



Contents lists available at ScienceDirect

# Construction and Building Materials

journal homepage: [www.elsevier.com/locate/conbuildmat](http://www.elsevier.com/locate/conbuildmat)

## A novel application of silicone-based flame-retardant adhesive in plywood

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### HIGHLIGHTS

- SI adhesive was used to replace conventional PU adhesive to provide a flame-retardant adhesive for plywood.
- SI/plywood showed high flame retardancy and thermal barrier efficiency as compared to PU/plywood.
- SI adhesive was also reinforced with CF or GF to prepare composite plywood with improved fire performance.
- SI/GF/plywood exhibited the most effective fire barrier among all plywood types.

### ARTICLE INFO

#### Article history:

Received 23 May 2018

Received in revised form 18 August 2018

Accepted 31 August 2018

#### Keywords:

Flame-retardant adhesive

Silicone-based adhesive

Plywood

Shear strength

Fire performance

Thermal barrier

### ABSTRACT

A silicone-based elastomer filled with vinyl-silane treated aluminum hydroxide was used to replace conventional polyurethane-based adhesive to provide a flame-retardant adhesive for plywood. The shear strength and fire performance of such a silicone-based (SI) adhesive glued plywood (SI/plywood) were investigated and compared to those of the polyurethane-based (PU) adhesive glued plywood (PU/plywood). The shear strength of the SI/plywood [(0.92 ± 0.09) MPa] was about 63% lower than that of the PU/plywood at room temperature, but it was less sensitive to water (62% reduction for the PU/plywood and 30% reduction for the SI/plywood after hot-water immersion at 63 °C for 3 h). The fire performance of plywood was assessed by a simulated match-flame ignition test (Mydrin test), lateral ignition and flame spread test, cone calorimetry, and thermocouple measurements. With a higher burn-through resistance and thermal barrier efficiency, and lower flame spread and heat release rate, the SI/plywood exhibited a superior fire-resistance and reaction-to-fire performance and improved fire-resistance as compared to the PU/plywood. The SI adhesive generated an inorganic protective layer on the sample surface that visibly suppressed glowing and smoldering of the plywood during combustion. The SI adhesive was also combined and reinforced with cellulosic fabric (CF) or glass fabric (GF) to prepare composite plywood (SI/CF/plywood and SI/GF/plywood) with improved fire performance. The cone calorimetry and thermocouple measurements indicated that the use of CF or GF in SI/CF/plywood and SI/GF/plywood, respectively, suppressed the delamination and cracking of the composite plywood and promoted the formation of an effective thermal barrier during smoldering and flaming combustion. Particularly, the SI/GF/plywood exhibited the most effective fire barrier with no crack formation, and the lowest heat release rate among the plywood types investigated in this study.

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### 1. Introduction

Wood-based products such as plywood, fiberboard, particleboard, laminated veneer lumber, and wood-plastic composites, provide a substitute for solid wood while retaining the requisite structural properties with favorable performance and low cost.

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They have been widely used in residential, commercial, and industrial buildings [1,2]. In North America, most residential housing and a significant portion of low-rise construction are built using wood-based products [3]. Meanwhile, mid-rise ( $\geq 6$  stories) and high-rise ( $\geq 12$  stories) timber constructions are becoming more common [4,5]. The replacement of non-flammable materials (e.g., concrete and steel) with wood-based products, which are intrinsically flammable, increases the fire load and fire hazard for these buildings. Therefore, there has been a surge in research in North America focused on assessing and improving the performance of wood-based products in structural fires [6–12], which is resulting in undated standards for the fire performance of engineered timber products specifically to address delamination [13].

Two key aspects of fire performance are fire-resistance and reaction-to-fire [14]. Fire-resistance refers to the ability of a material or system to withstand exposure to high temperatures based on thermal insulation and mechanical integrity evaluations. Reaction-to-fire defines how a material or system will contribute to fire development and refers to measurements including ignitability, energy released by combustion, and flame spread. Delamination in engineered timber drastically deteriorates fire-resistance and reaction-to-fire of the wood product. It directly affects the structural integrity of the product (due to the failure of the adhesive) and induces an increase in flame spread and heat release rate (due to the detachment of superficial charred insulating layer and consequent exposure of the virgin underlying material to direct flame impingement).

Three methods are commonly employed to provide wood-based products with improved fire-resistance and reaction-to-fire: chemical impregnation [2,15–18], incorporation of flame retardants into the adhesive [19–22], and flame-retardant coatings [23–27]. For chemical impregnation, the most widely used flame-retardant chemicals for treating wood-based products are inorganic salts that contain elemental phosphorus or boron. However, phosphates usually pose an adverse effect on the mechanical properties of wood products. Moreover, phosphates easily leach out due to their poor water resistance [18,28]. Boron compounds are not recommended due to their hygroscopicity that might affect the dimensional stability of wood [29,30]. Meanwhile, these chemicals could also leach out and be harmful to the environment and human health [31]. For the second method, the incorporation of flame retardants inevitably increases the viscosity and curing time, and decreases the bond strength of the adhesive [21]. Flame-retardant coatings, particularly intumescent coatings, are a convenient and effective way to decrease the flammability of the substrate materials, but they can be affected by aging and mechanical abrasion/impact [23,27]. Coatings also affect the aesthetics and appearance of wood. Therefore, there is a need for flame-retardant technologies for wood-based products that are durable, effective, environmentally friendly and preserve the wood aesthetics.

Silicones have been successfully used in the field of flame retardancy. Thanks to their high thermal stability, minimal sensitivity to external heat flux, low heat release rate, and low toxic gas generation during combustion [32–34]. Bourbigot et al. [35] used a phenyl-branched silicone coating as a thermal barrier on the backside of steel. They found that the silicone-based coating performed an effective fire barrier. The backside temperature of the coating protected steel was 130 °C lower than that of the non-protected steel in a fire-resistance test (the coating was exposed to an open flame around 1100 °C). They also reported that the chain length and crosslinking density of the silicone-based coatings affected their thermal stability and barrier performance (i.e., ability to prevent burn-through and thermally shield the substrate) [36]. Our previous research [37] showed that a silicone-based backcoating drastically increased the fire performance of the cellulosic fabrics.

The backcoating imparted the cellulosic fabrics with an advantageous combination of flaming/smoldering ignition resistance without affecting the original color and general appearance of the fabric's face. Silicone-based polymers can be used as adhesives and exhibit high flexibility, wetting capability, excellent chemical and weathering resistance, a relatively low curing temperature, and a benign toxicological profile [38–41]. More importantly, silicone-based adhesives are expected to be beneficial in terms of delamination resistance in wood engineered products due to their high thermal stability and high yield of inorganic thermally stable residue [42].

In this work, we investigate the use of a silicone-based elastomer filled with vinyl-silane treated aluminum hydroxide to increase delamination resistance and fire performance of plywood. The shear strength and fire performance of such a silicone-based adhesive glued plywood were investigated and compared to those of a conventional polyurethane-based adhesive glued plywood. The use of cellulosic or glass fabrics was also explored to further improve the fire performance of the resulting composite plywood.

## 2. Materials and methods<sup>i</sup>

### 2.1. Materials

All materials were used as received unless otherwise indicated. A two-component (a base and a curing agent) silicone-based elastomer crosslinked by platinum-catalyzed hydrosilylation (Sylgard 184) was purchased from Dow Corning (USA). A vinyl-silane modified aluminum hydroxide (VSATH) (Apyral 40 VS1) was provided by Nabaltec AG (Germany). A conventional ethyl acetate (Analytical reagent) was obtained from Macron Fine Chemicals (USA). The one-component polyurethane-based adhesive (Original type) for wood was purchased from Titebond (USA). The wood used in this work was scroll saw veneer sheets of aspen [*Populus grandidentata*, 1.6 mm thick, density of  $(0.526 \pm 0.002)$  g/cm<sup>3</sup>] and purchased from Ocooch Hardwoods (USA). The cellulosic fabric (CF) was 100% cotton [white plain weave of (19–33) threads/cm<sup>2</sup>, with an areal density of  $(115 \pm 1)$  g/m<sup>2</sup>]. The glass fabric (GF) was Style 104 [ECD 900 1/0, with an areal density of  $(19.0 \pm 0.1)$  g/m<sup>2</sup>] and supplied by Hexcel (USA).

### 2.2. Sample preparation

VSATH (20.43 g) was dispersed in Sylgard 184 base (10 g) and ethyl acetate (10 g) for 5 min by using a bladeless mixer (SpeedMixer, Flack Tek Inc, USA) at 2500 rotations/min. Then the curing agent (1 g) was added and mixed for another 1 min. The resulting silicone-based (SI) adhesive was applied by brush to both sides of a veneer specimen. Then, the adhesive-coated wood veneer was stacked between two uncoated wood veneers with the grain directions of two adjacent veneers perpendicular to each other (Fig. 1a). For the composite plywood containing CF (SI/CF-plywood) or the composite plywood containing GF (SI/GF-plywood), the CF or GF were used in combination with the SI adhesive (Fig. 1b). A single fabric layer was placed between two adjacent veneer layers. The adhesive was applied by brush to the veneer above and underneath the fabric. All the SI adhesive glued samples were prepared in accordance with such a methodology followed by hot-pressing (M-12-1, Grimco, USA) at 120 °C for 2 h with a pressure of 5 MPa. Complete evaporation of acetate occurred during curing and lead to a final content of VSATH in the SI adhesive of 65% by mass.

Samples glued by the conventional polyurethane-based (PU) adhesive were prepared with the same protocol, but the hot-pressing was performed at 80 °C. Before hot pressing, the mass ratio between adhesive and wood veneer in the PU adhesive and SI adhesive glued plywood samples was  $(20 \pm 1)$  % and  $(60 \pm 1)$  %, respectively (Table 1). Some adhesive was removed during the hot pressing and, as a result, the content of adhesive in the plywood decreased. The final plywood composition and the adhesive to wood mass ratio (after hot pressing) for all the plywood types are shown in Table 1. The presence of CF or GF did not significantly affect the content of adhesive in the samples after hot pressing.

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