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*Research Article*

# Preparation of Soy-Based Adhesive Enhanced by Waterborne Polyurethane: Optimization by Response Surface Methodology

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Response surface methodology was used to optimize the preparation conditions of soy-based adhesives (SBAs) in this work. The parameters such as the effects and interactions of waterborne polyurethane (WPU) addition level ($X_1$), temperature ($X_2$), and time ($X_3$) on wet shear strength ($Y$) were investigated. The regression model for SBA preparation was significant ($p = 0.0034 < 0.05$). The coefficient of determination ($R^2$) of this model was to be 0.9256. According to the results, WPU addition level ($X_1$) had a significant influence on the wet shear strength, whereas reaction temperature ($X_2$) and reaction time ($X_3$) were not significant. The optimal preparation conditions of SBA were 12 wt.% WPU addition level for 101 min at 76°C. Under the optimal conditions, the wet shear strength was 1.07 ± 0.08 MPa, which was in good agreement with the model predicted value. An analysis of FTIR spectra of WPU, soy flour, and soy-based adhesive further confirmed the validity of the model.

## 1. Introduction

Formaldehyde-based adhesives such as urea-formaldehyde (UF), phenol-formaldehyde (PF), and melamine-formaldehyde (MF) are widely applied in wood industry due to their excellent adhesion performance and low cost [1]. However, there are two challenges remaining for these types of adhesives. On the one hand, formaldehyde-based adhesives derive from fossil resources, which are limited and nonrenewable. On the other hand, formaldehyde emission caused by massive production and application of formaldehyde-based adhesives is harmful and toxic. Formaldehyde has already been classified as possibly carcinogenic to human health by the World Health Organization (WHO), and the use of formaldehyde-based adhesives may contaminate environment and damage health of workers and consumers, especially infants.

Therefore, the need to find an environment-friendly and sustainable substitute is urgent at present. Great efforts have been made to develop formaldehyde-free adhesives from natural biomaterials, especially soy flour. Soy flour is one of the research focuses because not only it has high content of soy protein, but also it is abundant, affordable, and readily available [2, 3]. Though soy flour is an attractive raw material for wood adhesives, the application of soy-based adhesives as an alternative of traditional formaldehyde-based adhesives is severely restricted by poor water resistance. Then, it is necessary to improve water resistance in order to broaden the application of soy-based adhesives [1, 4]. Soy protein mainly consisted of 20 different kinds of amino acids with various functional groups bonded in the side chains of polypeptide, including hydroxyl (−OH), carboxyl (−COOH), thiol (−SH), and amino ($-NH_2$) [5, 6]. Through reacting with these functional groups, the physical and chemical properties of soy-based adhesives can be changed by directional modification.

In the past years, many attempts have been carried out to improve water resistance of soy-based adhesives. Most efforts have focused on unfolding soy proteins initially, exposing hydrophobic subunits and then employing methods such as enzymatic modification [7], blending with other resins [8]. Besides the abovementioned methods, cross-linking modification is found to be more suitable and affordable [9–12]. So far, many high-effective cross-linking agents, including itaconic acid-based PAE (polyamidoamine-epichlorohydrin) [13], 2-octen-1-ylsuccinic anhydride [14], undecylenic acid [15], polyamidoamine resin [16], have been presented. However, most of them are usually not readily available and

expensive for massive industrialized production. Therefore, these kinds of cross-linking agents are not suitable for large-scale production.

Considering both modification effect and cost, waterborne polyurethane (WPU) resins are chosen as cross-linking agent in this paper. WPU resins not only have good mechanical properties and low water absorption that can meet the requirements of modification of soy-based adhesives, but also are an environmental-friendly cross-linking agent that can resolve the concerns in formaldehyde emission and environment contamination. In addition, WPU resins are widely applied in many fields of industrialized production, including adhesives, films, foams, plastics, coating, and so on. Few reports have been reports that used WPU to modify soy protein to prepare films and foams [17–19]. Based on the same idea, the application of WPU modified soy-based wood adhesives has been reported in the present.

Response surface methodology (RSM) is an effective experimental design methodology, which can explore the interactions between independent variables and one or more dependent variables and predict their responses under specified sets of conditions [20].

Therefore, the objective of this study was to research the interactions between independent variables (WPU addition level $X_1$, reaction temperature $X_2$, and reaction time $X_3$) and dependent variable (wet shear strength $Y$) and optimize the preparation conditions to prepare soy-based adhesives (SBAs) by applying RSM.

## 2. Materials and Methods

*2.1. Materials.* Soy flour (protein: 52%; moisture: 10%) purchased from Xianglin Food Co., Ltd, China, was used as received. WPU was a commercial product and purchased from Guanzhi New Material Technology Co., Ltd, with the name of AH-1610 (solid content about 38%, pH 7–9, and viscosity 100–200 MPa·s). All other chemicals were of analytical grade and purchased from Sinopharm Chemical Reagent Co., Ltd, China. Poplar wood veneers with the dimensions of 600 mm × 600 mm × 1.6 mm (width × length × thickness) were provided by Zhensheng Wood Industry Co., Ltd, China. The moisture content of the veneer samples was no more than 10%.

*2.2. Infrared Spectroscopy.* Fourier transform infrared spectroscopic (FTIR) data of WPU, soy flour, and SBA were recorded with a NICOLET-is 5 spectrometer (Thermo Fisher, USA). The samples of WPU and SBA were kept into the oven for 2 h at 140°C. The cured samples were ground into fine powder. A total of 16 scans were performed with a resolution of 4 $cm^{-1}$ in the range 650–4000 $cm^{-1}$, using an attenuated total reflectance accessory (ATR).

*2.3. Preparation of Soy-Based Adhesives.* A 500 ml three-neck flask equipped with a mechanic stirrer, a condenser, and a thermometer was charged with distilled water (190.5 g), ethylene glycol (1.5 g), sodium hydroxide (7.5 g), and urea (1.5 g). The solutions were blended together, and then, soy flour (99 g) was slowly added to the flask with rapid agitation. The mixture was heated to 80°C in 15–20 min and held for another 60 min. After addition of different amount of WPU into the SBA slurry, it was stirred for different times at different temperatures. The slurry was cooled to room temperature and adjusted to a pH value between 8.0 and 9.0 with 10 wt.% ammonium chloride solution.

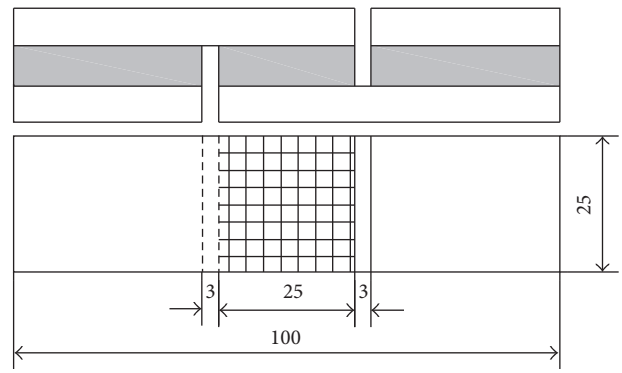


Figure 1: Test sample size of plywood.

*2.4. Preparation of Plywood Samples.* All obtained SBAs were applied to prepare duplicate samples of three-layer plywood panels by coating 350 g/$m^2$ of the adhesives on both sides of veneer. Then, the coated veneer was stacked between two uncoated ones with the grain directions of two adjacent veneers perpendicular to each other. These assembled plywood samples were cold-pressed at 0.8 MPa for 60 min at room temperature and then hot-pressed at 1.2 MPa and 85 s/mm at 120°C. After hot press, the panels were stored at ambient temperature for at least 24 h before tests.

*2.5. Wet Shear Strength Test of Plywood Samples.* Test samples shown in Figure 1 were cut from plywood panels along the face-grain axis in accordance with Chinese National Standard GB/T 9846.7-2004. The shear strength was measured according to Chinese National Standard GB/T 17657-1999. The process was as follows: the samples were firstly immersed into water at 63°C for 3 h, and then, these soaked samples were tested right in wet state after cooling for about 10 min at room temperature. Finally, the tension-testing machine (Jinan Shijin Co. Ltd., China) with a crosshead speed of 2.0 mm/min was applied for measuring the shear strength of the plywood samples.

*2.6. Single-Factor Design for Wet Shear Strength.* A single-factor experiment was carried out to determine the preliminary range of wet shear strength including WPU addition level (2~14 wt.%), reaction temperature (60~80°C), and reaction time (20~140 min). The wet shear strength was the dependent variable.

*2.7. Response Surface Methodology (RSM) Experimental Design.* The software Design Expert (Trial Version 10.0.1.0, Stat-Ease Inc., Minneapolis, USA) was employed for experimental design. Based on the previous single-factor

Table 1: Variables and levels used in RSM design.

| Coded and uncoded variables | Levels | | |
|---|---|---|---|
| Factors | −1 | 0 | 1 |
| WPU addition level ($X_1$, %) | 8 | 10 | 12 |
| Temperature ($X_2$, °C) | 60 | 70 | 80 |
| Time ($X_3$, min) | 80 | 100 | 120 |

experiment, the standard RSM design called BBD (Box–Behnken Design) was applied to research the influence of three independent variables s ($X_1$, WPU addition level; $X_2$, temperature; and $X_3$, time) at three levels on the dependent variable ($Y$) and optimize the conditions for wet shear strength. The independent variables and its levels are presented in Table 1. Similarly, the results of the whole design comprising 17 experimental points performed in randomized order are presented in Table 2.

The following second-order polynomial model was used to describe the relationship between the dependent variable and the independent process factors [21–23]:

$$Y = \beta_0 + \sum_{j=1}^{k} \beta_j X_j + \sum_{j=1}^{k} \beta_{jj} X_j^2 + \sum \sum_{i<j} \beta_{ji} X_i X_j, \quad (1)$$

where $Y$ is the predicted response; $\beta_0$ is a constant; $\beta_j, \beta_{jj}$, and $\beta_{ji}$ are the linear, quadratic, and interactive coefficients, respectively; $X_i$ and $X_j$ are the independent coded variables ($i \neq j$; $i$ and $j$ ranging from 1 to $k$); and $k$ is the number of independent parameters (in this research, $k = 3$).

The quality of the fit of the polynomial model equation was assessed by the coefficient of determination ($R^2$) and ANOVA. The significance of the regression coefficient was evaluated by checking $F$ value and $p$ value [24, 25].

## 3. Results and Discussion

### 3.1. Single-Factor Experimental Analysis

*3.1.1. Effect of WPU Addition Level on Wet Shear Strength.* The effect of WPU addition level on wet shear strength is shown in Figure 2. Wet shear strength was performed by applying WPU addition level ranging from 2 wt.% to 14 wt.% with the other reaction conditions as follows: the reaction temperature was 80°C and the time was 120 min. When the WPU addition level exceeded 6 wt.%, wet shear strength reached 0.76 ± 0.02 MPa that could meet the requirement for type II plywood applications in accordance with the Chinese Standard GB/T 9846.3-2004. In addition, the more the WPU addition level was, the greater the wet shear strength could be. However, the trend of wet shear strength was increasing even when the WPU addition level reached 14 wt.%. Considering the cost and modification effect, the optimal condition in the present experiment should be ranged from 8 wt.% to 12 wt.%.

*3.1.2. Effect of Reaction Temperature on Wet Shear Strength.* The reaction temperature was an important factor to fabricate soy-based adhesive. Thus, the effects of reaction temperature ranging from 30°C to 90°C were investigated under WPU addition level of 10 wt.% and reaction time of 120 min. As shown in Figure 3, when the temperature was below 70°C, wet shear strength increased with the increased in reaction temperature, which was 0.94 ± 0.04 MPa.

Table 2: Experimental scheme and results.

| Run number | WPU addition level ($X_1$, %) | Reaction temperature ($X_2$, °C) | Reaction time ($X_3$, min) | Wet shear strength ($Y$, MPa) |
|---|---|---|---|---|
| 1 | 10.00 | 80.00 | 120.00 | 0.97 |
| 2 | 8.00 | 70.00 | 80.00 | 1.03 |
| 3 | 10.00 | 60.00 | 120.00 | 1.02 |
| 4 | 10.00 | 70.00 | 100.00 | 1.18 |
| 5 | 10.00 | 70.00 | 100.00 | 1.13 |
| 6 | 12.00 | 70.00 | 80.00 | 1.06 |
| 7 | 8.00 | 80.00 | 100.00 | 0.98 |
| 8 | 10.00 | 70.00 | 100.00 | 1.14 |
| 9 | 8.00 | 60.00 | 100.00 | 1.09 |
| 10 | 10.00 | 70.00 | 100.00 | 1.12 |
| 11 | 12.00 | 70.00 | 120.00 | 1.12 |
| 12 | 12.00 | 60.00 | 100.00 | 0.97 |
| 13 | 12.00 | 80.00 | 100.00 | 1.14 |
| 14 | 8.00 | 70.00 | 120.00 | 0.93 |
| 15 | 10.00 | 60.00 | 80.00 | 1 |
| 16 | 10.00 | 70.00 | 100.00 | 1.13 |
| 17 | 10.00 | 80.00 | 80.00 | 1.05 |

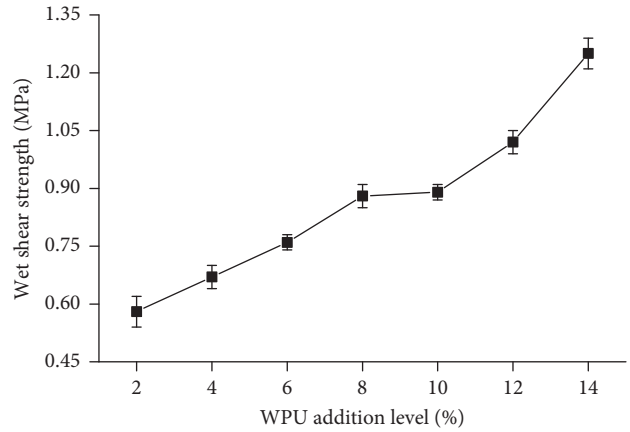


Figure 2: Effects of WPU addition level on wet shear strength.

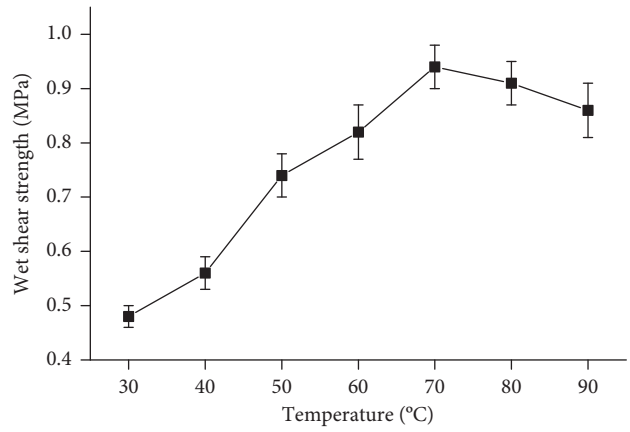


Figure 3: Effects of reaction temperature on wet shear strength.

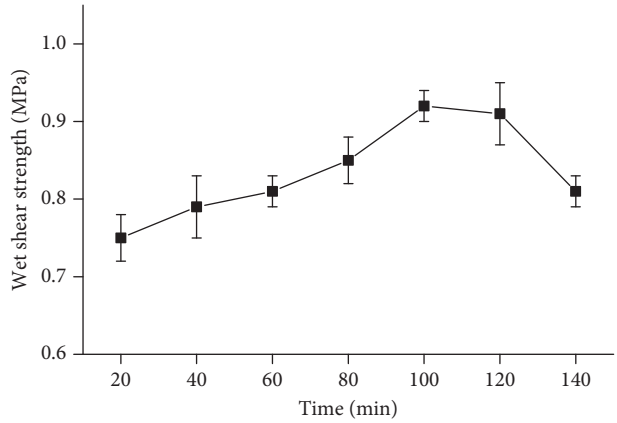


Figure 4: Effects of reaction time on wet shear strength.

Table 3: Variables and levels used in RSM design.

| Source | SS | *Df* | MS | *F* value | *p* value | Significance* |
|---|---|---|---|---|---|---|
| Model | 0.084 | 9 | $9.328 \times 10^{-3}$ | 9.67 | 0.0034 | ** |
| $X_1$ | $8.45 \times 10^{-3}$ | 1 | $8.45 \times 10^{-3}$ | 8.76 | 0.0211 | * |
| $X_2$ | $4.5 \times 10^{-4}$ | 1 | $4.5 \times 10^{-4}$ | 0.47 | 0.5165 | — |
| $X_3$ | $1.25 \times 10^{-3}$ | 1 | $1.25 \times 10^{-3}$ | 1.30 | 0.2924 | — |
| $X_1X_2$ | 0.02 | 1 | 0.02 | 20.33 | 0.0028 | ** |
| $X_1X_3$ | $6.4 \times 10^{-3}$ | 1 | $6.4 \times 10^{-3}$ | 6.64 | 0.0367 | * |
| $X_2X_3$ | $2.5 \times 10^{-3}$ | 1 | $2.5 \times 10^{-3}$ | 2.59 | 0.1514 | — |
| $X_1^2$ | $5.158 \times 10^{-3}$ | 1 | $5.158 \times 10^{-3}$ | 5.35 | 0.0540 | — |
| $X_2^2$ | 0.015 | 1 | 0.015 | 15.72 | 0.0054 | ** |
| $X_3^2$ | 0.021 | 1 | 0.021 | 21.40 | 0.0024 | ** |
| Residual | $6.75 \times 10^{-3}$ | 7 | $9.643 \times 10^{-4}$ | — | — | — |
| Lack of fit | $4.55 \times 10^{-3}$ | 3 | $1.517 \times 10^{-3}$ | 2.76 | 0.176 | — |
| Pure error | $2.2 \times 10^{-3}$ | 4 | $5.5 \times 10^{-4}$ | — | — | — |
| Correlation total | 0.091 | 16 | $9.328 \times 10^{-3}$ | — | — | — |
| $R^2$ | 0.9256 | — | — | — | — | — |

*Note.* *Significant at the 5% level ($p < 0.05$); **significant at the 1% level ($p < 0.01$).

However, with temperature further increased, the wet shear strength decreased slightly. Therefore, the optimal reaction temperature was 60°C to 80°C.

*3.1.3. Effect of Reaction Time on Wet Shear Strength.* By fixing the WPU addition level of 10 wt.% and reaction temperature of 70°C, respectively, the effects of reaction time ranging from 20 to 140 min were studied. Figure 4 shows that the wet shear strength changed under the different reaction time. When the reaction time was 100 min, the wet shear strength reached its peak value. Therefore, the optimal condition of reaction time was ranging from 80 min to 120 min.

*3.2. Statistical Analysis and Model Fitting.* The important independent variables ($X_1$, $X_2$, and $X_3$) influencing the wet shear strength ($Y$) and the optimum values of the parameters were determined by BBD, and the results are shown in Table 2. The coefficient of the independent variables ($X_1$, $X_2$, and $X_3$) for the dependent variable ($Y$) could be expressed by the following second-order polynomial equation:

$$\begin{aligned} Y = {} & 1.04 + 0.0325X_1 + 0.0075X_2 - 0.0125X_3 \\ & + 0.07X_1X_2 + 0.04X_1X_3 - 0.025X_2X_3 \\ & - 0.035X_1^2 - 0.06X_2^2 - 0.07X_3^2, \end{aligned} \quad (2)$$

where a positive or negative coefficient illustrates a synergetic and antagonistic influence, respectively [26, 27].

In order to determine whether the second-order polynomial model was significant, it was necessary to apply ANOVA. The results of the analysis of variance and fitness of the model are presented in Table 3. As could seen from Table 3, the $p$ value is 0.0034 that was much lower than 0.05, indicating that the second-order polynomial model was suitable to illustrate the effects of the independent variables ($X_1$, $X_2$, and $X_3$) on the wet shear strength. Meanwhile, the value of the coefficient of determination ($R^2$) was 0.9256, which indicated 92.56% of the total variation in wet shear strength attributed to the experimental variables. Therefore, it could conclude that the model could be applied to predict the wet shear strength in the response.

The lack of fit was used to evaluate the validity of the model. In this model, the $F$ value and $p$ value of the lack of fit

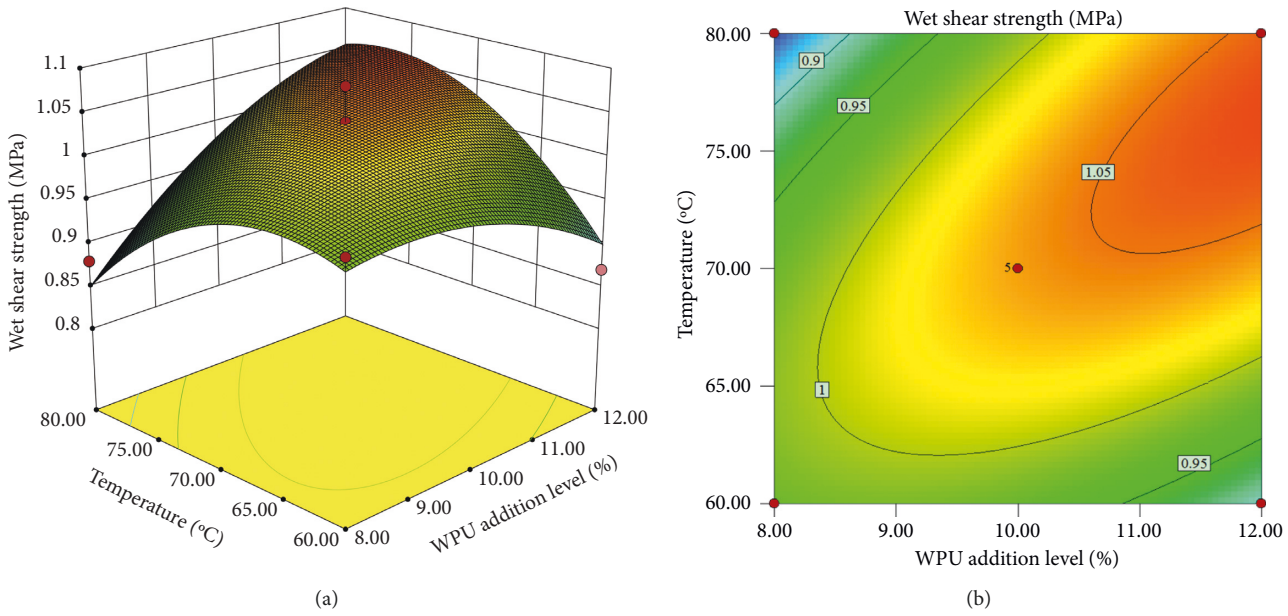


Figure 5: Response surface and contour plots for effect of temperature and WPU addition level on wet shear strength.

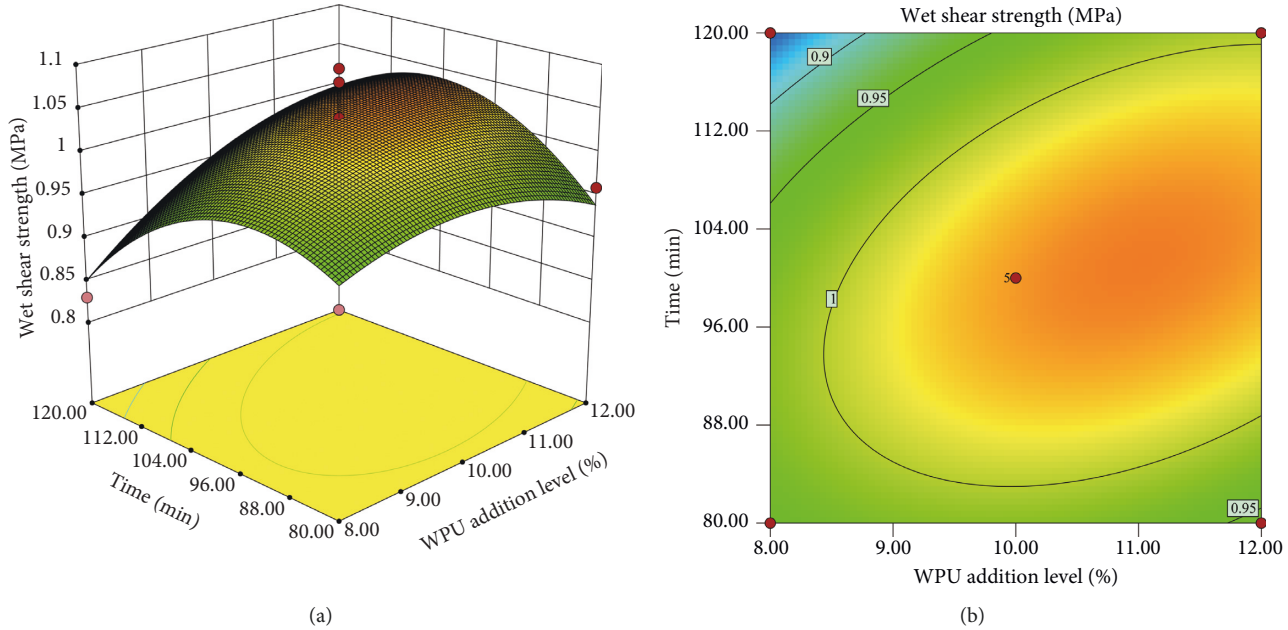


Figure 6: Response surface and contour plots for effect of time and WPU addition level on wet shear strength.

were 2.76 and 0.176, respectively, which implied the lack of fit was not significant relative to the pure error. From another point of view, the application of this model to explain the relationship between the dependent variable and independent variables was credible.

The WPU addition level ($X_1$) had a significant influence on the wet shear strength based on the analysis of significance of the model equation coefficient (Table 3). Meanwhile, the interaction terms ($X_1X_2$, $X_1X_3$) and the quadratic terms ($X_1^2$, $X_2^2$, $X_3^2$) were also significant ($p < 0.05$). However, the other variables, including reaction temperature ($X_2$), reaction time ($X_3$), and the interaction term ($X_2X_3$), were not significant. Based on the results above, the effects of these factors on wet shear strength were not simple linear relationship [28–30].

*3.3. Analysis of Response Surface.* By employing RSM, the influences of the $X_1$, $X_2$, and $X_3$ on $Y$ were depicted in 3D response surface plots and 2D contour plots, which are illustrated in Figures 5–7, respectively. As observed from Figures 5 and 6, it was evident that the independent variable $X_1$ had a significant influence ($p = 0.0211 < 0.05$) on $Y$. It was generally known that WPU containing both amino

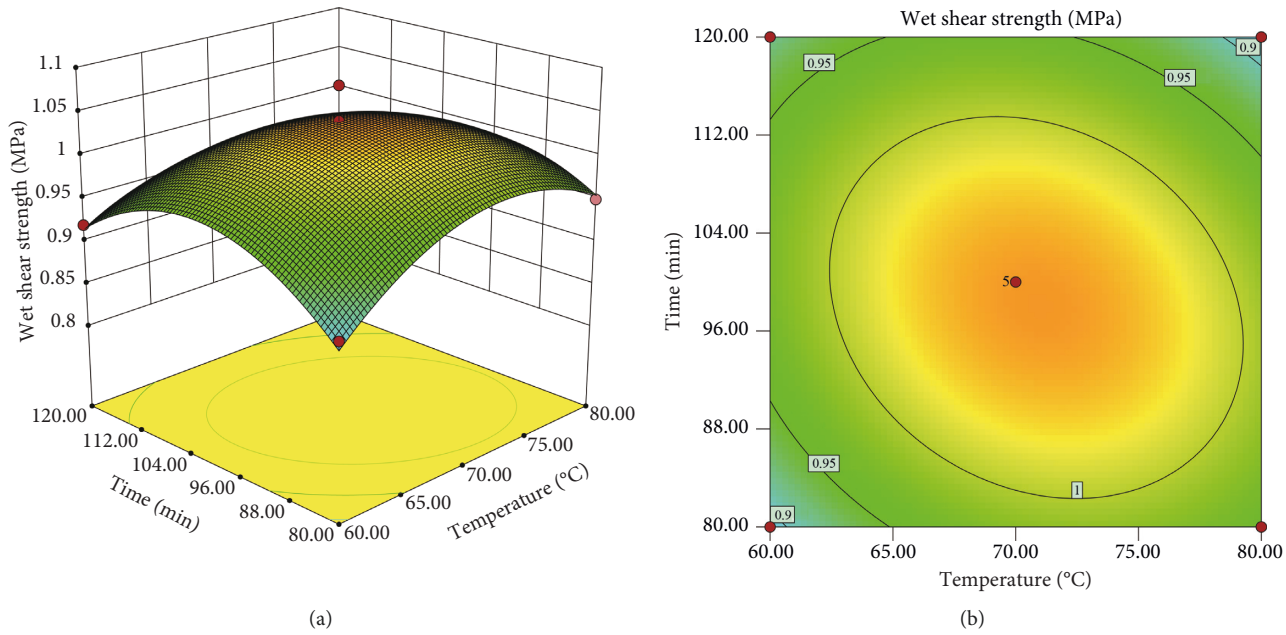


Figure 7: Response surface and contour plots for effect of time and temperature on wet shear strength.

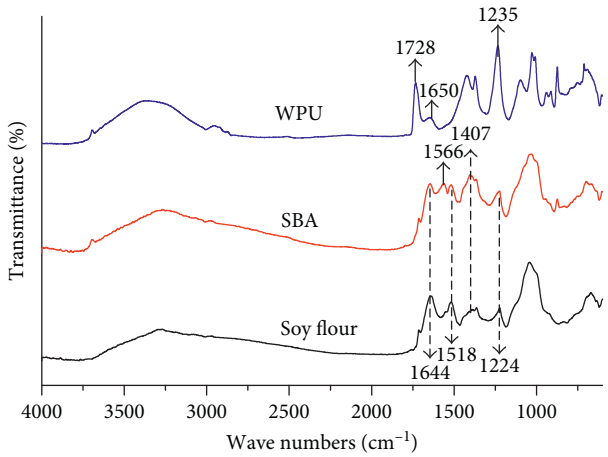


Figure 8: FTIR spectra of WPU, soy flour, and SBA with 12 wt.% WPU addition.

group ($-NH_2$) and hydroxyl group (–OH) performed as the cross-linker in soy-based adhesives. During the curing process, macromolecule of WPU could react with both soy protein and reducing sugar of defatted soy flour, and the active groups (–OH, –COOH, etc.) on wood macromolecule [16]. With the increase of $X_1$, the wet shear strength was improving significantly. The reason was that the solidified adhesive formed a water-insoluble cross-linking network structure, resulting in restricting the penetration of water into the glue layer and hence enhancing the water resistance of SBA with WPU modification [16, 31].

However, the influences of the $X_2$ and $X_3$ were not significant ($p > 0.05$). The $X_2$ on the $Y$ had a different trend that $Y$ increased at first and then decreased. In addition, the interaction effect of $X_1X_2$ on $Y$ was highly significant ($p = 0.0028 < 0.01$). Low reactivity in moderate temperature and high reactivity of WPU were responsible for this interaction effect [32, 33]. $Y$ was less influenced by $X_3$, and the effect was in the reverse direction. As shown in Figure 7, $Y$ decreased with the increase in $X_2$. The interactions between WPU addition level and the other two variables ($X_2$ and $X_3$) influenced $Y$ significantly, illustrating a positive effect on $Y$ when the $X_1$ varied from 8 wt.% to 12 wt.%. Meanwhile, the interaction effect of $X_2X_3$ on $Y$ was insignificant ($p > 0.05$).

*3.4. Optimization of Modification Conditions and Model Validation.* The predicted optimal conditions derived from surface response experiments were obtained by using (2) derived from RSM and illustrated as follows: WPU addition level of 12%, temperature of 76.29°C, and time of 101.68 min. Under the conditions, the predicted maximum value of wet shear strength was 1.06 MPa. In order to verify the accuracy of the optimization procedure, a validation test including three parallel experiments was performed by applying modified optimal conditions of 12% WPU addition level for 101 min at 76°C. The observed average value of wet shear strength was $1.07 \pm 0.08$ MPa, which is in good agreement with the model predicted value. The result confirmed the validity and accuracy of the models.

*3.5. FTIR Analysis.* Figure 8 provides the FTIR spectra of WPU, soy flour, and SBA. Soy flour and SBA both had infrared absorption bands of amide I, amide II, and amide III, the characteristic groups of soy protein at 1644 $cm^{-1}$, 1518 $cm^{-1}$, and 1224 $cm^{-1}$ attributing to C=O stretching, N-H bending, and C-N stretching vibrations, respectively [8, 16]. In addition, the characteristic absorption peaks of WPU from –NH bending vibration could be observed at 1235 $cm^{-1}$. The double bond in the polyol structure was observed at 1728 $cm^{-1}$ and 1650 $cm^{-1}$ [18, 34]. After curing process, the typical absorption

peak at 1566 $cm^{-1}$ appeared, implying reactions between soy protein and WPU molecule. Moreover, the absorption peak at 1235 $cm^{-1}$ of SBA compared with WPU decreased that indicated the reactions between soy protein and WPU molecule. What is more, the COO− absorption band (1368 $cm^{-1}$) of SBA was red-shifted to lower wavenumbers (1407 $cm^{-1}$) with the addition of WPU that also indicated the intermolecular bonds increasing between soy protein and WPU molecule. Therefore, the FTIR analysis confirmed the reactions between soy protein and WPU during curing process.

## 4. Conclusions

In this research, SBA with high water resistance was prepared and the preparation conditions were optimized by applying RSM. The statistical analysis illustrated a significantly good fit for the model that could be used to navigate the experimental design space. The regression model for SBA preparation was significant ($p = 0.0034 < 0.05$) and the coefficient of determination ($R^2$) of this model was to be 0.9256. Moreover, the WPU addition level had a significant effect on wet shear strength, whereas the reaction time and temperature were not significant. However, the combined effects of WPU addition level and the other two variables were significant. Moreover, the modified optimal conditions were 12 wt.% WPU addition level for 101 min at 76°C. The corresponding value of wet shear strength of the validation test was $1.07 \pm 0.08$ MPa, which was in good agreement with the predicted one. FTIR analysis of WPU, soy flour, and SBA further confirmed the validity of the model. This study may provide useful tools to prepare soy-based adhesives.

## Data Availability

The data used to support the findings of this study are available from the corresponding author upon request.

## Conflicts of Interest

The authors declare that they have no conflicts of interest.

## Acknowledgments

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