The effect of pre-freezing treatment on the color changes and wettability of teak wood (Tectona grandis Linn.F.)

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Abstract. Pre-freezing treatment is one of the methods used for increased the properties of wood. The pre-freezing treatment affects the physical properties of wood, extractive, and surface conditions, this will affect the wettability of wood and wood color. The purpose of this research was to evaluate the effect of pre-freezing treatment on the color changes and changes in contact angle and wettability of wood. The study was conducted on teak wood (Tectona grandis) with the temperature -16°C for 24 hours and 48 hours. The results of this study showed that the pre-freezing treatment made decreasing of brightness with a reduction of the k-value. Besides, the pre-freezing time affected the wettability of wood. The wettability of wood was declined with increased time of pre-freezing. Wood utilization as a raw material of composite needed more treatment to increase the wettability of wood.

1. Introduction
Teak wood (Tectona grandis) is one of the most important species of wood in the world [1]. Teak wood is an expensive species hardwood with physical properties, durability, and good esthetic [2,3]. The superior properties of teak wood increasing demand in the community cause decreasing in the availability of teak wood. To solve the problem, the community and Perhutani Corporation are maximizing wood production per unit area through genetic selection and silvicultural techniques to produce fast-growing teak [4]. Fast growing teak wood had weaknesses properties such as low dimensional stability, strength, and durability [5]. Therefore, treatment is required to improve the quality of teak wood. One of the alternatives treatment is pre-freezing treatment.

The pre-freezing treatment has been done in the some of wood species in the winter in America. Wood store at the negative temperature has advantages such as reduced the biodeterioration, drying defects, and, dimensional shrinkage, and rate of drying [6,7]. Pre-freezing treatment caused the water transfer from the cell wall to the lumen, so that the cell wall is thickening and cells become rigid [8]. Cell walls under pressure make the cell more rigid and tend to get steady. Pre-freezing treatment in a short time has a positive effect on the strength properties of wood. This was also stated by Szmukcu et al. [7] that fast pre-freezing results in better quality than products with long pre-freezing.

The study of pre-freezing treatment has been carried out on the several species of wood such as spruce wood [7], pinewood [8], and fast-growing species like rose gum and Gympie messmate [5]. Based on the results mentioned that wood has lower surface energy with higher pH value, decreasing weight loss, and increasing the water content. Furthermore, the pre-freezing treatment caused decreasing of MOE value and compression parallel to a grain of spruce wood [5]. Alteration of wood properties due to pre-freezing treatment is thought to affect wettability and the color of the wood. Changes of the wettability and color was occured on the same species after heat treatment [9,10].
Based on the research that the high temperature affected the ability wetting the water and the color of wood. After heat treatment the color of wood became darker. In addition, wood is also kepted in low temperatures. The wood that is stored at low temperatures effect to changes the components of wood, so it is thought to also affect the wetting and color of the wood. Therefore, this study was conducted on the fast-growing teak wood with pre-freezing treatment to evaluated the color change and the wettability of teak wood. The research aimed were to evaluate the effect of the pre-freezing treatment on the color change and the wettability of the teak wood fast-growing wood species.

2. Materials and Methods

2.1. Materials
The research was conducted at Wood Anatomy and Quality Improvement Laboratory, and Wood Design and Engineering Laboratory, Department of Forest Product, Faculty of Forestry, IPB University. The material used for this study was the teak wood with the size of the length, width, and thickness 10 x 10 x 2 cm. The color testing and wettability was perfomed three replication.

2.2. Methods

2.2.1. Pre-freezing treatment
Samples were scanned before and after pre-freezing treatment. Samples were ready to put into the freezer at a temperature of -16°C with a variation of time for 24 hours and 48 hours. After that, samples were drying in an oven at 60°C for 4 days. Dried were occured until the moisture content of wood under 20%. Dried samples put into desiccator for 15 minutes.

2.2.2. The color changes
The measurement of wood color was used the Cannon MP 145 Scanner which is connected to a laptop, then the data was processed used ImageJ which is produced using ImageJ which produces L*, a* and b* values. The measurement of the color change in the wood refers to the research of [11] using the Cannon MP 145 Scanner the data formed in a photograph of a wood surface. The color change was tested at nine different points as in Figure 1.

![Figure 1. Sketch of the sample for the color changes teak wood](image)

The color changes were measured before and after the pre-freezing treatment. The data obtained were processed using the ImageJ application. The color change was calculated using the L* a* b* formula [12]. L* was the change in the brightness from 0 (black) to 100 (white). a* showed the change in color red (positive) to green (negative). b* was indicated the change in color from yellow (positive) to blue (negative). The difference of color (∆E) was calculated by the CIELab formula [13,14].
\[
\Delta E = \sqrt{(\Delta L^*)^2 + (\Delta a^*)^2 + (\Delta b^*)^2}
\]

where,

- \(\Delta E\): Different of color
- \(\Delta L^*\): Different of light = \(L^*\) treatment – \(L^*\) control
- \(\Delta a^*\): Different of color red or green
- \(\Delta b^*\): Different of color yellow or blue

The value of changed or differenced in wood color before and after pre-freezing can be seen in Table 1.

### Table 1. The differences in color (\(\Delta E\)) [15]

| Different of color (\(\Delta E\)) | The influenced                                             |
|----------------------------------|-----------------------------------------------------------|
| 0.2 < \(\Delta E^*\)            | Invisible difference                                      |
| 0.2 < \(\Delta E^*\) < 2        | Small difference                                          |
| 2 < \(\Delta E^*\) < 3          | Color change visible with a high-quality filter           |
| 3 < \(\Delta E^*\) < 6          | Color change visible with medium-quality filter           |
| 6 < \(\Delta E^*\) < 12         | High color changes                                        |
| \(\Delta E^*\) > 12             | Different color                                           |

2.2.3. The measurement of the contact angle
Measurement of the contact angle between liquid and wood surface using the sessile drop method. The liquid used was distilled water. A lot of liquid that is used 0.02 mL was dripped on a wooden surface, then recorded using a portable microscope camera. The results obtained were analyzed using GOM player software to get photos every 9 seconds for 3 minutes thus obtained 19 photos for each point. The photos were measured its contact angle using Image-J (Figure 2). Each board is dripped at three different points (Figure 3). The measuring contact angle were conducted into the two sample. There sample were freeze treatment for 24 hours and 48 hours. Contact angles were tested on samples before and after pre-freezing treatment.

![Figure 2](image2.png)

**Figure 2.** The measuring of the contact angle

![Figure 3](image3.png)

**Figure 3.** Sketch of the sample for wettability test
Over time, the contact angle is formed will also change. Changes of the contact angle in the surface of wood was illustrated the ease of wetting. The change in contact angles was analyzed using the polynomial regression model based on Equation 1.

\[ y = ax^2 + bx + c \]  

The measuring of the equilibrium contact angle used PROC NLIN software from SAS 9.1 with a segmented regression model based on Equation 2.

\[ y = a + bX + cX^2 + E \]  

where, \( y \): contact angle; \( x \): time; \( a \), \( b \), and \( c \): constants; \( E \): error

2.2.4. The wettability based on the k-value

The contact angle value was determined based on a segmented regression equation between time (t) and contact angle (\( \theta \)) using PROC NLIN SAS. Parameter of k-value is determined based on the S/G model (Shi and Gardener 2001). The S/G model uses the formula

\[ \theta = \frac{\theta t, \theta e}{\theta i + (\theta e - \theta i) \exp[K(\theta e - \theta i)t]} \]  

where,

- \( \theta \) : Contact angle at the specific time
- \( \theta i \) : Contact angle at the initial time
- \( \theta e \) : Contact angle at the equilibrium
- \( t \) : Time
- \( K \) : The constant rate of change the contact angle

The k-value measuring from S/G model with the XLSTAT 2019 software.

3. Results and Discussion

3.1. The Color changes

Color is one of the determinants of wood quality that will affect customer perceptions of the product [16]. Color is shown visual perception that is determined by spectra light makeup reflected off the surface. The L* value was indicated the level of brightness of the coloring, the more positive the brighter the wood. But on the contrary, if the value of L* decreased was the darker the coloring result. The value of a* represents the color produced in red or green, while the value of b* represents yellow or blue [17].

The color changes of teak wood after pre-freezing treatment can be seen in Figure 4. The L* value of teak wood for 24 hours treatment before and after pre-freezing treatment is 80.18, and 76.95, respectively. The average brightness decrease of 4.02%. Meanwhile, wood brightness before 48 hours pre-freezing treatment is 83.28 and decreased to 77.78. The average brightness reduction of 6.60%. The a* and b* values of teak wood due to pre-freezing treatment tended to increase. The value of b* after 24-hour pre-freezing treatment was increasing by 18.23%, while the 48-hour pre-freezing treatment higher increasing by 22.31%. The percentage change in the value of b* 48-hour pre-freezing treatment is greater than the 24 hours treatment. This relates to changes in the color of teak wood which becomes darker as increasing of the cooling time. The decreasing trend that occurs in teak wood due to pre-freezing treatment be expected by changes in chemical composition such as degradation of hemicellulose. Karlinasari et al. [18] stated that the freeze treatment can be presented the highest variation in holocelulose content, wherein the decrease was from 68.44% to 63.55%. Degradation on the holocelulose effected to the color wood. This was stated by [15,10,19] on the species of sengon, teak, mangium, mahagony, shorea, merbau, oak trees, scotch pine trees, and silver bitch tress that the color change becomes darker is more influenced by the degradation of hemicellulose.
Figure 4. The color changes of teak wood pre-freezing treatment

The total color change was calculated by the value ∆E. ∆E is strongly influenced by temperature differences in the wood [12]. Figure 5 explains that the amount of color changes occurred on the surface of the wood. Figure 6 explains that the amount of color change can be quantitatively measured. The average color change of untreated 24 hours is (∆E) 5.647, this color change is classified as a moderate color change. The 48 hours pre-freezing treatment had a color change (∆E) of 7.157 and was classified as a high color change. Bárčik et al. [15] stated that if a change of 3 <∆E <6 that color changes with a medium quality filter, whereas if a change occurs in the range of 6 <∆E <12 that a high color change.

Color changes that occurred in wood due to pre-freezing treatment are caused by the degradation of the wood components. The decreasing trend of the brightness of wood that occurs in teak wood due to pre-freezing treatment is caused by changes in chemical composition such as degradation of hemicellulose [18]. Also, changes in the value of a* were affected by condensation, decomposition, and / or degradation of lignin content. Meanwhile, changes in the value of b* was affected by the lignin content in wood, lignin has a role in the presence of yellow in the wood [12]. This is following the research of Missio et al. [5] that the pre-freezing treatment effects to the chemical components of wood consisting of extractives, lignin, holocellulose. Particularly for teak wood, quinones are classified as colouring matters in plants and give yellow, orange, red and green or black. Based on the results of the study, Gympie messmate wood extractive substances have increased (1.16-1.23%), while the holocellulose content has decreased (64.15-62.83%). Therefore, the wood had a pre-freezing treatment will be darker.

Figure 5. Changes of the color wood (a) Untreated, (b) after 24 hours, (c) Untreated, (d) after pre-freezing 48 hours
Figure 6. The changes of ∆E values on the pre-freezing treatment for 24 hours and 48 hours

3.2. The contact angle and wettability

The wettability is the ability of a material to absorb the liquid [20]. Wettability was influenced by several factors related to adhesives (surface tension, temperature, and viscosity) and wood (density, porosity, extractives, and surface roughness) [18]. One of the criteria that can be used in determining the wetting of wood is the contact angle. Figure 7 shows the distillate water droplets on teak wood without pre-freezing treatment (untreated) and after pre-freezing treatment. If the contact angle value is more than 90° then wood wetting is not good. As a result, the surface of the wood is difficult to get wet by the liquid (hydrophobic) [21]. Changes in contact angle that occur based on changes in time indicate the wettability of a material [22]. The liquid that falls on the surface of the wood will spread and penetrate until reach a constant angle (θe).

Figure 7. The contact angle untreated and pre-freezing treatment for 24 hours

Figure 8 shows the change in the contact angle value of the teak wood treated 24 hours and 48 hours. Significant changes in contact angle occur in the initial 10 seconds. The contact angle without treatment is in the range between 80°-90°, while the contact angle after treatment is in the range between 114°-116°. The average contact angle untreated wood in the 0° second by 79° ± 7°, 30 second by 61° ± 6, and 180 second by 42° ± 4. The average contact angle treated wood in the 0 second by 102°
± 12°, 30 second by 65° ± 10, and 180 second by 44° ± 9. This shows that the contact angle of the pre-freezing treatment is higher than the contact angle without the pre-freezing treatment. Pre-freezing treatment can reduce wood wettability. This is influenced by wood factors such as species, wood anatomy (composition), wood chemicals (extractives, cellulose, hemicellulose, and lignin), and surface roughness [23]. Wood is a heterogeneous material that will have an impact on the results of varying wettability on each species [24]. Wettability of teak wood is quite poor, this is due to the high density of wood and some deposits can inhibit the process of water absorption. The type of red banana wood also has a deposit which caused a low wettability [24].

Wettability of teak wood is influenced by extractive substances in wood. As a result of pre-freezing treatment, it is assumed that the extractive moves to the surface so that the content of the extractive substance tends to be higher and cause lower wettability. This was also conveyed by Ilic [25] that pre-freezing treatment causes extractive substances to move to the surface of the wood so it causes the wood more rigid. The wood extractive substances make wood more hydrophobic [26]. Besides, the results of the research Karlinasari et al. [18] on wood species of rose gum and Gympie messmate explained that pre-freezing treatment affects the chemical content, especially wood extractive substances. The pre-freezing treatment affects the increase of extractive content of 1.16% to 1.23%. Some studies suggest that extractive substances affect wettability [23,27]. Extractive substances make the interaction between the adhesive and lignocellulose material less than optimal. As a result, the surface of the wood is difficult to pass by liquid.

![Figure 8](image_url)

**Figure 8.** The transformation of the contact angle pre-freezing treatment 24 hours and 48 hours

The liquid that falls on the surface of the wood will spread and penetrate the wood until a constricted contact angle (θe) is reached. The value θe is used to determine the rate of change in the constant contact angle (k-value). The value of k is an indicator of the value of wettability. The higher the value of k, the better the wettability of material, this indicates that the liquid is more easily spread and absorbed in the material [28]. Figure 9 shows the k-value treatment after pre-freezing treatment for 24 hours and 48 hours tends to decrease compared to without pre-freezing treatment. This indicates that pre-freezing treatment for 24 hours and 48 hours reduces the wettability of teak wood. When associated with wood absorption values, 24 hours and 48 hours pre-freezing treatments are lower than without pre-freezing treatment. This causes the longer wood to be pre-freezing, the better dimensional stability. According to the research of Szmuktu et al. [7] on spruce wood that the dimensional stability of wood is increasing with increasing pre-freezing time. The pre-freezing treatment is a method that makes the water turn into ice which causes the expansion of the lumen and constriction of the cell.
wall. This causes the wood absorption value to decrease and wood wettability decreases while the contact angle increased.

![Figure 9. K-value of teak wood from pre-freezing treatment for 24 hours and 48 hours](image)

4. Conclusion
The pre-freezing treatment occurred with the variation of time 24 hours and 48 hours. Based on the treatment, the color teak wood was darker with decreasing of L* value and ΔE value of teak wood is 5.65 for 24 h and 7.16 for 48 hours. The pre-freezing treatment caused decreasing the wettability with decreasing of the k-value while the contact angle was increased.

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