

Curing Behavior of Resorcinol Formaldehyde Resin Adhesives in Mangium and Pine Woods Bonding System under Various Curing Times

Eka M Alamsyah¹⁾, Masaaki Yamada²⁾, Kinji Taki²⁾

- 1) Department of Forest Products Technology, Faculty of Forestry, Winaya Mukti University, Jatinangor-Sumedang, Indonesia
- 2) Laboratory of Wood Adhesion, Department of Environment and Forest Resources Science, Faculty of Agriculture, Shizuoka University, Japan

Corresponding author: email ekaalamsyah@yahoo.com (Eka M Alamsyah)

Abstract

Effects of room-temperature curing time on the curing behavior of resorcinol formaldehyde (RF) resin adhesives by using thermo mechanical analysis spring method in relation to the mangium (*Acacia mangium*) and pine (*Pinus merkusii*) woods bonding system were examined. Total amount of liquid resin adhesives that inserted into the small wire spring was 10 mg for each curing time parameters. Results showed that the curves of modulus of elasticity (E') from resin adhesives that cured for 1 month, 2 months and 3 months was almost flat from room temperature to 200°C. By far E' curve of resin adhesives that cured for 3 months did not changed and adhesive predicted had cured completely. Results further showed that in *A. mangium* and *P. merkusii* bonding system all laminates bonded with RF in room temperature at least 1 month curing time is required to meet high bond strength with minimally 60 percent wood failure as one of requirements of good bonding performance classification.

Key words: *Acacia mangium*, bonding system, curing behavior, *Pinus merkusii*, resorcinol formaldehyde,

Introduction

Our previous study indicated that in normal condition the bonding performance of *A. mangium* and *P. merkusii* laminates bonded with resorcinol formaldehyde (RF) resin adhesive in room temperature (20°C and 65% RH) was distributed in bad and worst categories showing by low strength and wood failure percentage (Alamsyah *et al.* 2007). One of the reasons was predicted that the adhesive did not yet cure completely for 1 week curing time under the condition of 20°C and 65% RH. Results similar to those of our study were reported by Taki *et al.* (1979) and Liu (2005) for a study that used selangan batu and hinoki, respectively. To clarify this indication, an investigation on the curing

behavior of RF using thermo mechanical analysis (TMA) spring method have been conducted. This method had been used to study the curing behavior of some thermosetting resin such as phenol-formaldehyde (PF) and RF (Sato *et al.* 1997, Taki *et al.* 1998, Taki *et al.* 2001, Adachi & Taki 2007). They reported that this method is suited to study curing reaction behavior as the thermosetting resin progresses from liquid to gel and to solid state.

Previously, many investigators have studied the curing behavior of thermosetting resin by dynamic mechanical analysis method (DMA, TBA, etc.) (Mizumachi 1973, Mizumachi & Morita, 1975, Pizzi & Stephanou, 1994,

Umemura *et al.* 1995, 1998). However, TMA with a wire spring method had not been widely used to determine mechanical properties of RF resin adhesive. In this study, the effects of curing time on the curing behavior of RF resin adhesives in relation to the bonding performance of *A. mangium* and *P. merkusii* are discussed.

Materials and Methods

Measurement of curing behavior of RF resin

A commercial RF resin adhesive (J-6000, Dainippon Ink and Chemicals, Incorporated) with solid content of $58 \pm 3\%$ and viscosity of 6.0 ± 2 Pa.s was used in this study. Liquid adhesive specimen (a) was 100 parts by weight (pbw) resin mixed with 15 pbw paraformaldehyde (TD-473) as a catalyst. Total amount of liquid specimen that inserted into the small wire spring was 10 mg. The heating rate was $10^\circ\text{C min}^{-1}$. The amplitude cycle was 0.1Hz and the tensile amplitude load was 1.96×10^{-2} N. The curing behavior of resin was analyzed in elevating temperature condition (20°C - 200°C). In order to clarify that the resin has cured completely or not, specimens that cured in room temperature for 1 week (b), 2 weeks (c), 1 month (d), 2 months (e) and 3 months (f) were also prepared.

The curing behavior of resin was analyzed by using TMA apparatus (TMA-4000, Mac Science Co.). The top of the spring was caught in the hook of the terminal of the detector, and a container was fixed on the support tube with the hook as shown in Fig. 1. Then, a fixed vibration load was applied at the spring in every constant period with controlling temperature of the specimen through the atmosphere in the electric oven. When the load was applied, the spring caused a change in volume. The quantity of change in volume (amplitude) greatly depends on hardness of the specimen. The modulus of elasticity (E') and loss energy of each period were analyzed by vibration load (stress) and the delay of the phase of the vibration of the spring (strain).

Curing behavior is expressed in relative elasticity (E_r), which calculated using following equation:

$$E_r = \frac{(E_x - E_{min})}{(E_{max} - E_{min})}$$

Where E_r is the relative elasticity, E_x is the modulus of elasticity (E') at $x^\circ\text{C}$, E_{min} is the minimum modulus of elasticity, and E_{max} is the maximum modulus of elasticity.

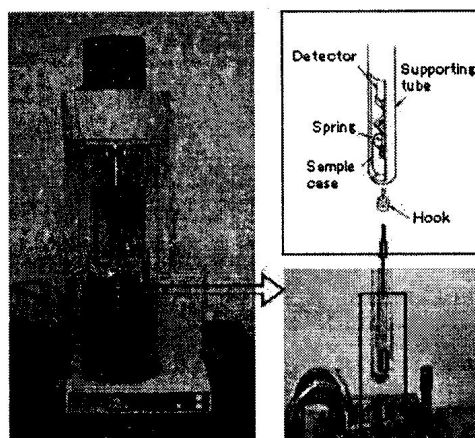


Figure 1 TMA apparatus (TMA-4000, Mac Science Co.)

Bonding performance

Two ply lamination of *Acacia mangium* Willd with 0.54 g cm^{-3} air-dry density and 9% MC and *Pinus merkusii* Jungh et De Vriese (density 0.59 g cm^{-3} and 9% MC) were bonded with RF resin adhesive at spread rate of 250 g m^{-2} with a roller. Pressure was applied at 1.18 MPa in 20°C and 65% RH room conditions for 24 h cold-pressing time. Then, the laminates were reconditioned at 20°C and 65% RH room for 1 week as a control (C), 2 weeks (W2), 1 month (M1), 2 months (M2) and 3 months (M3) curing time. Ten small block-shear specimens (20 mm in thickness \times 25 mm in width \times 30 mm in length) for each treatment were cut from the two-ply lamination. In block-shear test, an Amsler machine was used with a load rate of about 9.8 KN min^{-1} . Bond strength and wood failure under normal condition (N) were measured according to Japan Agricultural Standard for Structural Glued Laminated Timber (shear area about 6.25 cm^2) (JAS 2005) and those of wet conditions were also measured after accelerated aging test as described below, which were used for delamination test of glulam.

1. Cyclic boiling (CB) test: Specimen submerged in boiling water for 4 h \rightarrow submerged in cold tap water for 1 h \rightarrow dried at $70^\circ\text{C} \pm 3^\circ\text{C}$ for 18 h \rightarrow submerged in boiling water for 4 h and then tested when wet.
2. Vacuum pressure soaking (VPS) test: Specimen submerged in cold tap water for 1 h \rightarrow evacuated at 0.085 MPa for 5min \rightarrow pressurized at $0.51 \pm 0.03 \text{ MPa}$ for 1 h \rightarrow these step repeated and tested when wet.

Strength retention (SR) was calculated using this equation:

$$\text{SR (\%)} = \frac{\text{bond shear strength}}{\text{Solid shear strength}} \times 100$$

Results and Discussion

Curing behavior of RF resin adhesives

The effect of curing time on the curing behavior of RF in room temperature is shown in Fig. 2. At the beginning of test, the initial modulus of elasticity (E') of adhesive compounds is assumed as 0 and assumed as 1 when adhesive had cured completely. In liquid state (a), specimen that cured for 1 week (b) and 2 weeks (c), the curve of the storage modulus of elasticity (E') was flat from room temperature to 85°C . Beyond 85°C the E' curve increased until nearly 120°C , and the E' curve did not change in a hard glass state up to 200°C . It seemed that the reactive zone of the chemical and mechanical changes of liquid and specimen that cured for 1 week (b) and 2 weeks (c) was in the temperature ranges from 85°C to 120°C , over 120°C the adhesive predicted had cured completely. The initial temperature on the reactive zone in this study was 20°C higher than a previous study (Sato *et al.* 1997). This may be caused by the differences on the adhesive formulation. For the specimens that cured for 1 month (d), 2 months (e) and 3 months (f) the E' curve was almost flat from room-temperature to 200°C . However, the E' curve in specimen cured for 1 month (d) and 2 months (e) seemed to remained a view changed in a hard glass state to 200°C . In specimen that cured for 3 months (f) the E' curve did not change in a hard glass state from room temperature to 200°C and adhesive predicted had cured completely. A previous study had also reported that more than 1 month is needed for RF to meet adhesive cure completely when it applied in room temperature (Sato *et al.* 1997). Vick (1999) reported that the process of adhesion is essentially completed after transition of the adhesive from liquid to solid form.

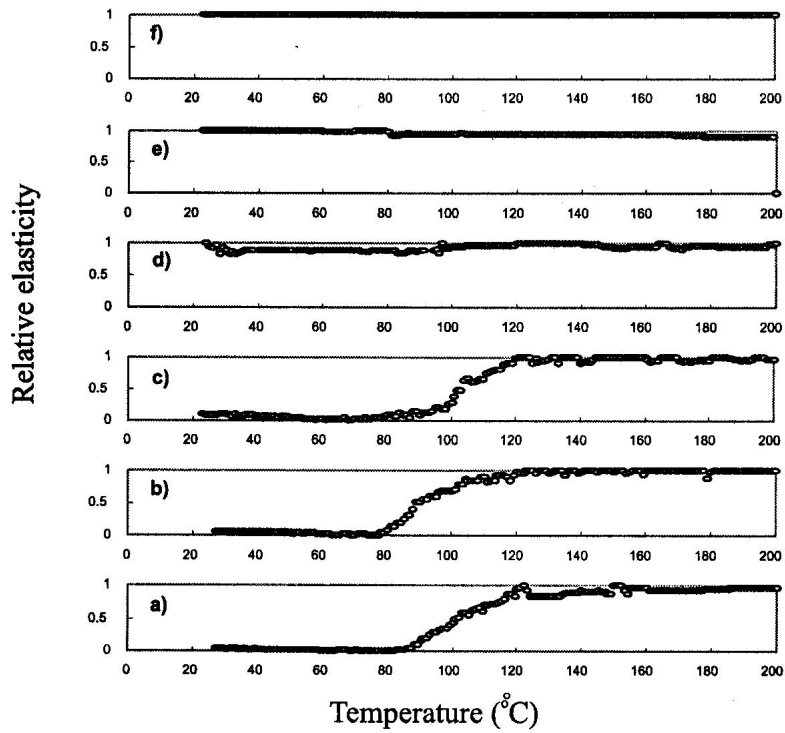


Figure 2 Curing behavior of RF resin adhesives for liquid (a), 1 week (b), 2 weeks (c), 1 month (d), 2 months (e) and 3 months (f) curing time

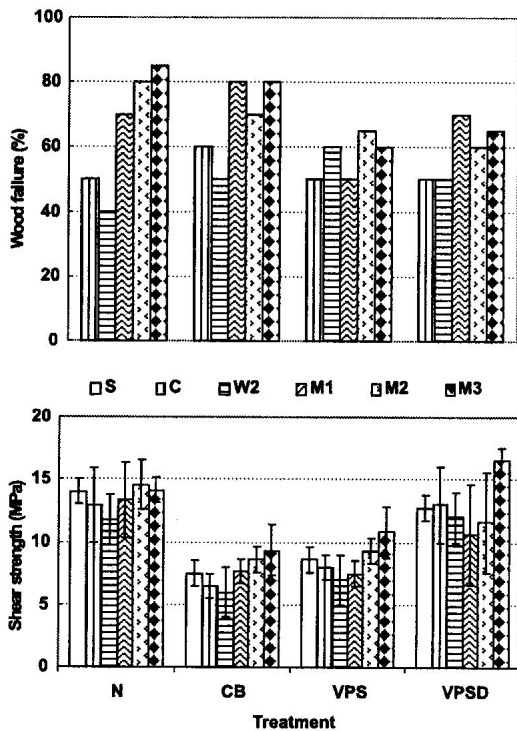


Figure 3 Shear strength and wood failure of *A. mangium*

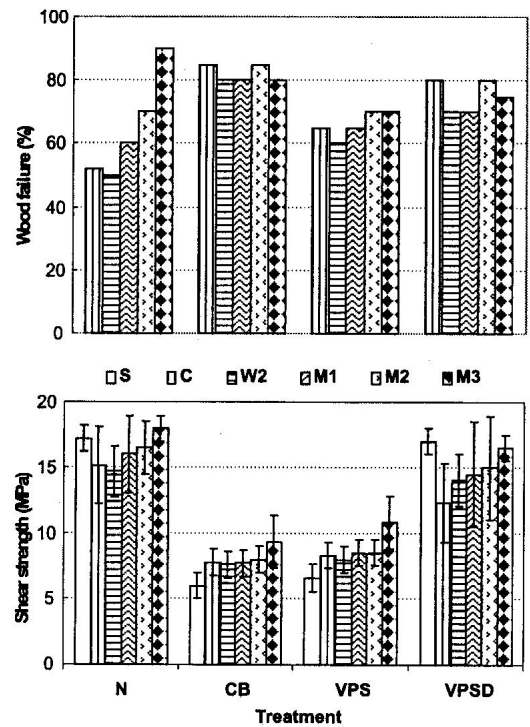


Figure 4 Shear strength and wood failure of *P. merkusii*

The effect of curing time on the shear strength and wood failure of *A. mangium* and *P.merkusii* bonded with RF in room temperature is shown in Fig. 3 and 4. In *A. mangium* results showed that for normal conditions, the highest shear strength was achieved by M2 (around 14MPa), while the highest wood failure was achieved by M3 (around 80%). After CB treatment, the highest shear strength was achieved by M3 (9.3 MPa) while the highest wood failure was achieved by M1 and M3 (80%). In VPS treatment, the highest shear strength was achieved by M3 (10.8 MPa) while the highest wood failure was achieved by M2 (65%). After VPSD treatment, the highest shear strength was achieved by M3 (16.5) while the highest wood failure was achieved by M1 (70%). In *P.merkusii* for normal conditions, after CB, VPS and VPSD results showed that the highest shear strength was achieved by M3 around 18.0, 9.3, 10.8, and 16.5 MPa, respectively while the highest wood failure in normal condition was achieved

by M3 (90%), after CB and VPSD was achieved by C and M2 (80%) while after VPS was achieved by M2 and M3 (70%).

Fig. 5 and 6 showed that the strength retention (SR) for *A. mangium* and *P. merkusii* in normal condition and after 1 week, 2 weeks, 1 month, 2 months and 3 months curing time were excellent showing by high RS percentages (more than 80% for each curing time parameters). However, high wood failure as shown by minimally 60% wood failure value was achieved by the laminates that cured for 1 month, 2 months and 3 months. These results indicated that in *A. mangium* and *P. merkusii* laminates bonded with RF in room temperature, at least 1 month curing time is required to meet the good bonding performance classification. By far, it was suggested that the bond shear strength and wood failure increased with the increasing of curing time, relatively.

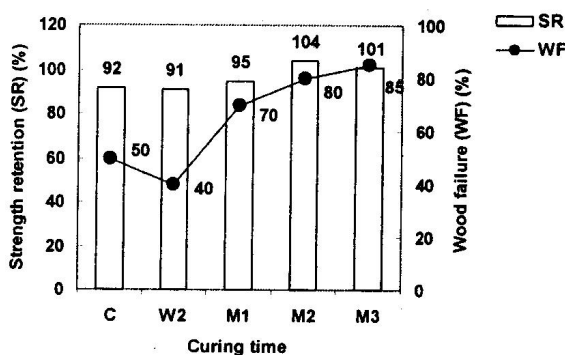


Figure 5 Strength retention and wood failure of *A. mangium* in normal condition

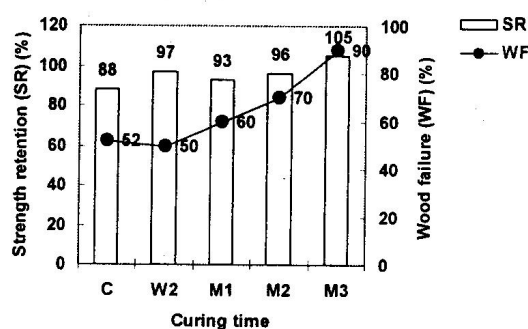


Figure 6 Strength retention and wood failure of *P. merkusii* in normal condition

Conclusions

Results showed that the curves of modulus of elasticity (E') from resin adhesives that cured for 1 month, 2 months and 3 months was almost flat from room temperature to 200°C. By far E' curve of resin adhesives that cured for 3 months did not changed

and adhesive predicted had cured completely. In normal condition, high bond strength with high wood failure wood failure was achieved by the laminates that cured for 1 month, 2 months and 3 months. These results indicated that in *A. mangium* and *P. merkusii* laminates bonded with RF in room temperature, at

least 1 month curing time is required to meet the good bonding performance classification.

References

- Adachi K, Taki K. 2007. Curing behavior of wood adhesives and bond quality. Di dalam: *Curing temperature of resin and methods and assessment of curing process*. Osaka: Science and Technology press. Pp. 175-187.
- Alamsyah EM, Liu CN, Yamada M, Taki K, Yoshida H. 2007. Bondability of tropical fast-growing tree species (I)- Indonesian wood species. *J Wood Sci*. 53(1): 40-46.
- [JAS) Japanese Agricultural Standard. 2005. *JAS No.235 for Structural Glued Laminated Timber*. Tokyo: The Ministry of Agriculture, Forestry and Fisheries of Japan.
- Liu CN. 2005. *Bond quality of moisture curing polyurethane adhesives* (in Japanese). [Thesis]. Shizuoka: Faculty of Agriculture, Shizuoka University.
- Mizumachi H. 1973. Activation energy of the curing reaction of urea resin in the presence of wood. *Wood Sci*. 7(4):14-18.
- Mizumachi H, Morita H. 1975. Activation energy of the curing reaction of phenolic resin in the presence of woods. *Wood Sci*. 7(3):256-260.
- Pizzi A, Stephanou A. 1994. Phenol formaldehyde wood adhesives under very alkaline conditions, part I: behavior and proposed mechanism, *Holzforschung*. 48(1):35-40.
- Sato Y, Taki K, Yoshida H. 1997. Thermal analysis and bond quality of thermosetting resin adhesives (in Japanese). *Proceedings of the 35th Annual Meetings of The Adhesion society of Japan*. Tokyo: *The Adhesion society of Japan*. Pp. 129-130.
- Taki K, Mizumachi H, Yamagishi Y. 1979. Bond quality of PVA-isocyanate reactive resin adhesives (II) (in Japanese). *Mokuzai Gakkaishi*. 25(3): 216-224.
- Taki K, Yamada M, Yoshida H. 1998. Thermal analysis and bond strength of thermosetting resin. *Book of abstract the 1st World Congress on Adhesion and Related Phenomena*, Germany. Pp. 167
- Taki K, Yoshida H, Yamada M. 2001. Thermal analysis and bond quality of phenol-formaldehyde resin adhesives. *Proceedings of Wood Adhesive 2000* No.7252. USA: USDA. Pp. 351-355.
- Umemura K, Kawai S, Nishioka R, Mizuno Y, Sasaki H. 1995. Curing behavior of wood adhesives under high-pressure steam I, phenolic resin. *Mokuzai Gakkaishi*. 41(9): 828-836.
- Umemura K, Takahashi A, Kawai S. 1998. Durability of isocyanate resin adhesives for wood I: thermal properties of isocyanate resin cured with water. *J Wood Sci*. 44(3): 204-210.
- Vick CB. 1999. Adhesive bonding of wood materials. In: *Wood hand book, wood as an engineering material*. Madison: Forest products society. Pp. 9-23.

Riwayat naskah (*article history*)

Naskah masuk (*received*) : 02 October 2009

Diterima (*accepted*) : 26 December 2009