# Modified High-density Polyethylene Film as the Adhesive for Veneer Overlaying of Wood-based Panel

Ying Wu, Jianying Xu,* Lishu Deng, Yu Su, Jiaying Liang, and Xinfeng Wu

Maleic anhydride grafted high-density polyethylene (MAH-HDPE) film was used instead of urea-formaldehyde resin adhesive to face wood-based panels (WBP) such as finger-jointed wood panel, medium-density fiberboard (MDF), high-density fiberboard (HDF), and three-ply plywood with decorative wood veneer. The effects of hot pressing conditions on the physical-mechanical performance (surface bonding strength, water immersion performance, and hot-cold cycling performance) of overlain veneered panels were evaluated. Rheology analysis, differential scanning calorimetry (DSC), and attenuated total reflection-Fourier transform infrared spectroscopy (ATR-FTIR) were used to study the characteristics of MAH-HDPE film. Scanning electron microscopy (SEM) was used to observe the microstructure of the bonding interface. The veneered board using MAH-HDPE film as adhesive showed excellent properties, especially on water resistant and weather resistant performance. The veneered boards with finger-joint wood panel substrate could meet the Chinese national standard of type I grade veneered board of GB/T 17657 (2006), and the optimum hot pressing parameter was under a pressure of 1.3 MPa at 160 °C for 4 min. The MAH-HDPE film also can be applied to various other wood-based substrate materials for preparing veneered board. No glue penetration was observed on the surfaces.

*Keywords: Veneered board; HDPE film; Overlaying; Decorative wood veneer*

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## INTRODUCTION

Wood-based panels are widely used in furniture manufacturing and interior decoration. Before applications, the surfaces of these panels are usually decorated with various materials, such as wood veneer, melamine-impregnate paper, vinyl film, and other finishes (Rowell 2012). Among these methods, wood veneer overlaying is one of the most often used methods for presenting the beautiful natural appearance of wood. During the veneer-overlaid process, a mixed adhesive composed of urea-formaldehyde (UF) resin and polyvinyl acetate is typically used to bond the veneer and wood-based panel (National Particleboard Association 1996). However, this adhesive has some disadvantages. UF emits formaldehyde, which is harmful to human health. The adhesive easily penetrates the porous veneer, which stains the surface and reduces the decorative effect. Therefore, it is important to look for a formaldehyde-free and non-aqueous adhesive to replace the traditional adhesive.

High-density polyethylene (HDPE) has a wide variety of applications on account of its low cost and versatile properties. As a thermoplastic material, HDPE is in a solid state, contains no formaldehyde, and has great water and weather resistance. HDPE particles can be mixed with wood powder to fabricate a wood-plastic composite (WPC). HDPE film has been used as the adhesive for manufacturing plywood. Fang *et al.* (2013) fabricated plywood with veneers and pure HDPE films by a hot and cold pressing successive process. Their study showed that the tensile shear strength of resulting plywood met the requirement of type II grade (water-resistance) plywood of Chinese national standard GB/T 9846.3 (2004). However, HDPE is a non-polar characteristic material, which leads to its poor cementation with polar materials such as wood and wood-based materials. To improve the interfacial adhesion of HDPE film and wood, many studies have been conducted. Tang *et al.* (2011, 2012) modified HDPE powder through *in situ* chlorinating graft copolymerization, and the results indicated that the tensile shear strength of the resulting plywood can meet the standard of Type I grade (weather-resistance) plywood. Fang *et al.* (2013, 2014) treated the surface of poplar veneer with silane A-171 (vinyl trimethoxy silane) by spraying to fabricated wood-plastic plywood with HDPE film and poplar veneer, which resulted in a 293.2% increase in shear strength, with a 34.6% and 40.8% reduction in water absorption and thickness swelling, respectively. While there have been significant efforts in using HDPE film to fabricate plywood, the authors did not find any record of research using HDPE film as an adhesive to face veneer on wood-based panel substrates.

In this study, MAH-HDPE (maleic anhydride grafted high-density polyethylene) was used as adhesive, and wood-based panels were overlain with decorative wood veneers. The effects of hot pressing conditions on the physical and mechanical properties (surface bonding strength, water immersion performance, and hot-cold cycling performance) of overlain veneered panels were evaluated.

## EXPERIMENTAL

### Materials

*Decorative wood veneers*

*Pinus radiata* veneer (1.0 mm thick) and mahogany veneer (0.2 mm thick) were purchased from Fuzhou, China. Both veneers were rotary cut, with the moisture content of 10%. The veneers were cut to the size of 300 × 300 mm.

*Substrates*

Finger-joint wood panels (species: Chinese fir; Hunan Fuxiang Wood Industry Co. Ltd., Yueyang, China) were 15 mm thick, with 10% moisture content. The MDF was purchased from Hunan Wanhua Wood Industry Co. Ltd. (Chenzhou, China), and was 9 mm thick, with 9% moisture content. The HDF was purchased from Dare panel Group Co., Ltd. (Danyang, China), and was 8 mm thick, with 9% moisture content. Three-ply plywood of 6 mm thickness was prepared in the lab with poplar veneers bonded with MAH-HDPE film; the moisture content was 10%.

*Adhesive*

The MAH-HDPE film was supplied by Qingxin Clarwood co., Ltd. (Fuzhou, China), with a thickness of 0.03 mm and a density of 0.97 g $cm^{-3}$. The film weighed 29 g per square meter.

Unmodified HDPE film (control) was supplied by Lvyuan Co., Ltd. (Fuzhou, China), with a thickness of 0.03 mm and a density of 0.91 g $cm^{-3}$. The film weighed 27 g per square meter.

## Veneered Board Fabrication

The surface of the substrate was covered with a piece of MAH-HDPE film, and the decorative wood veneer was placed on the film to form a mat. The mat was hot pressed and then immediately cold pressed at room temperature under the pressing pressure of 1.5 MPa for 10 min. The experiments were divided into two groups.

### *Veneered boards fabricated under various hot pressing conditions*

Using *Pinus radiata* veneer as finishing material, finger-joint wood panel as the substrate material, and MAH-HDPE film as adhesive, veneered boards were hot pressed under various temperature (130 to 180 °C), pressure (0.8 to 1.6 MPa), and time (1 to 5 min). HDPE film was used as adhesive to prepare veneered boards at one condition as a control. The veneered board was unloaded and immediately moved to a cold-press to consolidate the panel. The specific hot pressing conditions of the veneered board are shown in Table 1.

### *Veneered boards fabricated with different kinds of wood veneers and substrate materials*

*P. radiata* and mahogany wood veneer were used as finishing materials, and finger joint wood panel, MDF, HDF, and three-ply plywood were used as substrate materials to fabricate veneered boards. The hot pressing conditions were a pressure of 1.3 MPa, time of 4 min, and temperature of 160 °C.

## Veneered Board Evaluation

The physical and mechanical properties of veneered boards were investigated according to Chinese national standards GB/T17657-2013 (2013) and GB/T 15104-2006 (2006), including surface bonding strength (SBS), water immersion test, and hot-cold cycling treatment test.

### *The surface bonding strength of veneered board*

The SBS between veneers and substrates was evaluated following a procedure listed in GB/T17657-2013 (2013). The dimensions of the test specimen were 50 × 50 mm. The center of the specimen was bonded to a metal chuck with a contact area of 20 × 20 mm, and the wood veneer was cut along the edge of the chuck (Fig. 1). The SBS was then tested.

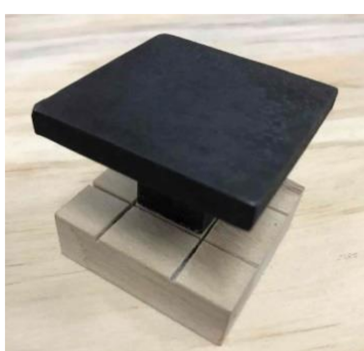

**Fig. 1.** The specimen bonded with a chuck

*Water immersion performance of veneered board*

The dimensions of the test specimen were 75 × 75mm. According to GB/T 15104-2006, veneered boards were divided into 3 types depending on their properties of water resistance.

The testing methods are as follows: for type I grade (weather-resistance) veneered board, specimens were immersed in boiling water for 4 h, oven-dried at 63 °C for 20 h, immersed again in boiling water for 4 h, and then oven-dried at 63 °C for 3 h. For type II grade (water-resistance) veneered board, specimens were immersed in 63 °C water for 3 h and then oven-dried at 63 °C for 3 h. The delaminated length at four edges of the specimen was measured. If the delaminated length of every edge is shorter than 25 mm, the veneered board is qualified, otherwise, it is unqualified.

*Hot-cold cycling treatment test*

The dimensions of the test specimen were 150 × 150 mm. The specimen was dried at 80 ± 3 °C for 2 h, followed by freezing at -20 ± 3 °C for 2 h. The drying/freezing cycle was carried out two times before returning to room temperature. Check, bubble, crimping, discoloration, and baldness are not allowed to occur on the surface, while the dimensions should remain stable.

*Statistical analysis*

All experiments were conducted in triplicate and the average values with standard deviation were used in the analysis. All data were evaluated by variance (ANOVA) using SPSS. To determine differences between treatments, Duncan tests were applied and significant differences were established at $p<0.05$.

## Analysis of MAH-HDPE and HDPE Films

*Differential scanning calorimetry analysis (DSC)*

DSC was performed using a Q100 differential scanning calorimeter (TA Instruments, New Castle, DE, USA). The sample of 5 mg in a flow of nitrogen, heated from 25 to 200 °C with a rate of 10 °C /min.

*Rheology analysis*

Rheology analysis was performed using a DHR-2 rotational rheometer (TA Instruments), in a flow temperature ramp mood. The temperature ranged from 130 to 180 °C, with a ramp rate of 5 °C/min, a velocity of 6.28 rad/s, and a sampling interval of 10 s/pt.

*ATR-FTIR analysis*

ATR-FTIR analysis was performed using a Nicolet IN10 MX infrared micro-spectrometer (Thermo Scientific, Waltham, MA, USA). The samples of 10 mm × 10 mm films were analyzed.

## Microstructure of the Bonding Interface

The samples of 0.8×0.4×0.4 cm cuboids were cut from the side face of C4 (Table.1) veneered board. The microstructure of bonding interface was observed in a Quanta 450 scanning electron microscope (Thermo Scientific).

## RESULTS AND DISCUSSION

### Characteristics of MAH-HDPE Film

*Melting points and viscosity of HDPE and MAH-HDPE Films*

As Fig. 2 shows, the melting points of HDPE and MAH-HDPE films were 126 °C and 121 ℃, respectively. Only when the hot pressing temperature was higher than the melting point was the film able to melt, flow, and penetrate the wood structures. While the viscosity of MAH-HDPE film decreased rapidly from 130 °C to 140 °C and remained almost the same value after 140 °C, the viscosity of HDPE film stayed relatively stable between 130 ℃ and 140 ℃ and had lower viscosity than MAH-HDPE film after 150 ℃. (Fig. 3). These may indicate that MAH-HDPE film responded to heat quicker and melted faster than HDPE film.

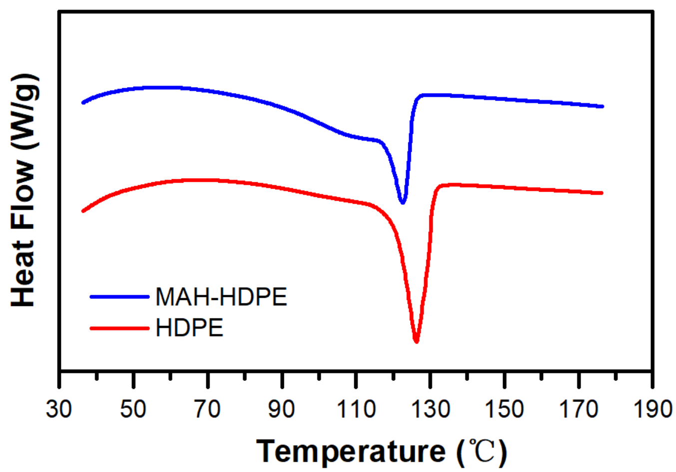


**Fig. 2.** DSC result of HDPE and MAH-HDPE films

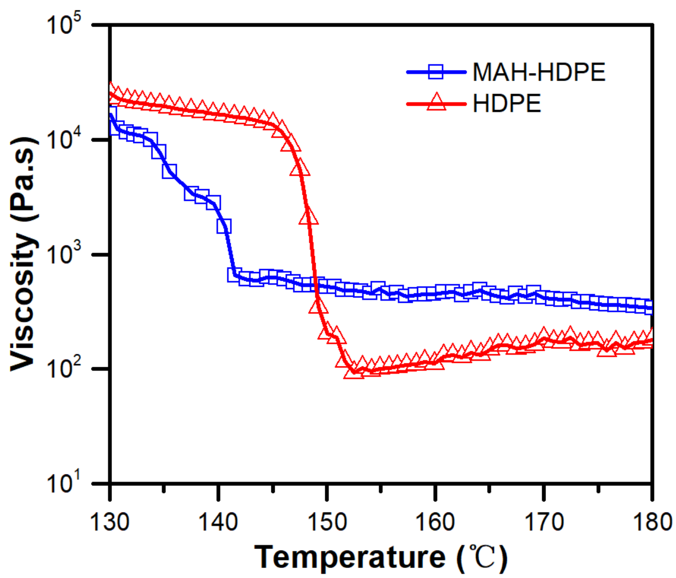


**Fig. 3.** Rheology analysis of HDPE and MAH-HDPE films

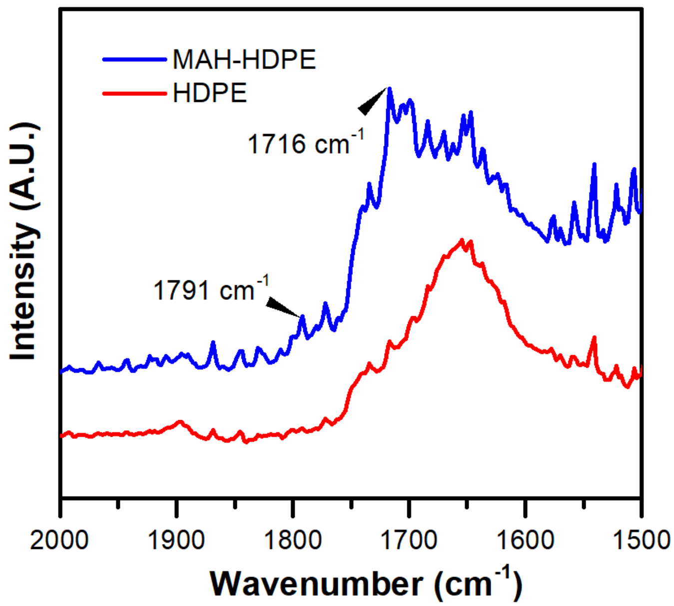


**Fig. 4.** The ATR-FTIR analysis of HDPE and MAH-HDPE films

*ATR-FTIR analysis of HDPE and MAH-HDPE films*

Figure 4 shows that there were new absorption peaks of carbonyl groups of circular anhydride at 1791 $cm^{-1}$ and carbonyl groups of carboxylic acid at 1716 $cm^{-1}$ after HDPE film was modified, which confirmed that MAH was grafted onto the molecular chain of HDPE (Ma *et al*. 2009).

## Effect of Hot Pressing Conditions on Physical-mechanical Properties of Veneered Board

*Surface bonding strength*

Figure 5 shows that the SBS increased when the hot pressing temperature was raised from 130 °C to 160 °C. Increasing the temperature to 180 °C did not greatly change the values. The reason may be related to the characteristic of MAH-HDPE film. The melting point of MAH-HDPE was 121 °C, and the viscosity decreased rapidly when the temperature increased from 130 °C to 140 °C (Figs. 2 and 3). Thus, the film was able to flow and penetrate into the wood cells when the pressing temperature was higher than the melting point. Figure 6 is the SEM photograph of the bonding interface of the veneered board. The MAH-HDPE penetrated into the cell lumens of the veneer surface and functioned as a glue, as previously noted (Chang 2014).

Figure 5 also shows that under the same pressing conditions, the SBS of veneered board bonded with MAH-HDPE film was 54% higher than that of with control HDPE film. The increase of SBS may also be related to the esterification reaction to create a covalent linkage between the carbonyl group of MAH-HDPE and the hydroxy group of wood-based panel/wood veneer (Fig. 7). Guo *et al*. (2016) also reported that when the WF/HDPE panel was covered with the veneer by using the MAPE intermediate film, a new peak of an ester group located at 1,741.46 $cm^{-1}$ was detected on the interface after pressing. A relatively low temperature is not enough to initiate the esterification reaction (Tang *et al*. 2011), considering the heat loss during hot pressing and when board transferring from hot-press to cold-press. The hot pressing temperature higher than 150 °C is required.

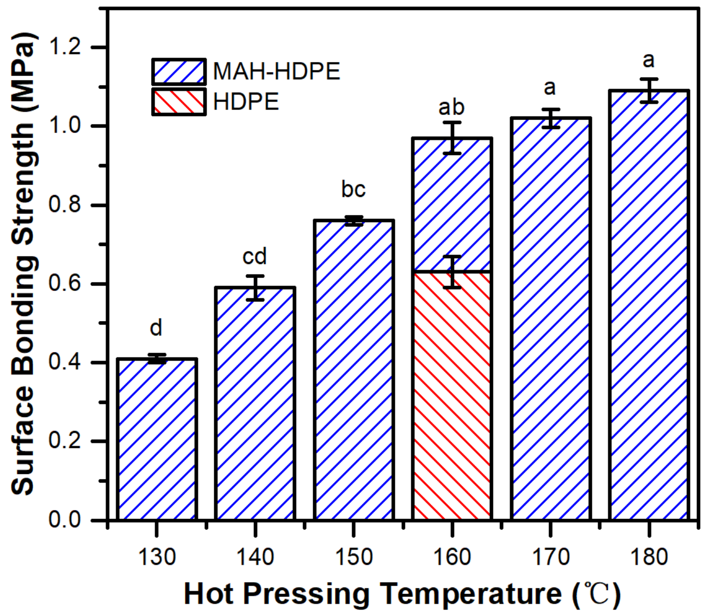


**Fig. 5.** Effect of hot pressing temperature on SBS of veneered board. Different letters in the figure indicate that means are significantly different ($p<0.05$）

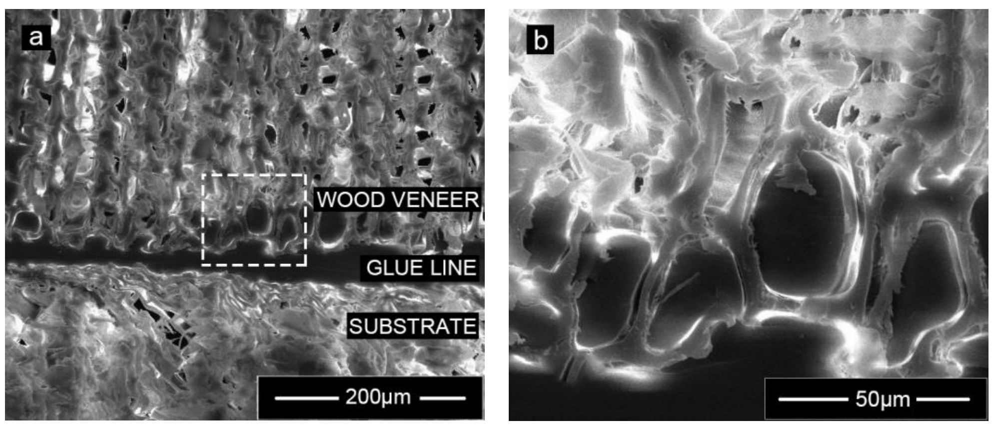


**Fig. 6.** The microstructure of bonding interface of veneered board

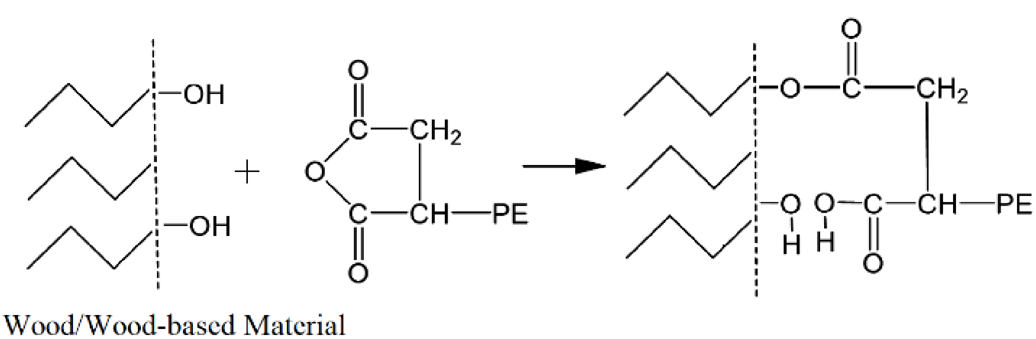


**Fig. 7.** The esterification reaction of MAH with hydroxyl groups of wood/wood-based materials

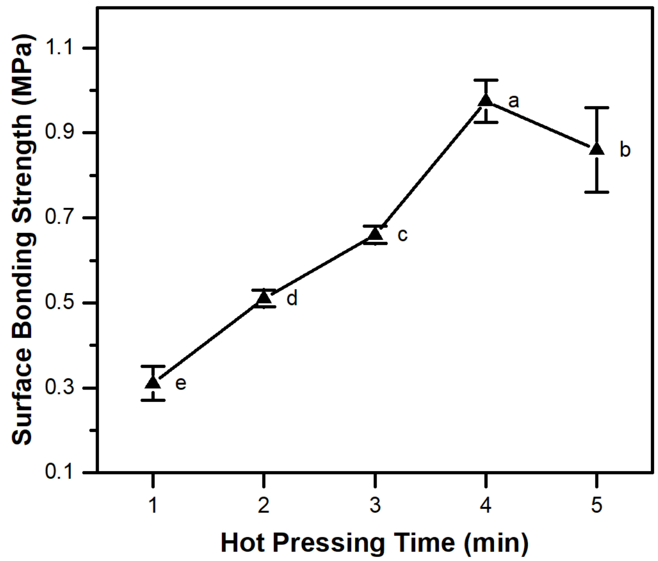


**Fig. 8.** Effect of hot pressing pressure on SBS of veneered board. Different letters in the figure indicate that means are significantly different (p<0.05)

The effect of hot pressing pressure on the SBS of veneered board is shown in Fig. 8. The SBS increased when the pressure increased from 0.8 to 1.3 MPa; however, the SBS value decreased when further increasing the pressure to 1.6 MPa. The role of hot pressing pressure is to make adequate contact between veneer and substrate through the bonding action of adhesive. In a certain range, increasing pressure made intimate contact between bonding surface; the melted HDPE film evenly distributed on the bonding surface and partly penetrated into the wood material, creating a good bonding condition. When the pressure is too high, however, the glue line might become much thinner even could not form a continuous film membrane, resulting in the reduction of surface bonging strength.

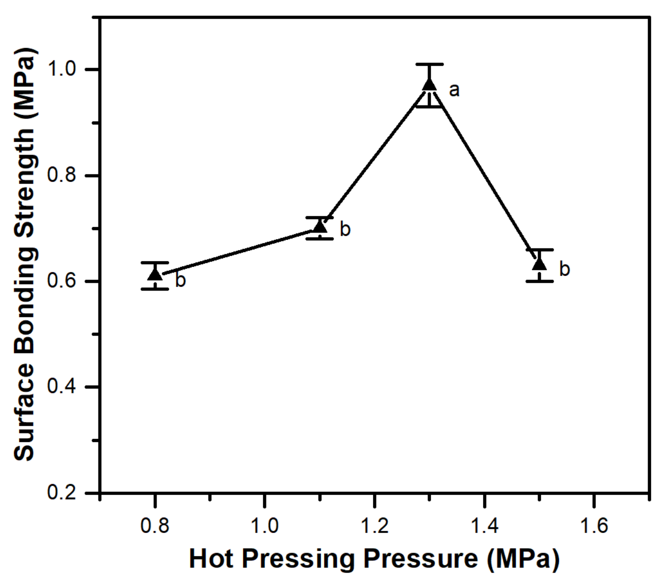


**Fig. 9.** Effect of hot pressing time on SBS of veneered board. Different letters in the figure indicate that means are significantly different (p<0.05)

**Table 1.** Effect of Hot Pressing Conditions on Physical-mechanical Properties of Veneered Board

| Number | | Hot Pressing Conditions | | | Physical-Mechanical Properties | | |
|---|---|---|---|---|---|---|---|
| | | Temperature (°C) | Pressure (MPa) | Time (min) | Surface Binding Strength (MPa) | Immersion Test | Hot-cold Cycling Treatment |
| A | 1 | 160 | 0.8 | 4 | 0.6 | type I | passed |
| | 2 | 160 | 1.1 | 4 | 0.71 | type I | passed |
| | 3 | 160 | 1.3 | 4 | 0.97 | type I | passed |
| | 4 | 160 | 1.5 | 4 | 0.63 | type I | passed |
| B | 1 | 160 | 1.3 | 1 | 0.31 | type II | failed |
| | 2 | 160 | 1.3 | 2 | 0.51 | type II | passed |
| | 3 | 160 | 1.3 | 3 | 0.66 | type I | passed |
| | 4 | 160 | 1.3 | 4 | 0.97 | type I | passed |
| | 5 | 160 | 1.3 | 5 | 0.85 | type I | passed |
| C | 1 | 130 | 1.3 | 4 | 0.4 | type II | failed |
| | 2 | 140 | 1.3 | 4 | 0.59 | type II | passed |
| | 3 | 150 | 1.3 | 4 | 0.75 | type I | passed |
| | 4 | 160 | 1.3 | 4 | 0.97 | type I | passed |
| | 4* | 160 | 1.3 | 4 | 0.63 | type II | passed |
| | 5 | 170 | 1.3 | 4 | 1.01 | type I | passed |
| | 6 | 180 | 1.3 | 4 | 1.09 | type I | passed |
| Number A3, B4, C4 is the same board. Number 4* is the board with HDPE film (control). | | | | | | | |

Figure 9 shows the effect of hot pressing time on the SBS of the veneered board. The SBS values increased noticeably when the hot pressing time increasing from 1 to 4 min. Under the pressing time of 4 min, the SBS value was 0.97 MPa, which was much higher than the minimum requirement (0.4 MPa) of GB/T 15104 (2006). Relatively longer time may contribute to the melting and even distribution of HDPE film between veneer and substrate. However, a too-long pressing time may cause the over-penetration of melting adhesive into the porous wood materials, resulting in starved glue lines. This may explain why a decreased SBS value was observed at the hot pressing time of 5 min.

## Water-resistance and Weather-resistance

As the great weather and water resistance associated with plastic is evident in WPC materials (Zhou *et al.* 2017), the veneered-board using MAH-HDPE film as bonding adhesive performed well in the water immersion and hot-cold cycling treatment tests. As shown in Table 1, most of the veneered board passed the type I water immersion test, except for the boards that were hot pressed under low temperature (130 to 140 °C) and short time (1 to 2 min). The veneered boards using control HDPE film as adhesive can merely pass the type II grade test with a 5 mm of delaminated length. Compared with the board bonded with other kinds of adhesive, the veneered board bonded with MAH-HDPE film in this study showed excellent water-resistant performance. They could bear a long-time boiling test without delamination, while the veneered board using traditional UF resin as adhesive could only pass type II grade test. Plywood using pure HDPE film as adhesive can merely pass the type II grade test (Tang *et al*. 2011) (Chang *et al*. 2018). Except the veneered board overlaying under poor hot pressing conditions (130 °C and 1 min), all boards passed the hot-cold cycling treatment.

The excellent water-resistance and weather-resistance properties shown by them might be due to the strong covalent linkage between the carbonyl group of MAH-HDPE and the hydroxyl group of wood-based panel/wood veneer, which was mentioned previously.

## Effect of Different Kind of Wood Veneers and Substrate Materials on Physical-mechanical Properties of Veneered Board

All veneered boards were successfully manufactured. No glue penetration was observed on the surface of all veneered boards, even when using 0.2 mm porous mahogany veneer as the decorative material.

**Table 2.** Properties of Veneer Board Fabricated with Different Kinds of Wood Veneers and Substrate Materials

| Number | Substrate Material | Wood Veneer | Surface Binding Strength (MPa) | Immersion Test | Hot-cold Cycling Treatment |
|---|---|---|---|---|---|
| 1 | finger joint wood panel | mahogany | 1.09 | type II | passed |
| 2 | MDF | mahogany | 0.47 | type II | passed |
| 3 | HDF | mahogany | 0.91 | type II | passed |
| 4 | three-ply plywood | mahogany | 0.89 | type II | passed |
| 5 | finger joint wood panel | *P. radiata* | 0.97 | type II | passed |
| 6 | MDF | *P. radiata* | 0.43 | type II | passed |
| 7 | HDF | *P. radiata* | 0.81 | type II | passed |
| Mahogany: 0.20 mm thick, *P. radiata*: 1.00 mm thick | | | | | |

Table 2 shows the properties of veneered board fabricated with various wood veneers and substrates. All veneered boards showed high SBS value (higher than 0.8 MPa, which was twice the standard requirement), except for the veneered board with MDF substrate. NO.2 and NO.6 veneer boards with MDF substrate recorded lower SBS values than the other substrates. During the SBS test, the veneered boards with MDF substrate were broken in substrate rather than the interface (glue line) of wood veneer and substrate. This was because the glue line strength was higher than the internal bond strength of MDF substrate. The failure happened in the weakest place during tests, which means the actual value is higher than the test value. Despite this effect, the test SBS value of the veneered board with MDF substrate met the minimum requirement of the standard. Compared with other substrates, MDF has lower internal bonding strength thus the failure occurred in the substrate itself, while the failure of veneered boards with other substrates happened in the glue lines.

Considering that MDF and HDF do not have fine water resistance, all water immersion tests were performed as type II. The immersion test results show that all veneered boards can meet the standard of type II, and all veneered boards passed the hot-cold cycling treatment test. It is feasible to use different kinds of wood veneers to face different kinds of substrate materials.

In addition, the MAH-HDPE film contains no formaldehyde which is a promising adhesive for veneer decoration of wood-based panels.

## CONCLUSIONS

1. The veneered board using MAH-HDPE film as adhesive showed excellent properties, especially on water resistant and weather resistant performance. The board with finger-joint wood panel substrate met the requirements of the Chinese national standard GB/T 17657-2006 for Type I grade decorative veneered wood-based panel. The optimum hot pressing conditions were a temperature of 160 °C, pressing time of 4 min, and pressure of 1.3 MPa.
2. MAH-HDPE film melted and was evenly distributed between veneer and wood substrate to act as an adhesive. Ester linkages between the carbonyl groups of MAH-HDPE and the hydroxyl groups of wood-based panel/wood veneer might contribute to the high bonding strength of the glue line.
3. MAH-HDPE film can be applied to various wood-based substrate materials for preparing veneered board.
4. No glue penetration was observed on the surface of the veneered board, even when using 0.2 mm porous thin veneer as decorative material.

## ACKNOWLEGEMENTS

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