

MANUFACTURING AND INTERFACIAL BONDING BEHAVIOR OF EVA FILM REINFORCED FLEXIBLE DECORATIVE VENEER¹

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Abstract. Polyethylene film reinforced flexible decorative veneer has attracted wide attention. However, its curling deformation has not been well solved due to the high processing temperature. Ethylene-vinyl acetate copolymer (EVA) has lower melting temperature and better flexibility than polyethylene. In this work, EVA film was selected to enhance Manchurian ash (*Fraxinus mandshurica*) aimed at preparing flexible decorative veneer with high strength and low curling degree. Box-Behnken experimental design method was applied to determine the optimum processing parameters. The secondary overlaying performance of the flexible veneer was evaluated. Under optimum conditions the EVA reinforced veneer obtained a maximum transverse tensile strength of 2.42 MPa. EVA can form a continuous interface with the veneer, which was helpful to improve the flexibility of the veneer. Curling degree of the EVA film reinforced veneer was far less than that of the polyethylene reinforced veneer even at the same processing temperature. EVA reinforced flexible veneer was good to be used to decorate wood-based panels without additional adhesives. Both immersion peel strength and surface bonding strength of the prepared decorative panel can meet the requirements of GB/T 15104-2006.

Keywords: Flexible decorative veneer, polyethylene film, EVA film, overlaying performance.

INTRODUCTION

Wood-based composites have been widely developed as alternative materials for furniture manufacturing or structure building board because of

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their stable physical properties and wide sources of raw materials (Zheng et al. 2021; Lee et al. 2011; Gao et al. 2020). In order to obtain decorative and high value-added wood products, a sliced veneer with a thickness of 0.05~0.8 mm prepared from naturally precious wood is usually employed to overlay on the surface of wood-based panels (Guo et al. 2018; Yu et al. 2020; Zhang et al. 2020; Wang et al. 2020). Common precious tree species included Manchurian ash, oak, elm, black walnut, maple, teak, red sandalwood, and all the rest of it. Due to the small thickness of decorative veneer, it is easy to crack in the process of storage, transportation and use. Decorative veneer is rarely used for surface decoration of complex curved surfaces because of its poor flexibility (Ohyanagi et al. 2004). This problem can be solved to a certain extent by compounding a piece of paper, non-woven fabric or plastic film on the back of veneer (Armin 1937; Zhang et al. 2014a; Peng et al. 2019b; Zeng 2003). At present, paper reinforced decorative veneer is rarely used in the market, mainly resulting from its poor water resistance and unsatisfactory flexibility. The production cost of non-woven reinforced flexible decorative veneer is relatively low, and its flexibility, tensile strength and water resistance are much better than paper. Fuller company in UK has successfully prepared waterproof flexible decorative veneer by compounding 30 g·m⁻² non-woven fabric with oak wood by continuous rolling method (Arthur 1998). The existing research showed that the peel strength of non-woven reinforced flexible veneers was significantly affected by the veneer type, the glue spreading amount and the hot-pressing temperature (Zhang et al. 2014a; Zhang et al. 2015). Non-woven reinforced veneers can be continuously produced on a large scale, but the adhesive is always needed when it is subsequently used for wood-based panels surface decoration, which could cause the formaldehyde pollution problem (Qin et al. 2020). In the process of sizing and pressing, adhesive overpenetration may also occur, resulting in a decrease in bonding strength.

Plastic film has received great research interest in the area of decorative wood veneer flexible modification in recent years, because of its good flexibility

and water resistance, light weight, as well as excellent environmental protection (Peng and Zhang 2016; Zhou et al. 2020; Zhang et al. 2014b). The plastic film is not only the flexible reinforcing material for the decorative veneer, but also served as the adhesive in the process of veneer modification. Since there is no need to add additional adhesive, its preparation and finishing process is simple, and the energy consumption is low. Peng et al. systematically evaluated the manufacturing process and properties of flexible decorative veneer which was reinforced by modified polyethylene (PE) film (Peng and Zhang 2019a; Peng and Zhang 2020; Peng and Zhang 2019b; Peng and Zhang 2018; Peng and Zhang 2017). The flexible decorative veneer can obtain excellent peel strength, transverse tensile strength and flexibility under the appropriate process conditions. However, the processing temperature of PE film reinforced flexible veneer was higher (about 150°C), and the compatibility between hydrophilic wood and hydrophobic PE was poor, which resulted curling deformation of the veneer. When plasma pretreatment was performed on the sliced veneer or/and the PE film, the hot-pressing temperature can be reduced to 120°C (Peng and Zhang 2019a; Peng and Zhang 2017). In addition, the curling deformation caused by the large thermal expansion coefficient difference between the two materials can also be effectively alleviated because of the increased compatibility. But modification will make the preparation process more complex and increase the cost.

Ethylene-vinyl acetate copolymer (EVA) has the advantages of relatively low melting temperature, remarkable flexibility, and aging resistance among various plastics (Savini and Orefice 2021). It has been widely used in packaging, furniture, electronics and other industries in the form of particles or rods hot melt adhesive (Remmers et al. 2018). EVA film was also used in many areas, for example, solar cell packaging, roof composite materials, safety glass (He et al. 2012; Serafinavicius et al. 2013; Carvalho de Oliveira et al. 2018). Nevertheless, little information about the use of EVA film in the wood industry was obtained based on the literature review. Therefore, EVA film was selected as reinforcing material for the Manchurian Ash

decorative veneer in this study. The effect of hot-pressing conditions on the properties of EVA film reinforced veneer was evaluated by the response surface method (Li et al 2018; Li et al 2020). To evaluate the secondary overlaying performance of the EVA film reinforced flexible veneer, MDF panel was selected to composite with the flexible veneer under different hot-pressing temperatures.

MATERIALS AND METHODS

Materials

Manchurian Ash (*Fraxinus mandshurica*) decorative veneer with dimension of 250 mm × 250 mm × 0.5 mm was purchased from Deqing Meilun decoration materials Co., Ltd. EVA film with a thickness of 0.1 mm and density of 0.91 g·cm⁻³ was used as the reinforced material (Fujian Furong new materials Co., Ltd). Release paper was cut into the same size as the decorative veneer (Suzhou Yuxuan Paper industry Co., Ltd). MDF panels with a thickness of 8 mm was obtained from Shanghai Aiwei Industrial Development Co., Ltd.

Thermal properties of EVA film

Differential scanning calorimetry analysis (DSC). The melting temperature of the selected EVA film was measured using DSC-250 (TA Instruments) under the N₂ atmosphere. 5 ~ 10 mg of EVA film was used for the test. (1) heating from 0°C to 180°C at a rate of 10°C/min, equilibration for 5 minutes, (2) cooling from 180°C to 0°C at a rate of 10°C/min, equilibration for 5 minutes, (3) heating from 0°C to 180°C at a rate of 10°C/min.

Thermogravimetric Analysis (TGA). TGA of EVA film was conducted using TGA-250 (TA Instruments). 5 ~ 10 mg of sample was tested under N₂ flow at a heating rate of 10°C/min from 25°C to 800°C.

Preparation of EVA film reinforced flexible decorative veneer

The decorative veneer was composited with a piece of EVA film under different process conditions. A Box-Behnken factorial design method in response

Table 1. Experimental range and coded levels of the independent variables for fabricating EVA film reinforced flexible decorative veneers.

Coded	Factors	Ranges and levels		
		-1	0	+1
A	Hot-pressing pressure (MPa)	0.5	1	1.5
B	Hot-pressing temperature (°C)	95	110	125
C	Hot-pressing time (s)	50	100	150

surface methodology (RSM) was employed to optimize the process parameters. Three critical parameters: hot-pressing pressure (A), hot-pressing temperature (B) and hot-pressing time (C), were selected as the independent variables based on the preliminary experiments, and flexibility (Y1) and transverse tensile strength (Y2) were considered as the dependent variable (response). The parameters were coded in three levels of 0, -1 and +1, which represented central, low and high (Table 1). Experimental results were analyzed by means of Design Expert 11.0 software through which a regression model was proposed.

Physical-mechanical characterization

Flexibility. The flexibility of decorative veneer was evaluated according to the standard of “Decorative micro veneer” (LY/T 2879-2017). Specimens were cut into 120 mm × 75 mm along the grain, and tested using different diameter steel rods. Flexibility was defined as the minimum steel rod diameter when the specimen began to crimp and crack.

Transverse tensile strength. The EVA film reinforced decorative veneer was cut along the vertical grain into 100 mm × 20 mm test pieces and the transverse tensile test was performed according to the standard of “Test methods of evaluating the properties of wood-based panels and surface decorated wood-based panels” (GB/T 17657-2013).

Overlaying performance. The EVA film reinforced decorative veneer was subsequently composited with the MDF panel under the hot-pressing pressure of 1 MPa, and the hot-pressing time of 100 s. The hot-pressing temperature was controlled to 95°C, 100°C, 105°C, and 110°C

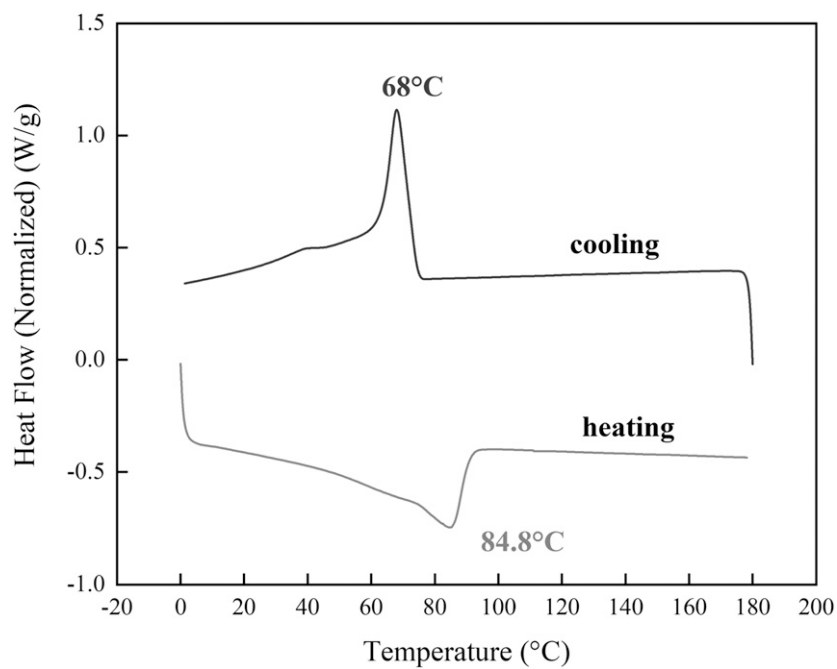


Figure 1. DSC curves of the EVA film.

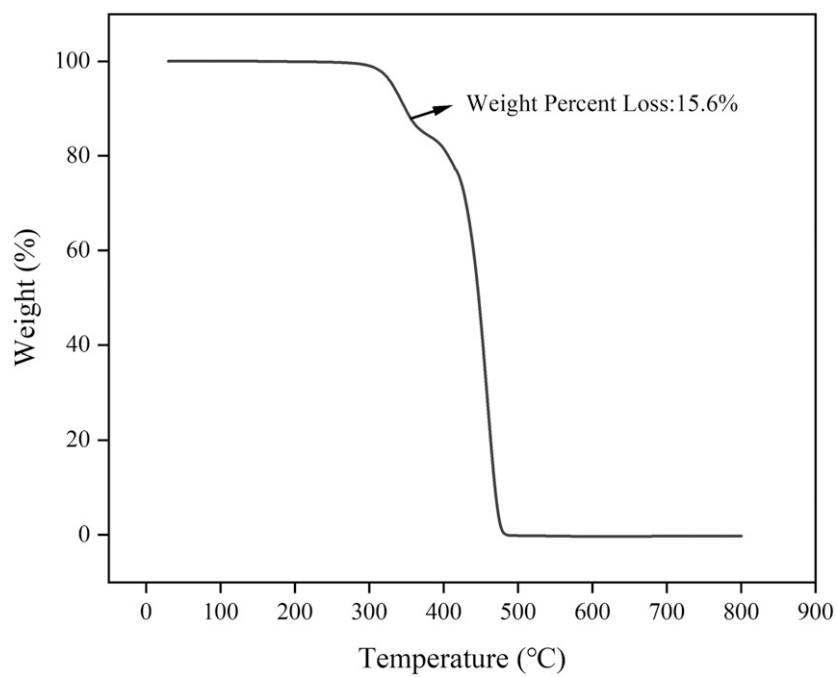


Figure 2. TGA curves of the EVA film.

Table 2. Experimental design and responses for RSM.

Run	Level A B C			Transverse tensile strength/MPa	Run	Level A B C			Transverse tensile strength/MPa
1	0	1	−1	1.26	10	1	−1	0	1.47
2	−1	−1	0	1.18	11	1	0	1	1.69
3	1	0	−1	1.58	12	0	−1	−1	0.93
4	0	0	0	2.31	13	0	0	0	2.33
5	0	0	0	2.42	14	0	0	0	2.37
6	−1	1	0	1.84	15	0	0	0	2.40
7	−1	0	1	1.90	16	1	1	0	1.55
8	−1	0	−1	1.58	17	0	−1	1	1.32
9	0	1	1	1.50	—	—	—	—	—

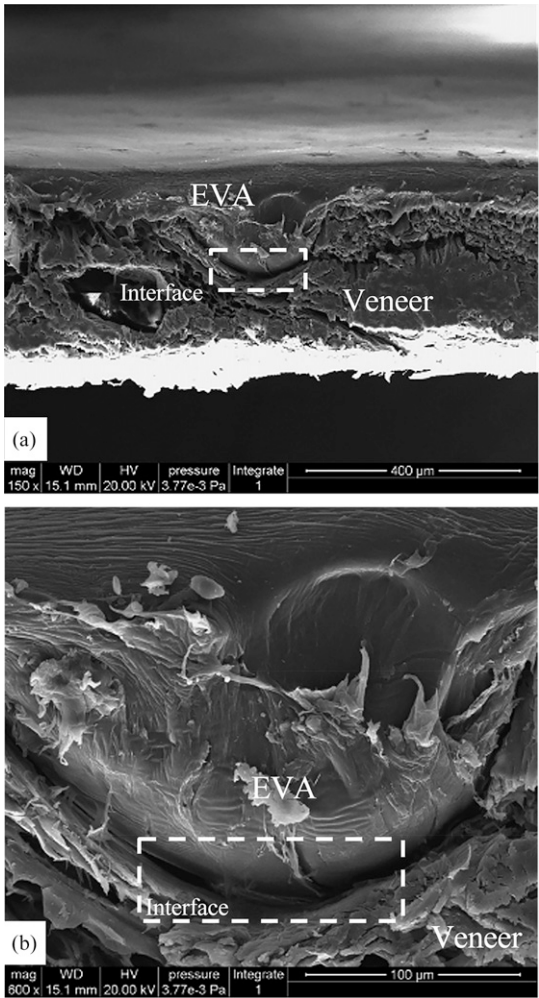


Figure 3. Bonding interface between EVA film and veneer under different magnifications (a) $\times 150$, (b) $\times 600$.

respectively. Overlaying performance, including surface bonding strength and immersion peel performance, was evaluated according to the standard of “Surface Decorated Wood-Based Panels” (GB/T 15104-2006).

Scanning electron microscopy (SEM). The bonding interface morphologies between EVA film and the decorative veneer, and the bonding intersection between MDF substrate and the flexible veneer, were examined under a Quanta-200 ESEM (Hillsboro, OR). The specimens were first coated with gold and then examined with an accelerating voltage of 15 kV.

RESULTS AND DISCUSSION

Thermal properties of EVA film

EVA is formed by bulk polymerization of ethylene monomer and vinyl acetate monomer under high pressure in the presence of initiator. EVA has lower crystallinity and better flexibility than PE due to the introduction of polar vinyl acetate (VA) monomer into the ethylene molecular chain. Representative DSC curve of the selected EVA was given in Fig 1. The first cooling stage in Fig 1 showed that the crystallization temperature of EVA was 68°C. Its melting temperature can be obtained from the second heating curve (Fig 1). In this paper, EVA film was not only a reinforcing material for *Fraxinus mandshurica* decorative veneer, but also acted as a wood adhesive. To form a good mechanical interlocking structure between EVA and decorative veneer, the hot-pressing temperature should not be lower than 90°C.

Table 3. ANOVA for experimental results.

Source	Sum of Squares	df	Mean Squares	F-value	P-value	Significance
Model	3.57	9	0.3972	108.41	<0.0001	**
A	0.0055	1	0.0055	1.50	0.2596	—
B	0.1953	1	0.1953	53.31	0.0002	**
C	0.1404	1	0.1404	38.34	0.0004	**
AB	0.0841	1	0.0841	22.96	0.0020	**
AC	0.0110	1	0.0110	3.01	0.1264	—
BC	0.0056	1	0.0056	1.54	0.2552	—
A ²	0.1866	1	0.1866	50.93	0.0002	**
B ²	1.75	1	1.75	478.88	<0.0001	**
C ²	0.9222	1	0.9222	251.72	<0.0001	**
Residual	0.0256	7	0.0037	—	—	—
Lack of fit	0.0171	3	0.0057	2.68	0.1824	—
Pure error	0.0085	4	0.0021	—	—	—
Std. Dev.	0.0605	—	—	—	—	—
C.V. %	3.47	—	—	—	—	—
Adjusted R ²	0.9837	—	—	—	—	—

Very significant ($p < 0.01$); * significant ($0.01 < p < 0.05$); — not significant ($p > 0.05$).

The properties of EVA varied substantially with the VA concentration. Here the VA content in the EVA film was roughly determined by TGA. As presented in Fig 2, EVA lost 15.6% weight at 379.9°C in the first pyrolysis stage. It was attributed to the loss of VA units via a deacylation process resulting in the formation of double bonds. The content of VA in EVA was 22.4% calculated by the polymer molecular formula. The second stage was between 420°C and 550°C, which was due to the degradation of the polyethylene. The mass loss of EVA was complete by the time the temperature reached 600°C.

Effects of process conditions on the transverse tensile strength of EVA film reinforced veneer

Transverse tensile strength of the flexible decorative veneer was observed in a total of 17 experimental points, including five replications of the central point. As shown in Table 2, the range of the transverse tensile strength was from the lowest of 0.93 MPa to the highest of 2.42 MPa. In the preparation of EVA film reinforced veneer, EVA melted and penetrated into the porous structure of the veneer (Fig 3), which has a great contribution to the improvement of the transverse tensile strength of the veneer. This was because the tensile strength

of EVA film was much greater than that of *Fraxinus mandshurica* veneer (0 ~ 0.9 MPa). Analysis of variance (ANOVA) was done to investigate these results, as presented in Table 3. On the basis of p values obtained, the developed quadratic model was found to be significant for transverse tensile strength. The model has the adjusted determination coefficient (R_{adj}^2) of 0.9837, suggested that the quadratic model could explain 98.37% of the total variations in the response. The method was further validated by prediction analysis, the experimental values were found to be within the confidence interval and the standard error predicted was small, which again suggested the model (Eq.1 in supplementary materials S1) was valid to explain and predict the transverse tensile strength of EVA film reinforced veneer.

The three selected factors were found to have some effect on transverse tensile strength after running the ANOVA test. On the basis of the p -value of A, B and C in Table 3, it can be seen that hot-pressing temperature was the most effective individual factor in transverse tensile strength of EVA film reinforced veneer. It was because that processing temperature affected the mobility and penetration depth of EVA. Hot-pressing time was another important factor. When the time was increased from 50 s to 100 s, a clear growth in

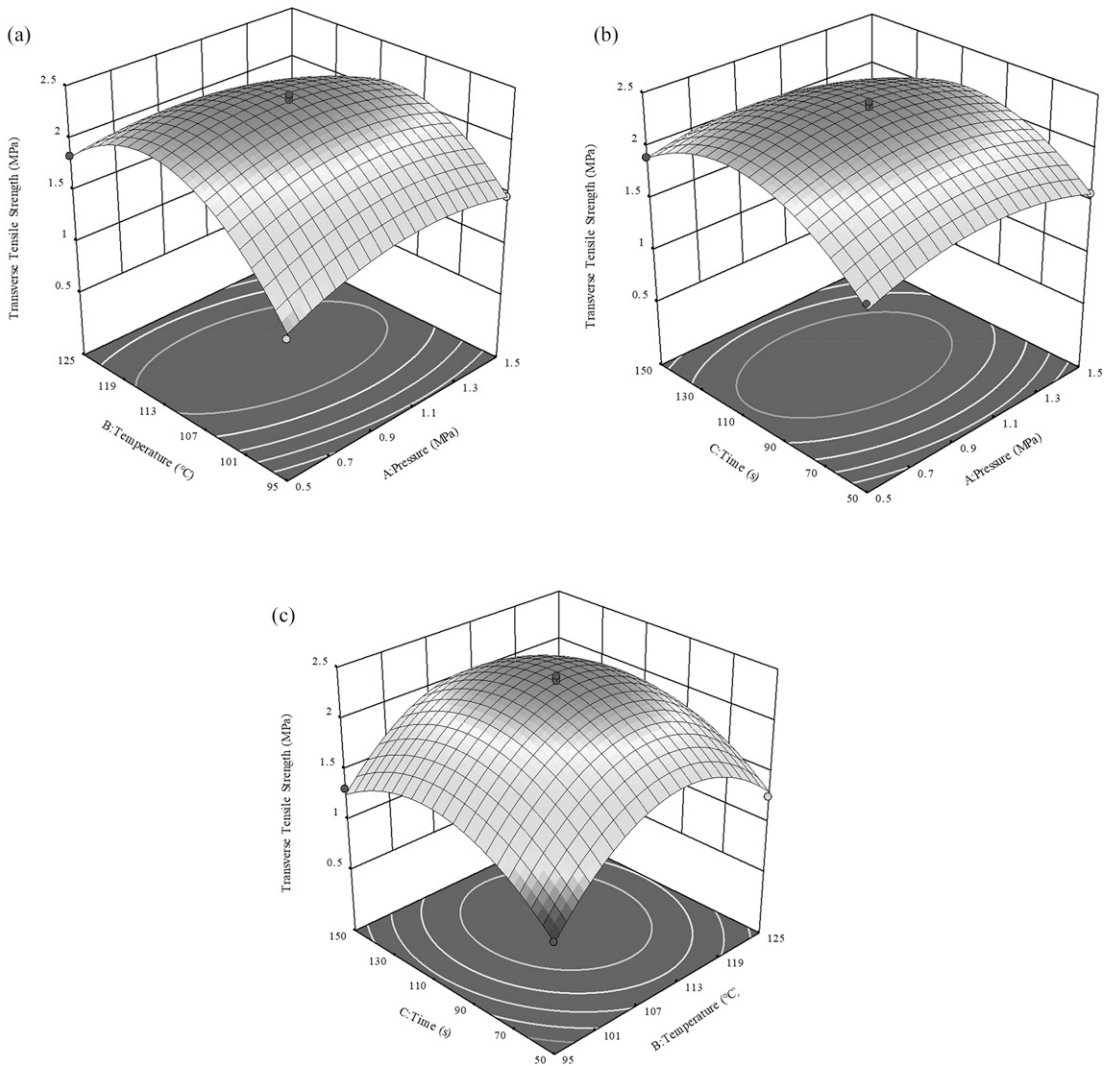


Figure 4. Three-dimensional plots obtained from the design of experiments approach for maximizing transverse tensile strength.

strength can be seen. One possible reason was that it took time for EVA to fully penetrate into the veneer and form a good mechanical interlock with the interior microstructure of decorative veneer. The importance of these factors and the interaction between the variable and the response were determined by illustrating the response surfaces as three-dimensional plots (Referred to Fig 4). Generally, the steeper the slope of the three-dimensional plots was, the greater the influence of factors on the response value was. It is clear that hot-pressing pressure and hot-pressing temperature were

interdependent on each other. The transverse tensile strength enhanced when the hot-pressing pressure was set at a low level and the hot-pressing temperature increased to a high level. When hot-pressing pressure was set at a high level (1.5 MPa), the impact of hot-pressing temperature was small.

The best hot-pressing conditions were obtained (112°C, 107 s, 0.94 MPa) by taking the first-order derivative of the best-fitted equation (Eq. 2 in S2). Under this condition, the measured value of

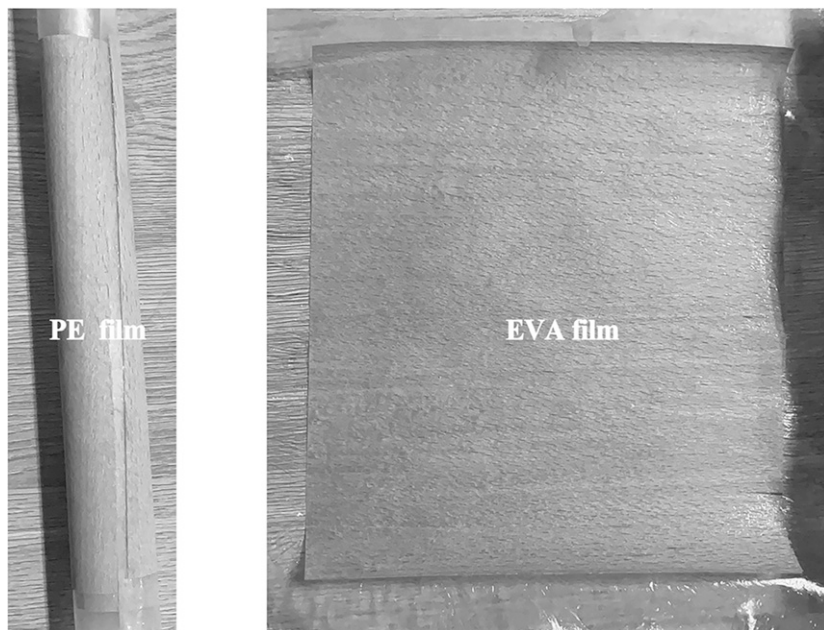


Figure 5. Curling deformation of PE film and EVA film reinforced veneer.

transverse tensile strength was 2.42 MPa. The error between the model and the theoretical value was 5.4%.

Effects of process conditions on the flexibility of EVA film reinforced veneer

The unmodified decorative veneer has poor flexibility and was easy to crack (S3). Its minimum steel rod diameter was greater than 14 mm, making it unsuitable to decorate special-shaped wood-based panels. When the decorative veneer was composited with EVA film, the veneer flexibility was effectively improved and no cracks were found on the veneer surface. This further confirmed the role of the mechanical interlock structure formed during the hot-pressing process. The function of this structure was similar to that of reinforcement in the soil-concrete structure, which was able to unload the bending stress and delaying the brittle fracture of slice veneer (Fang *et al.* 2021). The selected process conditions in Table 1 have little influence on the flexibility. The minimum steel rod diameter of the 17 groups of samples was kept between 4 mm to 6 mm. This was probably

attributed to the mild processing conditions. The selected EVA film (0.1 mm) was enough to form a continuous composite interface with the veneer after melting and penetration (Fig 3a). At the same time, the appropriate hot-pressing temperature and time were conducive to prevent the veneer embrittlement.

Due to the significant difference of thermal expansion coefficients between plastic and wood, curling deformation has become one of the key technical issues that limited the application of plastic film reinforced flexible veneer. Previous studies showed that high hot-pressing temperature was the most important factor to cause the curling deformation of PE film reinforced flexible decorative veneer (Peng and Zhang 2018). Interestingly, the curling degree of EVA film reinforced flexible veneer was much smaller than that of PE film reinforced flexible veneer. Figure 5 clearly showed the difference in curling deformation between the two flexible veneers. The hot-pressing temperature used for producing both flexible veneers was 130°C. This may be attributed to the polar component in EVA. It was much easier for EVA to form a good bonding interface with veneer at certain pressure

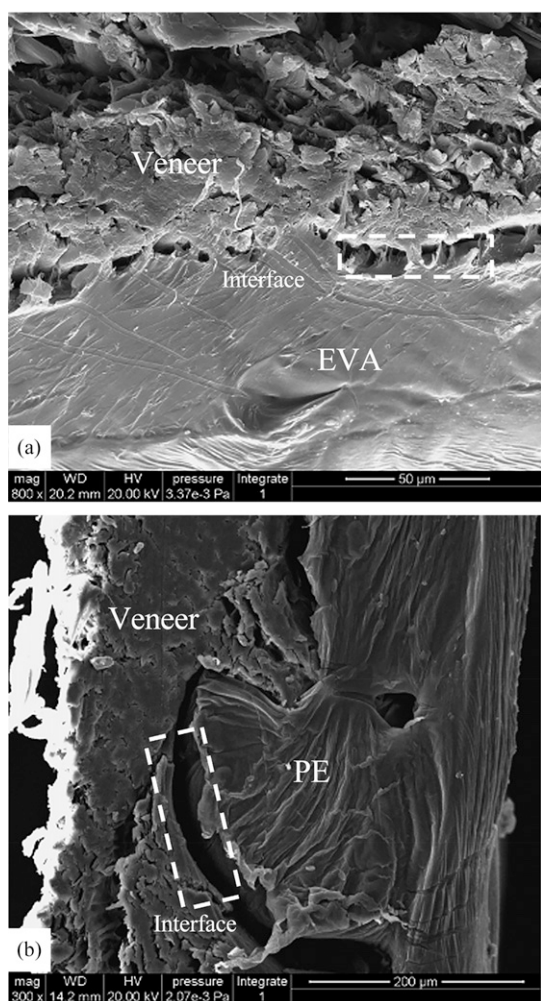


Figure 6. The interface between decorative veneer and (a) EVA film, (b) PE film.

and temperatures. As shown in Fig 6a, a large number of small nails formed at the bonding interface of EVA reinforced veneer, which can inhibit the shrinkage of EVA and alleviate the difference in the shrinkage displacement of the two different materials. However, the interface between PE and veneer was very smooth with a large gap (Fig 6b).

Overlaying performance of EVA film reinforced flexible veneer

The EVA film reinforced flexible veneers prepared under the optimized conditions were subsequently

used to decorate MDF panels. In this process, no additional adhesive was needed, since the EVA can be remelted and served as adhesive.

The immersion peel test of the decorated panel was conducted to evaluate the water resistance of the adhesion between EVA film reinforced veneer and MDF substrate. Six specimens (75 mm × 75 mm) for each condition were cut from three replicate veneers. Type II immersion peel test (soaking at 63°C for 3 h, then drying at 63°C for 3 h) was firstly performed on each sample. As presented in Fig 7, the average immersion peel strength was around 2 mm to 3 mm when the decorative MDF panels prepared at 95°C and 100°C, far exceeding requirements in GB/T 15104-2006 (no larger than 25 mm). With the increase of hot-pressing temperature, much more EVA can penetrate into the MDF substrate, and the bonding interface between EVA and MDF was enhanced. No delamination appeared in those veneer-covered specimens that were fabricated at 105°C and 110°C. However, due to the low softening temperature of EVA, and the relatively poor compatibility between EVA and MDF, the bonding between EVA film reinforced veneer and MDF substrate could not stand a hot and humid environment. Severe delamination occurred after the panels were soaked in boiling water for 4 h. The veneer completely separated from the MDF substrate, and most of the EVA remained on the MDF panel (S4).

Figure 7 showed the bonding strength between the flexible decorative veneers and substrate MDF panels. When the hot-pressing temperature reached the melting point of EVA, the EVA remelted and partially penetrated into the porous structure of MDF, forming a mechanical interlock with MDF. All of the decorated panels had surface bonding strength higher than 0.4 MPa, meeting the lowest strength requirement in GB/T 15104-2006. Figure 8 clearly showed that the glue nails and mechanical interlocking structure formed in the EVA film reinforced veneer in the first stage were not easy to be damaged. The amount of EVA that can be penetrated into MDF in the second melting process was limited. The maximum surface bonding strength was obtained when the hot-pressing temperature was 105°C. Further increasing the

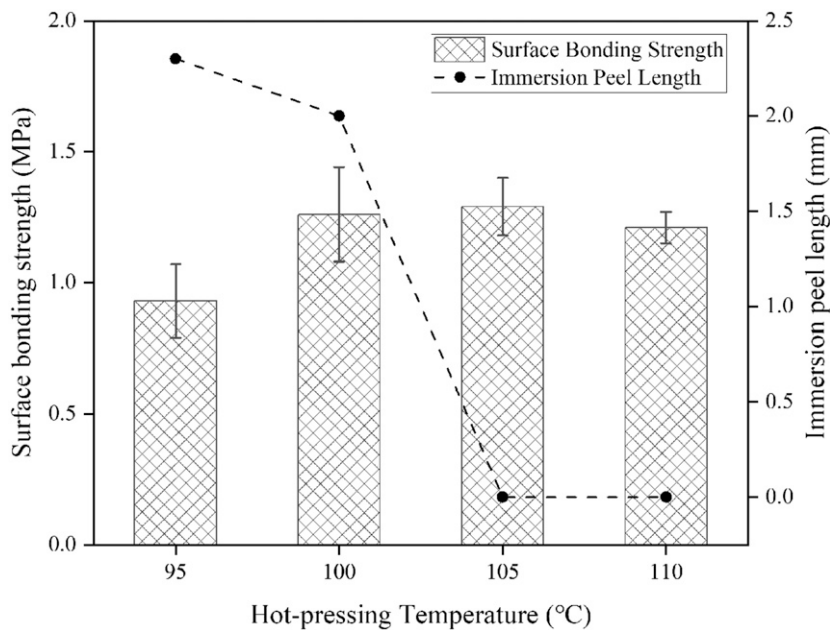


Figure 7. Surface bonding strength and immersion peel length of EVA film reinforced veneer decorated MDF panels as a function of hot-pressing temperature.

hot-pressing temperature, although the fluidity of EVA was improved, it was not enough to penetrate into the deeper part of MDF under a certain hot-pressing time and hot-pressing pressure. On the contrary, they may accumulate on the bonding interface, resulting in a decrease in surface bonding strength.

CONCLUSIONS

Decorative veneer with high strength and good flexibility has been successfully manufactured by composited with EVA film. It is attributed to the mechanical interlock structure formed during the hot-pressing process. Hot-pressing temperature, hot-pressing time and the interactions between hot-pressing temperature and hot-pressing pressure have a serious impact on the transverse tensile strength of the specimens. The optimum conditions for preparing the EVA film reinforced veneer are as follows: hot-pressing temperature 112°C, hot-pressing time 107 s, and hot-pressing pressure 0.94 MPa. The minimum steel rod diameter of the flexible veneer is kept between 4 mm to 6

mm, which is not affected by the process conditions. Because of the existence of polar components in EVA, curling deformation was not serious in EVA flexible veneer. EVA film reinforced flexible veneer can be directly used to decorate wood-based panels without adding adhesive. The maximum surface bonding strength of the flexible veneer decorated MDF panel was obtained when the hot-pressing temperature is 105°C. All of the decorated MDF panels can pass the type II immersion peel test and can be used in a humid environment.

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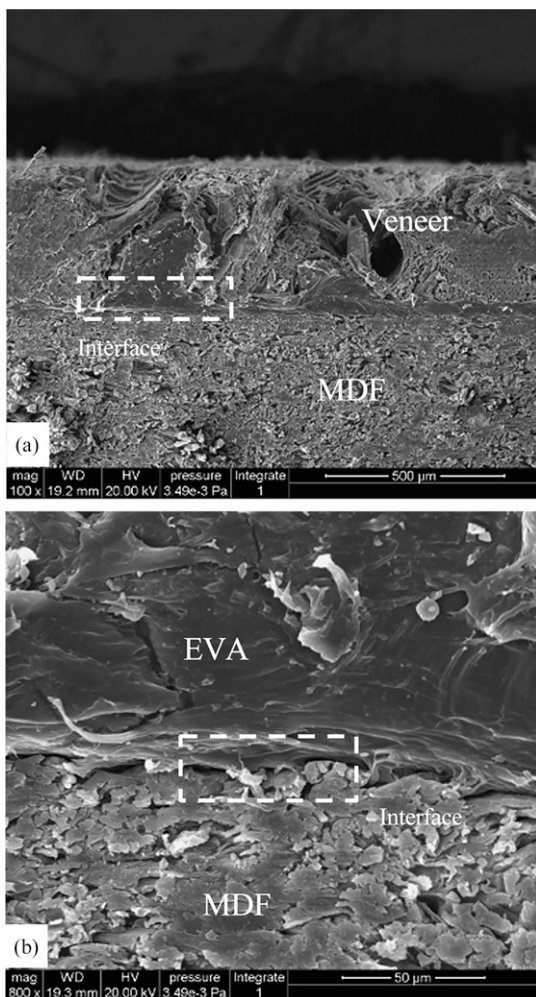


Figure 8. Bonding interface microstructure of the decorated MDF panel.

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