after soaking in 60 °C water for 3 h.

Observation by scanning electron microscopy (SEM)

The veneers pretreated with 1-min wetting and wire brush scratching were provided for SEM microscopic study to examine the relationship between surface roughness and shear strength. They were coated with platinum–palladium after drying at room temperature. A field emission scanning electron microscope (S4800; Hitachi, Japan) was employed with accelerating voltage at 1.0 kV for microscopic observation.

Results

12-h wetting and sandpaper scratching

When neither wetting nor scratching pretreatment was applied, the sugi veneers did not bond. Additionally, scratching pretreatment alone was not sufficient to make Kurokochi et al. J Wood Sci (2019) 65:15 Page 4 of 7

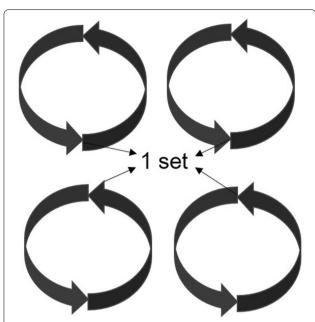


Fig. 3 The direction for circular scratching of a veneer. One set consists of four partial scratches to cover the surface area of the veneer

plywood, because delamination occurred during cutting of the resultant plywood. When both pretreatments were applied, plywood with a high self-bonding property could be manufactured in this study.

Figure 4 shows the dry and wet shear strengths of the plywood when the pretreatment combining 12-h wetting and sandpaper scratching was applied

(Table 1A-D). Comparing the shear strengths of A and B, longer pressing time did not result in a major increase of shear strength. It was suggested that 4-step pressing was a better method than 1-step pressing, as the shear strength of C exhibited a smaller standard deviation than that of B, although the average values were similar. The reason for this was that 4-step pressing could release vapor gradually and avoid explosive delamination during hot pressing. The shear strength of C was higher than that of D, reflecting the fact that a higher pressing temperature resulted in a higher self-bonding property. Nevertheless, sugi plywood could be bonded when pressed at 200 °C, although beech LVL pressed at the same temperature delaminated during soaking in water [2]. Sugi veneers successfully self-bonded at lower temperatures than the 240-260 °C range used for beech binderless plywood [1].

12-h wetting and wire brush scratching

Figure 5 shows the shear strengths of the plywood when the pretreatment combining 12-h wetting and wire brush scratching was applied (Table 1E–H). Comparing E–G, the wet shear strength tended to increase with increasing scratching time. It might be supposed that scratching treatment increased the bonding area. The shear strength of H was lower than that of E, despite the scratching number of times being more than E. This indicated that scratching in the direction to open lathe checks might be more suitable than that in the close direction (Fig. 2), for the purpose of increasing the bonding area. The shear strength of G was almost the same as that of C as shown in Fig. 4. It was considered that the wire brush could have the same effect as sandpaper with less scratching work.

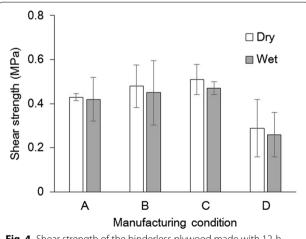


Fig. 4 Shear strength of the binderless plywood made with 12-h wetting and sandpaper scratching pretreatment under various conditions as indicated in Table 1A–D. The error bar shows the standard deviation

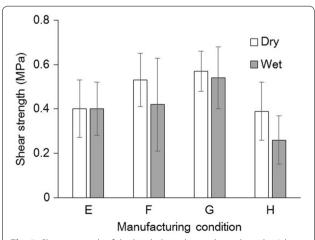


Fig. 5 Shear strength of the binderless plywoods made with 12-h wetting and wire brush scratching pretreatment under various conditions as indicated in Table 1E–H. The error bar shows the standard deviation

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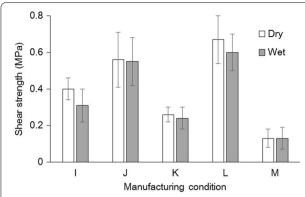


Fig. 6 Shear strength of the binderless plywood made with 1-min wetting and wire brush scratching pretreatment under various conditions as indicated in Table 1I–M. The error bar shows the standard deviation

1-min wetting and wire brush scratching

Figure 6 shows the shear strengths of the plywood when the pretreatment combining 1-min wetting and wire brush scratching was applied (Table 1I–M). The shear strength of I at 1-min wetting was lower than that of F at 12-h wetting (Fig. 5). However, the highest shear strength in our study was seen for L at 1-min wetting. This confirms that 12-h wetting is not always more effective than 1-min wetting.

Some tracheids and ray parenchyma cells were observed in the SEM image of the untreated surface of veneer (Fig. 7a). They could be still recognized where they were, even though the surface got rough because of wire brush scratching with the aim to open lathe checks (Fig. 7b). The surface roughness further advanced at the veneer after the pretreatment of circular wire brush scratching for 50 sets so that a tracheid was not observed (Fig. 7c). However, the surface roughness was not progressed, ever though the scratching number of times was raised to 100 sets (Fig. 7d). Excess scratching might rather whittle and flatten the uneven surface of veneer. The shear strength of J was higher than I and K. The reason might be that the bonding area became larger with increasing surface roughness.

Comparison of J, L, M indicated that a shorter pressing time increased the shear strength. High moisture content of only the outer veneer might dry quickly during hot pressing, so there is no need for 4-step pressing that releases vapor gradually, or a long pressing time. This trend was different from the samples made with the 12-h wetting pretreatment. As a result, the pretreatment of 1-min wetting made it possible to produce binderless plywood with a simple manufacturing process, requiring less scratching work and 1-step pressing for 20 min.

Discussion

High water resistance of binderless plywood made from sugi heartwood

Binderless laminated veneers have suffered from weak water resistance, with little research into wet shear strength values so far. Only thickness swelling after water soaking for 48 h was measured in binderless LVL to evaluate water resistance [2]. As a result, thickness swelling tended to decrease with increasing pressing temperature but was not related to dry shear strength [2]. In the present study, when pretreatment combining wetting and scratching of veneers was applied, the resultant plywood did not delaminate in water during the test procedure for wet shear strength measurement. Indeed, the water resistance of our plywood was high to such an extent that the wet shear strength was similar to the dry shear strength.

It was reported that wetting pretreatment for 24 h increased the water resistance of the birch binderless plywood [8]. Moisture in wood reduces both thermal softening temperature of lignin and thermal decomposition temperature of hemicellulose. These changes enable to self-bond lignocellulose materials during hot pressing. According to this mechanism, our self-bonding of sugi plywood was also improved by 12-h wetting pretreatment under relatively lower temperature as 220 °C, resulting in high water resistance. As the plywood made with 1-min wetting pretreatment also showed high wet shear strength, it was suggested that this bonding mechanism would work if at least the adhesive surface is wet.

It is generally known that the lignin content of sugi is approximately 33% [9, 10], higher than those beech and birch, whose veneers were used for binderless LVL or plywood, which are approximately 20% [11, 12]. Lignin has high water resistance, and has been suggested to contribute to self-bonding. Additionally, sugi heartwood is rich in extractives. The chemical components of sugi might be another reason for the high water resistance of the plywood.

Simple manufacturing conditions

We found that pretreatment by wetting only the surface of veneers was also effective in increasing the self-bonding of the plywood, along with shorter pressing time. Mansouri et al. [7] applied 225–250 °C of pressing temperature for 60–75 min and successfully produced binderless LVL from beech. Cristescu [1] found that a pressing temperature higher than 240 °C was needed to make binderless plywood. All these reports indicate that pressing at high temperature is necessary to make binderless laminated wood without a wetting pretreatment. When a relatively low temperature such as 160 °C

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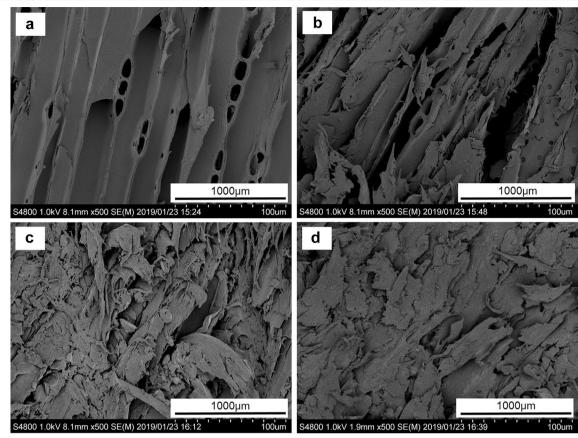


Fig. 7 SEM images of the surface of veneers (×500). **a** Untreated veneer with lathe checks. **b** The veneer which was pretreated with 1-min wetting followed by linear wire brush scratching to open lathe checks for 40–60 times. **c** The veneer which was pretreated with 1-min wetting followed by circular wire brush scratching for 50 sets. **d** The veneer which was pretreated with 1-min wetting followed by circular wire brush scratching for 100 sets

was used for pressing, a longer pressing time such as 2–4 h was required to make binderless LVL with wetting pretreatment [8]. In contrast, we found that neither a high temperature nor a long pressing time was needed for the self-bonding of sugi: sugi binderless plywood showed the highest wet shear strength when the veneers were pressed at 220 °C for 20 min. These mild pressing conditions might be advantageous when considering practical production in the future.

The highest score of wet shear strength in this study was 0.6 MPa, which was fairly close to the 0.7 MPa JAS requirement [6]. A previous study reported that sugi plywood bonding with sugi powder satisfied the requirement of JAS [4]. However, the operation required great effort, including the process of making fine sugi powder with a ball mill and spreading it equally over a veneer. In contrast, the method in our study requires pretreatment of wetting for just 1 min followed by scratching with a wire brush, which is much easier. Further study into the detailed manufacturing

conditions should raise the shear strength sufficiently to satisfy the JAS standard.

Conclusions

By applying a pretreatment combining wetting and scratching of veneers, 3-ply sugi binderless plywood could be manufactured without adding any resins or powders. Both dry and wet shear strengths were evaluated according to JAS standard testing [6]. It was found that the combination of pretreatment conditions affects shear strength:

- 1. Sugi binderless plywood showed sufficiently high water resistance, as wet shear strength exhibited a comparable value to dry shear strength.
- 2. Wetting pretreatment was effective for self-bonding irrespective of wetting time for 1 min or 12 h. Although 12-h wetting needed 4-step pressing with a long pressing time to release vapor gradually, 1-min

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- wetting made it possible to employ 1-step pressing with a shorter time.
- 3. Scratching pretreatment was effective for establishing bonding between the veneers laminated in perpendicular directions. Using a wire brush required less scratching work compared to sandpaper.
- 4. The wet shear strength showed the highest value of 0.6 MPa when the pretreatment combining wetting the surface and circular scratching with a wire brush was applied, followed by 1-step hot pressing at 220 °C for 20 min. This result was fairly close to 0.7 MPa, the requirement of JAS. Sugi binderless plywood could be manufactured with a simple pretreatment and mild pressing conditions.

Authors' contributions

MS made substantial contributions to conception and design of the study and assisted in the preparation of the manuscript. WH contributed to design of the study and acquisition of data. YK contributed to analysis and interpretation of data, and was a major contributor in writing the manuscript. All authors critically reviewed the manuscript. All authors agree to be accountable for all aspects of the work in ensuring that questions related to the accuracy or integrity of any part of the work are appropriately investigated and resolved. All authors read and approved the final manuscript.

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Competing interests

The authors declare that they have no competing interests.

Availability of data and materials

The datasets used and/or analyzed during the current study are available from the corresponding author on reasonable request.

Consent for publication

Not applicable.

Ethics approval and consent to participate

This article does not contain any studies with human participants or animals performed by any of the authors.

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