Effects of Heat Treatment on the Color Change and Dimensional Stability of *Gmelina arborea* and *Melia azedarach* Woods

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Abstract. This study aimed to improve the color properties and dimensional stability of gmelina (*Gmelina arborea*) and mindi (*Melia azedarach*) woods via heat treatment. Heat treatment was conducted using an electric furnace at 180°C and 210°C for 3 h, with a heating rate of 2°C/min. Wood samples were stacked with and without metal clamp. The effectsof temperature and clamping during heat treatment on the color change and dimensional stability were evaluated. The evaluation of color change was performed using the CIE-Lab color system and the evaluation of dimensional stability was conducted by measuring the equilibrium moisture content and water absorption. The results showed that the overall color changes(ΔE^*) in gmelina and mindi woods were mainly affected by the reduction in lightness (L^*) due to heat treatment. The ΔE^*increased with increasing treatment temperature, with a higher degree obtained in gmelina wood. Application of metal clamp during treatment limited the exposure of wood surface to the heated air, resulting in lower value of ΔE^* than the samples without metal clamp. Dimensional stability of gmelina and mindiwoods improved by heat treatment, showing lower equilibrium moisture content and water absorption than the untreated woods. Furthermore, heat treated mindi absorbed less water than gmelina. The results suggested that heat treatment could enhance the color properties and dimensional stability of gmelina and mindi woods for value added products.

1 Introduction

Plantation forests play an important role in providing timber supply in Indonesia. The Indonesian Ministry of Forestry (2014) reported that timber production in 2013 was 23.23 million m^3, with the majority of timber production (84% or 19.55 million m^3) was produced

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from plantation forests, while the supply from natural forests was only 16% or 3.68 million m^3 [1].The tree species that commonly planted in plantations are fast growing tree species including gmelina (*Gmelina arborea*) and mindi (*Melia azedarach*). These species are very promising to be developed both in industrial plantations and in community forests. However, fast-growing wood species usually has a high proportion of juvenile wood, low density, low strength and high longitudinal shrinkage [2, 3]. Our previous studies also revealed that gmelina and mindihave relatively low density, low durability, and low mechanical properties that are not suitable for structural timbers [4-6].

Some properties of gmelina and mindi woods can be improved by thermal modification. Thermal modification or heat treatment of wood is the introduction of heat to wood at temperature ranging between 160 - 260°C to improve the inherent properties of wood, to produce new materials and to acquire a form and functionality desired by engineers without changing the eco-friendly characteristics of the material [7].Heat treatment of wood has more advantages compared to other wood modification techniques such as chemical, impregnation, and surface modification because it is relatively simpler and more eco-friendly because no hazardous chemicals used in the process.

Our previous studies on heat treatment have been carried out using various process conditions to improve the properties of woods from temperate region as Korean white pine (*Pinus koraiensis*Sieb. &Zucc.) and royal paulownia (*Paulownia tomentosa* (Thunb.) Siebold &Zucc. ex Steud.) [8, 9] and high-density wood as okan (*Cylicodiscusgabunensis* [Taub.] Harms) [10-12]showing the improvement in wood color, equilibrium moisture content, and improves dimensional stability. However, study on heat treatment of tropical wood species as gmelina and mindi has not been performed. Therefore, we studied the heat treatment of gmelina and mindi woods and determinedits effects on the color properties and dimensional stability.

2 Materials and Methods

2.1 Materials

Logs of two fast-growing wood species i.e., gmelina (*Gmelina arborea*) and mindi (*Melia azedarach*)were converted into boards with dimensions of 200 mm (length) × 90 mm (width) × 20 mm (thickness).Only boards with straight grain and free of natural defects were selected. Selected boards were then air-dried and conditioned at 25ºC under relative humidity of 70-80% until reached equilibrium moisture content (EMC).

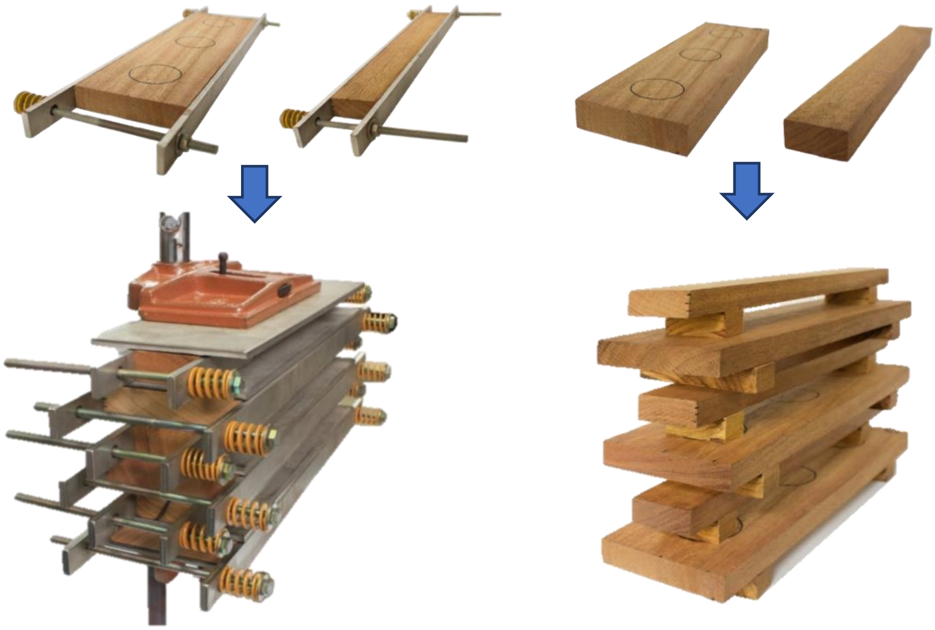
2.2 Heat Treatment

Sample boards were stacked with metal clamps with the arrangement as shown in Fig. 1. Another set of boards were stacked without clamps for comparison. Heat treatment was conducted using electric furnace with the following steps: 1) raise the furnacetemperature from 25°C to targeted temperature with a heating rate of 2°C/minute, 2) maintain target temperature of 180°C or 210°Cfor 3 hours, 3) lower the temperature until reaching room temperature and then take out the boards from the furnace. The heat-treated boards were then kept at conditioning room until they reached EMC.

2.3 Board Evaluation

Color change

Color change was measured using CIE-*Lab*color systems consisting of three color parameters as lightness (L^*), red/green chromaticity (a^*),andyellow/blue chromaticity (b^*)[13].The L^*parameter has a maximum value of 100 (white) and minimum value of 0 (black).



(a) (b)

Fig. 1. Board stacking during heat treatment: (a) with metal clamp and (b) without clamp.

The a^*parameter with positive value indicated red color($+a^*$) while negative value indicated green color ($-a^*$). $b^*$with positive value indicated yellow color($+b^*$) while negative value indicated blue color ($-b^*$).Color measurement was conducted using *chromameter* (CR-400, Konica Minolta Inc., Tokyo, Jepang) on the surface of the sample boards. The overall color change (ΔE^*) after heat treatment was measured using following equation:

$$\Delta E^* = (\Delta L^{*2} + \Delta a^{*2} + \Delta b^{*2})^{1/2} \quad (1)$$

where ΔL^* is the difference in lightness, Δa^* is the difference inred/green chromaticity, and $\Delta b^*$is the difference in yellow/blue chromaticity after heat treatment. The value of $\Delta E^*$where then used to determine the levels of perceived difference in color [14,15].

2.4 Physical Properties and Dimensional Stability

The physical properties and dimensional stability were tested by measuring the weight loss, volume shrinkage, density, equilibrium moisture content (EMC), water absorption, and wettability of the sample boards before and after heat treatment. Weight loss (WL) and volume shrinkage (VS) were calculated using following equations:

$$WL\ (\%) = 100 \times (m_1 - m_2)/m_1 \quad (2)$$

$$VS\ (\%) = 100 \times (V_1 - V_2)/V_1 \quad (3)$$

where $m_1$and V_1areoven-dried weight (g) and volume (cm^3) of sample before heat treatment, respectively. m_2 dan $V_2$are oven-dried weight (g) and volume after heat treatment, respectively.

Density of the boards was tested by measuring weight and volume of samples in oven dry condition in accordance with the KS F 2198 standard [16]. Equilibrium moisture content was tested according to KS F 2199 standard [17] by measuring air-dried and oven-dried weight of boards before and after heat treatment. Water absorption test was conducted according to KS F 2204 standard [18]by immersing wood samples in ambient water for two weeks and measuring the weight of samples before and after water immersion.

2.5 Wettability

Wettability of gmelina and mindi woods was tested by measuring contact angle on the surface of the untreated and heat-treated samples. A laboratory-scale contact angle analyzerwas developed with the design as shown in Figure.2, consist of digital camera with fixed lense, lamp, table to put sample, and a computer.

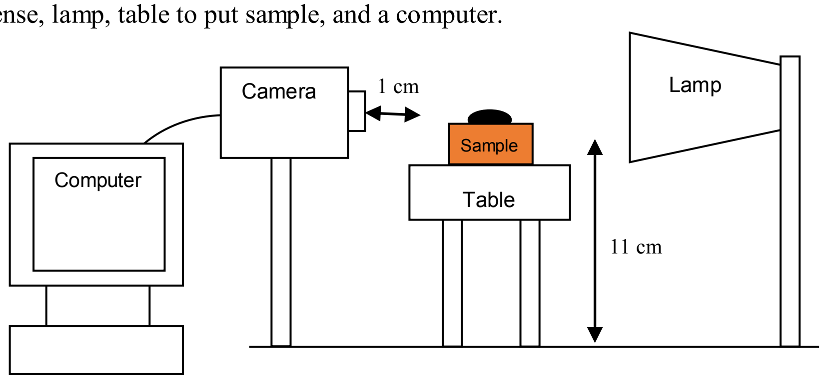


Fig. 2. Design of a lab-scale contact angle analyzer.

Wood sample was placed on the flat table and then a 5 mL liquid was dropped on the tangential surface of the samples. The contact angle formed by the liquid was measured using half-angle method (Figure 3) using the freely-available ImageJ software package [19].

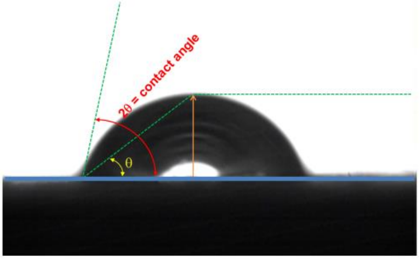


Fig. 3. Measurement of contact angle using Half-Angle Method.

3 Results and Discussions

3.1 Color change

The results showed that lightness (L^*) of untreated gmelina wood was slightly higher that mindi (Fig. 4). The L^* values in gmelina and mindi woods decreased after heat treatment with metal clamp and without metal clamp and the values decreased linearly along with the increase of the treatment temperature, showing similar trend with the previous study [10]. The heat treatment contributed a greater effect on the change of lightness in gmelina than in mindi wood as shown by higher decrease of L^*values in gmelina. Samples with metal clamp of heat-treated gmelina woodhave a higher L^* value compared to samples without metal clamp. In contrast, samples with metal clamp of mindi woodhave a lower L^* value compared to samples without metal clamp.

The untreated mindi wood has a higher red/green chromaticity value (a^*) compared to gmelina (Figure. 4). The a^*values in mindi tends to decrease with increasing treatment temperature while in gmelinathe a^*values increased with increasing treatment temperature. The use of metal clamps tends to increase the value of a^* in both types of wood. Yellow/blue chromaticity value (b^*) of untreated mindi wood was slightly higher than gmelina. The b^*values in mindi wood decreased with increasing temperature, where the sample with clamps shows a greater decrease compared to without clamp (Figure 4). The phenomenon that occured in gmelinawas quite different from mindi, indicated by an increase of the b^*value after heat treatment at 160°C and then decreased dramatically after heat treatment at 210°C with a smaller value compared to untreated wood.

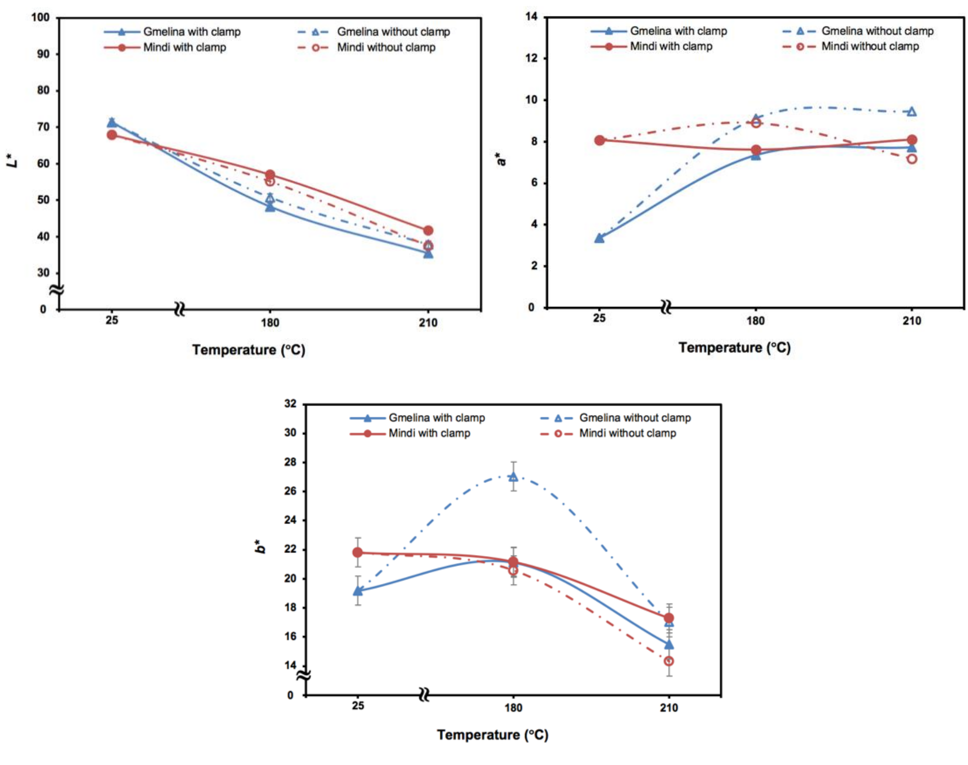


Fig. 4. Effects of temperature and clamping during heat treatment on the change of color parameters.

The overall color change (ΔE^*) increased with increasing treatment temperature, showing greater ΔE^*in gmelina than in mindi wood (Table 1). The results showed that the decrease ofL^* values contributed significantly on ΔE^*values obtained after heat treatment. [14] reported that ΔL^* is the most important parameter affecting ΔE^*. The decrease in L^* values is related to hemicellulose degradation that occurs when the heat treatment process takes place[20].Samples with metal clamp showed lower ΔE^*than samples without clamp, with the exception of gmelina wood heat-treated at 180°C. [12]) stated that the application of metal clamp during heat treatment protected the tangential and radial wood surfaces from direct exposure to hot air, where the the direct contact with hot air occured only in the cross section of the board. This caused the level of evaporation and oxidation of the chemical components of wood to be lower than in samples without metal clamps.

Table 2 shows that the color of gmelina was totally changed after heat treatment at 180°C and 210°C. The color changein mindi wood after heat treatment at 180°C was categorized as very appreciable and the color of mindi wood was totally changed after heat treatment at 210°C. Similar results were observed by [21] who stated that wood color changed drastically after heat treatment above 200°C [21]. [16] revealed that the temperature of heat treatment ranging from 180°C - 200°C are critical temperature points that causes significant color changes on woods [10].

Table 1. Overall color change (ΔE^*) after heat treatment.

Wood species	Temp. (°C)	$\Delta E^*$and the corresponding change[1]	
		With clamp	**Without clamp**
Gmelina	180	25.15 (Totally changed)	18.66 (Totally changed)
	210	36.17 (Totally changed)	38.44 (Totally changed)
Mindi	180	10.92 (Very appreciable)	11.88 (Very appreciable)
	210	27.25 (Totally changed)	36.24 (Totally changed)

[1]According to the ΔE^*classification [14, 15]: a) $0 < \Delta E^* \leq 0.5$ = negligible; b) $0.5 < \Delta E^* \leq 1.5$ = slightly perceivable; c) $1.5 < \Delta E^* \leq 3.0$ = noticeable; d) $3.0 < \Delta E^* \leq 6.0$ = appreciable; e) $6.0 < \Delta E^* \leq 12.0$ = very appreciable; and f) $\Delta E^* > 12,0$ = totally changed.

3.2 Physical Properties

The results showed that the weight loss and volume shrinkage in gmelina and mindi woods affected by heat treatment, showing a linear decrease in values with increasing treatment temperature (Table 2). The weight losing melina wood was higher than in mindi, with the exception of samples heat-treated at 210°C without clamp which showed a smaller value than in mindi. The results might related to differences in density and chemical composition of wood especially the extractives content, where gmelina wood has more extractives(3.0%) than mindi (2.6%) [22, 23]. This is in line with the results of research by [13] who stated that during heat treatment, extractives in wood are easily degraded and evaporated from wood when the heat treatment process takes place.

Table 2. Weight loss and volume shrinkage of gmelina and mindi wood after heat treatment.

Wood species	Temp. (°C)	Weight loss (%)		Volume shrinkage (%)	
		With clamp	Without clamp	With clamp	Without clamp
Gmelina	180	2.06 (0.20)	1.98 (0.20)	1.29 (0.23)	1.09 (0.37)
	210	6.64 (1.08)	7.74 (1.16)	1.95 (0.04)	3.99 (0.16)
Mindi	180	0.43 (0.10)	0.61 (0.15)	1.21 (0.58)	1.12 (0.18)
	210	6.13 (1.20)	10.73 (1.33)	2.34 (0.27)	4.25 (0.37)

*The means are averages of 3 measurements. Numbers in parenthesis are standard deviations.

The weight loss percentages in gmelina and mindi after heat treatment at 180°C could be categorized as low, with the values below 3% while the weight loss in both woodsafter heat treatment at 210°C reached above 3%. Weight loss is a very important parameter to determine the degree of thermal decomposition during heat treatment. [24] stated that a weight loss of at least 3% after heat treatment could increase the dimensional stability of wood while weight loss of at least 5% could increase the natural durability of wood [24]. [13] stated that weight loss and volume shrinkage after heat treatment at temperatures greater than 160°C occurred due to the degradation of extractive, hemicellulose, and a small portion of cellulose molecules in the amorphous region [25]. In other words, the main chemical components of wood constituent cells change in number and dimensions that contributed on the dimensional shrinkage and weight loss after heat treatment.

The untreated gmelina wood has a density of 0.46 g/cm^3, higher than the density of mindi wood of 0.50 g/cm^3(Figure. 5).The density of both wood tends to decrease after heat treatment, particularly in the samples without clamp due to higher degree of weight loss after heat treatment at 210°C. However, gmelinawood and mindi with metal clamp did not show significant changes after heat treatment. This may be due to a proportional decrease of the weight loss and volume shrinkage in both woods when the heat treatment process takes place.

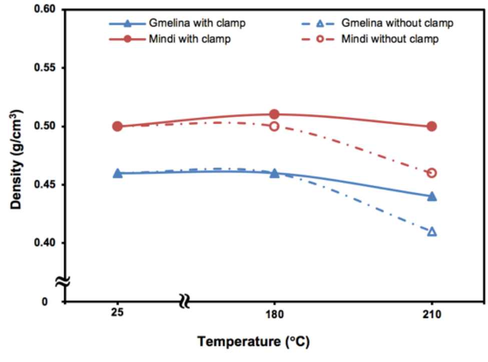


Fig. 5. Effects of temperature and clamping during heat treatment on the density of gmelina and mindi woods.

3.3 Dimensional Stability

The equilibrium moisture content (EMC) in gmelina and mindi woods without treatment was almost similar, *i.e.* 11.68% and 11.53%, respectively (Figure. 6). The values were in accordance with typical EMC conditions in Indonesia that ranging between 12%-19%. The results showed that EMC decreased with increasing treatment temperature, reaching a minimum value of 5.68% in gmelina and 4.08% in mindi. The decrease of EMC in gmelinawas higher than in mindi, or in other words the increase of hydrophobicity in gmelina after heat treatment was higher than in mindi wood. The application of metal clamp did not significantly affected the EMC changes. Research conducted by [23] reported a decrease in EMC in wood spruce from 10.6% to 5.9% and in Scots pine wood from 10.1% to 5.4% after heat treatment. This decrease is caused by the change in hygroscopic properties of the wood cell wall which become more hydrophobic.

Water absorption (WA) of untreated gmelina and mindi woods was 39.66% and 35.22% (Fig. 6). Heat-treated woods showed a remarkably lower WA values and the increase in treatment temperature further decreased the WA values of gmelina and mindi woods. The results obtained were in line with previous studies [8, 10, 12, 24], showing linear decrease of WA with the increase of treatment temperature. The decrease of WA in samples with metal clamp was lower than samples without metal clamp, showing similar results with previous study [9-11]. The decrease in WA was directly proportional to the weight loss, in which the higher weight loss contributed to a higher decrease in WA. The decrease in WA occurs due to the changes in hygroscopic properties, where the woods become more hydrophobic after heat treatment. These changes are caused by a decrease in the number of hydroxyl groups due to chemical reactions during heat treatment that resulted in lower water adsorption [26].

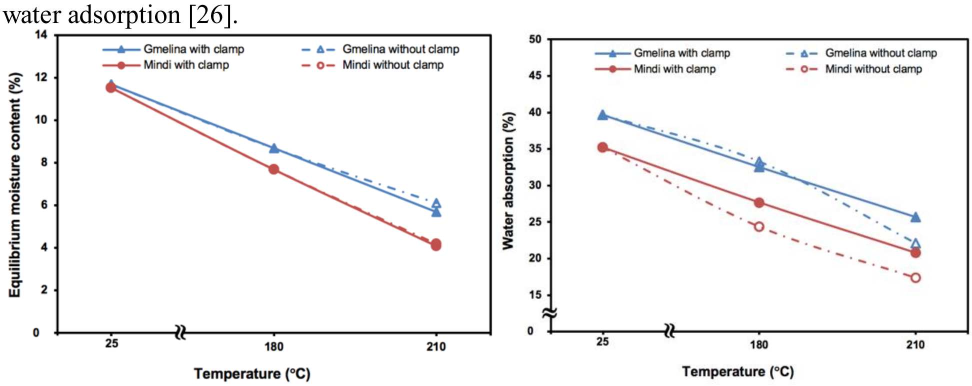


Fig. 6. Effects of temperature and clamping during heat treatment on the change of EMC and WA.

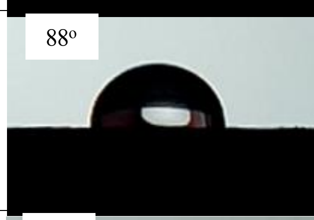
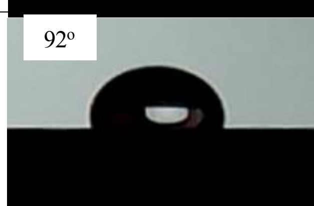
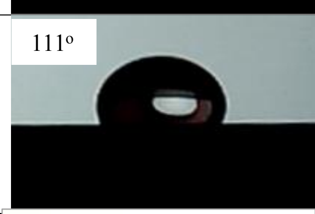
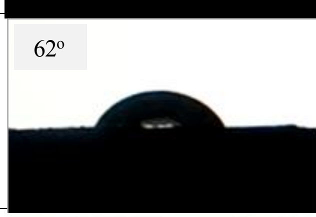
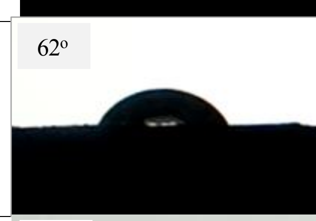
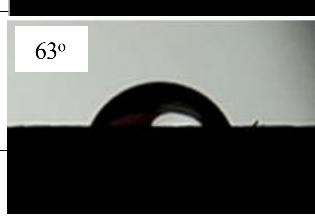
3.4 Wettability

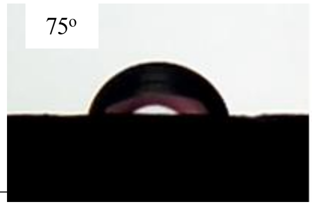
Contact angle represents the wettability of wood, where lower contact angle means higher tendency of wood to absorb water/moisture or in the other word, a higher wettability. The contact angle in gmelina and mindi before and after heat treatment is presented in Table 3. Untreated mindi wood had a greater contact angle than mindi or lower wettability. The contact angles of gmelina and mindi woods increased with the increase of treatment temperature, which means that the wettability of both woods decreased due to heat treatment. The increase of contact angle after heat treat in samples with metal clamps was lower than samples without clamp. The results were linearly related with the increase of weight loss after heat treatment, showing lower weight loss in samples without clamp.

{13] stated that the change of contact angle in wood is affected by the extractives content, where wood with more extractives generally has greater contact angles or low wettability. The observation of contact angle in untreated gmelina and mindiwoods showed opposite results, where gmelina with higher extractive content of 3.0% than mindi of 2.6% had lower contact angle. It was likely related to the density of wood, where gmelina wood has a lower density compared to mindi. The results of contact angles in gmelina and mindi after heat treatment were in line with the statement of [13], showing greater contact angles in gmelina than that observed in mindi wood.

The overall color change (ΔE^*) ingmelina and mindi woods was mainly affected by changes in lightness (ΔL^*) after heat treatment. The ΔE^* increased with the increase of temperature, where the highest ΔE^*observed in gmelina wood. Application of metal clamp during heat treatment limited the exposure of wood surface to heated air and contributed to a smaller ΔE^* value compared to samples without clamps. Dimensional stability of gmelina and mindi woods increased with heat treatment as shown by the decrease of equilibrium moisture content and water absorption with a higher decrease achieved by mindi wood.

Table 5. Visual of contact angles of gmelina and mindi wood before and after heat treatment.

Wood species	**Temp. (°C)**	**Contact angle (°)**	
		With clamp	**Without clamp**
Gmelina	Control		
	180		
	210		
Mindi	Control		
	180		

Wood species	Temp. (°C)	Contact angle (°)	
		With clamp	Without clamp
	210	 75°	 82°

4 Conclusion

Results showed that heat treatment could modify wood color and improve dimensional stability of gmelina and mindi for value added products.

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