Optimization of application of natural rubber based API adhesive for the production of laminated wood

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Abstract. The use of natural rubber latex (NRL) for wood adhesive has been identified since a long time ago, even though its adhesion quality is fair. Some modifications of the NRL, as well as the blending of NRL with other commercial wood adhesives, have already been reported. Another potential use of NRL in the wood adhesive formulation is a base polymer of isocyanate type adhesive, which is called aqueous polymer isocyanate (API) adhesive. This study aimed to optimize the aqueous polymer isocyanate (API) type adhesive made from natural rubber latex (NRL) using response surface methodology (RSM). Central composite design (CCD) was used for the RSM. Three factors have been optimized, namely the percentage of crosslinker (7.3–20.7% of the weight of base polymer), glue spread rate (112–298 g/m²), and pressing time (2–22 hours). Results of the study showed that the model generated from the optimization was fit and could be used to predict the relationship between the three factors and the bonding strength. Optimum conditions for the adhesive application for the production of laminated wood used 16.4% crosslinker, glue spread rate of 298 g/m² with a pressing time of 6 hours. Using these conditions, the shear strength of the laminated wood obtained was 8.8 MPa.

1. Introduction

Natural rubber latex (NRL) has been used for wood adhesive since a long time ago, even though its adhesion quality is fair. Some modifications of the NRL, as well as the blending of NRL with other commercial wood adhesives, have already been reported [1–7]. Another potential use of NRL in the wood adhesive formulation is a base polymer of isocyanate type adhesive, which is called aqueous polymer isocyanate (API) adhesive. The other name for API adhesive is emulsion polymer isocyanate (EPI) adhesive. This type of adhesive has some advantages over commonly used wood adhesives, such as urea formaldehyde (UF), melamine formaldehyde (MF), phenol formaldehyde (PF), resorcinol-formaldehyde (RF), and phenol resorcinol formaldehyde (PRF) because it does not emit formaldehyde, which means environmentally friendly. Besides, it can be cured at room temperature, which means less energy needed in the application process. The adhesive has comparable performance with conventional RF adhesive, but the price is lower than RF adhesive so that API adhesive has been widely used in timber manufacturing in developed countries [8]. The API adhesive consists of two parts, the base polymer which usually contains water-soluble polymers or emulsions, such as polyvinyl alcohol (PVA) and polystyrene-co-butadiene rubber (SBR) latex and the isocyanate crosslinker [9]. In this study, we used NRL as a backbone polymer for substituting the SBR and blended with PVA to produce a base polymer for API adhesive. Our previous study showed that the formulation of this base polymer of API adhesive could produce plywood with satisfied shear strength after the cyclic boiling test. However, we...
have not optimized the application conditions for laminated wood. Therefore, in this study, we intend to optimize the application condition of API type wood adhesive made from NRL for the production of laminated wood that can be cured at room temperature.

2. Materials and methods

2.1. Materials

The natural rubber latex (total solid content 61-62%) was obtained from Balai Penelitian Perkebunan Sembawa, South Sumatera. The concentrated NRL was diluted with distilled water to 50% total solid content. The PVA was purchased from a local chemical supplier in Bogor. It was dissolved in warm water (45-50 °C) to get PVA solution with 15% solid content. The isocyanate crosslinker (polymeric methylene diphenyl diisocyanate – p-MDI) was the Wannate PM-200 of Yantai Wanhua with isocyanate group (-NCO) content of 30.2-32%. Other chemicals were purchased from domestic chemical suppliers in Jakarta and Bogor. The wood used for the application of the adhesive was rubberwood, which was obtained from Tangerang Selatan, Banten. The rubberwood was air dried for about one month. Then, it was cut into sizes of 20 cm (L) × 8 cm (W) × 1 cm (T), and dried in an oven at 40-45 °C so that the moisture content of the wood was less than 10%. The wood samples were sanded using sandpaper to get smooth surfaces.

2.2. Methods

2.2.1. Preparation of adhesive. The base polymer of API adhesive was prepared by blending NRL and PVA with formulas of 1:1. Calcium carbonate (CaCO₃), which was served as a filler, was added to the polymer mixture at the amount of 15% of the polymer weight. The optimization of the production of laminated wood used Central Composite Design (CCD). Three factors have been optimized, namely the percentage of crosslinker (7.3-20.7% of the weight of base polymer), glue spread rate (112-298 g/m²), and pressing time (2-22 hours). The number of the experiment was 20, which consisted of 2³ factorial points, six center points, and 6-star points at the extreme levels (±α, where α = 1.682). These points are shown in table 1. The response variable was shear strength of laminated wood sample, which was obtained after the shear strength testing according to JAS for glued laminated timber [10].

Statistical analysis was conducted using Response Surface Methodology (RSM) with software Design Expert 11 (Trial Version). The quadratic polynomial model was constructed for the data using the following regression equation.

\[ Y = b_0 + b_{11}X_1 + b_{22}X_2 + b_{33}X_3 + b_{12}X_1X_2 + b_{13}X_1X_3 + b_{23}X_2X_3 \]  

(1)

where Y is response variable (shear strength, N/mm²); X₁ (crosslinker, g/g), X₂ (glue spread rate, g/m²), and X₃ (pressing time, hours) are independent responses; b₀ is coefficient of intercepts; b₁, b₂, b₃, b₁₁, b₂₂, b₃₃, b₁₂, b₁₃, and b₂₃ are coefficients of regression. Fitting quality of the model was tested by the value of R² and the lack of fit (lof). The significance of each factor was determined by the p value (p ≤ 0.05) in the ANOVA (Analysis of Variance).

2.2.2. Preparation of laminated wood. The API adhesive was spread on the surface of a rubberwood sample with adhesion area of 20 × 8 cm² at the specified glue spread rate. Then, another wood sample with the same size was put on the glued surface of the former wood. The two pieces of wood were put together, and cold pressed at 14 Mpa for the specified length. The laminated wood produced was then conditioned at room temperature for one week before it was tested for its bond performance (shear strength) according to Japan Agriculture Standard (JAS) for Glued Laminated Timber [10]. There were five specimens tested for each run of the optimization experiment.
Table 1. Optimization design of adhesive application for the production of laminated wood.

| Run | X1a | X2b | X3c | x1d | x2b | x3c | Yd  |
|-----|-----|-----|-----|-----|-----|-----|-----|
| 1   | 1   | 1   | 1   | 18  | 18  | 18  | 18  |
| 2   | 1   | 1   | -1  | 18  | 18  | 18  | 6   |
| 3   | 1   | -1  | 1   | 18  | 150 | 18  | 6   |
| 4   | 1   | -1  | -1  | 18  | 150 | 6   | 6   |
| 5   | -1  | 1   | 1   | 10  | 260 | 18  | 18  |
| 6   | -1  | 1   | -1  | 10  | 260 | 6   | 6   |
| 7   | -1  | -1  | 1   | 10  | 150 | 18  | 18  |
| 8   | -1  | -1  | -1  | 10  | 150 | 6   | 6   |
| 9   | +α  | 0   | 0   | 20.7| 205 | 12  | 12  |
| 10  | 0   | +α  | 0   | 14  | 298 | 12  | 12  |
| 11  | 0   | 0   | +α  | 14  | 205 | 22  | 22  |
| 12  | -α  | 0   | 0   | 7.3 | 205 | 12  | 12  |
| 13  | 0   | -α  | 0   | 14  | 112 | 12  | 12  |
| 14  | 0   | 0   | -α  | 14  | 205 | 2   | 2   |
| 15  | 0   | 0   | 0   | 14  | 205 | 12  | 12  |
| 16  | 0   | 0   | 0   | 14  | 205 | 12  | 12  |
| 17  | 0   | 0   | 0   | 14  | 205 | 12  | 12  |
| 18  | 0   | 0   | 0   | 14  | 205 | 12  | 12  |
| 19  | 0   | 0   | 0   | 14  | 205 | 12  | 12  |
| 20  | 0   | 0   | 0   | 14  | 205 | 12  | 12  |

a X1 and x1 = crosslinker
b X2 and x2 = glue spread rate
c X3 and x3 = pressing time;
d Y= shear strength

3. Results and discussion

The results of shear strength measurements from the 20 runs of the optimization experiment are presented in table 2. The results of the statistical analysis (table 3) show that the equation model is significant. Table 3 also shows that the three factors, crosslinker, glue spread rate, and pressing time, had a significant effect on the shear strength of laminated wood, while the interaction among those factors did not. The lack of fit value obtained in the optimization was not significant. It means that the model was good and fit for predicting the shear strength of laminated wood. The coefficient of the determinant ($R^2$) was also quite high (0.8641). Considering the significant model, the insignificant lack of fit, and the high $R^2$, we would suggest that the quadratic equation model generated in this experiment was fit and could be used to predict the shear strength of laminated wood.

Shear strength $= 3.49 - 0.98 X_1 + 1.50 X_2 + 0.51X_3 + 0.77X_1^2 + 0.96 X_2^2 - 0.21 X_1^3 + 0.56X_1 X_2 - ...$ (2)

The results of response surface analysis in the contour plot and surface plot are presented in figure 1A, 1B, and 1C. Figure 1A shows that either at low or high glue spread rate, the shear strength decreased due to an increase of crosslinker. The decreasing tendency was steeper at higher glue spread rate. Similarly, either at lower or higher crosslinker, the shear strength decreased due to an increase of glue spread rate. The decrease was much steeper at the higher crosslinker. Figure 1B shows that pressing time did not affect the shear strength very much, either at low or high crosslinker. However, the shear strength decreased due to an increase of crosslinker up to about zero point (14 g/m²); then, it increased when the crosslinker was getting higher. Figure 1C also shows that pressing time did not affect the shear strength very much, either at low or high glue spread rate. However, the shear strength increased due to the increase of glue spread rate, either at low or high pressing time.
Table 2. Shear strength values obtained from the optimization of application of natural rubber based API adhesive for producing laminated rubberwood.

| Run | Crosslinker (%) | Glue spread rate (g/m²) | Pressing time (hours) | Shear Strength (MPa) |
|-----|-----------------|-------------------------|----------------------|---------------------|
| 1   | 18              | 260                     | 18                   | 5.43                |
| 2   | 18              | 260                     | 6                    | 5.99                |
| 3   | 18              | 150                     | 18                   | 2.18                |
| 4   | 18              | 150                     | 6                    | 1.70                |
| 5   | 10              | 260                     | 18                   | 6.60                |
| 6   | 10              | 260                     | 6                    | 6.54                |
| 7   | 10              | 150                     | 18                   | 5.67                |
| 8   | 10              | 150                     | 6                    | 4.38                |
| 9   | 20.7            | 205                     | 12                   | 4.34                |
| 10  | 14              | 298                     | 12                   | 9.43                |
| 11  | 14              | 205                     | 22                   | 4.88                |
| 12  | 7.3             | 205                     | 12                   | 7.60                |
| 13  | 14              | 112                     | 12                   | 3.59                |
| 14  | 14              | 205                     | 2                    | 1.50                |
| 15  | 14              | 205                     | 12                   | 3.93                |
| 16  | 14              | 205                     | 12                   | 3.64                |
| 17  | 14              | 205                     | 12                   | 4.02                |
| 18  | 14              | 205                     | 12                   | 3.83                |
| 19  | 14              | 205                     | 12                   | 2.72                |
| 20  | 14              | 205                     | 12                   | 2.71                |

Table 3. Analysis of variance from the experiment of optimization of application of natural rubber based wood adhesive for producing laminated rubberwood.

| Sources                        | DF  | SS  | MS  | F   | p value | Effects |
|--------------------------------|-----|-----|-----|-----|---------|---------|
| Model                          | 9   | 72.67 | 8.07 | 14.42 | 0.0001 | S       |
| Crosslinker                    | 1   | 13.09 | 13.09 | 23.39 | 0.0007 | S       |
| Glue spread rate               | 1   | 30.63 | 30.63 | 54.71 | <0.0001 | S       |
| Pressing time                  | 1   | 3.54  | 3.54  | 6.33  | 0.0306 | S       |
| Crosslinker*Crosslinker        | 1   | 8.61  | 8.61  | 15.37 | 0.0029 | S       |
| Glue spread rate*Glue spread rate | 1 | 13.38 | 13.38 | 23.91 | 0.0006 | S       |
| Pressing time*pressing time    | 1   | 0.64  | 0.64  | 1.14  | 0.3115 | NS      |
| Crosslinker*Glue spread rate   | 1   | 2.48  | 2.48  | 4.42  | 0.0618 | NS      |
| Crosslinker*Pressing time      | 1   | 0.26  | 0.26  | 0.46  | 0.5145 | NS      |
| Glue spread rate*Pressing time | 1   | 0.64  | 0.64  | 1.15  | 0.3086 | NS      |
| Residual                       | 10  | 5.60  | 0.56  |       |         |         |
| Lack of fit                    | 5   | 3.79  | 0.76  | 2.09  | 0.2191 | NS      |
| Residual error                 | 5   | 1.81  | 0.36  |       |         |         |
| **Total**                      | 19  | 78.27 |       |       |         |         |

*DF= degree of freedom; SS = sum of square; MS = mean square; S= significant; NS = not significant

The optimum parameters of adhesive application and the optimum shear strength obtained from this experiment is presented in figure 1D. The optimum conditions were crosslinker 0.6 (16.4%), glue spread rate of 1.68 (298 g/m²) and pressing time of -1 (6 hours). The shear strength obtained was predicted 8.8 MPa. To see whether this value could meet the shear strength standard of laminated wood, we could compare this value with the standard mentioned in the JAS for Glue Laminated Timber [10]. In that particular standard, there are six wood classes, each with certain minimum value of shear strength, ranging from 5.4 to 9.6 MPa. There is no information about the criteria of wood classification in that
standard. It is probably based on the specific gravity (Sg) of the wood, from beech and oak, the highest, to cedar wood, the lowest. The list of specific gravity of wood could be seen in Miles and Smith [11]. The highest the Sg, the highest the minimum shear strength should be met, for example, minimum 9.6 MPa for beech or oak, and minimum 5.4 MPa for cedar. Rubberwood is not mentioned in the list of wood classification in that standard. Since rubberwood has Sg of around 0.6 [12, 13], which is close to the Sg of tamo or shioji wood (ash wood in English from the genus of Fraxinus), it is suggested that the laminated wood from rubberwood should have a minimum shear value of 8.4 MPa. Therefore, the value of shear strength at this optimum conditions from this experiment could meet the standard of shear strength stated in JAS for Glue Laminated Timber [10].

4. Conclusion

Figure 1. Contour plot obtained from the optimization of application of adhesive for producing laminated rubber wood, which show the effects of crosslinker and glue spread rate (A), the effects of crosslinker and pressing time (B), the effects of glue spread rate and pressing time (C), and shear strength obtained from one of optimum conditions of application of the adhesive for producing laminated wood (D).
Optimization of the application of natural rubber based API adhesive for the production of laminated rubber wood using the RSM has been successfully conducted and the model generated can be used for determining the shear strength of laminated wood bonded using the adhesive.

Acknowledgment
The authors gratefully acknowledge the Indonesian Institute of Sciences for funding the research through Program Unggulan LIPI in the fiscal year 2015.

5. References
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