ISSN: 0975-8585

Research Journal of Pharmaceutical, Biological and Chemical Sciences

Changes in The Features of Oil Heat Treated 18-Years Old Acacia Mangium.

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ABSTRACT

The changes in the oil heat treated 18 years old of Acacia mangium evaluated. The heartwood and sapwood of Acacia mangium were oil heat treated in palm crude oil at temperatures of 180, 200 and 220°C for 30, 60 and 90 minutes. The evaluation of the changes in the wood was performed by standards using a Minolta Chroma Meter, TAPPI standards, and static bending test, respectively. The relationship between the changes in the color, chemical composition and strength, were made using correlation analysis. The result showed oil-heat treatment reduced the lightness of the wood and darkened the both parts of the wood. In the chemical compositions, the percentages of the holocellulose, α-cellulose, hemicellulose and extractives contents decrease with the increase in treatment duration and temperature. The strength of the wood reduced slightly after the oil-heat treatment. The relationship between wood properties and treatment variables by in the correlation analysis which found that temperature is the dominant variable affected the properties of oil-treated wood.

Keywords: Acacia mangium, oil heat treatment, physical properties, colour changes, strength.

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INTRODUCTION

Deforestation and degradation of natural tropical forest are global issues that changed the landscape of wood industry [1]. The reduction in the deforestation and the protection of natural forest causes a drop in the supply of high quality logs and sawn timbers. This situation in return doubles the plantation forest globally to meet the demand for wood. In Malaysia, several timber species namely Tectona grandis, Pinus caribea, Khaya ivorensis, Hevea brasiliensis and Acacia mangium are planted in the forest plantation program. Acacia mangium has established as main plantation species due to its fast growth rate and adaptability to local soil condition [2]. This fast-growing species has some disadvantages. Among them are a high proportion of juvenile wood, poorly developed heartwood and fast rate of growth that resulted in wide growth rings which eventually will lower the density of wood thus dropped the dimensional stability and readily attacked by biological deterioration agent [3].

Increased in environmental awareness and effective policies that support the use of renewable resources and environmental-friendly chemicals had resulted in high interest for 'non-biocidal' substances. Heat treatment is one of the wood modification processes that are environmental-friendly as it does not use chemicals [4],[5]. The Acacia mangium an exotic timber species to Malaysia was chosen for this study. The study investigated the effect of heat treatment on properties of wood before and after the treatment. The properties that investigated are color, chemical and mechanical properties and the wood species used is Acacia mangium. The result of this study benefits immensely regarding improving technology in treated wood to produce good quality Acacia mangium products.

MATERIALS AND METHODOLOGY

Tree Harvesting

Acacia mangium tree with long straight bole, decay free and minimum branches harvested from Jeli plantation forest, in Kelantan. The trees have 30 cm diameter at breast height (DBH) and height of at least 12 m in height and about 18 years old. The wood segregated into three portions which are the bottom, middle and the top. Then they were into samples of 30 x 10 x 2.5 cm with both portion of sapwood and heartwood on every wood piece. The wood pieces were then condition to 12% moisture content in a conditioning chamber with a temperature of 20±2°C and relative humidity (RH) of 65±5% for two weeks.

Oil Heat Treatment

Three (3) treatment temperatures of 180, 200 and 220°C and treatment durations of 30, 60 and 90 minutes. were used in the process. Palm oil was used as a heating medium as it is organic in nature. A stainless steel tank was used to treat the wood. The tank was three-quarter filled with oil. The wood samples were placed into the tank when the oil reached a temperature of 80°C. The temperatures checked every 10 minutes. After each treatment period ends, the wood removed from the tank while the oil is still hot. The samples were then wiped with a clean cloth to avoid excessive oil from entering the tissue of the wood and then cooled down. The wood was then conditioned in a conditioning chamber set at 20±2°C and RH of 65±5%.

Color Analysis

The color measurement performed on the acacia wood at 180, 200 and 220°C after oil heat treated at durations of 30, 60 and 90 minutes. The wood surfaces were cleaned and marked for consistency at the cross-section surfaces of sapwood and heartwood. The changes in color of the wood surface due to oil heat treatment were recorded using a Minolta Chroma Meter CR-10. Readings were taken before and after the oil heat treatment process. The value of ΔL^*, ΔC^* and ΔE^* were used to calculate the color change before and after the treatment. ΔL^* measured for the change in lightness. The value of ΔC^* is chroma differences. While, ΔE^* indicates total color differences value.

Chemical Analysis

Chemical composition in Acacia mangium before and after heat treatment was analyzed. The chemical components examined were the holocellulose, hemicellulose, α-cellulose, Klason lignin and alcohol-toluene solubility for extractive. All the chemical analysis tests were conducted separately for heartwood and sapwood. The evaluation of chemical constituents of Acacia mangium carried out according to TAPPI standards [6],[7],[8] and Razak et al., (2011) [9].

Sample Preparation for Chemical Analysis:

The wood first chipped for grinding purposes. Samples grounded into powder to pass BS 40-mesh sieve and retained on BS 60-mesh sieve. They were air-dried for three (3) days.

Determination of Holocellulose, α-Cellulose and Hemicellulose, Klason Lignin and Alcohol-toluene Solubility:

TAPPI Standard T204 om-88 (1997) [6] and methods outlines by Razak et al., (2013)[5] were used in determining the holocellulose, α-cellulose and hemicellulose, klason lignin contents and alcohol-toluene solubility before and after undergoing the oil heat treated.

Strength Properties

Samples conditioned to 12% moisture content (MC) in a conditioning chamber set at temperature of 20±2°C and 65±5% RH for at two weeks before testing. The strength tests carried out was the bending strength tests by BS EN 310:1993 [10] using a Testomeric machine number 0500-11213. The specimen size for bending test was 5 cm x 29 cm with an effective span of 150 mm and force applied at the mid-span using a loading head. Samples loaded on the radial surface. The tests stopped when the samples started to break. The proportional limit and ultimate load and deflection recorded while the MOE and MOR value calculated automatically.

RESULTS AND DISCUSSION

Color Properties

After completing oil-heat treatment, oil remaining on the wood surface was absorbed by wood quickly while cooling down causing dry wood surface appeared instantaneously [11]. Oil-heat treatment had caused the surface color of wood to darken. This favorable outcome because it achieved the first objective for this research which is to enhance the color of wood surface especially in sapwood and to homogenize the color between heartwood and sapwood. The consistency of color varies due to variation especially in density and type of wood used [12]. Tables 1-6 showed the results of the color changes before and after undergoing the oil heat treated treatment.

Table 1: Color Values of Heartwood (Bottom portion)

Treatment		Sapwood								
Temp. (°C)	Dur. (min)	L*	a*	b*	L_t*	a_t*	b_t*	ΔC	ΔL	ΔE*
180	30	26.2	10.0	14.2	24.9	10.7	7.9	-4.1	-1.3	6.5
	60	25.1	8.3	8.6	21.7	4.0	2.9	-6.0	-3.4	7.9
	90	28.2	11.9	13.6	23.6	7.9	7.0	-7.6	-4.6	9.1
200	30	25.4	10.3	9.9	23.4	6.3	5.7	-5.8	-2.0	6.2
	60	28.3	10.4	11.1	23.7	5.3	5.5	-7.6	-4.7	9.2
	90	26.7	12.1	13.9	20.9	5.6	5.8	-10.4	-5.8	12.1
220	30	22.3	9.2	7.9	19.7	2.5	2.5	-8.6	-2.6	9.0
	60	21.2	10.2	8.3	15.7	1.6	2.3	-10.4	-5.5	11.9
	90	40.4	10.0	15.7	19.7	7.1	6.0	-11.8	-6.8	13.6

Change in Lightness (ΔL*)

The changes in lightness (ΔL*) for heartwood from bottom to top height portions occurred between -1.3 to -20.0 (Tables 1, 3 and 5). The negative value indicates the decreases in lightness after oil-heat treatment [13], [14]. This resulting from decrement in certain chemical component especially hemicelluloses and lignin

[15],[16],[5]. The values of ΔL* decreased with increased in duration and temperature where the wood surface darkened after treatment, and it is clearly visible to naked eyes. Izyan et al. supports this result. (2010)[4], Cao et al. (2012)[17], Razak et. (2011)[9] and Guller, (2012)[18] in their respective studies.

The heartwood treated for 30 minutes has the smallest change value of ΔL*. The changes increase for treatment duration at 60 and 90 min. The values are low in the bottom portion of the acacia wood which is -1.2, -1.1 and -1.3 for each treatment temp. The middle portions also showed small differences when treated at 30 and 60 min. The top part revealed steadily decreased in a change in lightness for each treatment duration. The values of lightness change decrease with increases in temp. The value of ΔL* more observable when the temperature is above 180°C. The value of ΔL* decreased with increasing in wood portion. For the bottom portions of the heartwood, the value of ΔL* range from -1.3 to -6.8, middle from -4.1 to -16.8 and top -8.4 to -20.0. This showed the wood height portion does not significantly affect the change in lightness.

The values of ΔL* for sapwood were between -14.3 to -27.0. The value of ΔL* for sapwood was much lower than heartwood (Tables 2, 4 & 6). Consequently, through oil-heat treatment, the color of sapwood was slightly darkened compared to the color of heartwood. The wood types (sapwood and heartwood) significantly affecting the ΔL* and proven by the correlation analysis shown in Table 10. The darker color produced caused by the polyphenols compounds that related to the dark color of heartwood that moves to the sapwood and changed the color of sapwood from light to dark [5]. Moreover, the darker color of the wood surface in sapwood is caused by colored staining in outer shell as a result of the accumulation of sugar near the surface during capillary phase [12].

Table 2: Color Values of Sapwood (Bottom portion)

Treatment		Sapwood								
Temp. (°C)	Dur. (min)	L*	a*	b*	L_t*	a_t*	b_t*	ΔC	ΔL	ΔE*
180	30	40.4	10.0	15.7	19.7	7.1	6.0	-9.3	-20.7	23.0
	60	48.6	12.3	22.7	25.5	10.5	8.0	-12.6	-23.0	27.4
	90	51.9	15.0	26.2	26.5	11.5	10.8	-14.4	-25.4	29.9
200	30	52.0	16.0	28.3	29.7	13.1	14.1	-13.3	-22.3	26.6
	60	44.2	13.6	21.9	20.5	6.0	3.7	-17.5	-23.7	30.9
	90	46.5	12.0	21.3	20.6	4.0	3.0	-18.9	-25.9	32.7
220	30	44.8	12.6	20.6	21.9	6.2	4.9	-16.3	-22.9	28.5
	60	40.4	13.4	18.2	15.6	1.6	2.0	-20.0	-24.8	31.9
	90	48.8	11.8	24.9	21.8	5.2	4.7	-21.2	-27.0	34.4

Table 3: Color Values of Heartwood (Middle portion)

Treatment		Heartwood								
Temp. (°C)	Dur. (min)	L*	a*	b*	L_t*	a_t*	b_t*	ΔC	ΔL	ΔE*
180	30	35.6	10.2	12.8	31.5	7.6	7.8	-5.5	-4.1	7.5
	60	37.2	9.7	12.8	32.6	6.9	6.0	-6.9	-4.6	8.8
	90	28.8	10.3	13.3	21.9	5.9	7.4	-7.4	-6.9	10.2
200	30	33.5	10.4	14.9	25.1	5.0	7.2	-9.4	-8.4	12.6
	60	28.4	10.9	14.1	19.6	3.2	4.6	-12.2	-8.8	15.1
	90	34.3	10.0	14.9	22.9	2.3	4.9	-12.6	-11.4	17.0
220	30	36.5	9.0	12.4	22.4	3.0	3.7	-10.5	-13.6	16.4
	60	31.8	11.4	13.4	17.5	2.8	3.0	-13.5	-14.3	19.7
	90	40.0	9.8	14.1	23.2	1.9	2.1	-14.3	-16.8	22.1

Table 4: Color Values of Sapwood (Middle portion)

Treatment		Sapwood								
Temp. (°C)	Dur. (min)	L*	a*	b*	L_t*	a_t*	b_t*	ΔC	ΔL	ΔE*
180	30	42.3	12.1	17.4	26.7	14.3	15.3	-7.0	-15.6	14.6
	60	43.8	12.0	18.0	26.2	13.9	14.1	-8.3	-17.5	18.1

	90	44.6	14.5	18.9	25.4	13.9	16.8	-11.0	-19.3	19.4
200	30	50.0	9.9	21.8	32.0	13.2	15.7	-13.0	-18.0	19.3
	60	40.7	10.5	19.0	20.9	4.1	5.6	-14.7	-19.8	24.7
	90	43.4	10.7	19.7	21.7	4.1	6.2	-16.0	-21.7	26.4
220	30	51.0	12.2	23.0	32.1	6.0	6.8	-16.9	-18.9	25.7
	60	41.2	12.7	23.6	20.2	6.2	6.4	-17.8	-21.0	27.9
	90	46.8	13.1	23.5	22.0	6.0	6.4	-19.1	-24.8	31.1

Table 5: Color Values of Heartwood (Top Portion)

Treatment		Heartwood								
Temp. (°C)	Dur. (min)	L*	a*	b*	L_t*	a_t*	b_t*	ΔC	ΔL	ΔE*
180	30	34.6	9.6	14.6	26.2	8.9	7.6	-5.2	-8.4	10.9
	60	35.3	9.8	15.3	24.0	8.8	9.0	-6.7	-11.3	13.1
	90	37.1	9.9	15.5	23.2	7.2	7.7	-7.8	-13.9	16.2
200	30	35.5	10.6	13.8	24.8	3.7	3.6	-12.1	-10.8	16.3
	60	33.9	10.9	15.9	19.7	4.2	4.7	-12.9	-14.1	18.5
	90	33.2	11.7	15.2	16.3	3.0	3.9	-14.3	-16.9	22.1
220	30	34.0	9.6	14.8	19.2	2.0	3.1	-13.9	-14.8	20.3
	60	37.1	9.9	16.8	19.2	2.6	2.7	-15.9	-17.9	24.0
	90	32.6	11.6	17.5	12.7	1.8	2.5	-17.8	-20.0	26.8

Table 6: Color Values of Sapwood (Top Portion)

Treatment		Sapwood								
Temp. (°C)	Dur. (min)	L*	a*	b*	L_t*	a_t*	b_t*	ΔC	ΔL	ΔE*
180	30	54.3	14.4	26.0	37.8	11.7	19.9	-6.7	-16.5	16.6
	60	41.2	12.6	20.1	22.9	8.2	12.9	-8.3	-18.3	20.1
	90	43.3	9.9	16.2	24.2	8.8	7.5	-9.8	-19.2	21.2
200	30	47.1	11.8	19.7	27.7	9.2	9.6	-9.7	-19.4	21.3
	60	51.3	11.7	22.5	30.6	8.8	8.1	-13.5	-20.7	25.4
	90	40.7	14.0	19.3	19.3	4.1	4.7	-15.7	-21.3	27.7
220	30	46.1	11.9	19.6	23.2	7.2	6.9	-13.0	-22.9	26.6
	60	53.0	11.4	20.2	29.0	4.9	3.5	-17.2	-24.0	30.0
	90	49.7	12.2	21.2	24.7	4.1	3.8	-18.8	-25.0	31.5

The values of ΔL* decreased with increased in the duration of treatment. Compared to heartwood, there were only small and steady differences in reduction of lightness with increasing duration. The lowest decreased in lightness occurred at a top part of the tree for 60 and 90 minutes with -0.6 differences. The comparison between temperature and ΔL* also shows a steady decrease in lightness, but there is a further darkening of wood that treated over 200^{o}C. This result supported by a study conducted by Bekhta and Niemz (2003) [15] and Razak et al., (2013) [5]. The color of sapwood and heartwood tended to be uniform when the temperature is higher than 200°C. It was observed that the wood of middle and the top portions after reaching 200^{o}C experiences decreases of ΔL* up to -3.8. The considerably decreased in lightness changes increases temperature rather than extending the duration of treatment. This is supported by Aydemir et al., 2012 [12] in their study on the effect of thermal treatment on the color of six different wood species. The changes in lightness varies between the tree height portions. The bottom portion has values between -20.7 to -27.0, middle -15.6 to -24.8 and top -16.5 to -25.0.

Change of Chroma Color (ΔC): The values of changes of chroma **(**ΔC) for the heartwood were -4.1 to -17.8. The value decreases with increasing in treatment duration. At the bottom portion, the declined were constant but not more than -2.0. The middle and top portion of the tree experiences slight decreases in ΔC with duration increment. The value also decreases with increasing in temperature treatment. The major dropped occurred when the treatment started at 180^{o}C. This is due to the values of a* and b* affected by temperature below 160^{o}C. The wood portion in height does not cause variation in the value of ΔC. The changes in hue indicate that there are changes in chromophores. It suggested that some molecules with chromophoric function loss and some increase in content and polymerization take place during the heat treatment [19].

The reddish color appearances caused by subsequent formation condensation or degradation products of quinine and quinone methide types where the by-products formed absorbs the complementary light of reddish color and contribute to the redness of wood [20]. It is also related to the greater amounts of Brauns' lignin and proanthocyanidins and extractives content of wood [14]. The yellowish color was caused by low molecular weight phenolic substances and tightly connected to the chemical component of cell wall which are cellulose, hemicelluloses and lignin [20],[14]. Whereas, the blue color is a sign of stain where it is due to the photo-oxidation of lignin and extractives with the production of colored quinine component [4].

The values of ΔC were between -9.3 to -21.2 for sapwood. The ΔC in sapwood is higher than the heartwood since it was originally lighter in color and eventually during the oil-heat treatment, the color changed into dark brown [17]. The difference in wood type is significantly affecting the ΔC according to correlation analysis (Table 10). The values increase with increasing duration treatment. At the bottom and the top portion, the decreases were higher when wood treated for 60 and 90 minutes where the declined are almost identical. Whereas, in the middle portion experiences only slight differences between each length of treatments. The values also decreased with increased in temperature. A significant change in color obtained for sapwood even when it was treated for a short period. The wood started to decreases at the beginning of treatment, and this trend continues at 200˚C where the decreases in ΔC were more evident. The same results were obtained for heartwood [17] where the values were between 170 to 200˚C

In comparison, treatment temperatures have more influence on ΔC than duration for both sapwood and heartwood. It may suggest that increases in temperature can reduce the length of heat treatment to obtain desired wood color. Besides, the slow color formation produced at a lower temperature can balance with prolonging the treatment duration [19].

Total Color Differences (ΔE):

The total color differences (ΔE) indicates the degree of overall chemical modification where the degradation products are hemicelluloses, lignin and extractives compounds participating in the color formation of the wood [19],[20]. The values of ΔE increases with increases in treatment duration. At the bottom and middle portions, only slight differences in the value of ΔE for 30, 60 and 90 minutes at 180°C. The values of ΔE increases with the increased temperature of oil-heat treatment. The increased in ΔE were more noticeable with increasing temperature for the middle and top portions.

The treatment temperature and duration influenced the value of ΔE. Temperature is significantly affecting the ΔE more than the treatment duration. The rate of change increases with increases in temperature under the same duration, but decreases when the duration extends even at a maintained temperature [17]. This explained by the small differences in color of wood treated at 30, 60 and 90 minutes at the same temperature. The value of ΔE increases with increasing temperature as observable in the middle and the top portions when treated at 200ºC and 220ºC rather than 180°C where the value of ΔE is larger. The heartwood has the ΔE around 6.5 to 26.8 which were lower than sapwood. This due to the original color of sapwood is lighter than heartwood. Like ΔL* and ΔC, the wood type (sapwood and heartwood) significantly affecting the ΔE. The oil-heat treatment turned the sapwood color more noticeable than the heartwood. No significance obtained on the ΔE between the tree portions sampling heights of the bottom, middle and top for both heartwood and sapwood are observed.

The ΔE caused by the formation of secondary condensation products and degradation products. The degradation products include the reactive compounds from cleavage of α- and β-aryl ether bonds in lignin and hemicelluloses [20]. It also correlated with holocellulose content since it is easily degraded during heat treatment process [16]. Table 7 showed the correlation between the total color differences with extractive, holocellulose, cellulose and hemicelluloses contents. Temperature significantly affected all the wood properties except for the change in lightness.

Table 7: Chemical Compositions of Heartwood

Treatment	Extractives (%)	Lignin (%)	Holocellulose (%)	Hemicellulose (%)	Cellulose (%)
Control	13.9	22.3	75.5	24.7	50.8
180°C, 30 min	12.9	24.3	73.5	24.0	49.5
180°C, 60 min	11.3	24.8	71.9	23.1	48.8
180°C, 90 min	8.8	25.5	68.9	21.2	47.7
200°C, 30 min	10.3	24.6	67.6	20.3	47.4
200°C, 60 min	8.4	25.2	66.4	19.5	46.9
200°C, 90 min	5.3	26.4	64.7	18.9	45.8
220°C, 30 min	8.8	25.5	64.5	19.2	45.3
220°C, 60 min	5.5	26.2	62.1	18.3	43.8
220°C, 90 min	2.0	27.3	61.1	17.9	43.2

Duration of treatment is an important variable that affected the oil-heat treated wood beside temperature but was less significantly affected the color properties after treatment where it was only significantly affected the chroma changes. It may lead due to shorter duration of treatment which insufficient to modify the wood properties. The type of wood which is sapwood and heartwood shows significant differences in all color properties. Color significantly affected by the type of wood because A. mangium have a different color of sapwood and heartwood and through oil-heat treatment, the color of sapwood is slightly darkened to match the color of heartwood thus it affecting the color properties.

Chemical Compositions

There exist a relation between oil-heat treatment and chemical composition of A. mangium where the chemical compositions either degrade or improved during the treatment process. Klason lignin improved in composition during treatment while the extractives, hemicelluloses, holocellulose and α-cellulose degrade. The change in chemical composition able to explain the change in color and mechanical properties of oil-heat treated wood.

Holo cellulose

The holocellulose content in the heartwood of A. mangium control samples were 75.5% and sapwood 72.2%. The oil-heat treatment caused a reduction in the holocellulose composition in the heartwood. The highest decreases in holocellulose content occurred when the wood was treated at 220°C for 90 minutes with 61.1% of holocellulose remained while the lowest reductions happened when the wood treated at 180°C for 30 minutes with 73.5% of the holocellulose remained.

Extending the treatment duration further decreases the holocellulose content. At 180°C, every increment in duration doubled the loss in holocellulose content. Treatments at 200°C and 220°C with increment in duration decrease the holocellulose content steadily. However, the treatment duration does not significantly affect the holocellulose content (Table 10).

The holocellulose contents decrease with increases in temperature [21]. When the temperature of treatment reaches 180°C, the holocellulose experiences loses of 8.7%, at 200°C 14.7% and 220% 19.1%. The significant changes in holocellulose content after 200°C also reported by Tumen et al., 2010. The lowest decreased in holocellulose occurred at 180°C for 30 minutes which was 2.8% and highest at 220°C for 90 minutes which has degradation of 16.9%. The loss of holocellulose increases with increases in treatment duration. However, only slight decreases in holocellulose content occurred at extended treatment duration. The highest difference in holocellulose loss was only 2.3%.

An increase in temperature degrades holocellulose. However, the degradation rate of holocellulose in sapwood is slower than heartwood. At 180°C, the highest loss in holocellulose is 8.2%, at 200°C is 11.2% and at 220% is 16.9%. The slower rate of holocellulose degradation may relate to the ability of holocellulose to degrade at a lower temperature as low as 100°C [5],[22]. The oil-heat treatment causes a decrease in holocellulose content significantly as was reported by [21].

The α -Cellulose

The control heartwood has α-cellulose content of 50.8%. The cellulose content in Acacia mangium ranged 34% to 47.2% [23]. Untreated Acacia mangium possess higher α-cellulose content than treated samples because thermal treatment causes cellulose to degrade due to the split of high-molecular-weight fractions [21]. The α-cellulose decreased with treatment durations and temperatures. The highest reductions occurred at 220°C for 90 minutes duration with 15% of α-cellulose degraded, and the lowest degradation took place at 180°C for 30 minutes.

At 180°C and 200°C, the decreases in α-cellulose at 30 and 60 minutes are almost the same and significant reduction in α-cellulose occurs at 90 minutes. While, at 220°C, declines in α-cellulose occurs almost at a steady rate where α-cellulose degrade for 10.8%, 13.8% and 15.9% for 30, 60 and 90 minutes, respectively. Gawron et al., (2011) [24] also observed the decreases in α-cellulose in a given duration., in research on beech wood.

Reduction in α-cellulose content was also affected by the increases in treatment temperature. Wood treated at 180°C possess lowest α-cellulose contents due to the losses in cellulose at 200°C which were more rapid and double the cellulose degradation at 180°C. Tumen et al., 2010 [25] also reported the similar observation from their research. This is due to the reduction in the degree of polymerization (DP) of cellulose which started at 150°C [26]. At 200°C the formation of volatile products such as levoglucosan, anhydroglucose, furan and furan derivatives occurred [27]. In this study, the controlled wood contains 49.8% of α-cellulose. The oil-heat treatment causes the α-cellulose to degrade up to 13.9% until only 42.9% of cellulose remained in 220°C for 90 min. treatment.

The losses of α-cellulose were related to treatment duration. There were slight differences in the percentage of α-cellulose between three (3) different treatment durations with the highest differences in 2.8% reduction. Reductions in α-cellulose were also affected by the temperature. Like heartwood, a significant decrease in α-cellulose occurs when the temperature of treatment was at 200°C. The losses of α-cellulose which was related to the degradation of the less orderly area in crystalline cellulose and deterioration of the amorphous area in cellulose when the temperature was up to 200°C [28]. The cellulose degradation is only slightly affected by thermal treatment due to the high stability of cellulose and its high molecular weight [29],[9],[36]. Cellulose degrades at a higher temperature than hemicelluloses, pectins, and starch [9]. This proven by the largest percentage of α-cellulose degradation at 15% while the highest hemicelluloses loss at 27.5%.

Hemicellulose

The control wood of A. mangium heartwood contains about 24.7% of hemicelluloses. The degradation of hemicelluloses occurred with increasing duration of treatment. However, only slight decreases occurred for an extending of treatment duration and the highest declines recorded at only 1.9% of holocellulose difference [24]. The hemicellulose contents were also affected by the increases in temperature during treatment. Hemicellulose started to degrade at 180°C was gave significant changes [5]. Degradation of hemicellulose is due to the formation of formic and acetic acid that formed mainly from O-acetyl-galacto-gluco-mannan under the influence of temperature [24].

`In sapwood of oil treated A. mangium, the untreated samples contain 22.4% of hemicelluloses. The oil-heat treatment had affected the hemicelluloses to degrade, and the highest degradation of hemicelluloses occur at 220°C at 30 minutes where only 17.1% of hemicelluloses remained after the loss about 23.7% of hemicelluloses content.

The increase in duration caused decreased in hemicellulose content. However, there were only slight decreased in hemicelluloses content with increasing duration. For 180°C and 200°C, the significant reduction in hemicellulose occurs at 60 minutes may due to the lower temperature of treatment thus need longer treatment duration for the reduction in hemicellulose. While at 220°C, there are not many differences in loss of hemicellulose between each duration of treatment [30]. Treatment at 200°C significantly decreases the hemicellulose content where the decreases almost tripled compared at 180°C. The reduction in hemicelluloses contents continued at 200°C and 220°C.

Klason Lignin

The Klason lignin content in the heartwood of A. mangium is 22.3% whereas in sapwood is 20.1%. The lignin content in wood is between 18 to 35% [31]. However, a study by Mohammed et al., (2011) [23] stated that lignin content in Acacia magnum is between 10.5 to 20.5%. The oil-heat treatment had caused an increase in Klason lignin content. The low increment in Klason lignin content for heartwood occurred at 180ºC for 30 minutes which were 9.0% and highest at 220ºC for 90 minutes which are 22.4%. The Klason lignin content increased with increased duration of oil-heat treatment. There is a slight increase in Klason lignin content for 30 and 60 minutes treatment duration. Considerable increase in Klason lignin content when the temp. Increases from 200ºC to 220ºC. Yildiz et al., (2006)[27] and Tumen et al., (2010)[25] made similar observations which supported this finding in their studies separate studies. There was a substantial alteration in lignin structure occurred [32].

Polycondensation reactions occur during heating by increased cross-linking with separated substances from hemicellulose and cellulose that leads to stabilization or increase in lignin proportion [33],[25],[26],[34]. However, lignin degradation also occurs during oil-heat treatment but resulting in the formation of a conjugated structure that contributes to the color formation which eventually caused increased in lignin content [20]. The Klason lignin content in sapwood increased with an increment of treatment temperature. Even though there is only a slight increased, the treatment temperature still becomes a major factor for lignin content increment rather than duration because high temperature of treatment helps to degrade wood materials and increase the Klason lignin content [35].

Extractives

Based on the experiment conducted, the heartwood of untreated A. mangium contains 13.9% of extractive while the sapwood there were 9.1% of extractives. Normally, the extractive content in wood is around 20% [31].

The oil-heat treatment causes the decreases in extractive content. The lowest dropped is the wood treated at 180ºC for 30 minutes which were 12.9% extractive remained after declined by only 7.2%. While wood treated at 220ºC for 90 minutes has the highest decreased in extractives where only 2.0% extractive remained after shrank by 85.6%. The percentage of extractive decreased with increased in the duration of treatment. Increased in duration at the same temperature almost doubled the loss in extractives. This can be observed from treatment at 180°C where percentage decline is 7.2%, 18.7%, and 36.7% for 30, 60 and 90 minutes respectively. For the wood treated at 200°C and 220°C, the trend for percentage losses varies.

The amount of extractive content decreases rapidly with the increases in temperature during oil-heat treatment. Considerable reductions in extractive content occurred the most when the temperature of treatment was between 180ºC and 200ºC. The declined in the content were the highest at this point since at this temp., the wood started to undergo chemical transformation after the loss of free and bound water [36]. Moreover, the temperature used in the treatment are enough to eliminate natural resins during term rectification process which caused the extractives to degrade [37].

Even though most original extractives disappear from wood with heat treatment, the extractive content increases significantly with the mass loss than decreased. The significant increment was due to the water and ethanol extractives as a result of polysaccharide degradation [36].

The lowest reduction in extractive was at 180ºC for 30 minutes which 7.4% extractive remained after loss for about 18.7%. The wood treated at 220ºC for 90 minutes have the highest reduction in extractives where only 1.7% remained after reduction of 81.3%.

Extractives of wood mostly disappeared or degrade during the thermal treatment especially volatile substances. However, new compounds can also be extracted from wood that resulted from degradation of cell wall structural components. These compounds are waxes, carbohydrates, tannins, resins and small amounts of hemicelluloses. While substances like fats and waxes moves along the axial parenchyma cell to the surface of the wood and expected to disappear [36].

The heartwood has higher extractive content than the sapwood. This is due to the nature of the wood where extractive mainly located in the dead part of the tree [38]. The treatment temperature influences extractive degradation significantly [36]. This is proven through correlation analysis where the temperature was significantly affecting the extractive whereas duration does not affect the extractive content (Table 10).

Strength Properties

The oil-heat treatment some reduction in the strength of the acacia wood in both the Modulus of Rupture (MOR) and Modulus of Elasticity (MOE). This is due to the degradation of chemical contents especially cellulose which responsible for wood strength due to its long parallel and straight molecules packed together [19]. By manipulating the temperature of treatment, the reduction in wood strength can be avoided or minimized.

Modulus of Rupture (MOR)

Table 9 shows the MOR of oil-heat treated A. mangium wood. The MOR of control samples of taken at the bottom, middle and top portions of this study is 108.2N/mm^2, 105.4 N/mm^2 and 105.0 N/mm^2 at 12% MC respectively. The increases in duration of treatment decrease the MOR of the wood. In the bottom and middle portions of the wood, increases in duration not affecting the MOR substantially where the highest percentage change is only 6.5% loss when wood treated at 60 and 90 minutes. Whereas at the top part of the tree, the decreased in MOR is higher with increased in treatment duration with at least 4% of MOR decreased. However, similar to bottom and middle part reduced in MOR is significant when wood is treated at 60 minutes. Kubojima et al., (2000)[39] studied on the bending strength of Sitka spruce also observed the similar outcome. Nonetheless, even though duration affected the properties of wood, it is less significant.

At the bottom portion of the tree, MOR loss 5% when wood treated at 180°C and 200°C. While in middle and top parts, MOR loss doubled at every increased in treatment temperature. Increased in temperature significantly affected the MOR of treated wood (Table 9). Kubojima et al., (2000)[39], Poncsak et al., (2006)[40]; Korkut & Hiziroglu, (2009)[41]; Sahin, (2010)[42] and Kesik et al., (2014)[43] supports this where they suggested further heating will decrease the MOR significantly.

At the bottom and middle height of wood, the values of decreased in MOR were almost the same with the highest percent change of 18.9% and 16.6% respectively. While at the top part, the largest percentage change occurred at 220ºC for 90 minutes where 24.9% of MOR reduction. This is in agreement with Sundqvist, (2004) [19], where strength loss in thermally treated wood was between 0 to 30% depending on the process, and at 220ºC, the strength loss is rapidly decreased up to 30%, the MOR decreased by about 24% with heat treatment. It is presumed that changes in mechanical properties are related to the formation and accumulation of organic acid in wood which emits to the surrounding atmosphere during the process [19].

Table 8: Chemical Compositions of Sapwood

Treatment	Extractives (%)	Lignin (%)	Holocellulose (%)	Hemicellulose (%)	Cellulose (%)
Control	9.1	20.1	72.2	22.4	49.8
180°C, 30 min	7.4	22.6	70.2	21.9	48.3
180°C, 60 min	6.8	23.1	69.6	21.5	48.1
180°C, 90 min	6.6	24.4	68.7	20.8	47.9
200°C, 30 min	5.3	23.2	66.3	19.6	46.7
200°C, 60 min	3.0	24.1	64.6	18.7	46.0
200°C, 90 min	2.8	24.2	64.1	18.4	45.7
220°C, 30 min	2.8	25.1	62.1	17.8	44.3
220°C, 60 min	2.2	25.7	60.9	17.4	43.5
220°C, 90 min	1.7	26.5	60.0	17.1	42.9

The heat treatment had caused a reduction in strength to the treated samples due to thermal degradation and loss of substances due to depolymerization reaction of wood polymer especially changes in hemicelluloses and cellulose contents [44],[40],[44]. Moreover, viscosity and plasticity of wood are believed to affect the wood rupture in treated wood [39]. In this study, the reduction of MOR occurred because the heat treatment was done over 180^{o}C. The effect on decreased in strength and wood brittleness were believed to start when the temperature of treatment over 175^{o}C [27]. However, a study by Cao et al., (2012) [17] suggests that mechanical properties can enhanced by lower temperature and shorter duration of treatment.

Modulus of Elasticity (MOE)

The average MOE of oil-heat treated Acacia mangium was 6992.4N/mm^2. The strength of the wood, however, varied between the wood height portions. Table 9 shows the MOE of oil-heat treated A. mangium wood. MOE of treated wood decreased with extended duration. However, in each sampling height, there are only slight differences between increased in the duration of treatment which is not more than 3% strength loss. The duration does not cause a reduction in MOE of treated wood. While for temperature, in each sampling height, increased in temperature from 180°C to 200°C almost tripled the MOE loss. However, the rate of loss in MOE slightly slowed when wood treated at 220°C. Therefore, temperature significantly affected the oil-heat treated wood.

Table 9: Modulus of Rupture (MOR) and Modulus of Elasticity (MOE)

Treatment		Bottom		Middle		Top	
Temp. (ºC)	Dur. (min)	MOR (N/mm^2)	MOE (N/mm^2)	MOR (N/mm^2)	MOE (N/mm^2)	MOR (N/mm^2)	MOE (N/mm^2)
Control		108.2	6876.7	105.4	6988.6	105.0	7111.8
180	30	102.8	6665.9	102.6	6835.5	101.0	6966.7
	60	101.1	6500.7	100.7	6795.9	97.0	6897.6
	90	98.4	6332.3	98.5	6662.4	91.5	6866.7
200	30	98.4	6296.7	98.3	6261.7	99.2	6660.9
	60	92.0	6164.9	95.3	6174.2	93.8	6587.0
	90	89.6	6006.8	93.8	6037.1	85.0	6570.5
220	30	94.4	6001.0	91.4	5902.8	95.1	6333.1
	60	91.5	5894.5	89.6	5853.1	85.1	6220.4
	90	87.8	5736.4	87.9	5745.1	78.9	6209.9

The oil-heat treatment had caused MOE to decrease because MOE starts to decrease when the temperature is over 100^{o}C [15]. However, the reduction in MOE varied with the only slight reduction that depending on species and treatment schedule [41].

In bottom and middle part of the tree, there was considerably decreased in MOE at different temperature and duration, but for the top part, the decreased in MOE were almost the same throughout the duration and temperature of treatment. For all parts, the highest reduction in MOE occurred at 220^{o}C for 90 minutes where the percent losses were 16.6%, 17.8% and 12.7% for the bottom, middle and top. Sampling height significantly affected the value of MOE (Table 4.1). The reduction in MOE of heat treated wood is consistent with research carried out by Tankut et al., (2014)[45] which state that loss of MOE in heat treated wood is between 19% and Esteves et al., (2009)[31] and Sahin, (2010)[42] suggest that the reduction in MOE can be around 1 to 72%.

Similar to the MOR, reduction in MOE caused by the thermal degradation results in loss of chemical substances after oil-heat treatment where it is due in large part to de-polymerization reaction of wood polymers especially hemicellulose. Hemicellulose is essential for wood strength but at high temperature, it can degraded due to its lower molecular weight, thus it reduces the wood strength [38]. Other chemical components also degrade but it takes place at a higher temperature than hemicellulose, and they are responsible for the loss of wood strength.

Correlation Analysis

Correlation analysis was used to study the relationship between the 18-year-old acacia wood properties with color, chemical and mechanical changes in the oil heat treatment process (Table 10). Changes in lightness correlated with all wood properties except lignin and MOE. The chemical composition of wood was highly correlated with the lightness of wood especially hemicellulose where decreases in lightness primarily caused by degradation of hemicellulose particularly pentosans [15],[16]. Moya et al., (2012)[46] and Feher et al., (2014)[14] stated a relation between lightness and extractive content in wood. Todorovic et al., (2012)[47] also suggest that MOR correlated with lightness in wood.

Table 10: Correlation between color, chemical composition and mechanical properties of 18-years old A. Mangium

	ΔL*	ΔC	ΔE	Extractives	Lignin	Holo-cellulose	Cellulose	Hemi-cellulose	MOR	MOE
ΔL*	1.000	0.774**	0.961**	0.852**	-0.244^{ns}	0.730**	0.692**	0.737**	0.704**	0.125^{ns}
ΔC		1.000	0.901**	0.918**	-0.508*	0.910**	0.880**	0.907**	0.818**	0.515**
ΔE			1.000	-0.895**	0.371^{ns}	-0.817**	0.794**	0.838**	0.688**	0.607**
Extractives				1.000	-0.315^{ns}	0.853**	0.809**	0.865**	0.739**	0.656**
Lignin					1.000	-0.630**	-0.692**	-0.547*	-0.802**	-0.790**
Holocellulose						1.000	-0.984**	-0.984**	0.930**	0.919**
Cellulose							1.000	0.937**	0.921**	0.925**
Hemicellulose								1.000	0.907**	0.885**
MOR									1.000	0.561**
MOE										1.000

ns = not significant, * = significant at p≤0.05, ** = significant at p≤0.01.
ΔC = chroma changes, ΔL* = changes in lightness, ΔE = total color differences.

Unlike lightness, chroma changes correlated with all the wood properties. Lignin is related to the red color in the wood [48],[14]. While, yellow related with the photochemistry of chemical composition in cell wall including cellulose and hemicellulose, and wherein lignin was caused by lignin parts with quinonoid and stilbene structure [19],[14]. It also caused by the organometallic complex in extractive [19]. Besides that, grayish appearances in wood are caused by cellulose and hemicellulose [49],[14].

The total color difference correlated with each wood properties except lignin. For this reason, the total color difference can be an indicator of chemical contents especially holocellulose [20],[16]. Bekhta & Niemz, (2003)[15] and Todorovic et al., (2012)[47], also stated that the total color differences were related to the wood strength, especially MOR.

Holocellulose positively correlated with all wood properties. Degradation of holocellulose content are caused by hemicellulose and cellulose because they positively correlated [5]. Cellulose also positively correlated with all wood properties especially in wood strength where the structure of cellulose which are long, parallel, straight molecules that packed together are necessary to the wood strength [40]. Therefore, break-up in cellulose structure lowers the MOR of wood.

Hemicellulose also correlates with all wood properties. In the heat treatment, the color of the wood modified due to the released of by-products from hemicellulose degradation [3]. The decline of hemicellulose caused decreased in lightness [14]. Besides, loss in hemicellulose cause increase in the degree of polymerization in wood thus related to the degradation or rearrangement of amorphous cellulose [17]. In mechanical properties, it plays an important role especially glucomannan where degradation of hemicellulose caused mass loss hence reduce the wood strength particularly MOR [42],[50],[51],[52].

Lignin correlated with all wood properties except lightness, total color differences, and extractive content. Even though lignin only related with chroma color, degradation of lignin would cause color change [53]. Lignin also correlates with hemicellulose and cellulose content wherein heat treatment degradation of hemicellulose and cellulose increase the lignin content [26].

Extractive content positively correlated with all wood properties except lignin. The color of treated wood is primarily determined by extractive content where extractive is the first chemical transformation occur in treated wood even at a low temperature [54],[14].

MOR of the wood positively correlated with all wood properties, but MOE positively correlated with all wood features except lightness. The color of the wood has a strong correlation with wood strength [15]. While, in heat treatment, depolymerization of cellulose and hemicellulose caused the wood to turns brittle and lowered the mechanical strength of the wood depending on treatment level [55].

CONCLUSIONS

The oil heat treatment process enhanced the color of sapwood to match the heartwood of 18-year old A. mangium.

Acacia wood treated at 200°C and 220°C for the duration of 60, and 90 minutes showed considerable changes in the chemical composition. Holocellulose, hemicellulose, α-cellulose, extractive degraded and the Klason lignin increases in contents after the oil-heat treatment process.

The MOR and MOE of A. mangium wood decreased after treatment. The MOR and MOE decrease with increases in temperature. Durations of treatment affected the MOR values.

The oil heat treatment process at 200°C for 60 min. is recommended for acacia mangium wood as it improved the color of A. mangium and standardized the color of sapwood and heartwood. The loss in strength at this temperature and duration is acceptable as the treated wood only loss up to 15% strength in MOR and 10.7% in MOE.

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