Optimization of Wood-Plastic Composites by Response Surface Method

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To improve the material properties of wood-plastic composite, poplar fiber and polyethylene powder were used as the main components, a hot-press experiment was conducted using response surface methodology, and the relationship between processing parameters (wood/plastic ratio, hot-press pressure, and time) and experimental result (internal bond strength and thickness swelling) were explored. According to the experimental results, the increasing wood/plastic ratio led to the lower internal bond strength and higher thickness swelling. However, with the increase of both hotpress pressure and time, internal bond strength increased first and then decreased, and thickness swelling decreased first and then increased. Meanwhile, two mathematical models were developed with high feasibility, and the significance of the influence of each term in the models was also analyzed. The models were able to predict and optimize internal bond strength and thickness swelling. Finally, optimal processing parameters were determined as wood/plastic ratio of 1.09, hot-press pressure of 198.38 MPa, and hot-press time of 8.31 s, with respect to the higher internal bond strength and the lower thickness swelling. This work hopes to provide scientific support for the industrial processing of wood-plastic composite.

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INTRODUCTION

Wood-plastic composites (WPCs) stand out as a class of versatile material that seamlessly blends the aesthetics of wood with the durability of plastic (Gardner *et al.* 2015). The WPC is crafted by combining wood fibers or flour with thermoplastics such as polyethylene, polypropylene, or polyvinyl chloride (Ashori 2008; Zor *et al.* 2018). The manufacturing process, often involving extrusion or compression molding, results in a composite material with distinct advantages relative to either of its main components; such advantages have propelled their widespread adoption in various industries (Wang *et al.* 2021).

The appeal of the WPC lies in its ability to replicate the natural appearance and texture of wood while enhancing durability. Its composition often incorporates recycled materials, aligning with sustainability goals and reducing dependence on virgin wood (Bhaskar *et al.* 2012). Resistant to decay, insects, and environmental factors, WPC finds versatile applications in construction and manufacturing. A WPC's versatility extends across a myriad of applications, from outdoor decking and railing systems to wall cladding, flower boxes, and seating structures (Chaharmahali *et al.* 2010). Suitable for both

residential and commercial construction, the WPC has emerged as an attractive and durable alternative to traditional materials (Khan *et al.* 1999).

According to the related work, there are numerous material performance indicators for evaluating WPC (Gozdecki and Wilczyn 2015; Kord et al. 2022). Internal bond strength (IBS) and thickness swelling (TS) emerge as critical parameters in evaluating WPC's performance (Lopez et al. 2021). The IBS measures the cohesive forces within the WPC material, assessing the bonding strength between wood fibers and the thermoplastic matrix. High IBS is pivotal for ensuring the long-term durability and load-bearing capacity of WPC products. A robust bond contributes to structural integrity, preventing delamination and structural failure (Benthien and Thoemen 2021). The TS quantifies dimensional changes in WPC when exposed to moisture, expressed as a percentage increase in thickness after immersion in water. Low TS indicates enhanced resistance to swelling, reflecting improved dimensional stability. It is crucial, especially in applications where WPC is exposed to environmental moisture. Continual research efforts focus on optimizing IBS and minimizing TS in WPC. Mathematical models were developed by Kord and Kiaeifar (2010), and these can be used for the prediction for the changes in WPC material properties during the hygroscopic swelling process. Meanwhile, the physical and mechanical properties of WPC were held in focus by Leu et al. (2012); they found that the WPC material properties were affected by many factors, such as lubricant content, wood flour particle size, coupling agent dosage, and so on. A nuanced understanding of these factors is pivotal for advancing WPC's performance characteristics and broadening its applications across diverse environments.

Recently, in the industrial production of WPC, poplar has been widely used as the wood fiber due to its wide range of resources (Nörnberg *et al.* 2014). Meanwhile, the typical resin adhesives are being replaced by polyethylene, polypropylene, polyvinyl chloride, and so on. Among those polymers, polyethylene, which has high chemical resistance, flexibility, and durability, is extensively adopted in the WPC preparation (Kajaks *et al.* 2018). The mostly widely used varieties of this plastic are high-density polyethylene (0.945 to 0.96 g/cm³ density, 125 to 137 °C melting point), linear low-density polyethylene (0.925 g/cm³ density, 120 to 125 °C melting point), and high pressure low-density polyethylene (0.918 g/cm³ density, 105 to 115 °C melting point).

In this work, the composition and processing conditions of WPC were explored, Poplar fiber and polyethylene powder were adopted as the main components, and main attention was given to the changes in IBS and TS at different wood/plastic ratio, hot-press pressure, and time. Meanwhile, the optimal processing technique was determined, and this paper hopes to provide scientific guidance for the processing of high-quality WPC.

EXPERIMENTAL

Materials

Poplar fiber and polyethylene powder were used as the main components for WPC, which were manufactured by Yixing Dayang Wood Fiber Co., Ltd. (Wuxi, China) and Xinbeiyang Engineering Plastics Co., Ltd. (Suzhou, China), respectively. The additive of melamine-modified urea-formaldehyde was provided by Dennuo New Materials Technology Co., Ltd. (Shandong, China).

Equipment

A high-speed mixer (HEM, Tongsha Plastic Machinery Co., Ltd., Suzhou, China) was used for the blending and processing of poplar fiber and polyethylene powder. Meanwhile, the extrusion-type hot press was used for the production of WPC, which was manufactured by Zhangjiagang Yawei Machinery Co., Ltd. (Suzhou, China).

Experiment Design

The response surface method (RSM) (Yu *et al.* 2023) with Design-Expert (V.12, Stat Ease, Inc., Minneapolis, MN, USA) was used in this work. It is a statistical technique used in experimental design and analysis to model and optimize complex processes (Zhu *et al.* 2022). It involves conducting a series of experiments with varying input factors, and then using statistical methods to analyze the responses and develop predictive models (Song *et al.* 2023; Zhu *et al.* 2023). The detailed experiment design is given in Table 1, including experimental parameters of wood/plastic ratio (R), hot-press pressure (P), and time (T), and results of IBS and TS.

Runs	Wood/Plastic Ratio <i>R</i>	Hot-Press Pressure <i>P</i> (MPa)	Hot-Press Pressure Time <i>T</i> (s)	IBS (MPa)	TS (%)
1	50:50	180	6	2.46	1.46
2	50:50	160	8	2.43	1.34
3	50:50	180	10	2.60	0.39
4	50:50	200	8	2.73	0.73
5	60:40	180	8	2.38	1.92
6	60:40	200	10	2.08	1.54
7	60:40	180	8	2.38	1.92
8	60:40	160	10	1.82	5.01
9	60:40	180	8	2.38	1.92
10	60:40	160	6	1.52	6.86
11	60:40	180	8	2.38	1.92
12	60:40	200	6	2.19	3.98
13	60:40	180	8	2.38	1.92
14	70:30	200	8	1.84	5.96
15	70:30	180	6	1.59	8.88
16	70:30	180	10	1.72	5.51
17	70:30	160	8	1.66	8.75

Table 1. Experiment Design and Results of IBS and TS

Measurement

The IBS and TS were measured based on the GB/T 24137 (2009) standard of Wood-plastic Composite Decorative Boards. The IBS was acquired by using bonding strength measurement instrument (Rongjida Instrument Technology Co., Ltd. Shanghai, China), and TS was calculated using Eq. 1 (Wu *et al.* 1999),

$$T = \frac{t_2 - t_1}{t_1} \times 100\%$$
(1)

where T is the TS (%), t_1 is the thickness of WPC before immersion (mm), and t_2 is the thickness of WPC after immersion (mm).

RESULTS AND DISCUSSION

IBS Mathematical Model

According to the RSM result listed in Table 1, a mathematical mode for internal bond strength was developed, as given in Eq. 2. As shown in Table 2, the values of standard deviation (Std. Dev.) and coefficient of variation (CV%) (Jiang *et al.* 2022) were equal to 0.06 and 2.63. They both were judged to have low values. The Predicted-R² of 0.84 is in reasonable agreement with the Adjusted-R² of 0.98, *i.e.*, the difference is less than 0.2, and they were all close to 1. Meanwhile, Adequate precision measures the signal to noise ratio (Xu *et al.* 2022). A ratio greater than 4 is desirable. The ratio of 27.38 indicates an adequate signal. In all, those results of fit statistics showed the developed mode has high precision, which can be adopted for the prediction and optimization of IBS. The internal bond strength was fitted as follows,

$$IBS = -22.32 - 0.07R + 0.21P + 1.49T - 0.23 \times 10^{-2} RP - 0.19 \times 10^{-2} RT$$

-0.22×10⁻² PT - 0.05R² - 0.49×10⁻³ P² - 0.07T² (2)

where R is wood/plastic ratio, P is hot-press pressure (MPa), and T is hot-press time (s).

Table 2.	Fit	Statistics	for	Internal	Bond	Strength
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Std. Dev.	CV%	Adjusted-R ²	Predicted-R ²	Adeq Precision
0.06	2.63	0.98	0.84	27.38

The analysis of variance (ANOVA) for internal bond strength model is given in Table 3. A significance level of 5% ($\alpha = 0.05$) was used. If the p-value was less than 0.05, it indicates the term is significant (Zhu *et al.* 2020 a, b). In this model, the model F-value of 77.18 implies that the model is significant. There is only a 0.01% chance that an F-value this large could occur due to noise. In this case, the values of *R*, *P*, *T*, *PT*, *P*², and *T*² were lower than 0.05, *i.e.*, the parameters of wood/plastic ratio, hot-press pressure, and hot-press time, two-level interaction effects of hot-press pressure and time, and products of hot-press pressure and time, all had significant contribution to the IBS. Meanwhile, the values of *RP*, *RT*, and *R*² were higher than 0.05, *i.e.*, the two-level interaction effects of wood/plastic ratio and hot-press time, and product of wood/plastic ratio and hot-press time and product of wood/plastic ratio and hot-press time and product of wood/plastic ratio and hot-press time and product of wood/plastic ratio

Source	SS	DF	MS	F-value	p-value	Remark
Model	2.23	9	0.25	77.18	< 0.0001	Significant
R	1.45	1	1.45	452.71	< 0.0001	Significant
Р	0.23	1	0.23	70.95	< 0.0001	Significant
Т	0.02	1	0.02	6.23	0.04	Significant
RP	0.36 × 10 ⁻²	1	0.36×10 ⁻²	1.12	0.32	Insignificant
RT	< 0.0001	1	< 0.0001	0.78×10 ⁻²	0.93	Insignificant
PT	0.03	1	0.03	9.54	0.02	Significant
R2	0.17 × 10 ⁻²	1	0.17×10 ⁻²	0.52	0.49	Insignificant
P2	0.16	1	0.16	49.87	0.02×10 ⁻²	Significant
T2	0.30	1	0.32×10 ⁻²	93.84	< 0.0001	Significant
Residual	0.02	7				
Cor total	2.25	16				

Table 3. ANOVA Results of Internal Bond Strength

Changes in IBS at Different Processing Conditions

Single factors of wood/plastic ratio, hot-press pressure, and time on the internal bond strength are given in Figs. 1a through 1c. It can be found that the increased wood/plastic ratio led to the lower internal bond strength of WPC. An increased wood/plastic ratio implies a higher content of poplar fiber and a decrease in polyethylene powder content. Polyethylene powder serves as a filler in WPC, filling the gaps between wood particles and enhancing the bonding performance of the WPC. When the content of polyethylene powder decreases, the filling effect weakens, leading to a weaker bond between wood particles. The interfacial adhesion strength between wood fiber and polyethylene powder is crucial for the performance of the composite material.



Fig. 1. Effect of (a) wood/plastic ratio, (b) hot-press pressure, and (c) time on the internal bond strength; two-level interaction effects of (d) wood/plastic ratio and hot-press pressure, (e) wood/plastic ratio and hot-press time, (f) hot-press pressure and time on the internal bond strength

A reduction in the content of polyethylene powder results in a decrease in the interface area between wood particles and polyethylene powder, thereby weakening the

interfacial adhesion strength and impacting the internal bonding strength of the WPC. Therefore, as the wood/plastic ratio increases, the internal bonding strength of the WPC decreases. Furthermore, with the increase of hot-press pressure and time, internal bond strength increased first and then decreased. This is because with the increase of hot-press pressure and time, initially, the bonding between wood fiber and polyethylene powder is strengthened, improving the internal structural stability and strength of the WPC. However, when the hot-press pressure and time exceed a certain threshold, excessive pressure and time may weaken the interfacial adhesion between wood powder and polyethylene powder, leading to phenomena, such as thermal degradation of wood powder, thereby causing a decrease in the internal bond strength of the WPC. Therefore, appropriate hot-press pressure and time are crucial to ensure the internal bond strength of WPC.

Based on the ANOVA results in Table 3 and Fig. 1d through1f, there are two-level interactions affecting the results of internal bond strength. Response surface and contour plots can visually display the individual effect of a single factor on the interactive effect of two factors on experimental results. Through observing the distribution density of contour lines along the axes, it can determine whether a certain factor plays a dominant role in the interaction. If the contour lines of that factor are denser along the axes, it indicates that the result is mainly influenced by this factor in the interaction. According to the response surface and contour plots given in Fig. 2d through 2f, wood/plastic ratio played a dominant role in both interactive effects of wood/plastic ratio and hot-press pressure, and wood/plastic ratio and hot-press time. Meanwhile, hot-press time dominated in the interaction of hot-press pressure and time.

TS Mathematical Model

Based on the results for TS at different conditions listed in Table 1, a mathematical mode for thickness swelling was obtained, as given in Eq. 3. As shown in Table 2, the values of Std. Dev. and CV% were equal to 0.39 and 11.19, which were both low. The Predicted-R² of 0.86 is in reasonable agreement with the Adjusted-R² of 0.98, and their difference was less than 0.2, which were all close to 1. Meanwhile, Adeq precision measures the signal to noise ratio. A ratio greater than 4 is desirable. The ratio of 28.83 indicates an adequate signal. In all, those results of fit statistics showed the developed mode has a low error level, it can be used for the prediction and optimization of TS, as shown by Eq. 3,

$$TSR = 114.39 + 8.08R - 1.12P - 3.75T - 0.04RP - 0.43RT$$
$$-0.37 \times 10^{-2}PT + 2.24R^{2} + 0.32 \times 10^{-2}P^{2} + 0.29T^{2}$$
(3)

where TSB is the thickness swelling (%), R is the wood/plastic ratio, P is the hot-press pressure (MPa), and T is the hot-press time (s).

Std. Dev.	C.V%	Adjusted-R ²	Predicted-R ²	Adeq Precision
0.39	11.19	0.98	0.86	28.83

Table 4. Fit Statistics for Thickness Swelling

Table 5 shows the ANOVA results of TS. In this model, the model F-value of 86.75 indicates the model is significant. There was only a 0.01% chance that an F-value this large could occur due to noise. In this case, the values of R, P, T, RP, RT, R^2 , P^2 , and T^2 were lower than 0.05, *i.e.*, the parameters of wood/plastic ratio, hot-press pressure and hot-press time, two-level interaction effects of wood/plastic ratio and hot-press pressure, and

wood/plastic ratio and hot-press time, products of wood/plastic ratio, hot-press pressure and hot-press time, all significantly affected the changes in TS. While only the two-level interaction effects of hot-press pressure and time was bigger than 0.05, which had an insignificant contribution to the TS.

Source	SS	DF	MS	F-value	p-value	Remark
Model	121.79	9	13.53	86.75	< 0.0001	Significant
R	79.25	1	79.25	508.10	< 0.0001	Significant
Р	11.88	1	11.88	76.18	< 0.0001	Significant
Т	9.53	1	9.53	61.08	0.01×10 ⁻²	Significant
RP	1.19	1	1.19	7.62	0.03	Significant
RT	1.32	1	1.32	8.48	0.02	Significant
PT	0.09	1	0.09	0.56	0.48	Insignificant
R2	4.16	1	4.16	26.66	0.13×10 ⁻²	Significant
P2	6.91	1	6.91	44.31	0.03×10 ⁻²	Significant
T2	5.53	1	5.53	35.47	0.06×10 ⁻²	Significant
Residual	1.09	7	0.16			
Cor total	122.88	16				

Table 5. ANOVA Results of Thickness Swelling

Changes in TS at Different Processing Conditions

The single factor of wood/plastic ratio impacting the extent of thickness swelling is given in Fig. 2a. The increase of wood/plastic ratio increased the thickness swelling of WPC. The proportion of wood fiber and polyethylene powder determines the internal structure and properties of the WPC. When the content of wood fiber increases, the proportion of wood components in the WPC increases, whereas the proportion of polyethylene powder decreases accordingly. Wood fiber has a higher water absorption capacity, so when the content of wood fiber increases, the water absorption capacity of the WPC also increases, leading to an increase in thickness swelling. Additionally, the interfacial adhesion strength between wood fiber and polyethylene powder weakened with the increase of wood fiber content, further affecting the moisture absorption and swelling properties of the WPC. Therefore, the increased wood/plastic ratio resulted in a higher thickness swelling. However, based on Fig. 2b and 2c, with the increase of both hot-press pressure and time, the thickness swelling decreased first and then increased. As the hotpress pressure and time increase, initially, it strengthens the bond between wood fiber and polyethylene powder, reducing the pores and air leakage paths in the WPC, thereby reducing the rate of water penetration and causing the thickness swelling to decrease. However, with further increases in hot-press pressure and time, excessive pressure and time lead to an increase in the unevenness of the internal structure of the WPC, and even cause thermal degradation of wood powder, thereby increasing the water penetration and causing the thickness swelling to increase again. Therefore, with the increase of hot-press pressure and time, the thickness swelling of WPC first decreases and then increases.

As given in Table 3 and Fig. 2d through 2f, there are also two-level interactions affecting the results of thickness swelling evaluation. According to the response surface and contour plots given in Fig. 3d through 3f, wood/plastic ratio dominates in the interaction of wood/plastic ratio and hot-press pressure, and wood/plastic ratio and hot-press time. Meanwhile, hot-press time played a dominant role in the interactive effect of hot-press pressure and time.



Fig. 2. Effect of (a) wood/plastic ratio, (b) hot-press pressure, and (c) time on the thickness swelling; two-level interaction effects of (d) wood/plastic ratio and hot-press pressure, (e) wood/plastic ratio and hot-press time, (f) hot-press pressure and time on the thickness swelling

Optimization and Verification of Production Technology for WPC

Improving the IBS and TS of WPC is critical to its application (Raut *et al.* 2022). The IBS directly impacts the structural integrity and stability of the material. A strong IBS ensures the material is resistant to delamination and peeling under stress, thereby enhancing its overall strength and durability. The TS refers to the extent to which the material expands after being exposed to moisture, affecting its dimensional stability and aesthetic appearance. Reducing the TS can minimize deformation and damage in moist environments, thereby prolonging its lifespan (Soury *et al.* 2009; Martins *et al.* 2017).

In this work, the processing parameters of wood/plastic ratio, hot-press pressure and time were optimized for the targets of the highest IBS and the lowest TS. Based on the developed RSM models of Eqs. 2 and 3, the highest IBS of 2.74 MPa and the lowest TS of 0.25% can be obtained at the combination of 1.09 wood/plastic ratio, 198.38 MPa hot-press pressure, and 8.31 s time (Fig. 3). To verify the feasibility of the optimization results, a validation experiment was conducted, and its result is given in Table 6. With the combination of 1.09 wood/plastic ratio, 198.38 MPa hot-press pressure, and 8.31 s time, the predicted results of IBS and TS were equal to 2.74 MPa and 0.25%, and the measurement results of IBS and TS were equal to 2.95 MPa and 0.22%. The error values between the predicted and measurement results were -7.12% and13.6%, respectively, which were in a reasonable range. Thus, the developed RSM models had high feasibility, and can be used for the optimization of processing parameters. Meanwhile, the optimal processing parameters were determined as wood/plastic ratio of 1.09, hot-press pressure of 198.38 MPa, and hot-press time of 8.31 s, in terms of the higher IBS and the lower TS.

Both IBS and TSM were considered in this study. However, additional parameters, such as modulus of elasticity and modulus of rupture, are also important performance indicators for WPC materials. To better improve the performance of WPC materials, it is necessary to further consider them in future research.



Fig. 3. Optimized processing parameters of (a) wood/plastic ratio, (b) hot-press pressure, and (c) time, and the optimal results of (d) internal bond strength and (e) thickness swelling

Experiment	R	P (MPa)	T (s)	IBS (MPa)	TS (%)
Prediction	1.09	198.38	8.31	2.74	0.25
Measurement	1.09	198.38	8.31	2.95	0.22
Error (%)	/	/	/	-7.12	13.6

Table 6. Validation Experiment and Results

CONCLUSIONS

Both internal bond strength (IBS) and thickness swelling (TS) are important indicators for evaluating the performance of WPC. In this work, RSM was adopted to study the IBS and TS at different conditions of wood/plastic ratio, hot-press pressure, and time. The main conclusions are listed as follows:

- 1. Mathematical models were developed by RSM with high feasibility, it was verified that they can be used for the prediction and optimization of WPC material properties of IBS and TS. Meanwhile, an analysis of the significance of each term on the experimental results was conducted.
- 2. With the increase of wood/plastic ratio, IBS of the WPC decreased. With the increase of hot-press pressure and time, IBS increased first and then decreased. Meanwhile, as the wood/plastic ratio increased, TS showed an increased trend. With the increase of both hot-press pressure and time, TS decreased first and then increased.

Appropriate hot-press pressure and time are crucial to ensure the material properties of WPC.

- 3. With the optimal target of the highest IBS and the lowest TS, the optimal processing parameters were determined, which were wood/plastic ratio of 1.09, hot-press pressure of 198.38 MPa, and hot-press time of 8.31 s.
- 4. In future research, more material properties of WPC will be considered, based on chemical and physical principles as well as microscopic perspectives, such as hardness, tension strength, modulus of rupture, modulus of elasticity and so on.

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