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RELATIONSHIP BETWEEN CELL LUMEN AREA AND LIGNIN CONTENT OF ALKALINE-TREATED DENSIFIED TIMBER OF *PARASERIANTHES FALCATARIA*

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ABSTRACT

In this study, low-density plantation timber, *Paraserianthes falcataria* was pretreated with 3%, 6% and 9% NaOH before densification process. Alkaline pretreatment leads to lignin reductions and cell wall structure becomes more porous. Densification was done by crushing the cell wall with hot-press machine, resulting in reduction of thickness to about 60%. Scanning electron microscopy images were captured and processed through ImageJ software. As to support the data, lignin content determination was conducted according to TAPPI T222 and the correlation coefficient between cell lumen areas and lignin content were studied statistically.

KEYWORDS: Densification, alkaline pretreatment, scanning electron microscopy, *Paraserianthes falcataria*, low-density timber.

INTRODUCTION

Wood densification is one of the effective methods in enhancing the mechanical properties of wood, as wood is a viable constructional material (Shi et al. 2020). The same authors also stated that, it is possible to undergo both chemical treatment, such as alkaline pre-treatment or Klason pre-treatment, and wood densification to maximize the value of density of woods. Wood with high porosity has a pyramid arrangement, which causes density and strength to deteriorate. According to Cencin et al. (2021), after the cell walls have crumbled under heat condition, low-density wood can be densified to achieve a greater density. Laine et al. (2013a,b) mentioned that hardness increases with densification, therefore densified wood would be advantageous in a variety of applications, such as flooring, where high hardness is required.

According to Kim et al. (2015), to pass through both chemicals and physical obstructions can be an efficiently pre-treatment. Pre-treatment can levitate the enzymatic digestibility of biomass through suitable application of chemical reagents/outer substances, catalysts. Shi et al.

(2020) stated that as to remove lignin and/or hemicellulose, a strategy of optimal treatment reagent/catalysts will be set. Pre-treatment at an extreme temperature within short period of time of reaction for instance, can be diluted sulfuric acid, ARP, or steam explosion (Kim et al. 2003).

Following densification, low-density timber species can be used as an alternative to hardwood species (Kutnar and Sernek 2007). The higher mechanical properties of densified wood products allow their use in diverse and advanced applications (such as jigs and tooling in the construction, aerospace, and automotive industries). For example, Anshari et al. (2012) utilised the moisture-dependent swelling and improved mechanical properties of densified wood as a reinforcement material in glulam beams. The authors reported a bending stiffness increase of up to 46% compared to that of unreinforced glulam. Riggio et al. (2016) used densified wood plates and dowels to replace steel plates and dowels in a timber beam-column connection. These connections utilized the moisture-dependent swelling effect of densified wood materials to create a tight fit in the connections. Additional benefits of densified wood include recyclability, reusability and relatively lower density when compared with steel.

MATERIALS AND METHODS

Paraserianthes falcataria timber were processed into 300 mm (length) X 50 mm (width) X 20 mm (thickness) lamina. Suryono and Pranoto (2021) used such lamina thickness in their study, between 6.5 mm to 20 mm. In this study, laminas with initial moisture content ranging 12 - 15% and initial density 309 - 345 $kg \cdot m^{-3}$ were used for alkaline pretreatment at different NaOH concentrations of 3%, 6%, and 9% (w/w), and untreated as control before the densification procedure. The laminas were cooked using soda pulping technique in boiling temperature (±100°C) for 30 min at 10 : 1 liquid to wood ratio, and then neutralized with acetic acid. Densification was done on untreated and treated laminas using a hot press at 105°C and 6 MPa pressure for 30 min, followed by 10 min of cooling to temperatures below 100°C while maintaining the pressure using the hot press. The densified laminas were cut into 5 mm (length) X 5 mm (width) using a sharp razor knife to get perfect transverse sections and gold-coated for Scanning electron microscope at magnification of 1.00 Kx with accelerating voltage of 15 kV. Thirty replicates scanned images of transverse section for each concentration were processed using ImageJ software. Lignin content determination was also conducted after soda pulping, according to standard T 222 with n = 30, as to justify the lignin content in the cell structure after treatment and compared with control. Pearson's correlation coefficient was done between cell lumen areas and lignin content of the untreated and treated densified *P. falcataria*.

RESULTS AND DISCUSSION

Fig. 1 shows that 9% NaOH has the largest cell lumen area compared to other concentrations with mean value of 10.53 µm^2, followed by 6% NaOH with 5.18 µm^2 of lumen area and 0% NaOH with 3.87 µm^2. The lowest lumen area achieved by 3% NaOH with 2.06 µm^2. Alkaline pre-treatment helped in increasing the maximum densification in 3% NaOH as the area of cell lumen collapsed more than other concentrations. Fig. 1 also shows that 9% NaOH has the lowest lignin content with 5.77%, while 0% NaOH has the highest lignin content with 12.83%, with slightly different in mean values compared to 3% NaOH with 12.44%, followed by lignin content of 6% NaOH with 7.98%.

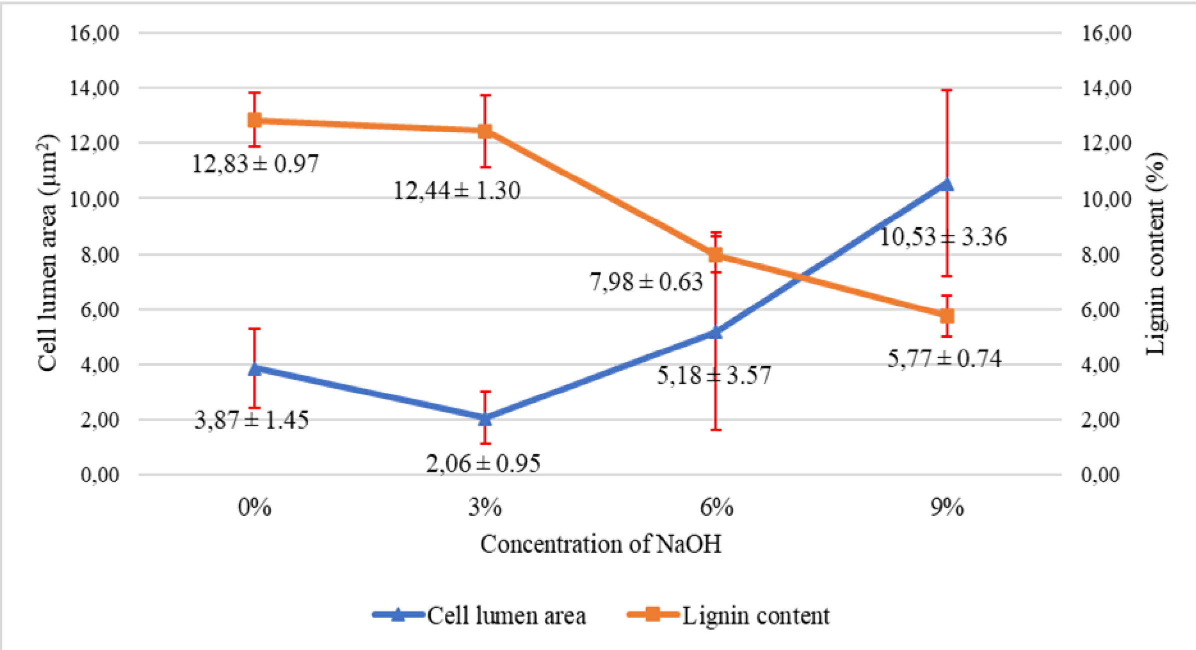

Fig. 1: The mean values and standard deviation (±) of cell lumen areas and lignin contents of densified and treated P. falcataria timbers at different concentrations of NaOH (3%, 6% and 9%) with densified and untreated P. falcataria timbers as control (0%).

Fig. 2 shows the SEM images of untreated and treated densified *P. falcataria* using different concentrations of NaOH (3%, 6%, 9%) and 0% NaOH as the control. Fig. 2a revealed the cell lumen structure of 0% NaOH after densification were not fully collapsed and numerous gaps remaining in between the cell walls, while Figs. 2b-d show more crushed cell wall because of the absence of lignin as the binder. However, 3% NaOH has the most packed lumen compared to 6% and 9%. Figs. 1 and 2 show higher concentration of NaOH leads to higher removal of lignin which caused the wood to become more porous and brittle. The cell wall itself had been degraded by the chemical modification. Moisture can be easily absorbed from the surrounding environment, which cause cell relaxation and cell lumen expanded.

Kutnar et al. (2009) had stated that the cell walls must be elastic in order to improve the properties of densified wood (Blomberg et al. 2005). The following factors are most likely to influence the alleviation and derogation phases: temperature, moisture, and time (Wolcott et al.

1990). Pelit et al. (2014) reported that the expansion of cell was structure and releasing internal tensions may cause the densified laminas to revert to their original state. Kutnar et al. (2009, 2015) 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. The comparison in thickness between untreated (0% NaOH), treated (3%, 6%, 9%), densified and undensified laminas showed in Fig. 3.

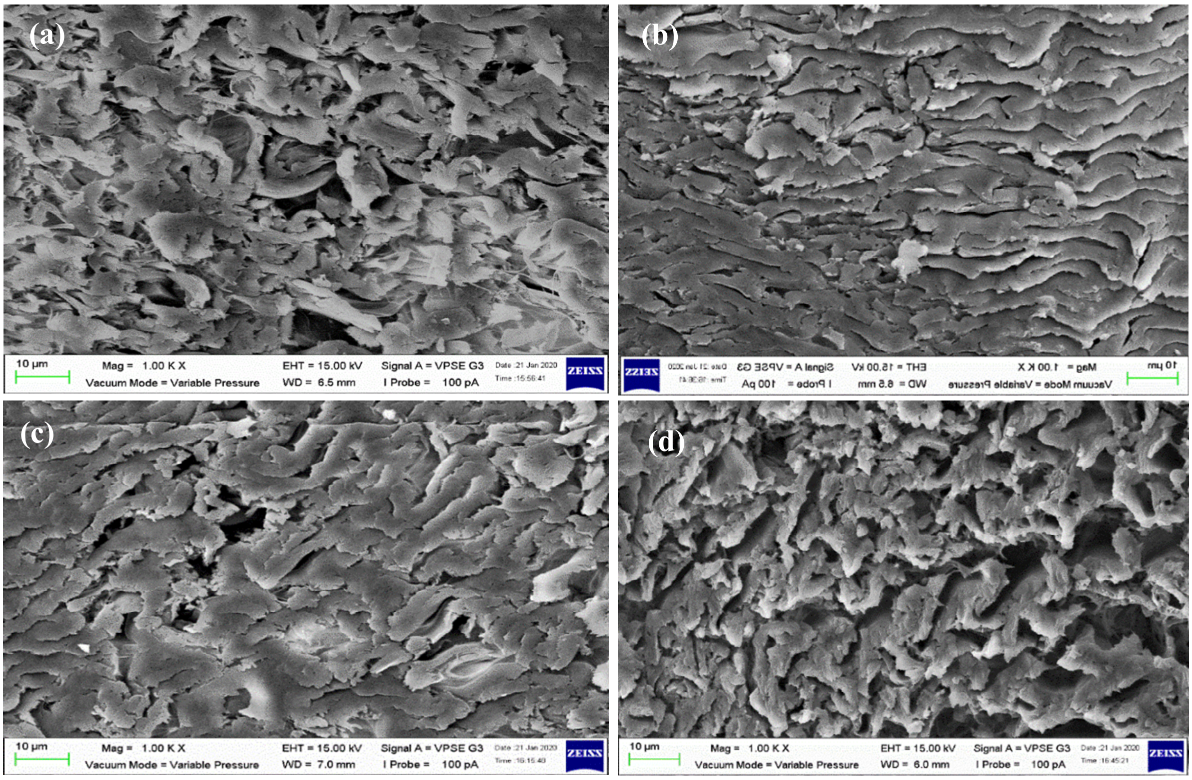

Fig. 2: SEM images of untreated and treated densified P. falcataria (a) 0% NaOH, (b) 3% NaOH, (c) 6% NaOH, (d) 9% NaOH). Magnification: 1.00 Kx.

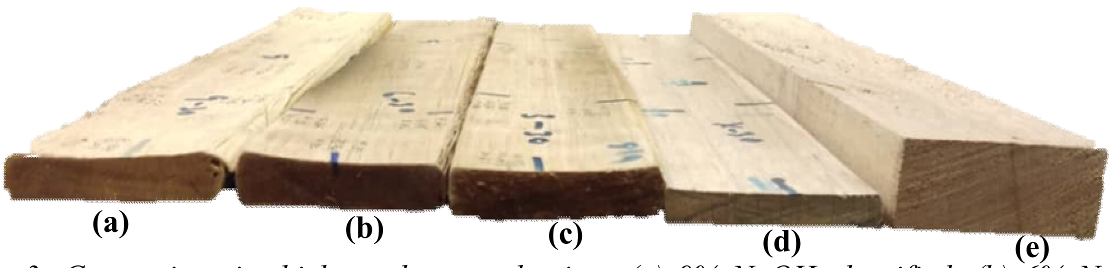

Fig. 3: Comparison in thickness between laminas (a) 9% NaOH, densified, (b) 6% NaOH, densified, (c) 3% NaOH, densified, (d) 0% NaOH, densified, (e) 0% NaOH, undensified.

Lenth and Kamke (2001) mentioned in their findings that densification of wood causes permanent and temporary changes that alter both physical and mechanical properties of woods. Lumen in cell walls get packed in densified woods after alkaline pretreatment, according to studies by Song et al. (2018) and Kutnar and Sernek (2007). As a result, after being flattened to improve hardness, the packed cell walls bonded to one another. Kamke and Rathi (2011) had mentioned in their findings that densification of wood brings to permanent and temporary changes, which affect both physical and mechanical properties of woods. However, according to Santos et al. (2012), Nairin (2006), Islam et al. (2014) and Silva and Kyriakides (2007), the effects of many elements on the mechanical properties of densified woods include wood species, treatment conditions, pressing conditions (such as temperature, pressure, and cooling), and conditioning conditions. However, Schrepfer and Schweingruber (1998) mentioned in their discoveries that earlywood sections were simpler to distort than latewood sections. In additional, the changes in wood properties depend on its characteristics (Bami and Mohebby 2011).

According to the authors' previous published paper on density in Raman and Liew (2020), the density of treated wood showed significant change, where the highest concentration (9% NaOH) has the lowest density 587.2 $kg \cdot m^{-3}$, while control has the highest density at 909.2 $kg \cdot m^{-3}$, which also caused lamina reduced in thickness for about 40% after densification process. In this study, lignin reduction helped the densification process after the cell wall structure become porous and brittle, as Wolcott et al. (1989) had stated that hemicellulose, lignin, wax, and oils in the fibre cell wall were eliminated after an alkaline pre-treatment. Because lignin is needed as a binder, withdrawing it may damage the features. Cells collapse depending on the test circumstances and the type of cell wall components, such as elastic distortion, plastic flexibility, or brittle suppression, cells collapsed. According to Islam et al. (2012), the adhesion and compatibility between wood components such as fibers and polymer had been increased after the lignin removed and cellulose reacted with NaOH. According to previous studies by Kollmann et al. (1975), Blomberg et al. (2004), Pelit et al. (2014, 2015), had mentioned that densified wood by compression would return to the measurement before compression when the wood had been soaked in water or exposed to high moisture environment.

Fig. 4 shows the coefficient correlation between lignin content and cell lumen areas of untreated (0% NaOH) and treated (3%, 6%, 9% NaOH) densified *P. falcataria* laminas had shown the range of correlation coefficient values and the corresponding levels of correlation. The relationship between lignin content and cell lumen areas of 0% NaOH was found to have very weak negative linear correlation, with r-value of $r(28) = -0.09$, $p < 0.64$, where higher lignin content leads to lower cell lumen areas in the cell structure. Meanwhile, the relationship between lignin content and cell lumen areas 3% NaOH shows very weak positive linear correlation, with r-value of $r(28) = 0.06$, $p < 0.74$. The relationship between lignin content and cell lumen areas 6% NaOH also reported very weak positive linear correlation, with r-value of $r(28) = 0.15$, $p < 0.42$. Both results indicated that decrease in lignin content very weakly increased the cell lumen areas in cell structure. The correlation between lignin content and cell lumen areas of 9% NaOH was found to have weak negative linear correlation, with r-value of $r(28) = -0.34$, $p < 0.06$. This result shows that decrease in lignin content increased the cell lumen areas. These interpreted results were statistically insignificant, as the p-values were greater than 0.05. After wood conditioning, cell lumen areas tend to expand after absorbing moisture from surrounding. This is caused by partially reduced lignin from the cell wall structure as the main function of the lignin is to bind the cell wall from another. Kutnar and Sernek (2007) stated in their findings that woods release glass state at a sped-up rate of moisture content and temperature (rigid and delicate). It will bounce at an elevated rate of moisture content and temperatures, in addition to an elevated rate of moisture content and temperatures. As a result, the glass transition temperature is the point at which temperatures change from glazed to bouncy behaviours.

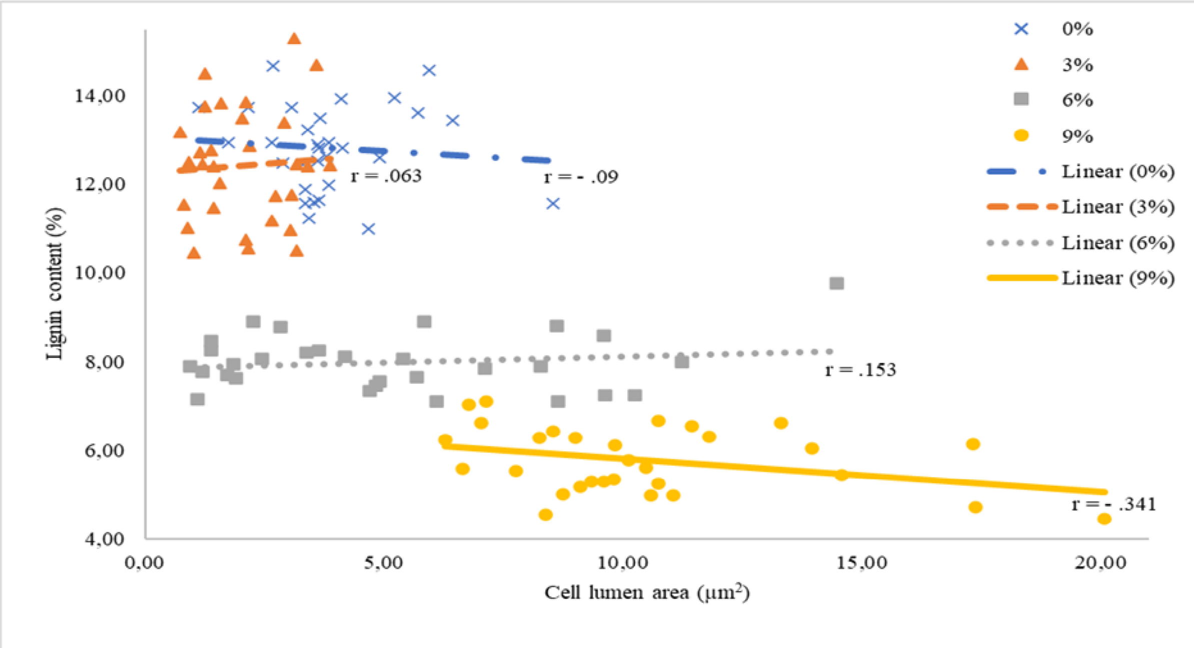

Fig. 4: Coefficient correlation between lignin content and cell lumen areas of untreated and treated densified P. falcataria laminas.

CONCLUSION

Alkaline pretreatment had altered the cell wall structure by removing partial of lignin, followed by the wood densification process which had compacted the void in the porous structure of cell wall. However, results showed that alkaline pretreatment with higher NaOH concentration leads to higher lignin content reduction in cell wall which causes cell lumen areas to expand after densification. Spring-back occurs when wood had been exposed to moisture or high room humidity. Higher concentration leads to higher lignin reduction which caused in more voids in cell wall as the cell lumen areas expanded.

Expansion of cell wall structure and loosening internal stresses caused for the samples to recover to the initial state as before densification. Meanwhile, results were proved by correlation coefficient analysis, where 9% NaOH leads to higher lignin reduction. Untreated shows the lowest cell lumen areas after densification and highly believed that less lignin reduction occurred in the cell wall structure helps the bonding within compositions, as the main function of lignin in cell wall structure is to hold the structures together. In other hand, by reducing the lignin in the structure would lead to highest spring-back after densification and special treatment or method would be needed to apply during wood modification as to eliminate the spring-back.

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