Effects of Hot-Pressing Parameters on Shear Strength of Plywood Bonded with Modified Soy Protein Adhesives

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The heating rate of the core layer of plywood during hot-pressing is of great importance to the final quality of the plywood and is affected by many factors, such as the hot-pressing temperature (T_{HP}), hot-pressing pressure (P_{HP}), hot-pressing time (t_{HP}), veneer layers, and moisture content. In this study, multi-plywood using modified soy protein (MSP) adhesives prepared to investigate the effects of T_{HP}, t_{HP}, and veneer layers on the core layer temperature during hot-pressing. The results indicated that all the core layer temperature curves were divided into four stages. The first constant temperature stage and the slow warming stage were decisive with respect to the time needed for the core layer to reach the T_{HP}. The time of moisture vaporization was approximately 400 s in the 3-layer plywood and approximately 900 s in the 5-layer plywood. In order to get an ideal strength the t_{HP} should greater than the time of moisture vaporization; therefore in theory, the optimum parameters of the 3-layer plywood production were t_{HP} of 600 to 720 s and T_{HP} of 120 to 125 °C. The research provides a theoretical basis for optimizing the hot-pressing of plywood.

Keywords: Modified soy protein adhesives; Plywood; Shear strength; Core layer temperature

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INTRODUCTION

Despite their low price and diverse applications, the market for petroleum-based adhesives has been threatened by their inevitable increase in price due to their limited reserves and worldwide environmental concern (Mo *et al*. 1999). The alternative use of biomass-based, biodegradable adhesives can solve these problems, as they can be degraded in the environment by humidity and microorganisms (Zhong and Sun 2001). Bio-based soybean proteins are potential adhesives with utility in the wood composites, packaging, and labeling industries (Zhong and Sun 2001; Zhong *et al*. 2002). Adhesives based on soybean meal have been in use since the 1920s (Huang and Sun 2000), but they can only be used in a narrow field, and their development is limited by their poor water resistance and bonding strength (Qi and Sun 2011).

Soy protein consists of 18 amino acid monomers. Some side chains can interact with organic or inorganic substances and cellulose fibers. These side chains can be chemically, physically, or enzymatically modified to achieve desired properties (Sun and Bian 1999). Substantial research has been conducted in the last few decades to improve the water resistance of soy proteins via chemical modification (Kalapathy *et al*. 1995; Cheng *et al*. 2004; Liu and Li 2004; Rogers *et al*. 2004; Garcia and Cloutier 2005; Qi and Sun 2010; Jang *et al*. 2011; Gao 2013; Li *et al.* 2014a,b). Chemical modification can enhance the general performance of soy protein adhesives but also bring about problems such as higher production costs, the need for high hot-pressing temperatures, high viscosity, and low solid content (less than 36 percent). The performance of soy protein adhesive plywood can be improved by adjusting and optimizing the hot-pressing process.

Hot-pressing is a key process in plywood production, as it affects the quality and cost of plywood manufacture. The main parameters affecting plywood quality include the hot-pressing temperature (T_{HP}), hot-pressing time (t_{HP}), and unit hot-pressing pressure (P_{HP}). At certain T_{HP} and P_{HP}, the t_{HP} depends on the curing time of the adhesives during which are farthest away from the hot plate (Hua and Zhang 1986). Most existing research has focused on heat and mass transfer in hot-pressing of particleboard and fiberboard. Garcia and Cloutier (2005) determined the gas permeability of a mat as a function of density and characterized panel properties, temperature, and evolution of the gas pressure in the mat during hot-pressing as a function of press closing strategy, panel density, and moisture content. Effects of particleboard density, t_{HP}, T_{HP}, and storage time of the wet soy-coated wood particles on the internal bond strength (IB), the modulus of rupture (MOR), and the modulus of elasticity (MOE) of particleboard have been investigated (Prasittisopin and Li 2010). Hot-pressing is the final stage in medium density fiberboard (MDF) manufacture in which the mat of fibers is compressed and heated to promote resin curing (Carvalho *et al*. 2001). The balance of properties of the resulting panel is primarily affected by the press cycle, so rigorous control of all processing parameters is necessary to improve product quality and shorten pressing time. A theoretical model for heat and moisture transfer in wood composite mats during hot-pressing was developed from the basic principles of mass conservation, momentum of gas flow, energy conservation, and resin curing kinetics (Dai and Yu 2004). Effective measures to perfect the hot-pressing process of particleboard were put forward based on influential factors on heat transfer during hot-pressing, such as target density, panel thickness, hot-pressing temperature, and moisture content of the slab before hot-pressing (Liu *et al.* 1995).

However, there have been few reports dealing with the heat transfer of plywood, especially plywood bonded with soy protein adhesives. In this study, we prepared multi-plywood using modified soy protein (MSP) adhesives and investigated the effects of hot-pressing parameters (T_{HP}, t_{HP}, and veneer layers) on shear strength. We also obtained the relationship between core layer temperature and shear strength of multi-plywood using experiments and analysis, which provide a theoretical reference for plywood preparation.

EXPERIMENTAL

Materials

Soy flour with an average protein content of 45.2% and moisture content of 5.0% was purchased from Sanhe Hopefull Group Oil Grain Food Co. Ltd. (Beijing, China). Glycerol polyglycidyl ether (GPE) and polyvinyl alcohol (PVA) were analytical grade and obtained from Beijing Chemical Reagents Co. Ltd. (Beijing, China). Urea-formaldehyde (UF) resins with an average solid content of 50% and viscosity 27 mPa·s and pH 7.5 were purchased from Beijing Chemical Reagents Co. Ltd. (Beijing, China). The poplar veneer was obtained from Wen'an (Wen'an, China) County with a moisture content of 8.0% and dimensions of 410 mm × 410 mm × 1.5 mm (width × length × thickness).

Preparation of MSP Adhesives

MSP adhesives: PVA solution (40 g, 5%), tap water (35 g), SF (25 g), and GPE (10 g) were sequentially added to a three-neck flask and stirred at room temperature. The basic physical parameters of MSP adhesives were viscosity 10 mPa·s, solid content 30%, and pH 5.3.

Preparation of Plywood

Poplar veneers with dimensions of 410 mm × 410 mm × 1.5 mm were used. The multi-plywood (3-layers, 5-layers, 7-layers, 9-layers) were made under the following conditions: 188 g/m^2 of glue spreading; 1.0 MPa of P_{HP}; and different specified values of t_{HP} and T_{HP}. After hot-pressing, the panels were stored at ambient temperature for at least 24 h prior to cutting it into specimens for evaluation of the wet shear strength.

Core Layer Temperature Testing

The veneers were oriented in alternating directions, layer-by-layer, so that the grains of the middle panel were perpendicular to the grains of the top and bottom veneers. A 5 mm × 150 mm (width × length) groove was opened in the middle of the core veneer to install the thermocouple (PT 100 thermocouple, length 200 mm, compensation conductor 3 m, and diameter 2.0 mm, Beijing Kunlun Tianchen Instrument Technology Co., Ltd., China).

The assembled veneers were conditioned for 15 min at room temperature and then hot-pressing with the 302 × 2/15 150 T universal hot press machine (Suzhou Xin Xieli Machinery Manufacturing Co., Ltd. Zhejiang, China) under various conditions. Data were collected when the surface veneer contacted pressure. The data acquisition interval was 10 s and lasted for 900, 1500, 2100, and 2700 s. When the unloading pressure plate dropped to the lowest position, the test was ended and the data were saved to a computer.

Wet Shear Strength Measurement

The shear strength of plywood was measured in accordance with China National Standard GB/T 9846.3-2004 (Standardization Administration of the People's Republic of China 2004) for type II plywood. The panels bonded with the MSP adhesives were balanced for at least 24 h under room conditions before being cut into six pieces with dimensions of 100 mm × 25 mm (glued area of 25 mm × 25 mm). Then the pieces were immersed in water at 63 ± 3 °C for 3 h, removed from the water, and cooled at room temperature for 10 min prior to the measurement of shear strength using a common tensile machine operating at a speed of 10.0 mm/min. The reported strength data were averaged from six pieces.

RESULTS AND DISCUSSION

Core Layer Temperature Analysis

The core layer temperature curves of the 3-layer control plywood and plywood prepared with different adhesives at 120 °C are shown in Fig. 1(a). Clearly, the heating rate of the control plywood was significantly higher than that of those bonded with UF adhesives and MSP adhesives, as the hot-pressing time was prolonged from 10 to 60 s (Fig. 1(c)). Compared with the other two groups, the core layer temperature curve of the control plywood did not contain an obvious constant temperature stage (60 to 200 s), and it reached the T_{HP} significantly earlier. These phenomena attributed to the small amount of moisture contained inside the veneers (about 8% on each single veneer). Thus, the small amount of moisture vaporization in veneers was negligible. The heating rate and moisture vaporization temperature of plywood bonded with UF adhesives were higher than those of plywood bonded with MSP adhesives, probably because the UF adhesives contained far less moisture than the MSP adhesives. Moreover, the UF adhesives released more heat than the MSP adhesives while curing.

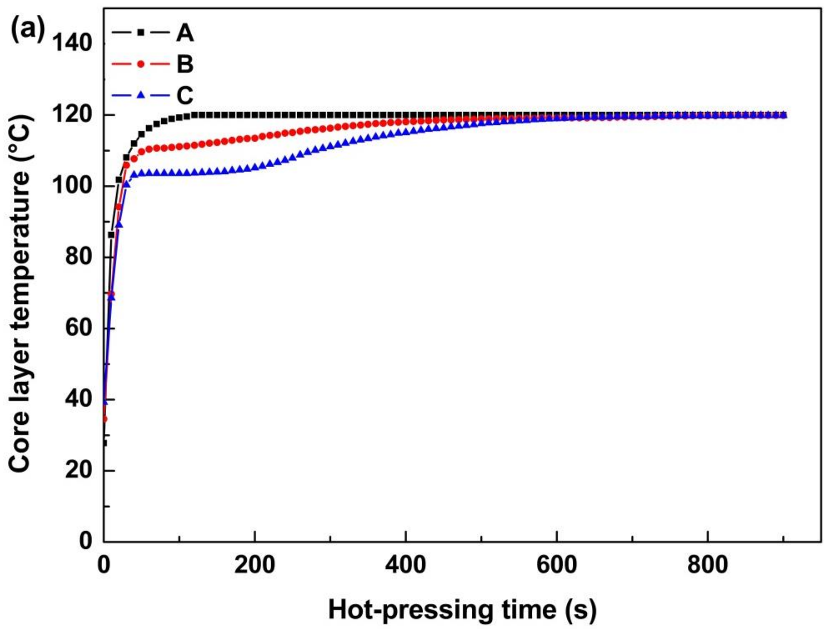

Fig. 1. **(a)** Core layer temperature curves of 3-layer plywood prepared with different adhesives at 120 °C (A: no addition of adhesives; B: urea-formaldehyde adhesives; C: MSP adhesives)

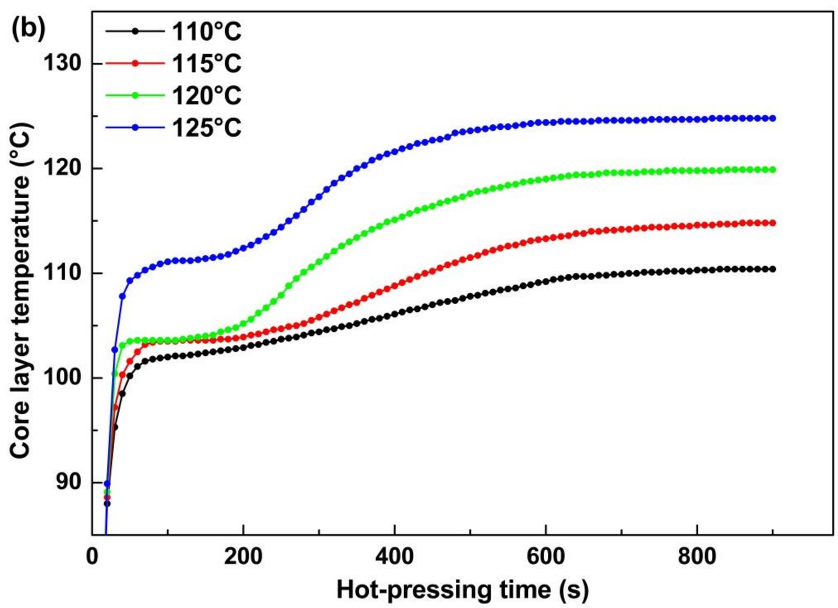

Fig. 1. **(b)** Core layer temperature curves of 3-layer plywood bonded with MSP adhesives at various hot-pressing temperatures

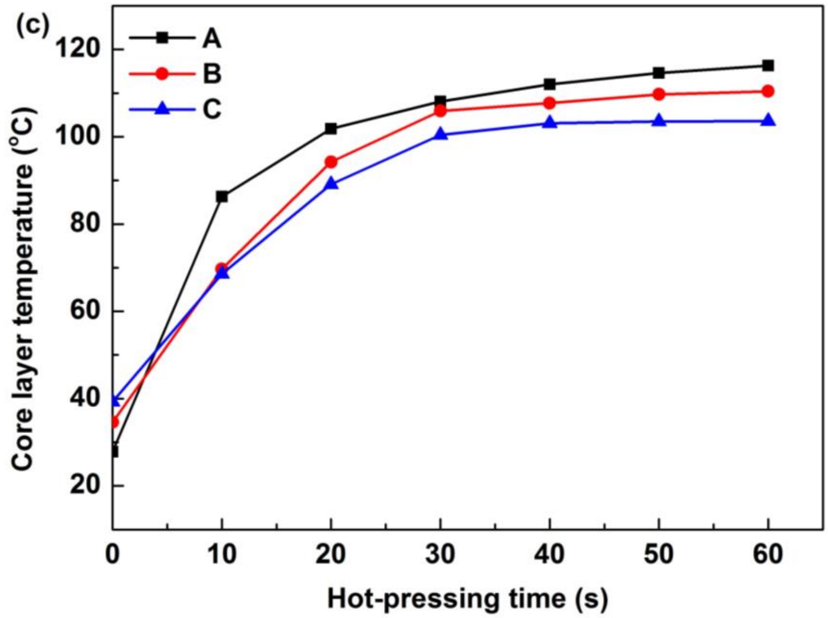

Fig. 1. (c) Core layer temperature curves of 3-layer plywood prepared with different adhesives at 120 °C during 0-60s (A: no addition of adhesives; B: urea-formaldehyde adhesives; C: MSP adhesives)

The solid content of UF adhesives is about 50%, in general, so the time needed for moisture vaporization in the resulting plywood was relatively shorter (Fig. 1(a)). However, the solid content of MSP adhesives is low, normally about 30%, so the soy protein adhesives contain high amounts of moisture. Because the moisture vaporization crucially affects the t_{HP}, further detailed study is necessary to reveal the core layer temperature of plywood bonded with MSP adhesives.

Figure 1(b) shows the core layer temperature curves of 3-layer plywood bonded with MSP adhesives prepared at various temperatures. Clearly, the core layer temperature curves of 3-layer plywood during hot-pressing can be divided into four stages: the rapid warming stage before moisture vaporization; the first constant temperature stage during moisture vaporization; the slow warming stage after moisture vaporization; and the second constant temperature stage after moisture vaporization. With moisture content and veneer layers kept constant, a higher T_{HP} implies that less time was taken by the core layer to reach the moisture vaporization temperature, but the difference of time was smaller. Table 1 shows that the time needed for the core layer of 3-layer plywood to reach 100 °C gradually decreased as the T_{HP} was raised from 110 °C (50 s) to 125 °C (30 s). When the T_{HP} was 120 and 125 °C, the tangent slopes of curves at the first stage were obviously larger than those at 110 and 115 °C, indicating that at higher $T_{HP,}$ the core layer spent less time in the rapid warming stage. When T_{HP} was 110 and 115 °C, the changes from the first constant temperature stage to the slow warming stage were not obvious. The temperature increased slowly from when the core layer temperature reached the moisture vaporization temperature until the core layer temperature was close to T_{HP}. Meanwhile, the first constant temperature stage was short-lived when T_{HP} was 120 and 125 °C (Table 1). In addition, the temperature rose rapidly during the slow warming stage, so the 3-layer plywood core layer reached the balance temperature quickly. This was probably because the increased temperature gradient catalyzed the heat transfer between the hot plate and the core layer. The temperature range in this study is commonly used in plywood production. The results can be used to estimate the core layer temperature in plywood production and to set the hot-pressing parameters.

Table 1. Time Needed for Core Layer of 3-Layer Plywood Bonded with MSP Adhesives to Reach 100 °C and Equilibrium Temperature

Hot-pressing temperature (°C)	Time to reach 100°C (s)	Time to reach equilibrium temperature (s)
110	50	470
115	40	430
120	30	370
125	30	340

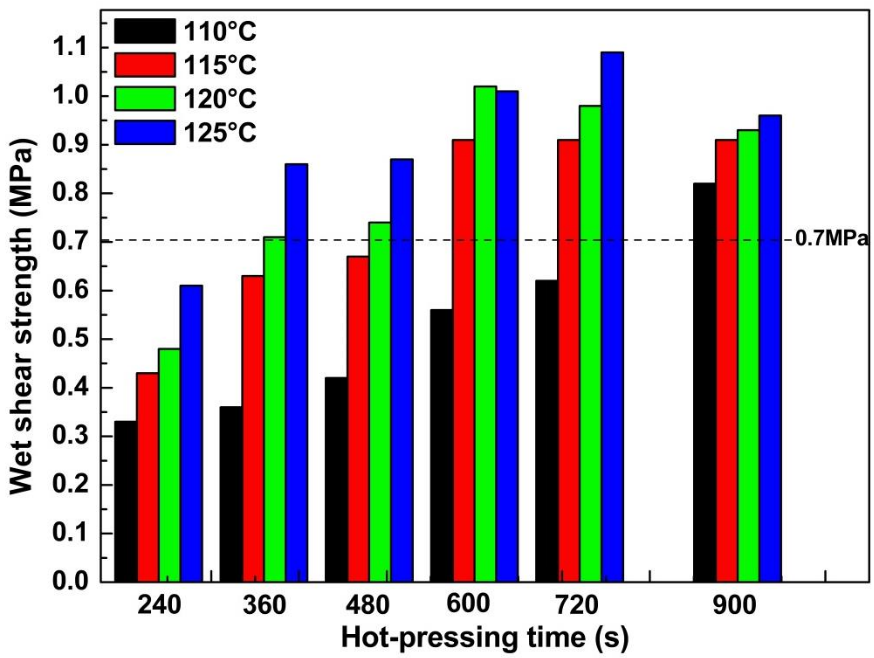

Fig. 2. Wet shear strength of 3-layer plywood prepared at various hot-pressing temperatures for various durations at 1.0 MPa

Figure 2 shows that the wet shear strength of 3-layer plywood gradually increased with the increase of T_{HP}, under the same P_{HP} (1.0 MPa), T_{HP}, and different t_{HP} (240, 360, 480, 600, and 720 s). This is because a higher T_{HP} resulted in faster moisture vaporization, which is more conducive to adhesion. On the contrary, at low T_{HP}, the MSP adhesives had less fluidity and the moisture couldn't evaporate rapidly, inhibiting effective adhesion. At higher T_{HP}, the rapid warming stage was shorter and the time of the plywood core layer spent in the first constant temperature stage was greater (Fig. 1). Consequently, the MSP adhesives could cure more completely. When T_{HP} and P_{HP} were kept constant, the wet shear strength of plywood improved with the increase of t_{HP}, probably because a longer t_{HP} resulted in better-cured MSP adhesives. However, at a t_{HP} of 900 s, the wet shear strength of plywood decreased as the T_{HP} increased from 115 to 125 °C, probably because the increased t_{HP} caused aging of the glue layer. Furthermore, the compression ratio and production cost of plywood increased at prolonged t_{HP}, which is not conducive to the industrialization of plywood production. At a T_{HP} of 110 °C, the wet shear strength of plywood improved with increasing t_{HP}, even to 900 s; when T_{HP} was too low to eliminate the moisture quickly, the 3-layer plywood required more time to complete the moisture vaporization period. Therefore, at low T_{HP}, a longer t_{HP} would improve the wet shear strength of plywood.

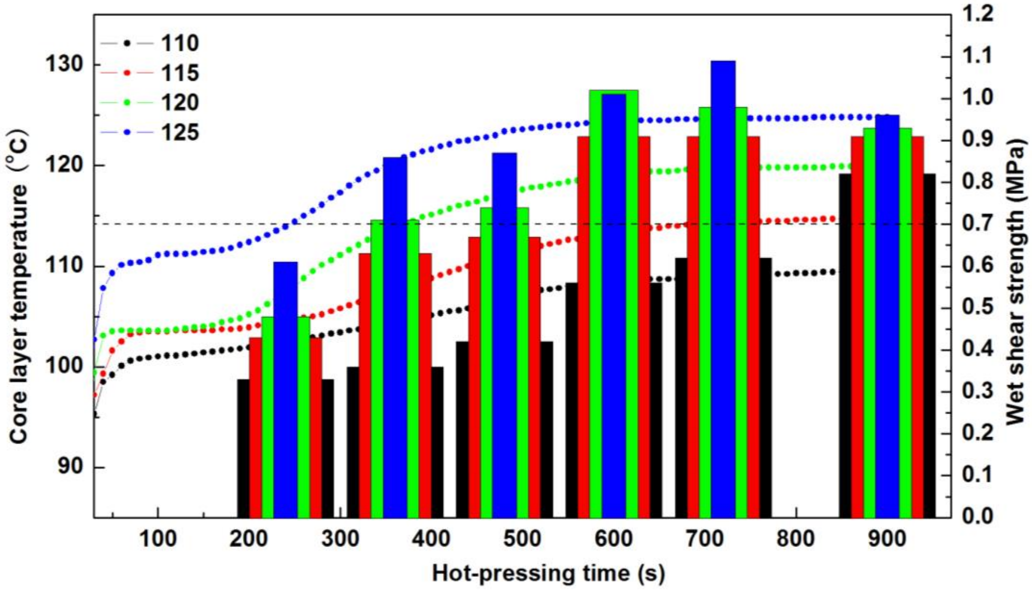

Fig. 3. Core layer temperature curves and wet shear strength of 3-layer plywood (black bar: Hot-pressing temperature 110 °C; red bar: Hot-pressing temperature 115 °C; green bar: Hot-pressing temperature 120 °C; blue bar: Hot-pressing temperature 125 °C)

Figure 3 shows that at a t_{HP} of 240 s, the wet shear strength of 3-layer plywood improved with the increase of T_{HP}, but all samples failed to meet GB/T 9846.3-2004 (≥ 0.7 MPa) for type II plywood. This was because a greater amount of time was needed for the MSP adhesives to eliminate moisture and complete the curing process. For the entire duration of the heat transfer between the panel and the hot pressure plate, the system remained in the heating stage of the veneer processing. The heat was transmitted from outside to inside, and the plywood gradually achieved T_{HP} from the surface layer to the core layer. The internal temperature of the plywood would fail to meet the curing temperature of MSP adhesives as needed when the t_{HP} was too short. This meant that at $t_{HP} \leq 240$ s, the core layer was at the first constant temperature stage, so the moisture was still being vaporized, regardless under which T_{HP}. The MSP adhesives were in the process of eliminating moisture and did not reach the curing phase, so the wet shear strength was low. When the t_{HP} was 360 s, the moisture vaporization phase of the core layer was completed, and the slow warming stage began. In this case, when the core layer temperature exceeded 120 °C, the wet shear strength could meet the standard requirement (≥ 0.7 MPa). The wet shear strength showed a maximum of 1.09 MPa when the 3-layer plywood was hot-pressing at 125 °C and 1.0 MPa for 720 s. When the t_{HP} exceeded 600 s and the core layer temperature was at least 115 °C, the wet shear strength of all samples was enough to satisfy the standard requirement (≥ 0.7 MPa). This phenomenon illustrated that, even if the whole panel reached the T_{HP}, a certain period of time still was needed to cure the adhesives completely. Plywood prepared at any T_{HP} could meet the standard requirement (≥ 0.7 MPa) when the t_{HP} was 900 s. However, when the t_{HP} was too long, it caused aging of the glue layer and reduced the wet shear strength. Also, at high t_{HP}, the compression ratio and the production cost of plywood are increased.

In consideration of the resultant quality and cost of 3-layer plywood preparation, the optimum parameters were a t_{HP} of 600 to 720 s and a T_{HP} of 120 to 125 °C.

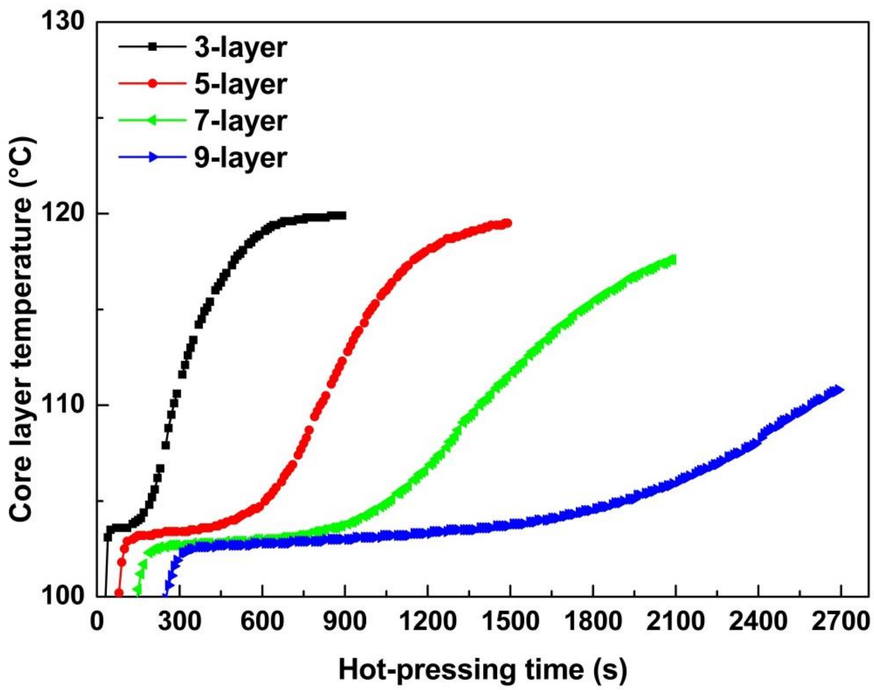

Fig. 4. Core layer temperature curves of multi-plywood prepared at 120 °C

Table 2. Time Needed for Core Layer of Multi-Plywood to Reach 100 °C and Duration of First Constant Temperature Stage

Veneer layers	Time to reach 100°C (s)	Duration of first constant temperature stage (s)
3	30	90
5	80	360
7	150	700
9	250	1240

Figure 4 shows the core layer temperature curves of multi-plywood prepared at 120 °C. The heating rate decreased significantly with the increase of veneer layers, not only in the rapid warming stage but also in the slow warming stage (Fig. 4 and Table 2). With the increase of veneer layers, the time for the core layer to reach 100 °C and the duration of the first constant temperature stage (moisture vaporization) increased (Table 2), as did the time for the core layer to reach T_{HP} (Fig. 4).

The moisture vaporization temperature declined with the increase of veneer layers, while the period of moisture vaporization was prolonged obviously, probably because more heat was needed for moisture vaporization as a result of the increase of veneer layers. However, the supply of heat quantity per unit time was limited, and the steam pressure dropped, leading to the decrease of moisture vaporization temperature. In addition, from the perspective of fracture density distribution, the center layer density is smaller for thicker plywood (Wang 1982; Xu 1995). This means that the center layer was less resistant to moisture vaporization; thus the moisture vaporization temperature was lower. With the increase of veneer layers, the total moisture content of the plywood increased, prolonging the water vaporization period.

The heat transfer rate was reduced with the increase of veneer layers, so the core layer required more time to reach the balance temperature. Moreover, the difference between the balance temperature and T_{HP} also increased (Fig. 4). When there were too many layers of plywood during the hot-pressing, the actual core layer temperature was not the T_{HP} (with unlimited extension of the t_{HP}, core layer temperature could only get very close to the T_{HP}). In actual production, much attention should be paid to matching the theoretical parameters, such as the adhesive curing temperature and the actual core layer temperature of plywood, to ensure complete curing of the adhesives.

Wet Shear Strength Analysis

Figure 5 shows that under the same hot-pressing conditions (120 °C, 1.0 MPa, 5 layers), the wet shear strength of the core and surface layers increased as t_{HP} increased. The wet shear strength of the surface layer was higher than that of the core layer and was high enough to meet the requirement of GB/T 9846.3-2004 (≥ 0.7 MPa) for type II plywood for all tested t_{HP} from 480 to 1500 s, while the core layer failed for t_{HP} of 480 and 600 s. This was because the surface layer reached the curing temperature of MSP adhesives earlier than the core layer did, so the MSP adhesives on the surface layer had enough time to cure sufficiently. Furthermore, the density of the surface layer was higher than that of the core layer, which might be another reason for this phenomenon.

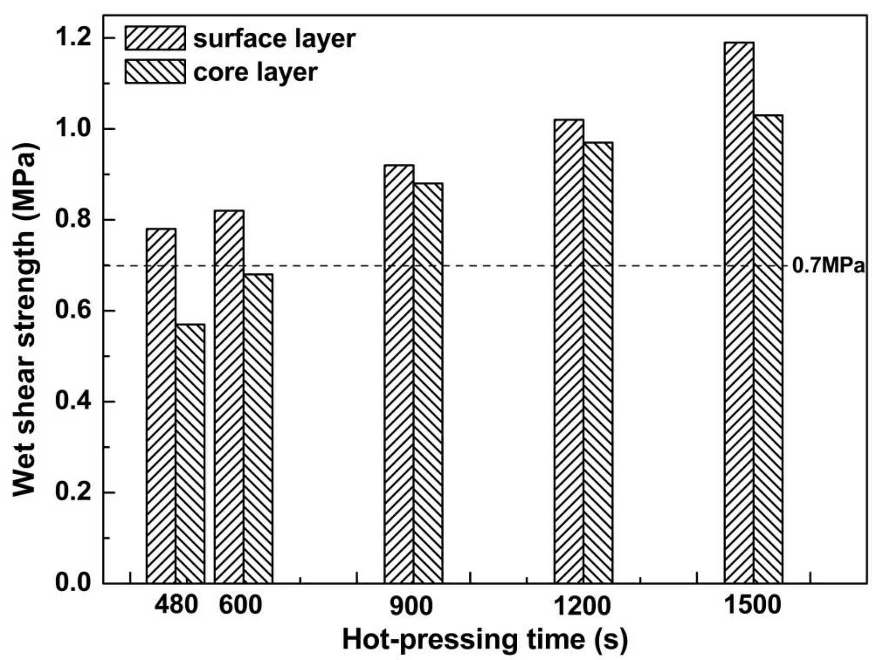

Fig. 5. Wet shear strength of 5-layer plywood prepared for various durations at 120 °C

Figure 6 shows that under the same hot-pressing conditions (900 s, 1.0 MPa, 5 layers), the wet shear strength of the core and surface layers was improved with the increase of T_{HP}. The wet shear strength of the surface layer was greater than that of the core layer, and the wet shear strength peaked at a T_{HP} of 125 °C (1.2 MPa at the surface layer, 1.1 MPa at the core layer). The wet shear strength of 5-layer plywood could meet the requirement of GB/T 9846.3-2004 (≥ 0.7 MPa) when the T_{HP} was 115 °C. This result showed that the 5-layer plywood could meet the standard requirement only with $T_{HP} \geq$ 115 °C and with $t_{HP} \geq 900$ s.

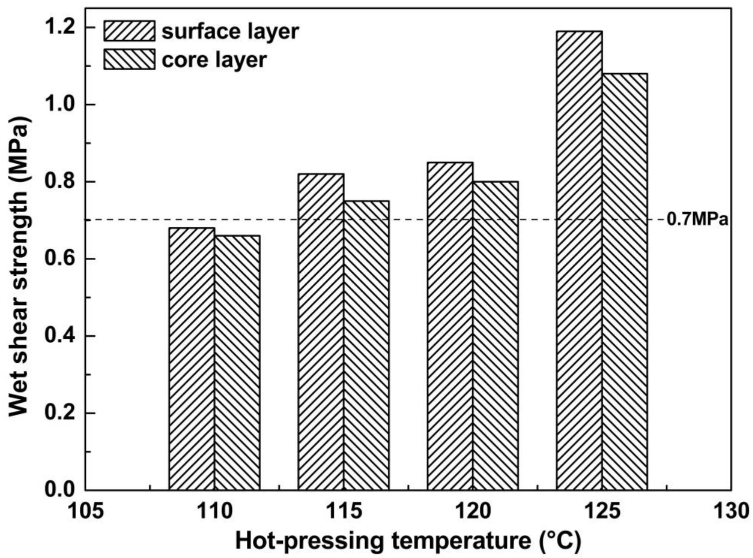

Fig. 6. Wet shear strength of 5-layer plywood prepared at various hot-pressing temperatures for 900 s

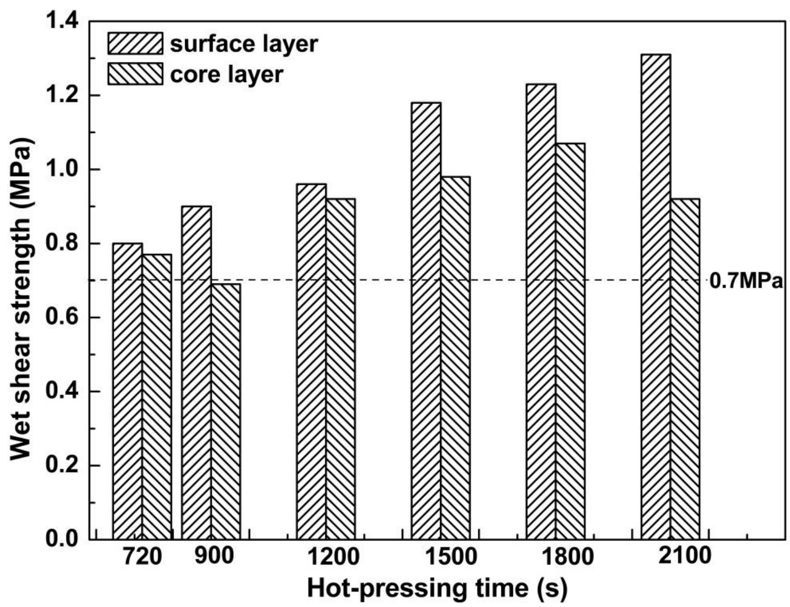

Fig. 7. Wet shear strength of 7-layer plywood prepared for various durations at 120 °C

Figure 7 shows that under the same hot-press conditions (120 °C, 1.0 MPa, 7 layers), the wet shear strength of the surface layer increased with the increase of t_{HP}, while the wet shear strength of the core layer increased generally. The wet shear strength of the surface layer was higher than that of the core layer, which was consistent with the result for 5-layer plywood. Again, this was because the surface layer reached the curing temperature of MSP adhesives earlier than the core layer did, so the MSP adhesives on the surface layer had enough time to cure sufficiently. The other reason was that the density of the surface layer was greater than that of the core layer. With increasing veneer layers, the linear dependence relation between the wet shear strength of the core layer and the t_{HP} was markedly reduced. This was probably because the increase in veneer layers resulted in slower heat transfer, so the adhesives were unable to flow freely, inhibiting effective adhesion. Moreover, the factors influenced the wet shear strength and the uncontrollable factors of the test were inevitably increased.

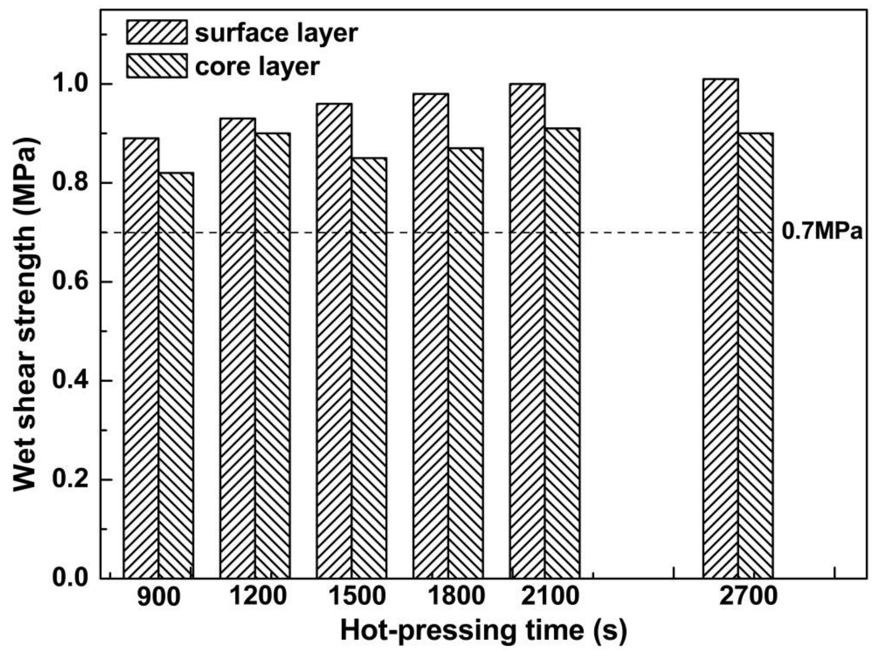

Fig. 8. Wet shear strength of 9-layer plywood prepared for various durations at 120 °C

Figure 8 shows that under the same hot-pressing conditions (120 °C, 1.0 MPa, 9 layers), the wet shear strength of the surface layer improved with increasing t_{HP}, while the wet shear strength of the core layer increased generally. The increase in the wet shear strength of 9-layer plywood was obviously smaller, compared with the 5-layer and 7-layer plywoods, because the 9-layer plywood was very thick, resulting in higher total moisture content and lower heat transfer efficiency. Therefore, the period of moisture vaporization prolonged with the increase of veneer layers, and the wet shear strength without noticeably improved by t_{HP} increased from 900 to 2700 s. Consequently, the adhesives were unable to flow freely, inhibiting effective adhesion. Moreover, the factors influenced the wet shear strength and the uncontrollable factors of the test were inevitably increased. Therefore, further research should be performed.

CONCLUSIONS

1. The temperature curve of the core layer of plywood bonded with MSP adhesives during hot-pressing was divided into four stages: the rapid warming stage before moisture vaporization; the first constant temperature stage during moisture vaporization; the slow warming stage after moisture vaporization; and the second constant temperature stage after moisture vaporization. The first constant temperature stage and the slow warming stage composed a large percentage of t_{HP}, so they were

decisive with respect to the time needed for the core layer to reach the T_{HP}. The rapid warming stage accounted for a small percentage of the t_{HP}.

2. The core layer temperature of plywood largely depended on the veneer layers. The heating rate decreased noticeably with the increase of veneer layers. The effect of an increased number of layers mainly was seen in the extension of the first constant temperature stage and the increase of time needed for the core layer temperature to reach the T_{HP}. The heating rate of the plywood core layer increased with the increase of T_{HP}.
3. The wet shear strength of plywood was the highest when the T_{HP} was equal or greater than 120 °C and the t_{HP} was greater than the time required for vaporization of the moisture within the plywood. All such samples could meet China National Standard (GB/T 9846.3-2004, ≥ 0.7 MPa) for type II plywood. The time of moisture vaporization was about 400 s for 3-layer plywood and 900 s for 5-layer plywood, which could be set as a theoretical basis of the plywood hot-pressing; thus, the unnecessary cost in practical production could be avoid. The core layer temperature of 7-layer and 9-layer plywoods changed slowly, and the linear relationship between the core layer's wet shear strength and the t_{HP} was very weak.

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