DOI: 10.4067/s0718-221x2023000100408

# EFFECTS OF GLUE SPREADING RATE AND VENEER DENSITY ON SUGI (*Cryptomeria japonica*) PLYWOOD ADHESIVE PENETRATION

Ayuni Nur Apsari<sup>1</sup>

https://orcid.org/0000-0002-1003-8219

Takashi Tanaka<sup>2,</sup>\*

https://orcid.org/0000-0002-7074-7703

# ABSTRACT

An optimum adhesive penetration is needed to provide satisfactory bonding strength at the veneer-veneer interface. The effect of veneer density and glue spreading rate on the phenol formaldehyde adhesive penetration plot profile was determined. An X-ray apparatus was used to visualize the adhesive penetration of the plywood. The heartwood and sapwood veneer of *Cryptomeria japonica* with low, medium, and high veneer densities were made into plywood. The glue spreading rate was applied from 75 g/m<sup>2</sup> to 225 g/m<sup>2</sup> for the heartwood plywood and up to 300 g/m<sup>2</sup> for the sapwood plywood (plus 75 g/m<sup>2</sup> at every level of glue spreading rate). An X-ray apparatus with a low tube voltage successfully visualized the adhesive penetration plot profile. Based on the half-width calculation, the adhesive penetration depth ranged from 0,3 - 0,9 mm. The mean half-width was 0,5 mm. The adhesive concentration increased with increasing glue spreading rate. In contrast, it also showed that using different veneer densities and increasing glue spreading rates does not affect the half-width value as the adhesive penetrates deeper.

Keywords: Adhesive penetration, phenol-formaldehyde, spreading rate, veneer density, X-ray densitometry.

### **INTRODUCTION**

Plywood is a wood product that made from wood veneers glued together with their grains in a perpendicular direction. The important thing of the quality and durability of plywood depends on the adhesive used in manufacturing (Asif 2009). Phenol formaldehyde (PF) is one of the most common moisture-durable adhesives used in plywood. Phenol formaldehyde adhesives are usually used in plywood manufacturing for structural applications in exterior conditions (Kurt 2010, Kurt and Cil 2012). The flow of PF adhesive into the wood cellular structure and infiltration into the cell walls create the wood adhesive interphase (Jakes *et al.* 2015). The pressure applied on the PF adhesive will affect the adhesive penetration (Cognard 2005).

It is important to investigate the adhesive penetration through the adhesive interphase on plywood. Tanaka (2018) developed a geometrical model of wood cells and adhesive penetration at the veneer-veneer interface of plywood. Adhesive penetration is categorized into four scales of penetration, namely, macroscopic penetration, microscopic penetration, nanoscale penetration, and angstrom-scale penetration (Laborie 2002). The depth

<sup>&</sup>lt;sup>1</sup>Gifu University. The United Graduate School of Agricultural Science. Science of Biological Resources. Gifu, Japan.

<sup>&</sup>lt;sup>2</sup>Shizuoka University. Faculty of Agriculture. Department of Bioresource Sciences. Shizuoka, Japan.

<sup>\*</sup>Corresponding author: tanaka.takashi@shizuoka.ac.jp

Received: 01.02.2022 Accepted: 25.09.2022

2

of adhesive penetration is influenced by adhesive parameters (viscosity), substrate parameters (grain angle; earlywood or latewood), and processing parameters (bonding pressure; open time) (White 1975). Optimum adhesive penetration information is important of the efficiency in using the adhesive (Kurt and Cil 2012).

X-rays can be used to investigate adhesive penetration until microscopic penetration. Since the last century, X-ray has been used for wood inspection (Worschitz 1932, Fisher and Tasker 1940, Tomazello *et al.* 2008) consequently, considering the X-ray apparatus as a potential tool for quantitative analysis. Based on research conducted by Tanaka and Shida (2012), the X-ray apparatus can be used to quantitatively measure the moisture content distribution in plywood. However, limitation of knowledge regarding the quantitative analysis of adhesive penetration in plywood requires further research. In a study by Ferrtikasari *et al.* (2019), it was stated that the qualitative analysis of the X-ray computed tomography image can clearly show the penetration of PF adhesive into lathe check on plywood at a glue spreading rate of 225 g/m<sup>2</sup> and elucidate the relationship between increasing glue spreading rate of plywood and its thermal conductivity (Ferrtikasari *et al.* 2019). Research conducted by Tanaka *et al.* (2015) stated that the PF adhesive in wood can be visualized clearly because the mass attenuation coefficient is different between wood and PF. Thus, the PF adhesive visible to visualize in the wood using X-rays (Tanaka *et al.* 2015).

This study focused to understand PF adhesive penetration size which define as half-width. The effect of increasing glue spreading rate, different wood veneer density, and wood part (heartwood and sapwood part) on the half-width size were investigated. The method to calculate half-width was developed. Other following def **ImageJ** inition to avoid confusion is term peak-height as PF adhesive concentration.

### MATERIALS AND METHODS

#### Materials

Five-ply sugi (*Cryptomeria japonica*) plywood was made from low-, medium-, and high-density heartwood with density values of 0,281-0,309 g/cm<sup>3</sup>; 0,319-0,343 g/cm<sup>3</sup> and 0,346-0,372 g/cm<sup>3</sup>, respectively. The medium-density sugi sapwood plywood was made with range of density at 0,320-0,340 g/cm<sup>3</sup>. PF adhesive with solid content 48 % was used in this study. Heartwood plywood was made using three glue spreading rates: 75 g, 150 g and 225 g. Sapwood plywood was made using four glue spreading rates: 75 g, 150 g, 225 g and 300 g Plywood manufacturing was performed by cold-pressing and hot-pressing at 1 MPa for 20 min (Ferrtikasari *et al.* 2019) (The properties of the X-ray plywood sample are shown in Table 1. Samples for X-ray assessment were cut with dimension 10 mm × 100 mm x plywood thickness for all plywood.

Veneer type	Veneer's density group	Spreading rate (g/m <sup>2</sup> )	Dimension (mm)			Weight (g)	Plywood density (g/cm <sup>3</sup> )
	Low		10.03	17.31	100.42	6.11	0.351
Heartwood	Medium	75	9,39	17,74	98,28	6,54	0,400
	High	1	10,22	17,89	100,62	7,75	0,421
	Low		9,36	16,59	100,00	5,92	0,381
	Medium	150	10,39	17,13	100,68	7,92	0,442
	High		10,63	17,41	100,34	7,89	0,425
	Low		10,33	15,58	100,33	6,92	0,428
	Medium	225	10,06	16,59	100,80	7,45	0,443
	High		10,23	16,75	100,73	8,36	0,484
		75	9,33	18,08	100,51	5,77	0,340
Sapwood	Medium	150	10,77	17,66	100,48	6,73	0,352
		225	10,13	17,48	100,47	6,61	0,372
		300	10,25	16,90	100,30	6,52	0,375

Table 1: The dimensions and densities of the X-ray plywood samples.

#### X-ray scanning of plywood

An X-ray apparatus, SR-1010 (Softex Co., Ltd., Ebina, Japan) equipped with a digital X-ray sensor (NX-04H, Softex Co., Ltd.), was used. The sample was scanned on a bondline section with 21,5 kV tube voltage, 3 mA tube current, exposure time of 50 s, and a 416-mm-thick polytetrafluoroethylene (PTFE) filter. A schematic of the X-ray photography set-up is shown in Figure 1.



Figure 1: Schematic of the X-ray photography condition.

The X-ray image of the plywood sample was divided into seven lines using ImageJ 1.48v software to obtain seven plot profiles. The gray value of the poly (methyl methaceylate) (PMMA) block area was used to obtain the equation for the PMMA-equivalent thickness (Figure 2). This equation was used to convert the gray values of the plot profiles. The converted plot profile was calculated using a certain mechanism to obtain the adhesive penetration and adhesive concentration in plywood.



Figure 2: Equation of the PMMA-equivalent thickness.

# Statistical analysis

The effects of the glue spreading rate and veneer density on the half-width were analyzed using two-way analysis of variance (ANOVA).

# **RESULTS AND DISCUSSION**

#### X-ray image of adhesive penetration in sugi plywood

Adhesive penetration was successfully visualized using an X-ray apparatus. Figure 3a clearly shows the plywood X-ray image. The X-ray setting is one of the determinants in the clear and successful visualization of X-ray images. In this study, the low X-ray tube voltage in the X-ray setting was used to visualize the adhesive penetration in heartwood and sapwood plywood with low, medium, and high veneer densities using various levels of glue spreading rates (75 g/m<sup>2</sup>, 150 g/m<sup>2</sup>, 225 g/m<sup>2</sup> and 300 g/m<sup>2</sup>).

The seven-plot profile from seven representative lines on each X-ray image was obtained using ImageJ. The peak that represented the glue line was pointed out, and the rest were the latewood peaks (Figure 3a). The adhesive that penetrated the lathe check area was estimated to make the glue line peak broader than the glue line, which has no lathe check penetration (Figure 3b). However, the adhesive inside the lathe check has its own peak, which was not part of the general glue line peak.



Figure 3: (a) X-ray image of high-density heartwood plywood plot profile with 225 g/m<sup>2</sup> glue spreading rate with seven adhesive plot profile with the black arrow pointed on the adhesive peak; (b) Lathe check gray value peak pointed by red arrow which separated for adhesive peak.

### PF adhesive penetration in sugi plywood

The seven-plot profile was converted into a PMMA-equivalent thickness value. This was done in order to understand the penetration of the adhesive in the plywood and the concentration of the adhesive in relation to the glue spreading rate. An example of a successful converted plot profile is shown in Figure 4.



**Figure 4:** The plot profile of line 6 in high density heartwood plywood with 225 g/cm<sup>2</sup> glue spreading rate that successfully converted using PMMA equation from PMMA calibration curved.

After converting the plot profile, the mechanism for calculating the penetration depth of the glue was developed in this study and is shown in Figure 5.



Figure 5: The mechanism of calculated the peak height and half-width.

The base point of the peak should be determined based on the horizontal inequality position of the base point. After the peak height was obtained, the height of the graph was divided by two, which was the way to obtain the middle line of the graph. Then, the length of the middle line of the graph, which is defined as the half-width occurred. The half-width of the peak was considered as the adhesive penetration depth. The peak height was considered as the adhesive penetration depth. The peak height was considered as the adhesive concentration. The half-width and peak-height were obtained from the calculation of the glue line peak (Figure 6). The half-width (adhesive penetration) value obtained from various glue spreading rate and veneer density was investigated to see the relations between them (Figure 7).



**Figure 6:** Seven points of peak height and half-width of low, medium, and high veneer density heartwood plywood with 75, 150, 225 g/m<sup>2</sup> glue spreading rate; and medium veneer density sapwood plywood with 75, 150, 225, and 300 g/m<sup>2</sup> glue spreading rate.



Figure 7: PF adhesive half-width of (a) low density heartwood plywood, (b) medium density heartwood plywood, (c) high density heartwood plywood, and (d) medium density sapwood plywood.

The half-width was within the range of 0,3 mm to 0,9 mm and was approximately 0,5 mm on average, regardless of the glue spreading rate or veneer density. This finding is similar to that reported by Modzel *et al.* 2011 that the penetration depth of the adhesive in the oak, Douglas-fir, and poplar were 400  $\mu$ m, 100  $\mu$ m, and 400  $\mu$ m, respectively (Modzel *et al.* 2011). These results support the idea that glue penetration into the veneer in plywood manufacturing is constant, regardless of the glue spreading rate, veneer density, or the wood species of the veneer. The smallest half-width value (= 0,3 mm) occurred near the boundary between earlywood and latewood (Figure 8), suggesting that the existence of latewood in the veneer might limit the adhesive penetration. This limitation may lead to the consistency of the average half-width value without regard to the glue spreading rate or veneer density.



**Figure 8:** The X-ray image and its plot profile which indicates the smallest half-width value (0,3 mm). (a) A plot profile of medium veneer density of sapwood plywood with 150 g/m<sup>2</sup> glue spreading rate and (b) its X-ray image, and (c) a plot profile of high veneer density of heartwood plywood with 75 g/m<sup>2</sup> glue spreading rate and (d) its X-ray image.



Figure 9: PF adhesive peak height of (a) low density heartwood plywood, (b) medium density heartwood plywood, (c) high density heartwood plywood, and (d) medium density sapwood plywood.

The adhesive concentration (peak height) increased along with increasing the glue spreading rate. The peak height of the heartwood plywood with low, medium, and high veneer densities showed the same trend (Figure 9a, Figure 9b and Figure 9c). The peak height increased with increasing glue spreading rate. In addition, regarding the sapwood-veneer plywood, the maximum peak height was obtained at the maximum spreading rate of 300 g/m<sup>2</sup> (Figure 9d). These results indicate that the concentration of the adhesive at the bondline is increased by increasing adhesive spreading rate.

The statistical analysis using two-way ANOVA method did not reveal any significant effect of glue spreading rate, veneer density, or their interactions on the half-width value (Table 2).

<b>Table 2:</b> Results of the two-w	ay ANOVA of veneer	density and glue	spreading rate	in relation with PF
adhesive	penetration (half-width	n) on Sugi heartwo	ood plywood.	

Variation factor	Sum of squares	Degree of freedom	Mean of Squares	F value		F critical (5%)
Veneer Density	0,009294	2	0,004647136	0,511127732	<	3,18
Spreading rate	0,002184	2	0,001091854	0,120090442	<	3,18
Interaction	0,086737	4	0,021684153	2,384989672	<	2,56
Residue	0,463688	51	0,009091927			
Total	0,561903	59				

## CONCLUSIONS

We successfully evaluated the concentration profile of sugi plywood with a low X-ray tube voltage. The PF adhesive penetration in the sugi plywood had values of 0,3-0,9 mm, with an average of 0,5 mm. In this experiment, using sugi plywood, it was shown that the existence of latewood might limit the PF adhesive penetration. On the other hand, the PF adhesive concentration in the glue line increases with increasing veneer density and glue spreading rate. Findings from this study would be useful in the analysis of a plywood's quality and durability.

### ACKNOWLEDGEMENTS

This study was supported by JSPS KAKENHI grant numbers 17H05032 and 20K06163. We thank J-Chemical Co., Ltd. for supplying the phenol formaldehyde adhesive, and NODA Corporation for supplying the sugi veneer. We are grateful to Ms. Nindya Ferrtikasari for preparing the plywood samples. We thank Ms. Aoi Oishi for statistical analysis.

# REFERENCES

Asif, M. 2009. Sustainability of timber, wood and bamboo in construction. Chapter 2. In *Sustainability of construction materials*. Khatib JM (ed.). Woodhead Publishing, Sawston: United Kingdom: 31-54. https://doi.org/10.1533/9781845695842.31

**Cognard, P. 2005.** Technical characteristics and testing methods of adhesives and sealants. Chapter 2. In: *Handbook adhesive and sealants*. Cognard P (ed.), Elsevier Oxford: United Kingdom. 21-99. https://doi.org/10.1016/S1874-5695(02)80003-3

**Ferrtikasari, N.; Tanaka, T.; Yamada, M. 2019.** Relationship between thermal conductivity and adhesive distribution of phenol-formaldehyde visualized with x-ray computed tomography on sugi (*Cryptomeria japonica* D.Don) heartwood plywood. *IOP Conference Series: Materials Science and Engineering.* The 14th Pacific Rim Bio-Based Composites Symposium. October 2018, South Sulawesi Province, Indonesia. vol. 593: 29-31. http://dx.doi.org/10.1088/1757-899X/593/1/012003

Fisher, R.C.; Tasker, H.S. 1940. The detection of wood boring insects by means of X-rays. Annals of Applied Biology 27(1):92-100 https://doi.org/10.1111/j.1744-7348.1940.tb07480.x

Jakes, J.E.; Hunt, C.G.; Yelle, D.J.; Lorenz, L.F., Hirth, K.; Gleber, S.C., Vogt, S.; Grigsby, W.; Frihart, C.R. 2015. Synchrotron-based X-ray fluorescence microscopy in conjunction with nanoindentation to study molecular-scale interactions of phenol-formaldehyde in wood cell walls. *ACS Applied Materials & Interfaces* 7: 6584-6589. https://doi.org/10.1021/am5087598

**Kurt, R. 2010.** Possibilities of using poplar clones and boron compounds to manufacture fire resistant laminated veneer lumber. In: Project No: 106O556 progress report. Turkish Scientific Research Council: Kavaklıdere - Ankara, Türkiye.

Kurt, R.; Cil, M. 2012. Effect of press pressures on glue line thickness and properties of laminated veneer lumber glued with phenol formaldehyde adhesive. *BioResources* 7(4): 5346-5354. https://ojs.cnr.ncsu.edu/index.php/BioRes/article/view/3200

Laborie, M.P.G. 2002. Investigation of the Wood/Phenol-Formaldehyde Adhesive Interphase Morphology. PhD Dissertation, Virginia Polytechnic Institute and State University, Virginia, The United State of America.

Modzel, G.; Kamke, F.A.; De Carlo, F. 2011. Comparative analysis of a wood: Adhesive bondline. *Wood Science and Technology* 45: 147-158. https://doi.org/10.1007/s00226-010-0304-z

Tanaka, T. 2018. Simple geometrical model of thermal conductivity and bound-water diffusion coefficient in resin-rich regions of softwood plywood. *Wood Science and Technology* 52: 331-342. https://doi.org/10.1007/s00226-018-0985-2

Tanaka, T.; Adachi, K.; Yamada, M. 2015. X-ray mass attenuation coefficient for some common wood adheives in the tube voltage range from 15 kV to 100 kV. *Mokuzai Gakkaishi* 61(5): 308-315. https://doi.org/10.2488/jwrs.61.308

Tanaka, T.; Shida, S. 2012. Changes of Through-thickness Moisture Distribution in Wood and Woodbased Materials in Adsorption Phase III. Nondestructive measurement of moisture content distribution in plywood and sheathing insulation fiberboards. *Mokuzai Gakkaishi* 58(5): 271-278. https://doi.org/10.2488/ jwrs.58.271

**Tomazello, M.; Brazolin, S.; Chagas, M.P.; Oliveira, J.T.S.; Ballarin, A.W.; Benjamin, C.A. 2008.** Application of x-ray technique in nondestructive evaluation of eucalypt wood. *Maderas. Ciencia y Tecnologia* 10(2): 139-149. https://doi.org/10.4067/S0718-221X2008000200006

White, M.S. 1975. Influence of Resin Penetration on the Fracture Toughness of Bonded Wood. PhD Dissertation, Virginia Polytechnic Institute and State University, Virginia, The United State of America.

**Worschitz, F. 1932.** L'utilization des rayos. In: Congrès IUFRO, X en vue de l'etude de la qualité du bois. Paris. France: 459-489.