Density profile of alkaline-treated and densified 3-layered *Paraserianthes falcataria* composites

V Raman and K C Liew*
Faculty of Tropical Forestry, Universiti Malaysia Sabah, Jalan UMS, 88400, Kota Kinabalu, Sabah, Malaysia

* Corresponding author: liewkc@ums.edu.my

**Abstract.** Wood densification is one of the wood modification methods that had been invented in the 1900s. However, this method was discontinued due to lack of knowledge to completely understand the process. Findings indicated that the procedure of wood densification had been remediated in recent years due to increase competition in construction materials and the needs of quality structural materials. In this study, low-density plantation timber, *Paraserianthes falcataria* was pretreated with alkaline before undergoing densification process. The aim of this paper was to determine the density profile of the treated and untreated densified 3-layered *P. falcataria* composites. Alkaline pretreatment using NaOH was done according to soda pulping method to remove partially lignin from the timber used. Wood densification was done according to hot-pressing mathematical modelled by previous researchers, to enhance the properties of timber, such as density, by compressing the timber cell wall structure to eliminate voids between walls. In this study, 3%, 6% and 9% NaOH were used with 0% NaOH as control and the results for density profile showed that densified composites treated with 6% NaOH has the highest density profile, while densified composites treated with 0% NaOH (control) has the lowest density profile.

**Keywords:** Densification; alkaline pretreatment; low-density timber; density profile; *Paraserianthes falcataria*.

1. **Introduction**
Timber has been and remains an extensively utilized structural and environmentally friendly material [1-3]. UNECE [4] had reported sawn wood production in Europe and North America was 122 million m$^3$ and 128 million m$^3$, respectively, in 2016. Wood is a cost-effective alternative to other regularly used building materials due to its high specific stiffness and strength [5]. However, their mechanical properties vary widely due to their natural origin. Thus, their mechanical qualities also vary greatly.

Densification is advantageous for low-density wood species, as it helps to enhance density while also enhancing the value of the wood. The procedure of compression to the grain needs to be done erectly without any cell fractures during wood densification. In order to control compression deformation, [6] said that the cell walls must be elastic in order to improve the properties of densified wood [7-8]. The following factors are most likely to influence the alleviation and derogation phases: temperature, moisture, and time [9-10].

According to [11] and [12], hardness increases with densification, therefore densified wood would be advantageous in a variety of applications, such as flooring. The differences are due in part to a tree's natural traits (e.g., presence and size of knots, grain slope) and growth conditions (e.g., soil type, availability of water and nutrients) [13-14]. Densified wood products' superior mechanical qualities enable them to be used in a wide range of advanced applications, such as jigs and tools in the construction, aerospace, and automotive industries. Low-density wood can be used as a replacement and alternative for hard species after being modified into high-density wood for a superior performance.
product [15]. High density wood species can still be transformed through densification [16]. According to [17-18], filling or a combination of filling and compression of solid woods improved the qualities of the wood, such as hardness.

The processing method of wood compression at a high temperature with chemical treatment was developed by [19] to levitate the compressed woods. The chemical treatment involves the functions of compounded sodium hydroxide and sodium sulphide. This is done to remove portions of lignin and hemicellulose from the wood before wood compression. This will only be of benefit to the wood treatment and the most often used was alkaline pre-treatment. Non-corrosive and non-polluting chemicals are encouraged to be used throughout this operation. In comparison to acid pre-treatment, alkaline pre-treatment required a gentler approach. According to [20], the efficacy of alkaline pretreatment in lignin removal is higher as alkali react with lignin. This study was done to determine density profile and the effect of alkaline pre-treatment of untreated and treated densified *Paraserianthes falcataria* composites.

2. Materials and methods

In this study, laminas from *Paraserianthes falcataria* (Local name: Batai) were used for alkaline pre-treatment prior to densification process. 3%, 6% and 9% NaOH concentrations were produced by diluting NaOH on weight basis (w/w) with control 0% NaOH (untreated). Soda pulping method (Pulp & Paper Technology method) with 10:1 ratio of liquid to wood was used to cook the laminas for 30 minutes before being neutralized using 10ml acetic acid. Hot press was used for the densification of untreated and treated laminas at 105°C with 6 MPa pressure for 30 minutes, followed by 10 minutes of cooling under 6 MPa pressure to temperature less than 100°C. Laminas were laminated into 3-layered composites using Polyvinyl acetate (PVAc) with 24 hours of adhesive recovery time before trimmed into 50 × 50 × 24 mm. 30 replicates of test pieces were prepared, marked, cut, warped by plastic material and conditional under temperature of 23 ± 2 C and at 65% relative humidity. Test pieces for density profile were prepared and determined for density distribution characterization. GreCon densitometer Model DA-X 5000 was used to analyze the density profiler along the thickness, whereas X-ray technology was used to measure the raw density profile. Before the testing conducted, the instrument had undergone self-calibration procedure on appreciation was carried out before running. The test pieces were measured and arranged in the chamber, as shown in Figure 1, with optimized measuring source, at speed from 0.05 mm to 1 mm per second, involving X-ray combination. The density distribution profile was illustrated by setting the device operated under voltage at 33 kV, current of 0.25 micro sievert radiation dosage, temperature at 23 ± 2 °C and 65% relative humidity, respectively.

![Figure 1. Test piece arrangement in the chamber. (Image source: FIDEC [21]).](image)

3. Results and discussion

Density profile properties for untreated and treated densified 3-layered composites from *Paraserianthes falcataria* were measured and the mean values of density profile were tabulated in Table 1 and presented using scatter chart in Figure 2.
Table 1. Mean values and standard deviation for density profile of untreated (0% NaOH) and treated (3%, 6%, 9% NaOH) of 3-layered P. falcataria composites.

| Concentration of NaOH (%) | Total thickness of 3-layered composites (mm) | Density profile (kg/m$^3$) |
|---------------------------|---------------------------------------------|-----------------------------|
| 0                         | 24                                          | 656.46 ± (21.96)            |
| 3                         |                                             | 737.08 ± (26.67)            |
| 6                         |                                             | 854.41 ± (7.58)             |
| 9                         |                                             | 721.77 ± (18.64)            |

Figure 2. The mean values of density profile properties for layered test piece of untreated (0%) and treated (3%, 6%, 9%) densified P. falcataria composites at different concentrations of sodium hydroxide, NaOH.

The density profile in Figure 2 shows that the untreated (0% NaOH) has the lowest density with mean value of 656.46 kg/m$^3$ compared to treated layered test pieces. However, the results for treated test pieces shows that highest concentration of NaOH (9%) has the lowest density with mean values of 721.77 kg/m$^3$ compared to 3% NaOH and 6% NaOH with mean values of 737.08 kg/m$^3$ and 854.41 kg/m$^3$, respectively. Density profile for 3-layered P. falcataria woods with different NaOH concentrations varied with different depth of thickness. During the densification process, density profile was formed, and the analysis of the profile was impacted by various NaOH concentrations. The density profile and the density profile formation limitation changed the characteristics of the products, including densification time, temperature, humidity, densification force, pressure, adhesive curing, and wood conditioning were used to assign density profile development in wood. Untreated and densified P. falcataria woods resulted in lowest peak through the relative thickness of 3-layered composites. 6% NaOH has the highest peak and the density profile for 9% NaOH had dropped. 9% NaOH shows increases in thickness, as in this study, the thickness of laminas was reduced from 20mm to 8mm after the densification. Figure 3 shows the relationship of density profile for glulam and cell lumen areas of 0%, 6% and 9% NaOH has very weak positive linear correlation, indicating the increase in cell lumen areas very weakly increased in density profile. Figure 3(a) show the cell wall lumen with 0% NaOH was not fully compressed with the present of lignin, while Figure 3(b) shows that 3% NaOH has the more packed cell lumen, followed by Figure 3(c) and Figure 3(d), where the cell wall lumen with 6% and 9% NaOH started to expand and fibrous which may cause by the wood rheological properties, such as relaxation of cell. Previous study [19] reported that cell wall lumens got compacted and packed in densified woods. However, by eliminating partial lignin, cell lumen become more porous and caused the cell lumen to expand after densification. This was caused by the irreversible swelling when the densified laminas were exposed to humidity [16].
Figure 3. Correlation coefficient between cell lumen areas and density profile of untreated (0%) and treated (3%, 6%, 9%) densified *P. falcataria* composites at different concentrations of sodium hydroxide, NaOH; (a) SEM of 0% NaOH, (b) SEM of 3% NaOH, (c) SEM of 6% NaOH, (d) SEM of 9%.

Raman and Liew [22] reported that highest concentration of NaOH leads to higher density for lamina of *P. falcataria*, but after 7 days, the density of treated and densified *P. falcataria* began to lower compared to untreated and densified wood. According to the authors, the laminas were densified from 20mm to 8mm, as in Figure 4. However, density profile for 3-layered *P. falcataria* shows that treated and densified *P. falcataria* woods has higher mean values of density profile.
Figure 4. The thickness comparison before and after densification. (a) the untreated (0% NaOH) and densified wood lamina (8 mm), (b) the untreated (0% NaOH) and undensified wood lamina (20 mm). (Image source: Raman and Liew [22]).

Common technique such as density profile analysis always applicable to solid wood composites or engineered wood panels (EWPs), i.e., glulams, CLTs and particleboards. The formation of density profile was built during the densification process, where the analysis of profile was influenced by different concentrations of NaOH. According to [23-25], properties of products were modified by the density profile and the limitation of density profile formation. As mentioned by [26], there were samples that only inner layers of the samples were densified while outer layer was not compressed. This was most likely influenced by thermal conductivity properties. However, [27] reported that factors, such as type of wood and direction of densification, and wood behaviors were influencing the thermal conductivity. Cell wall properties were anticipated to have persistent density, thus thermal conductivity rising with density [28]. Kutnar [8] mentioned that alteration in densification modulus with relative thickness, respectively, influenced by moisture content, gas pressure and transient temperature differences in the mat, which caused by heat and mass transfer. Hence, the authors also mentioned density profile was produced when some areas in the mat were densified more than other areas. Density profile development in wood composites was allocated after densification with hot-press machine by time, temperature, humidity, densification force, pressure, adhesive curing, and conditioning of woods [9,29-30].

4. Conclusion
From this study, the results obtained shows that the higher density profile was achieved by 6% NaOH, while 9% NaOH shows low density profile for treated and densified P. falcataria composites. Treated and densified 3-layered P. falcataria shows higher gradient after alkaline pre-treatment, while untreated and densified 3-layered P. falcataria shows low density profile gradient. Hence, the study on wood density profiles was important in improving the composites quality.

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