Effect of Soy Protein on the Toughness and Bonding Performances of Cold-Setting Melamine-Urea-Formaldehyde Resin Adhesive

Jiankun Liang,[a,#] Bengang Zhang,[b,#] Hui Yang,[a] Tong Meng,[c] Chuchu Chen,[c] Huali Li,[c] Cheng Li,[a,]* and Zhigang Wu [c,]*

The effects of adding defatted soybean flour (SF) at different stages of the "alkali-acid-alkali" process were studied relative to the viscosity, impact toughness, bonding strength, and water resistance of melamine-urea-formaldehyde (MUF) resin. The results showed that the influence of SF on the strength properties and water resistance of the resin exhibited distinct stage-specific and dosage-dependent characteristics. In the first stage, an appropriate amount of SF (4% to 6%) significantly improved the resin's viscosity, impact toughness, and bonding strength, demonstrating good toughening and strengthening effects. In the second stage, the addition of SF led to more complex and somewhat unstable effects on viscosity and impact toughness. In the third stage, a moderate amount of SF (2% to 6%) evidently enhanced bonding strength, but high dosages (8% and 10%) resulted in a decrease in water resistance and impact toughness.

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*Contact information: a: College of Civil Engineering, Kaili University, Qiandongnan 556011, China; b: Forest Products Industry Research Institute, Jiangxi Academy of Forestry Sciences, Nanchang 330013, China; c: College of Forestry, Guizhou University, Guiyang 550025, China; #: These two authors contributed equally to this work; *Corresponding authors: licheng_730@163.com; wzhigang9@163.com*

INTRODUCTION

Glued wood is capable of retaining the intrinsic structural characteristics of solid wood to the greatest extent. Through a series of processing steps during manufacturing—such as drying, defect removal, and precision machining—its overall performance is significantly enhanced compared to that of natural solid wood. Glued wood can exhibit superior properties in terms of compressive strength, tensile strength, and material uniformity. It effectively achieves the utilization of small-dimension and lower-grade timber for structural purposes, thereby promoting a more rational and efficient application of wood in construction and substantially improving the overall percentage of material utilization (Brandner *et al*. 2016; Martins *et al*. 2019; Dias *et a*l. 2020; Autengruber *et al*. 2021; Yin *et al*. 2022; Faircloth *et al*. 2024). As a result, in many technologically advanced countries, glued wood has been widely adopted and promoted, becoming one of the predominant forms of modern timber construction. Adhesives are a critical factor in determining the water resistance, durability, and weatherability of glued wood.

Currently, melamine-urea-formaldehyde (MUF) co-condensation resin represents a focal point in the development and application of adhesives for glulam structures (Sauget *et al*. 2014; Lei and Frazier 2015; Yang *et al*. 2024). However, the cured MUF resin possesses relatively short internal network chains, high cross-linking density, and significant steric hindrance, which limit the flexibility of its molecular chains. As a result, the cured resin exhibits high rigidity, pronounced brittleness in the adhesive layer, and poor impact resistance (Zhang *et al*. 2022; Song *et al*. 2022; Li *et a*l. 2023; Xu *et al*. 2024). When applied in glued wood, these characteristics may pose potential safety risks.

Recent research on the toughening of MUF resin has primarily focused on chemical modification approaches. These methods aim to increase the distance between triazine rings and reduce cross-linking density, thereby enhancing the flexibility of molecular chains and ultimately improving impact strength, bending strength, and overall toughness (Karakaya *et a*l. 2007; Liang *et al*. 2020; Huang *et al*. 2024). Soy-based products—including defatted soybean flour (SF), soybean protein concentrate (SPC), and soybean protein isolate (SPI)—are considered ideal renewable raw materials and modifiers for environmentally friendly adhesives, as they emit no volatile organic compounds (VOCs). The amino acid side chains of soy proteins contain functional groups such as amino, carboxyl, hydroxyl, sulfhydryl, and phenolic hydroxyl groups, which can chemically react with active groups present in MUF resin (Liu *et al*. 2022; Chen *et al*. 2023; Yu *et al*. 2024; Hou *et a*l. 2025). On the one hand, the long-chain protein molecules interpenetrate the MUF network, not only enhancing the overall flexibility of the adhesive but also reinforcing the cross-linked structure and improving cross-linking density. The cured adhesive layer forms a dense and compact network structure, which effectively prevents water intrusion, thereby significantly improving the water resistance of the adhesive. On the other hand, the toughening effect of these long-chain protein molecules helps alleviate the brittleness induced by excessive cross-linking in the MUF system and moisture evaporation during the hot-pressing process, thus preventing deterioration of bonding performance (Qu *et al*. 2015; Liu *et al*. 2018; Bacigalupe *et al*. 2020; Zhang *et al*. 2020; Li *et a*l. 2024). From a macroscopic perspective, the modified adhesive exhibits excellent bonding strength and water resistance.

This study investigated the effects of incorporating SF at different stages of the "alkali–acid–alkali" process on the viscosity, impact toughness, bonding strength, and water resistance of MUF resin. The modification of synthetic resin using a natural material such as SF not only leads to significant enhancements in the performance of conventional MUF resin, but it also offers a novel strategy and pathway for the development of new, more environmentally friendly wood adhesives.

EXPERIMENTAL

Materials

Melamine, urea, NaOH, formic acid, and formaldehyde (37 wt%) used were analytically pure and were purchased from Sinopharm Chemical Reagent Co., Ltd., Shanghai, China. Defatted soybean flour (100 mesh, protein content 53.4%) was supplied by Shandong Yuxin Soy Protein Co., Ltd. Rubberwood panels, with a density of 0.65 g/cm^3, were obtained from Jingshan Timber Business Department, Baoshan District, Shanghai, China.

Preparation of Melamine-Urea–Formaldehyde Resins

An appropriate amount of formaldehyde was added into a three-neck round-bottom flask equipped with a mechanical stirrer, thermometer, and reflux condenser. The contents were heated in a water bath condition at 50 °C. The pH of the system was adjusted to 9.0, followed by the addition of the first portion of urea (U_1 = 75.20 g) and the first portion of melamine (M_1 = 12.21 g). The mixture was then heated to 90 °C to initiate the first stage of the reaction. Subsequently, the pH was adjusted to 5.0 and the temperature was maintained for 60 min to proceed with the second stage. After that, the pH was readjusted to the range 8.7 to 8.9, and the second portion of melamine (M_2 = 69.19 g) was added, followed by continued reaction at constant temperature for 100 min (third stage). Then, the pH was raised to 9.0. Once the temperature dropped to 45 °C, the second portion of urea (U_2 = 17.10 g) was added. After maintaining the reaction for 10 min, the pH was adjusted to 8.0 to complete the reaction. The resulting product was collected and stored, yielding the control melamine-urea-formaldehyde resin, with a molar ratio of n(F): n(M+U) = 1.9:1 (Zhang *et al*. 2022).

For the preparation of SF-modified MUF resins, varying proportions of SF (2%, 4%, 6%, 8%, and 10% based on the total mass of urea) were added at different stages of the synthesis as follows: SF was added concurrently with the first addition of urea (U_1) and melamine (M_1) during the first stage; during the second stage, SF was introduced after adjusting the pH to 5.0; in the third stage, SF was added together with the second melamine portion (M_2). The viscosity of the adhesives was measured in accordance with the GB/T 14074-2006 standard.

Impact Toughness Test of MUF

In accordance with the standard GB/T 1043.1 (2008), the resin was cured into a notch-free specimen with a width of 10 mm and a thickness of 4 mm. Subsequently, the impact strength of the specimen was evaluated using a simply supported beam impact testing machine.

Preparation of Glued Wood and Test of Shear Strength

According to the GB/T 26899 (2011), glued wood was prepared using a cold-pressing method using control MUF and experimental group. The specific procedure was as follows: rubberwood specimens with dimensions of 30 mm × 25 mm × 10 mm were coated with adhesive and left at room temperature for 20 min. The specimens were then subjected to compression curing using a flat-plate vulcanizing press, and their shear strength was subsequently tested. The processing parameters were as follows: a pressure of 3.5 MPa was applied for 2 h; the adhesive loading was 300 g/m^2; manual application of adhesive was used; and the bonding area was 25 mm × 25 mm.

Water Resistance Test of Glued Wood

Rubberwood specimens with dimensions of 75 mm × 50 mm × 10 mm were bonded using the prepared glued wood. In accordance with the GB/T 26899 (2011), the bonded specimens were subjected to delamination tests involving immersion in water at room temperature for 24 h and in boiling water for 4 h, respectively.

RESULTS AND DISCUSSION

Viscosity Analysis of MUF Resin

The "alkali–acid–alkali" process is a classical preparation method for MUF resin. The first stage involves a hydroxymethylation reaction, generating hydroxymethyl urea and hydroxymethyl melamine, which serve as the foundation for subsequent resin crosslinking. In the second stage, dehydration condensation occurs between low-molecular-weight hydroxymethylated products in an acidic medium, leading to a gradual increase in molecular weight. The third stage involves further crosslinking reactions between hydroxymethyl groups and amino groups, resulting in the formation of a more complex three-dimensional network structure (Xu *et al*. 2024). The viscosity changes of the resin with different amounts of SF added at each of the three stages are shown in Table 1.

From the viscosity results across the three stages, it is evident that the addition of SF significantly influenced the resin viscosity in a stage-dependent manner. In the first stage, the addition of SF markedly increased the resin viscosity, indicating its prominent role in promoting the hydroxymethylation and initial crosslinking reactions. In the second stage, the effect of SF on the resin viscosity was relatively small, suggesting that SF may interfere with the condensation reaction to some extent. This interference could limit the rate and extent of the condensation reaction, thus affecting the growth of resin molecular chains and the formation of the crosslinked network. In the third stage, the addition of SF again significantly increased the resin viscosity, particularly at higher addition levels, indicating its strong promotional effect on the crosslinking reaction under alkaline conditions.

Table 1. Effect of Adding Various Amounts of SF on the Viscosity of MUF Resin at Three Stages of Preparation

SF (%)	Viscosity at the First Stage (mPa·s)	Viscosity at the Second Stage (mPa·s)	Viscosity at the Third Stage (mPa·s)
0	20.13		
2	27.29	24.29	23.92
4	46.82	32.90	45.23
6	115.97	45.48	61.02
8	132.03	38.46	119.70
10	140.13	62.72	139.89

Toughness Analysis of MUF Resin

Toughness refers to the ability of an adhesive to absorb energy and deform without undergoing brittle fracture when subjected to external forces. The toughness of an adhesive plays a crucial role in bonding performance, as it not only enhances bonding strength, impact resistance, and durability, but it also improves processing performance and adapts to complex application environments (Zhang *et al*. 2022; Li *et a*l. 2023). Materials with good toughness generally exhibit higher impact strength, as they can absorb more energy upon impact without undergoing brittle fracture. This energy absorption capability is a key manifestation of toughness. During the preparation of MUF resin, several reactions occur upon the addition of SF: the reaction of SF with formaldehyde; the hydroxymethylation of urea with formaldehyde; the reaction between hydroxymethyl urea and melamine; and the reaction between hydroxymethyl protein and melamine. The crosslinking reaction between SF and MUF forms sufficient chemical bonds and a compact structural backbone, improving the bonding strength of the resin. Additionally, by introducing soybean protein structures with flexible groups into the network of molecular chains formed during the final crosslinking, the toughness of MUF resin is further enhanced. Figures 1 to 3 show the impact strength test results of MUF resin before and after modification.

During the modification process in the first stage (Fig. 1), the impact strength of the blank MUF resin was 6.40 kJ/m^2. When 2% SF was added, the impact strength of the modified resin increased to 6.79 kJ/m^2, an increase of 0.39 kJ/m^2, or approximately 6.1%. This result indicated that introducing a small amount of SF in the first stage can significantly improve the impact strength of the resin. Further increasing the SF content to 4% and 6% resulted in the resin reaching an impact strength of 6.88 kJ/m^2, an increase of 0.48 kJ/m^2 compared to the blank MUF. This suggests that within the 4% to 6% addition range, the incorporation of SF led to a relatively stable high value for the resin's impact strength. This is likely due to the good compatibility between SF and the resin in this range, allowing for effective integration into the resin system and the formation of a stable crosslinked structure, thereby maximizing the resin's toughness.

However, when the SF content continued to increase to 8% and 10%, the impact strength of the modified resin gradually decreased. This could be due to the difficulty in achieving adequate compatibility between excessive SF and the resin system, which interferes with the microstructure of the resin, disrupting the uniformity of the crosslinking network and thereby reducing the toughness. Additionally, excess SF may form aggregated structures within the resin, which act as stress concentration points. Under external impact, these aggregated structures are more likely to cause brittle fracture, resulting in a decrease in the resin's impact strength.

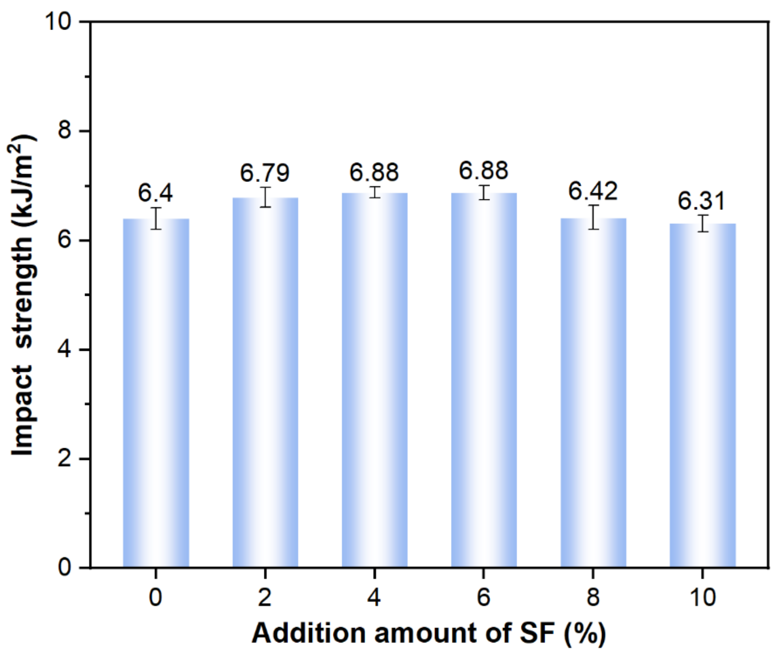

Fig. 1. Effect of adding various amounts of SF on the impact strength of MUF resin in the first stage

During the modification process in the second stage (Fig. 2), as the SF content increased, the impact strength of the resin exhibited an unstable trend, initially decreasing, then increasing, and subsequently decreasing again. This complex trend reflects the intricate interactions between SF and the resin under the reaction conditions of the second stage. This is because the addition of SF during the acidic stage of resin synthesis can influence the condensation reaction process. As the catalytic effect of the acidic conditions on the condensation reaction is quite sensitive, the incorporation of SF may interfere with the normal condensation pathway, preventing the resin from reaching the desired degree of polymerization. This insufficient polymerization directly affects the molecular structure and crosslinking density of the resin, leading to instability in its impact strength. Moreover, the inadequate polymerization may also weaken the resin's water resistance and bonding strength.

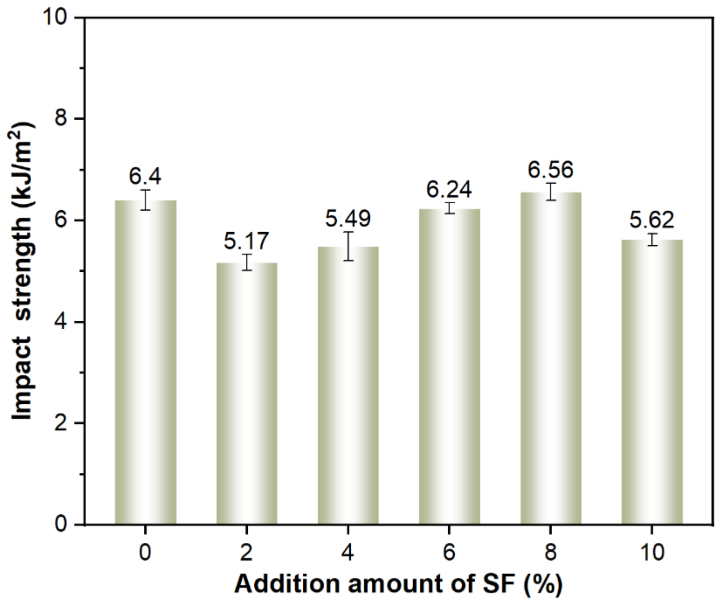

Fig. 2. Effect of adding various amounts of SF on the impact strength of MUF resin in the second stage

During the modification process in the third stage (Fig. 3), when the SF amount was varied from 2% to 6%, its effect on the resin's impact strength was not significant. This may be because, within this range, SF exhibited good compatibility with MUF resin, effectively integrating into the resin system. However, its impact on the resin's crosslinking structure and molecular arrangement was relatively minor, and therefore, its effect on improving impact strength was not pronounced. This also suggests that, under the reaction conditions of the third stage, the crosslinking structure of the resin was already relatively stable, and the addition of SF has limited ability to significantly optimize it. However, when the SF content increases to 8% and 10%, the impact strength of the MUF resin gradually decreases. This may be due to the excessive SF forming aggregated structures within the resin (Qu *et al*. 2015; Liu *et al*. 2018).

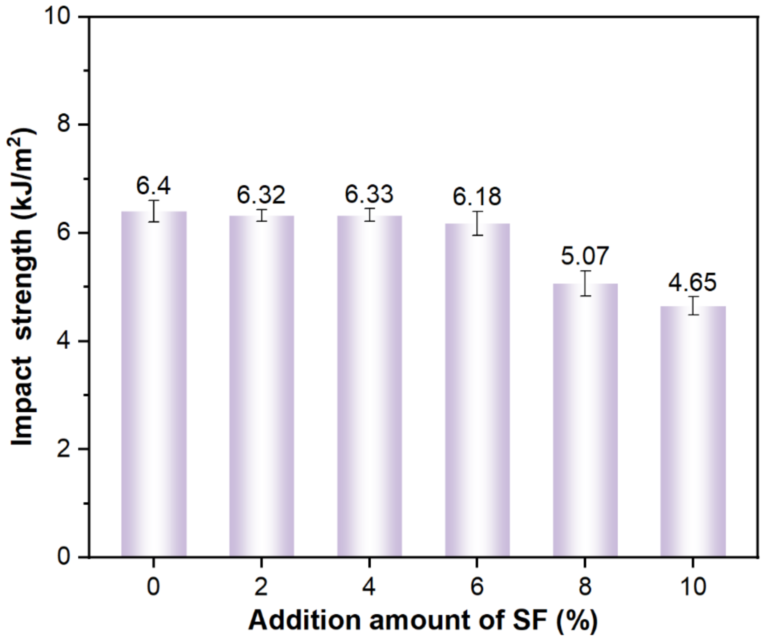

Fig. 3. Effect of adding various amounts of SF on the impact strength of MUF resin in the third stage

Overall, the modification results across the three stages reveal that SF's effect on the impact strength of MUF resin exhibited clear stage-specific and dosage-dependent characteristics. In the first stage, a moderate amount of SF (4% to 6%) significantly enhanced the impact strength of the resin. In the second stage, the effect of SF on impact strength was more complex. In the third stage, the influence of SF on impact strength was minimal, but at higher SF addition levels, it led to a decrease in impact strength.

Bonding Strength Analysis of Glued Wood Prepared with MUF Resin

Good toughness enables an adhesive to dissipate energy through deformation, thereby reducing stress concentration, and effectively preventing brittle fracture during mechanical loading. This enhances the bonding strength between the adhesive and the adherend. Therefore, materials with higher toughness generally exhibit greater bonding strength. Figures 4 to 6 present the bonding strength test results of MUF resins before and after modification.

During the first-stage of modification process (Fig. 4), the bonding strength of the unmodified MUF resin was 4.93 MPa. Upon the addition of 2%, 4%, and 6% SF, the bonding strengths increased to 6.09, 6.03, and 6.43 MPa, respectively. All of these values met the requirements of the national standard GB/T 26899-2011 for bonding strength (≥6.0 MPa), indicating that the incorporation of an appropriate amount of SF at this stage significantly enhanced the adhesive performance of the resin. However, as the SF content was further increased to 8% and 10%, the bonding strength began to decline. At 8% SF, the bonding strength dropped to 5.88 MPa, although this still represented an improvement of 0.95 MPa (approximately 19.3%) over the unmodified MUF. When the addition reached 10%, the bonding strength further declined to 4.90 MPa, which was comparable to that of the unmodified resin.

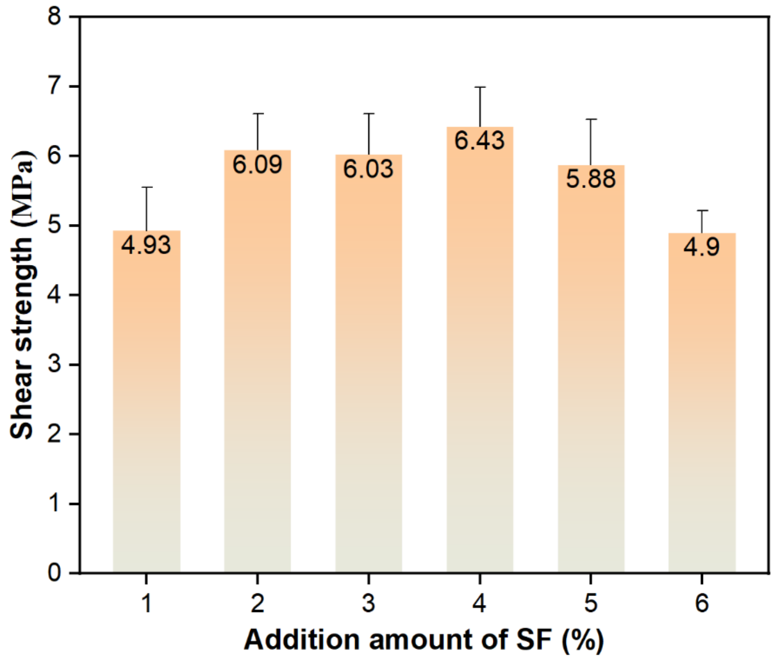

Fig. 4. Effect of adding various amounts of SF on the shear strength of MUF resin in the first stage

During the second-stage of modification process (Fig. 5), the bonding strength exhibited a relatively complex variation trend. This trend showed certain similarities with the results of impact toughness, suggesting a degree of correlation between the two performance indicators with respect to the amount of added SF. However, notable differences were also observed. When an appropriate amount of SF was introduced (4% and 8%), the bonding strength was significantly enhanced. Specifically, with the addition of 4% SF, the bonding strength increased from 4.93 MPa (unmodified MUF) to 5.74 MPa, representing an increment of 0.81 MPa, or approximately 16.4%. Similarly, with 8% SF, the bonding strength rose to 5.67 MPa, an increase of 0.74 MPa (approximately 15.0%) over the unmodified MUF. These results indicate that, at this stage, a moderate addition of SF can effectively improve the crosslinking structure of the MUF resin, thereby enhancing its shear resistance and bonding performance.

However, when the SF content was either too low (2%) or too high (10%), the bonding strength tended to decrease. Notably, at an addition level of 10% SF, the bonding strength increased markedly to 6.72 MPa—an improvement of 1.79 MPa or 36.3% over the unmodified MUF. Yet, the impact toughness dropped significantly to 5.62 kJ/m^2, which was much lower than the 6.40 kJ/m^2 of the unmodified resin. This phenomenon suggests that, in the second stage, the effects of SF addition on bonding strength and impact toughness are not necessarily aligned. A possible explanation is that, under the acidic conditions of the second stage, SF primarily influences the resin’s crosslinking reactions and density, rather than contributing to improvements in toughness.

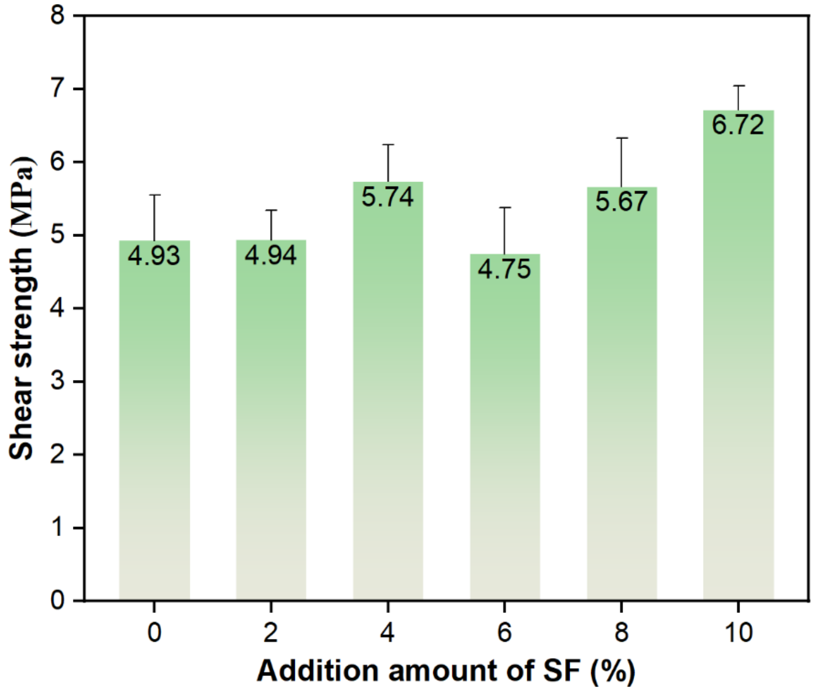

Fig. 5. Effect of adding various amounts of SF on the shear strength of MUF resin in the second stage

During the third-stage of modification process (Fig. 6), the bonding strength of the resin was significantly enhanced, particularly when 4% to 6% of SF was added. Under these conditions, the bonding strength reached a maximum of 9.26 MPa, representing an increase of 4.33 MPa, or approximately 87.8%, compared to the unmodified MUF resin (4.93 MPa). This substantial improvement indicates that SF addition at this stage greatly promotes the crosslinking reactions of the resin, thereby markedly enhancing its shear resistance and bonding performance. However, this outcome stands in sharp contrast to the observed trend in impact toughness. In the third stage, the impact toughness significantly decreased when SF levels were at 8% and 10%, suggesting a notable deterioration in material toughness. This apparent contradiction indicates that the addition of SF during the third stage exerted a more pronounced influence on the crosslinking density of the resin, while its effect on toughness is comparatively limited. Although a higher crosslinking density enhances the material's shear resistance and consequently its bonding strength, it simultaneously reduces the material's energy dissipation capacity, leading to a decline in toughness. This phenomenon may be attributed to the fact that, under the alkaline conditions of the third-stage reaction, the interaction between SF and the resin tends to favor the formation of a dense crosslinked network. While such a structure improves the material's resistance to shear forces, it restricts the mobility of molecular chain segments, thereby impairing their ability to dissipate mechanical energy under external impact and ultimately resulting in reduced toughness.

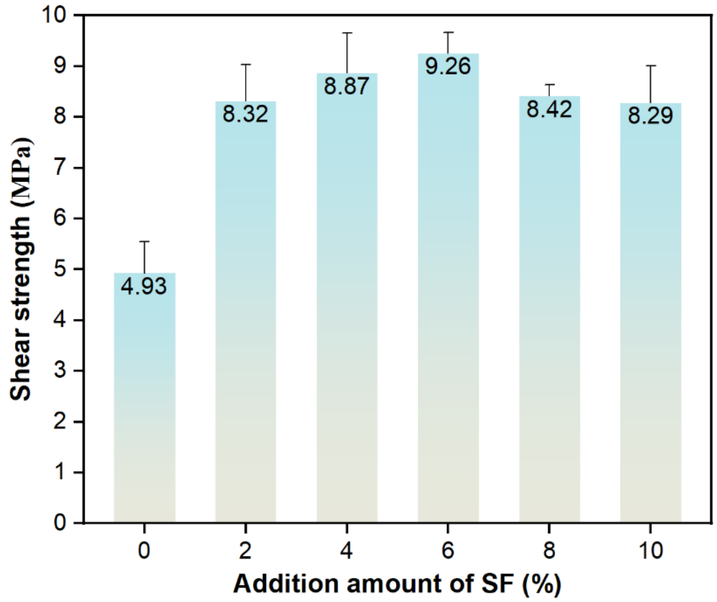

Fig. 6. Effect of adding various amounts of SF on the shear strength of MUF resin in the third stage

Based on the modification results across the three stages, the effects of SF levels on the impact toughness and bonding strength of the resin exhibited clear stage-dependent and dosage-dependent characteristics. In the first stage, a moderate addition of SF (4% to 6%) significantly enhanced both toughness and bonding strength, demonstrating a favorable synergistic effect. However, in the second and third stages, although the bonding strength was markedly improved at certain levels of SF addition, the impact toughness showed varying degrees of decline.

Analysis of Water Resistance of MUF Bonded Glued Wood

Water resistance is a critical indicator for evaluating the ability of glued wood to withstand moisture intrusion under practical application conditions. Tough adhesives, due to their inherent flexibility, are better equipped to resist water molecule penetration under varying environmental conditions (fluctuations in temperature and humidity), thereby enhancing overall water resistance. According to the standard GB/T 26899-2011, the total delamination rate after room-temperature water immersion and boiling water immersion must each be less than 5%, with the maximum delamination rate for any single glue line not exceeding 25%.

During the first-stage of modification process (Table 2), under room-temperature water immersion, the addition of 2%, 4%, 6%, and 8% SF resulted in both total and maximum single glue line delamination percentages of 0%, indicating that these additive levels had no adverse effect on the water resistance of the glulam. However, when the SF addition reached 10%, the total delamination increased to 2.34%, and the maximum single glue line delamination rate rose to 4.72%. Although these values remained within the permissible range (total delamination < 5%, maximum single glue line delamination < 25%), the increase in delamination suggests a slight reduction in water resistance. Under the condition of boiling water immersion, the addition of 2% and 4% SF maintained total and single glue line delamination at 0%, demonstrating a significant stabilizing effect on the water resistance of the glulam. However, as the SF content was increased, a decline in water resistance was observed. At 6% SF addition, the total delamination reached 4.05%, and the maximum single glue line delamination rate was 8.11%, still within the acceptable limits. When the SF content was increased to 8%, the total delamination rose to 5.92% and the maximum single glue line delamination was 11.84%, approaching the upper limit specified by the standard. A further increase to 10% SF resulted in a significant rise in total delamination rate to 12.17% and in maximum single glue line delamination rate to 24.31%, exceeding the standard threshold and indicating a substantial deterioration in water resistance.

Table 2. Effect of SF Addition Amount in the First Stage on the Water Resistance of Glued Wood

SF (%)	Delamination Rate in Room Temperature Water (%)		Delamination Rate in Boiling Water (%)	
	Total	Maximum at Glue Line	Total	Maximum at Glue Line
0	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.00
6	0.00	0.00	4.05	8.11
8	0.00	0.00	5.92	11.84
10	2.34	4.72	12.17	24.31

During the second-stage of modification process (Table 3), under both room-temperature water immersion and boiling water immersion conditions, the addition of 6% and 8% SF resulted in total delamination rates and maximum single glue line delamination percentages of 0%. However, other addition levels (2%, 4%, and 10%) failed to meet the water resistance requirements specified by the standard. Under boiling water conditions specifically, the addition of 2%, 4%, and 8% SF met the standard criteria for both total and single glue line delamination rates, while the remaining formulations did not satisfy the standard requirements for water resistance. These findings indicate that, during the second-stage modification process, the water resistance of the modified resin exhibits considerable instability.

Table 3. Effect of SF Addition Amount in the Second Stage on the Water Resistance of Glued Wood

SF (%)	Delamination Rate in Room Temperature Water (%)		Delamination Rate in Boiling Water (%)	
	Total	Maximum at Glue Line	Total	Maximum at Glue Line
0	0.00	0.00	0.00	0.00
2	16.47	27.46	5.33	2.48
4	7.44	16.79	3.63	7.47
6	0.00	0.00	7.11	14.17
8	0.00	0.00	4.07	8.15
10	14.42	18.92	33.55	66.67

During the third-stage of modification process (Table 4), under room-temperature water immersion conditions, the addition of 2%, 4%, 6%, and 8% SF resulted in both the total delamination rate and the maximum single glue line delamination rate remaining at 0%, indicating that these addition levels had no adverse effect on the water resistance of the bonded wood. However, when the SF content reached 10%, the total delamination increased to 2.12% and the maximum single glue line delamination rose to 4.24%. Although these values remained within the acceptable limits specified by the standard (total delamination rate < 5%, maximum single glue line delamination rate < 25%), the observed increase suggests a decline in water resistance performance. Under boiling water immersion conditions, the addition of 2% and 4% SF also resulted in 0% delamination for both metrics, demonstrating a significant enhancement in water resistance. However, as the SF addition continued to increase, water resistance performance began to deteriorate. When 6% SF was added, the total delamination rate rose to 1.55% and the maximum single glue line delamination rate to 3.11%, still within the standard limits. At 8% SF, the rates further increased to 2.10% and 4.19%, respectively, approaching the upper limits of the standard. When the SF content reached 10%, the total delamination rate sharply increased to 71.7% and the maximum single glue line delamination rate soared to 85.6%, far exceeding the standard requirements, indicating a significant decline in water resistance.

Table 4. Effect of SF Addition Amount in the Third Stage on the Water Resistance of Glued Wood

SF (%)	Delamination Rate in Room Temperature Water (%)		Delamination Rate in Boiling Water (%)	
	Total	Maximum at Glue Line	Total	Maximum at Glue Line
0	0.00	0.00	0.00	0.00
2	0.00	0.00	0.00	0.00
4	0.00	0.00	0.00	0.00
6	0.00	0.00	1.55	3.11
8	0.00	0.00	2.10	4.19
10	2.12	4.24	71.66	85.63

In summary, throughout the three stages of modification, the influence of SF on the water resistance of MUF resin exhibited clear stage-dependent and dosage-dependent characteristics. In the first stage, moderate SF additions (2% to 6%) significantly enhanced water resistance, with both the total delamination and the maximum single glue line delamination under room-temperature and boiling water immersion conditions meeting the requirements of the national standard. Among them, the 6% addition demonstrated the most optimal water resistance performance. In contrast, the second stage showed considerable instability in water resistance. In the third stage, water resistance exhibited favorable stability overall; however, at high addition levels (10%), the delamination under boiling water conditions rose sharply, indicating a substantial deterioration in water resistance.

CONCLUSIONS

1. Defatted soybean flour (SF) addition in the first stage markedly increased resin viscosity and promoted crosslinking; its effect was less pronounced in the second stage, while high SF loadings in the third stage also significantly increased viscosity.
2. Addition of SF at 4% to 6% levels in the first stage significantly improved the resin's impact toughness. In the second stage, the effect of SF on impact toughness was more

complex and exhibited a degree of instability. In the third stage, moderate SF levels had minimal effect on impact toughness, but higher additions led to a notable decline in toughness.

3. Moderate SF addition at 4% to 6% levels in the first stage significantly enhanced the bonding strength of the resin. In the second stage, moderate additions (4% and 8%) also led to significant improvements, although some instability was observed. In the third stage, SF additions between 2% and 6% significantly increased bonding strength, but higher additions (8% and 10%) caused declines in both water resistance and impact toughness.
4. Addition of SF at 2% to 6% in the first stage had a positive and stabilizing effect on water resistance. In the second stage, water resistance exhibited considerable variability. In the third stage, SF additions between 2% and 6% notably improved water resistance, whereas a high addition level (10%) resulted in a substantial decline in water resistance.

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