THERMAL PROPERTIES OF ACACIA MANGIUM CROSS LAMINATED TIMBER AND ITS GLUELINES BONDED WITH TWO STRUCTURAL ADHESIVES

Norwahyuni Mohd Yusof 1, Paridah Md Tahir 1*, Lee Seng Hua 1*, Fatimah Athiyah Sabaruddin 1, Redzuan Mohammad Suffian James 1, Mohd Asim Khan 1, Lee Ching Hao 1, Adlin Sabrina Muhammad Roseley 2

1 Institute of Tropical Forestry and Forest Products, University Putra Malaysia, 43400 UPM Serdang, Selangor Darul Ehsan, Malaysia.
2 Faculty of Forestry and Environment, University Putra Malaysia, 43400 UPM Serdang, Selangor Darul Ehsan, Malaysia.

* Corresponding authors: parida@upm.edu.my; lee_seng@upm.edu.my

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ABSTRACT

The properties of CLT can be affected by the type of adhesives used. The thermal properties of the adhesive that joins the timber together is essential to determine the thermal endurance of the CLT product. In this study, two types of adhesives were used to join the cross laminated timber (CLT) manufactured from Acacia mangium namely phenol resorcinol formaldehyde (PRF) and one component polyurethane (PUR). The thermal properties of the adhesives, A. mangium wood and the gluelines were determined via Thermogravimetric Analysis (TGA) and Dynamic Mechanical Analysis (DMA) tests. The TGA test showed that PRF adhesive had higher degradation temperature at 530 °C compared to PUR adhesive at 430 °C. Meanwhile, the PRF adhesive as a glueline in CLT also showed better thermal resistance where a higher amount of residue of 20.94 % was recorded at temperature up to 900 °C compared to PUR glueline with 18.26 % residue. The integrity of the CLT over temperature were determined via DMA test and the results showed that PRF adhesive as glueline had superior properties, indicating better interfacial bonding with the woods.

Keywords: Acacia mangium, dynamic mechanical analysis, one component polyurethane, phenol resorcinol formaldehyde, thermogravimetric analysis.
INTRODUCTION

Cross-laminated timber (CLT) is a wood panel composite that joining at least three layers of solid-sawn timber by using adhesive (Sutton et al. 2011). This material has been considered as high demand products that used as a constructional material for buildings. It enables a rapid installation for wall and floor structure that suitable for most finishes (Van De Kuilen et al. 2011). According to Dieste et al. (2019), CLT is a highly value-added products compared to that of the other wood products. The mass production of CLT is formed in a similar way to “glue-laminated timber” beams using permanent adhesives which the limitations such as knots, checks, splits, warping and weathering can be removed to reduce variability and enhanced its structural properties. Almost all the studies of CLTs reported are constructed from softwood species such as spruce, larch, white fir, silver fir, Douglas fir, pine and yellow poplar with density ranging from 350 kg/m³ to 700 kg/m³ (Engineering Toolbox, 2004). For instance, Wang et al. (2018) fabricated CLT from spruce-pine-fir lumber pieces to study the effects of edge-gluing and gap size in the cross layers.

The rapid industrialization and increasing number of populations in the world causes depleting of forest wealth at a rapid rate. The issue has urged the manufacturers to make use of fast grown plantation species for various timber applications (Shukla 2019). To relieve the pressure of the continuous extraction of logs from local natural forest, the establishment of plantation forests is a matter of utmost urgency. In 2005, a Forest Plantation Programme has been implemented by Ministry of Plantation Industries and Commodities (MPIC) where a total of 375000 ha of forest plantation has been targeted to be developed at the end of 2020. The programme aims to ensure the sustainability of the raw materials supply for the domestic
timber industry. Acacia spp. (mangium /hybrid) and rubberwood (Timber Latex Clone) are the two major species out of nine selected species under this programme.

A. mangium is one of the fast-growing and sustainable species for potentials timber production. It has a wood density ranging from 420 kg/m³ to 483 kg/m³ for green soaked volume and 500 kg/m³ to 600 kg/m³ in dry condition (Kasim et al. 2014). In another study, the density of A. mangium was reported to fall within a range of 290 kg/m³ to 675 kg/m³. The density increases as the age of tree increased from 2 years to 20 years (Nordahlia et al. 2013). The density of wood is the best method to determine the quality of wood and correlated to its product's strength and shrinkage (Nugroho et al. 2012; Miranda et al. 2007; Lim et al. 2003).

Wood is a renewable material and it has many applications. However, wood is easily degraded by sunlight, moisture and temperature due to the organic structure and the most important challenges of the components of wood is thermal decomposition of cellulose, lignin and hemicellulose at the low temperature (100 °C to 150°C) (Aydemir et al. 2016). Hence, wood have different degradation points and is highly dependent on their specific chemical compositions.

The selection of the adhesives to manufacture the CLT is very important as it needs to meet the characteristics and the application of the CLT itself. Polyurethane (PUR) adhesive glue is well known as a formaldehyde-free adhesive for exterior grade structure application for GLULAM and finger-joint in several European countries (Richter et al. 2006). Other than that, the thermal properties of the CLT are also very important but there is very limited information on this topic been reported. According to Asim et al. (2018), excellent thermal and fire-retardant behaviour of phenol resorcinol formaldehyde (PRF) adhesive glue allowed
it to be used in building structural materials and automobile industry. Some studies have been conducted to compare the performance of PRF and PUR adhesives in CLT fabrication. Norwahyuni et al. (2019a) compared both PRF and PUR adhesive in bonding CLT from A. mangium wood. The authors reported that PRF adhesive performed better than that of PUR adhesive as it led to higher shear bond strength and wood failure percentage. In another study, Norwahyuni et al. (2019b) evaluated the mechanical and physical properties of A. mangium CLT bonded with PRF and PUR. The results revealed that the CLT bonded with PRF adhesive exhibited higher mechanical properties compared to that of the PUR-bonded CLT. Nevertheless, information on the thermal stability of PRF and PUR in CLT manufacturing are rather limited.

Therefore, the objective of this study is to evaluate the thermal properties of neat PRF and PUR adhesive, A. mangium wood as well as the glueline formed between wood and adhesive. The glueline formed between two adjacent layers of wood is important as it might impart some thermal resistance to the wood itself. Through the thermal studies, the thermal degradation, weight loss, final residue, decomposition temperature of each component samples was assessed based on peak DTG curves, storage modulus, loss modulus and damping factor of the samples.

MATERIALS AND METHODS

Manufacturing Cross Laminated Timber (CLT) species

Twenty-year-old A. mangium wood with a density of 673 kg/m³ and moisture content of 12 % ± 3 % was obtained from a local lumber mill located at Bukit Rambai, Melaka,
Malaysia. The wood was sawn, trimmed and planed into 1000 mm long by 70 mm wide and 18.2 mm thick lumber. In this study, a 3-layer CLT of 1000 mm × 280 mm × 54.5 mm in size was produced by gluing three pieces of lumbers parallel and perpendicular to each other with edge bonding with 90° alternating transverse CLT layers. Boards were glued using two types of adhesive, namely phenol resorcinol formaldehyde (PRF) and one component polyurethane (PUR). Both types of adhesives were applied to the samples at a spreading rate of 250 g/m² within a short time to avoid possible oxidation and dimensional instability. The assemblies were then subjected to a pressure of 1.5 N/mm² at 30 °C for 90 minutes using a compressive machine. In the next stage, the laminated panels were conditioned at 65 % ± 5 % RH and 20 °C ± 2 °C for 2 weeks before cutting them into specimens for properties evaluation. Two types of adhesives were used in this study i.e., phenol resorcinol formaldehyde (PRF 1734 AkzoNobel) and one component polyurethane (1C-PUR or PUR, Jowapur 687,22). Hardener 2734 (commercial code) was also used in the preparation of PRF at a ratio of 100 to 25 parts by weight of PRF to hardener.

Characterization of the samples

Thermogravimetric Analysis (TGA)

Thermal stability of both PRF and PUR adhesives, *A. mangium* wood and glueline formed between wood and adhesives after pressing were characterized using Thermogravimetric analyser (TGA Q 500 TA Instrument, USA) at Institute of Tropical Forestry and Forest Product (INTROP), Universiti Putra Malaysia (UPM). In order to eliminate the effect of initial mass, 10 mg of sample was used for each experiment and placed in an alumina crucible. Non-isothermal TGA was conducted with the temperature raised from 10 °C to 900 °C with a rate of 10 °C/min in an oxygen atmosphere. The gas was purged at a constant flow rate of 30 mL/min. Three replications were used for every adhesive type tested.
A total of 15 specimens (3 x [PRF and PUR adhesives + A. mangium wood + glueline formed by both adhesives]) were tested in this study.

**Dynamic mechanical analysis (DMA)**

Dynamic mechanical analysis (DMA) was executed to determine the viscoelastic behaviour of specimens from 30 °C to 150 °C with a heating rate of 5 °C/min and controlled sinusoidal strain. DMA test was performed by employing TA (DMA Q 800) instrument at INTROP, Universiti Putra Malaysia, and operating in a three-point bending mode with 1 Hz oscillation frequency under controlled amplitude. The specimens having dimensions of 60 mm × 12 mm × 6 mm (l × w × t) were used for the testing. For DMA, only PRF and PUR gluelines were tested with three replications each. Therefore, a total of 6 specimens were evaluated.

**RESULTS AND DISCUSSION**

**Thermogravimetric Analysis (TGA)**
Heat resistance of a material is one of the most important criteria's in any building construction. Since wood is combustible, the application of wood as building material becomes very limited. Thus, thermal evaluation is an essential method to determine the capability of the material to resist heat. In general, TGA is used to observe the material's thermal stability and degradation behaviour. TGA analysis on the neat adhesive (PRF and PUR), glueline between wood and the adhesive and *A. mangium* wood are demonstrated in Figure 1 and Table 1.

**Figure 1:** TGA curve of PRF, PUR, PRF glueline, PUR glueline and *A. mangium* wood.

During thermal degradation, drying of free water and major mass loss due to devolatilization and thermal debonding has been observed. All samples experienced a slight weight loss at temperature below 100 °C as shown in Figure 1 due to evaporation or dehydration of the water molecules (Sanyang *et al.* 2015; Nadirah *et al.* 2012; Johar *et al.* 2012). Significant weight loss (26.8 %) was observed for PRF adhesive glue at a temperature less than 100 °C due to the high amount of volatile free formaldehyde and phenol other than free water removal. This finding was aligned with the findings from previous studies done by Asim *et al.* (2018) and Liu *et al.* (2017). Both adhesives showed similar initial degradation temperature which occurred at 220 °C.
However, PUR adhesive showed a higher weight loss of 74.40% compared to PRF adhesive with the amount of weight loss of 47.92% during the main degradation process. This finding could be attributed to the availability of the thermally unstable urethane bond in PUR adhesive that leads to rapid degradation (Lee et al. 2002).

**Table 1:** Thermogravimetric analysis (TGA) results.

<table>
<thead>
<tr>
<th>Sample</th>
<th>Initial degradation temperature (°C)</th>
<th>Final degradation temperature (°C)</th>
<th>Weight loss (%)</th>
<th>Final residue at 900 °C (%)</th>
</tr>
</thead>
<tbody>
<tr>
<td>PRF adhesive</td>
<td>220</td>
<td>530</td>
<td>47.92</td>
<td>39.37</td>
</tr>
<tr>
<td>PUR adhesive</td>
<td>220</td>
<td>430</td>
<td>74.40</td>
<td>13.24</td>
</tr>
<tr>
<td>A. mangium wood</td>
<td>140</td>
<td>380</td>
<td>72.38</td>
<td>16.72</td>
</tr>
<tr>
<td>PRF Glueline</td>
<td>140</td>
<td>360</td>
<td>65.43</td>
<td>20.94</td>
</tr>
<tr>
<td>PUR Glueline</td>
<td>130</td>
<td>420</td>
<td>72.57</td>
<td>18.26</td>
</tr>
</tbody>
</table>

Meanwhile, the weight loss of the *A. mangium* wood sample starts to occur between 30 °C up to about 130 °C, due to hydrophilic nature of wood which resulted in high moisture absorption (Crespo et al. 2015; Angelini et al. 2009). The main degradation of *A. mangium* wood occurred at 140 °C up to 380 °C and this low thermal stability performance is attributed to the degradation of lignocellulosic components of the wood (hemicellulose, cellulose and lignin) (Manya et al. 2003). Higher lignocellulosic components caused a higher mass loss and lower initial degradation temperature (Lee et al. 2018; Lee et al. 2017). The first peak was attributed to the decomposition of hemicellulose, while the second peak can be attributed to the cellulose while degradation of lignin occurred at the wide temperature range and overlapping with the degradation of other components (Di Blasi 2008; Mészáros et al. 2004).

On the other hand, when both adhesives were applied on *A. mangium* wood (PRF glueline and PUR glueline), thermal degradation was expected to be similar to the degradation of pure *A.
Mangium wood. This is because only a small portion of the specimen's mass was being replaced by higher thermal stability adhesive glue. Although the use of adhesive does not improve the specimen's degradation temperature, the glue application slightly reduces the mass loss. PRF glueline specimen was reported to have better integrity structure at temperature range up to 900 °C with 20.94% of total residue compared to pure A. mangium wood and PUR glueline with the mass residue of 16.72% and 18.26%, respectively. This result could be attributed to good bonding properties between PRF adhesive and A. mangium wood compared to PUR adhesive.

Derivative thermo-gravimetric (DTG) analysis of both PRF and PUR adhesives, A. mangium wood and glueline of both PRF and PUR are shown in Figure 2. DTG analysis is used to study the rate of degradation of the materials up to a certain temperature (Ridzuan et al. 2016). Three peaks were observed for all specimens which is below 100 °C, 280 °C to 380 °C and 420 °C to 520 °C. A low first degradation peak responsible for free water removal except for PRF adhesive glue. A maximum decomposition rate at 4.83%/min for PRF was observed due to the presence of hydroxyl molecule in PRF adhesive glue (Asim et al. 2016). All specimens demonstrate the maximum rate of weight losses in the second peak except PRF adhesive glue. A. mangium wood showed a higher DTG value at the second peak among all the samples. Meanwhile, the application of adhesive glues (PRF and PUR) on A. mangium wood seems to reduce the rate of weight loss as part of the specimen’s mass replaced by higher thermal stability materials. No significance changes showed at the third region of DTG peaks for all samples.
Figure 2: DTG curve of PRF, PUR, PRF glueline, PUR glueline and A. mangium wood.

Dynamic mechanical analysis (DMA)

DMA is one of the most functional analysis to investigate the morphological and viscoelastic properties of a material. It helps to analyse some other parameters such as primary relaxations, storage or loss compliance, dynamic fragility, cross-linking density, creep compliance and dynamic viscosity (Ornaghi et al. 2012; Pistor et al. 2012; Joseph et al. 2010, Qazvini and Mohammadi 2005). It is a technique where small deformation is applied in a cyclic manner.

In this section, only the gluelines formed by PRF and PUR adhesives were tested and discussed. Figure 3 illustrates the storage modulus of PRF and PUR adhesive gluelines of CLT made by A. mangium wood. According to Saba et al. (2016), there are three regions found for viscoelastic materials, namely glassy region, transition region and rubbery region. High storage modulus value indicated the glassy region where the components are in tightly packed and drop as it reached the glass transition temperature ($T_g$). This is because slipping of polymer chain above $T_g$ reduces its modulus and performances. As the temperature increase, the component
tends to increase in mobility correspond to rubbery state transition (Jawaid and Khalil 2011; Hameed et al. 2007; Jacob et al. 2006). The investigation of various storage modulus with a temperature of CLT was found decrease with temperature increment. At low temperature, storage modulus PRF glueline is higher but close to PUR glueline, suggesting that PRF glueline has better stiffness but lower mobility and flexibility (Chartoff et al. 2009). This finding was synchronized with TGA results reported in the above section where PRF glueline has better interfacial bonding between wood and PRF glue. Besides, a sudden drop of storage modulus at about 100 °C for both gluelines represents glass transition temperature, $T_g$ zone. 

**Figure 3:** Storage Modulus of PRF glueline and PUR glueline.

Meanwhile, the loss modulus measurement is carried out on the energy dissipation response under reciprocate deformation of a material (Jawaid et al. 2012). Loss modulus of PRF and PUR adhesive glueline of CLT made by *A. mangium* wood are illustrated in Figure 4. Both curves at maximum value indicates maximum dissipation of mechanical energy at lower temperature and decreased dramatically at higher temperature due to the free movement of polymer (Saba et al.
From Figure 4, both gluelines demonstrated a wide peak at 60 °C to 100 °C and the $T_g$ observed for PRF glueline is slightly lower than PUR glueline. This is because the inter-molecular friction in the PRF glueline was higher and increases the dissipation of energy within CLT (Hameed et al. 2007). PRF glueline resulted in a higher loss of modulus due to the increased amount of energy needed to disentangle a better interfacial bonding.

**Figure 4:** Loss Modulus of PRF glueline and PUR glueline.

Damping factor determines the ability of material on absorbing energy at a range of temperature or frequency. Damping is important when designing advanced structural especially when vibration and noise control involved. Besides, it is used to study the fatigue life and impact resistance of structural materials and monitor damage (Melo and Radford 2005). Figure 5 depicts the damping factor curve trend of the CLT for both types of gluelines with the temperature at frequency of 1 Hz. At temperature below $T_g$, low tan $\delta$ corresponds to close pack molecular structure. Whilst the temperature sweep, the tan $\delta$ increased indicate both gluelines become more viscous.

A wide area of tan $\delta$ peak for PUR glueline observed in this test reflecting higher dynamic fragility, due to poor interfacial bonding between adhesive and wood. On the other hand, the
lower tan δ peak of PRF glueline showed a good interfacial adhesion similar to the results of storage modulus (Jawaid and Khalil 2011). From the tan δ curve in Figure 5, $T_g$ of PRF glueline was also observed to be slightly lower than PUR glueline. George et al. (2003) evaluated two commercial PRF and PUR adhesives using thermomechanical analysis (TMA) and found that wood joints bonded with PUR exhibited a significant temperature-dependent creep. The finding suggested that PUR is not suitable for structural application. As in this study, similar conclusion can be drawn, where PUR is an inferior adhesive compared to PRF in the manufacturing of CLT.

**Figure 5:** Damping Factor of PRF glueline and PUR glueline.

**CONCLUSIONS**

In this study, thermogravimetric analysis was performed on both neat PRF and PUR adhesives, *A. mangium* wood as well as gluelines of both PRF and PUR formed after CLT fabrication process. On the other hand, dynamic mechanical analysis was conducted to characterize the PRF and PUR gluelines. Generally, it was found that PRF adhesive glue was more thermally stable than PUR adhesive glue, with a higher percentage of residue in TGA test. Both adhesive glues start the degradation at the same temperature of 220 °C but PRF
adhesive showed higher degradation temperature at 530 °C at the second stage with a lower percentage of weight loss of 47.92%. As expected, A. mangium wood specimen has the lowest thermal properties due to low thermal stability of hemicellulose. The application of adhesives glue on A. mangium did improved the thermal stability of the wood by reducing weight loss during degradation process. In comparison, PRF glueline has better thermal stability compared to that PUR glueline as indicated by its higher residue at temperature up to 900 °C.

In DMA, the storage modulus of PRF glueline is higher but close to PUR glueline, suggesting that PRF glueline has better stiffness but lower mobility and flexibility. A sudden drop of storage modulus at about 100 °C for both gluelines, indicating glass transition temperature, $T_g$ and major main chain slipping were found. The $T_g$ of PRF glueline observed from tan δ and loss modulus curve was slightly lower than that of PUR glueline. Besides, PRF glueline resulted in a higher loss of modulus due to increased amount of energy needed to disentangle a better interfacial bonding. Similar trend was also found in loss modulus and tan δ curves. Based on results obtained, it can be concluded that PRF adhesive is a better binding agent for A. mangium wood in the manufacturing of CLT as it resulted in better interfacial bonding and a promising thermal performance at higher working temperature compared to PUR.

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