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# Influence of silane surface modification of veneer on interfacial adhesion of wood–plastic plywood



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#### ABSTRACT

In this study, wood–plastic plywood was fabricated with high density polyethylene (HDPE) film and poplar veneer by hot-pressing. To improve the interfacial adhesion between the wood veneer and HDPE film, silane A-171 (vinyltrimethoxysilane) was used to treat the surface of poplar veneer by spraying. The effects of silane agent on the veneer surface properties as well as the physical–mechanical performance of wood–plastic plywood were evaluated. The adsorption of several prehydrolyzed alkoxysilanes onto the veneer surface and the existence of a covalent bonding between the wood veneer and silane agent were confirmed using FTIR, XPS and contact angle. Silane surface treatment resulted in enhancement of shear strength and water resistance. When one layer HDPE film was used as adhesive, it caused 293.2% increase in shear strength, 34.6% and 40.8% reduction in water absorption and thickness swelling, respectively. In addition, the wood failure also increased from 5% to 100% due to the silane modification. Dynamic mechanical analysis (DMA) results showed that treated plywood have higher storage modulus, lower tan  $\delta$  peak value with respect to untreated plywood. Experimental results of interfacial morphology by SEM further revealed better interaction between silane A-171 treated veneer and HDPE film.

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#### 1. Introduction

The rising concern towards environmental issues and, on the other hand, the need for more versatile plywood has led to increased interest in development of formaldehyde-free wood adhesives [1–3]. Plastic film is innovated as new generation formaldehyde-free adhesives for plywood (wood-plastic plywood) since it is inexpensive, relatively tough and has good dimensional stability when exposed to moisture [4]. Compared with conventional urea-formaldehyde (UF) plywood, wood-plastic plywood offers several advantages such as relatively high bending strength, low water absorption and thickness swelling, and most importantly environmental superiority [5]. However, the usually hydrophilic wood has inherently low compatibility with hydrophobic plastic, especially polyolefin plastic like polypropylene (PP) and polyethylene (PE) [6,7]. The incompatibility may cause insufficient wetting of plastic on the surface of wood veneer and result in weak interfacial adhesion between the two phases, which will adversely affect their mechanical properties and therefore make these materials can only find way in some nonstructural applications such as floors, door frames and interior parts of automobile.

In recent years, various methods have been studied to improve the interfacial adhesion between wood and plastic by modifying the wood surface, including physical or chemical means according to the way they modify the wood surface [8–15]. Frequently used approaches were by adding chemical coupling agents. Keener et al. [16] reported that, use of maleic anhydride grafted polyethylene coupling agent at 3% loading can double the tensile strength and triple the impact properties compared to non-coupled blend of wood and polyethylene. Karmarkar et al. [17] have also reported improved mechanical properties by using a novel compatibilizer with isocyanate functional group. Goriparthi et al. [18] used alkali, permanganate, peroxide and silane to promote adhesion between the jute fiber and polylactide matrix and have also shown that the adhesion between the natural fiber and the matrix played an important role in the final mechanical properties, thermal stability as well as the wear performance of the composites. There is, however, little knowledge about treatment on wood veneers to promote their interfacial adhesion with nonpolar thermoplastic polymer [8,13,19] because most of the work reported, so far, has been concerned with the use of thermoplastic polymer as matrix for wood fiber (wood flour), rather as wood adhesives for wood veneers.

Silanes are recognized as efficient coupling agents extensively used for improving fiber-matrix bonding [20–22]. The

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improvement in mechanical properties is believed to be due to their bifunctional structures, which can create a chemical bridge between the wood fiber and plastic matrix [23–25]. Since both wood fibers and wood veneers bear reactive hydroxyl groups, these methods have also been of interest in applying on wood veneer for wood–plastic plywood except that the application methods may be a little bit different and their effects on the eventual properties of veneer or plywood were unknown. Therefore, in this research work, silane A-171 (vinyltrimethoxysilane) was used to treat the wood veneer by spraying to improve the compatibility between polar veneers and HDPE films and to develop wood–plastic plywood for exterior applications. The influences of silane treatment on veneer surface properties such as wettability, surface chemistry, and their relationship with physical–mechanical properties of wood–plastic plywood were established.

#### 2. Material and methods

#### 2.1. Materials

Poplar veneers were purchased from Zuo gezhuang (Hebei, China) with dimensions of  $300 \times 300 \times 1.6$  mm and their moisture content was conditioned to 6–8%. The veneer has a pH value of 5.62 and a average roughnees (*R*<sub>a</sub>) of 4.81 µm (parallel grain) and 10.82 µm (across grain). HDPE film provided by Environment-Protecting Co., Ltd. (Shanghai, China) has a thickness of 0.06 mm and a density of 0.92 g/cm<sup>3</sup>. The plastic film was cut into the same dimension with veneer.

Silane A-171 (vinyltrimethoxysilane) provided by Guangzhou Zhongjie Chemical Technology Co., Ltd. and dicumyl peroxide (DCP) purchased from JK Chemical were used as modifying chemicals for the poplar veneers.

#### 2.2. Surface treatment

Silane solutions were prepared by mixing with ethanol and deionized water in 9:1 ratio at a pH value of 3–3.5. 2% silanes (wt% compared to the veneers) and a tiny part of DCP (0.05% of the HDPE dosage) were added in the solution until the silane solution concentration reached 4%. The silane was hydrolyzed for 1 h in the solution. After spraying the silane solution on the veneer samples, the veneers were air-dried for at least 24 h and finally treated at 120 °C in the convection oven for 2 h.

#### 2.3. Infrared spectroscopy (FTIR)

The surface chemistry of treated poplar veneer was obtained using Thermo Scientific Nicolet iN 10 FT-IR Microscope (Thermo Nicolet Corporation, Madison, WI) equipped with a liquid nitrogen cooled MCT detector. All sample spectra were recorded in the  $4000-650 \text{ cm}^{-1}$  range with a resolution of  $4 \text{ cm}^{-1}$  and 128 scans. Three replicated measurements were carried out for each sample.

#### 2.4. X-ray photoelectron spectroscopy (XPS)

The XPS spectra of the untreated and silane-treated poplar veneers were recorded on a Thermo ESCALAB 250 Xi spectrometer using Al  $K\alpha$  ( $h\nu$  = 486.6 eV) at 150 W. Binding energies were calibrated with C<sub>1s</sub> of 284.8 eV and the analysis of the spectra was performed using a commercial curve fitting software.

#### 2.5. Contact angles of poplar veneer

Wood veneer was cut into small strips ( $70 \times 10 \times 1.5$  mm). Contact angles (CA) of wood veneer were measured with OCA 20 (Data Physics Instruments, Germany) using distilled water. The CA was determined for each image by digital image analysis software. The room temperature and relative humidity were 25 °C and 50%, respectively. Ten samples were tested for both untreated and treated veneers with 3 drops on each sample.

#### 2.6. Scanning electron microscopy (SEM)

Interface morphology of the plywood samples were first coated with gold and then examined using a S-4800 scanning electron microscope at two different magnification factors ( $200 \times$  and  $1000 \times$ ).

#### 2.7. Dynamic Mechanical Properties (DMA)

DMA measurements were conducted on the sandwich specimens of about  $60 \times 12 \times 3 \text{ mm}$  (length  $\times$  width  $\times$  thickness) in three-point bending mode at a span of 50 mm on a TA Instruments Q2980 at a frequency of 1 HZ and at constant amplitude of 0.03 mm. Temperature ramp experiments were performed at heating rate of 3 °C/min from room temperature to 200 °C with triplicates.

#### 2.8. Plywood Fabrication and testing

Five-layer wood–plastic plywood was firstly assembled with HDPE film between every two veneers (treated or untreated). The plywood was prepared with opposing tight-side and loose-side veneers in all the glue lines, and was stacked with the grain directions of the two adjacent veneers perpendicular to each other. The HDPE film dosage was varied from 1 layer to 4 layers. Wood–plastic plywood was hot-pressed at 1 MPa pressure and 160 °C for 8 min followed by a cold-press stage that were performed at room temperature for 5 min, which was used to reduce the distortion and stress of the plywood.

Physical-mechanical properties commonly taken into consideration in the general usage areas of plywood panels were investigated according to Chinese National Standard GB/T 17657-1999 [26]. Thickness swelling (24 h-TS), water absorption (24 h-WA), shear strength (Immersion in boiling water for 4 h, then drying at  $63 \pm 3$  °C in the drying oven for 20 h, then immersion in boiling water for 4 h) were evaluated, respectively.

Mechanical properties were measured using a Multi-Function Mechanical Testing machine (Jinan Shijin Co., Ltd, 10 kN), with a crosshead speed of 5 mm/min. Before testing, all samples were conditioned at  $25 \,^{\circ}$ C and 65% humidity for 1 week.

#### 3. Results and analysis

#### 3.1. Infrared spectroscopy (FTIR)

In order to confirm the chemical reaction between silane coupling agent and poplar veneer, the spectra in the 4000–650 cm<sup>-1</sup> range, for both of the untreated and treated veneers are shown in Fig. 1. The presence of a new absorption bands in the regions of 765–770 cm<sup>-1</sup> in the treated sample can be seen in curve *b* of Fig. 1, which can be identified as -Si-C symmetric stretching bond [24] formed due to the condensation of silanols during the hydrolysis process. Also, a slight increment at approximately 1100 cm<sup>-1</sup> in treated plywood should be noted. The growth of this band could be attributed to the formation of covalent -Si-O-C- bond [24,27] as a result of the grafting reaction between silanols and wood veneer under heating condition.

#### 3.2. XPS

Fig. 2a and b shows the X-ray photoelectron spectroscopy of electron intensity as a function of binding energy for untreated

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