

The Characterization of Black-streaked Heartwood in Teak: Inter-tree Variation

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Abstract

The objective of this study was to investigate the variation in the color and chemical characteristics of black-streaked heartwood of teak and explore the relationship of these chemical properties with the degree of blackening. The samples used were outer heartwood parts from 11 trees with black streaks both thin and thick and 7 trees with normal heartwoods for comparison. The colorimetric analysis in CIEL*a*b* system was used to determine the brightness values (L^*) of black-streaked heartwood of teak ranging from 40~49 and a thick portion produced appreciably higher average values of extractive contents including n-hexane, ethyl acetate, and total extractive content as well as tectoquinone contents and pH values but lower squalene content compared to those in normal wood. The degree of blackening in the black-streaked heartwood was highly correlated to its extractive contents, especially the ethyl acetate soluble extractive content ($r = -0.94$) while moderate correlations were measured between the brightness index and tectoquinone content (negative) and squalene (positive). Moreover, no significant difference was found in the ash and individual inorganic elements contents between the group. The increase in pH values was observed to have corresponded with a decrease in brightness ($r = -0.75$). Therefore, the blackening was assumed to be due to the polymerization of quinones in weakly acidic conditions.

Keywords: darkening wood, quinones, extractives, inorganic materials, pH values.

Introduction

The discolorations in timber and lumber are one of the most serious defects in numerous economically important species of wood from tropical regions. An example of this is teak which is widely used for furniture or parquet and observed to have black streak discolorations leading to considerable degradation and economic loss. These black-streaked trees are locally known as *doreng* in Indonesia with the streak generally in the form of an annual ring in the transversal section as shown in Figure 1. The cause of this phenomenon remains unknown but has been related to the edaphic factor by Suhaendi *et al.* (1998).

Wood color is generally ascribed to the composition and structure of the extractives (Hillis 1987). This is evident in a previous report by Lukmandaru *et al.* (2009) which compared the color, pH, extractive content and components, and inorganic materials between normal and black-streaked wood in a radial position using partially black-streaked heartwood in only one sample. The relationship between the darkening part and the chemical properties of wood has also been observed in other species with different results (Kubo and Ataka 1998; Minato and Morita 2005; Romagnoli *et al.* 2013). Therefore, this study clarified the color and chemical characteristics of the black streak portion using a larger number of trees having black streak from the inner towards

the outer heartwood part. It also examined the relationship between chemical properties and the degree of blackening.

Materials and Method

Sample Preparation

The trees used in this study were collected from the Perhutani plantation, Randublatung, Central Java which is a with black calcareous soil and characterized by the frequent occurrence of black-streaked trees. The trees were between 25~45-years old, straight-stemmed, and sound with 5 cm discs obtained at different heights from the butt end of the trunks. Furthermore, wood powder at 40~60 mesh of the discolored trees was obtained from the outer heartwood part (0.5 cm from sapwood heartwood border in cardinal directions) for color and chemical analysis. Different black-streaked disks were collected using visual inspection and the brightness (L^*) value was measured by spectrophotometer after which they were classified into thick with $L^* < 45$ having six individuals and thin with $L^* = 45$ to 50 containing five individuals. In addition, seven disks were obtained from different trees with normal heartwood in the same sites with $L^* > 50$. The sample conditions are, therefore, described in Table 1.



Figure 1. Black-streaked heartwood of a teak tree (cross section)

Color Properties

The color of the specimens was measured with an NF777 spectrophotometer (Nippon Denshoku Ind. Co Ltd) having a sensor head of 6 mm in diameter and using illuminant D65, 10° viewing angle. The CIE color data (L^* , a^* , b^*) were obtained in triplicate where L^* is for lightness representing the position on the black-white axis ($L = 0$ for black, $L = 100$ for white), a^* = green-red coordinate and b^* = blue-yellow coordinate. The color saturation (C) was calculated using $C = \sqrt{[(a^*)^2 + (b^*)^2]}$ while the tonality angle (hue) was determined according to $H = \arctan(b^*/a^*)$.

Chemical Analyses

The extractive content was determined using 2 g wood meal based on oven dry weight in a Soxhlet apparatus and extracted using a 6-h sequence of *n*-hexane, ethyl acetate (EtOAc), and methanol (MeOH). The solvents were evaporated in vacuo after which they were dried and weighed to determine the *n*-hexane, EtOAc, and MeOH based on an oven-dry wood meal (m/m) and the Total Extractive Content (TEC) value, which was obtained by summing all the values for each of the contents. Meanwhile, the solubility in cold- and hot-water (ASTM D-1110-1984) was determined separately.

The contents of the main extractive components in the ethanol-benzene extracts were measured using Hitachi G-3500 GC and Shimadzu QP-5000 GC-MS with the detector being FID, the column was 30 m NB-1 bonded capillary, column temperature was 120~300°C (programming 4°C min⁻¹), detector and injector temperature at 250°C and held at 300°C for 15 min, while the carrier gas was helium after which one μL of the solution at 100 mg mL⁻¹ concentration was injected manually into the gas chromatograph. Moreover, for the quantification of individual substances, calibrations were made using known amounts of standard tectoquinone (2-methyl anthraquinone) while compounds were identified by comparing their mass spectra with data

from previous studies (Perry *et al.* 1991; Lemos *et al.* 1999) and the injection of standards. GC-MS (JEOL XS mass spectrometry at 70 eV) was used for gas chromatographic separations.

The ash content was measured according to the ASTM D-1102-1984 standard method. Moreover, the wood powder samples at 0.2 g were prepared for elemental analysis using a nitric acid-perchloric acid (5:3, v/v) solution using a digestion procedure. The amount of silica was also determined by filtering the solution while the contents of some inorganic materials such as calcium (Ca), magnesium (Mg), potassium (K), iron (Fe), and phosphorous (P) were measured through atomic absorption spectrophotometry (Hitachi Z-5000) and flame-photometry. Furthermore, 1 g per part of the wood powder was submerged overnight in 20 mL distilled water and the pH of the filtrate was measured with a pH meter (Horiba). Three measurements were made for each part.

Statistical Analysis

Data were tested to confirm a normal data distribution (Kolomogorv-Smirnov test). The effects of grouping (brightness level) were calculated by analysis of variance (One-way ANOVA) GLM procedures. The effects were taken into account only when significant at the 95% level using Type III Sums of Squares. The relationships between the brightness index and other independent characteristics were studied with Pearson's or Spearman's correlation analysis. All statistics were calculated with SPSS 10.0 software.

Results and Discussion

Color Properties

The brightness measurement in Table 1 shows the samples were simply divided into three groups of normal, thick, and thin black-streaked woods. The brightness values

(L*) of black-streaked heartwood were classified in the range 40–49 while the lowest value for normal heartwood was 51. Previous studies on the teak grown in India (Bhat *et al.* 2005) and Togo (Kokutse *et al.* 2006) were recorded to have brightness index ranged between 48 and 58 while Moya and Berrocal (2010) reported 46 to 76 for heartwood from teak trees growing in Costa Rica. Even though different color equipment was used as well as the effect of other factors such as genetics, edaphic, tree age, and others on

the color properties, the ranges showed the possibility of finding black streak in each locality. As expected, the black streak areas provided significantly lower average values in yellowness (b*), hue, and chroma as shown in Table 1 while no significant differences were observed between the thick and thin samples using Duncan test. Moreover, the redness values (a*) in the black-streaked samples were found to be higher than in normal wood but Duncan test showed the difference is not statistically significant.

Table 1. Description of the sample conditions and colour properties in teak heartwood of black-streak and normal.

Samples		Tree Ring Number	Disc Diameter	Color properties				
				L*	a*	b*	Hue	Chrome
Thick black streaked								
No tree	1	35	33.0	40.50	7.30	21.10	70.96	22.67
	2	29	23.8	41.40	6.80	20.50	71.78	21.61
	3	29	24.0	42.00	7.10	21.70	71.94	22.80
	4	28	24.7	43.00	6.80	21.20	72.12	22.30
	5	37	27.0	44.80	7.90	23.00	71.10	24.36
	6	37	31.8	44.99	7.20	22.30	72.07	23.47
			Average	42.78 a	7.18	21.63 d	71.66 f	22.86 h
		Std. dev.	1.82	0.40	0.90	0.50	0.95	
Thin black streaked								
No tree	7	33	22.1	45.60	6.90	21.70	72.44	22.74
	8	33	27.2	45.90	9.20	24.20	69.16	25.94
	9	32	35.2	47.50	7.20	23.50	72.97	24.60
	10	26	25.6	47.50	7.60	22.50	71.32	23.70
	11	30	31.8	49.00	7.40	23.20	72.35	24.38
			Average	47.10 b	7.60	23.02 d	71.64 f	24.27 h
		Std. dev.	1.38	0.89	0.95	0.50	1.18	
Normal								
No tree	12	44	27.3	51.30	8.10	24.10	71.46	25.46
	13	27	33.5	52.10	7.80	25.10	72.82	26.23
	14	37	39.5	51.70	7.80	24.70	72.57	25.89
	15	31	32.0	57.70	5.50	26.20	78.25	26.79
	16	32	26.0	58.60	5.00	25.20	78.68	25.66
	17	30	23.5	57.30	7.90	28.20	74.39	29.27
	18	30	25.5	61.30	4.60	27.90	80.41	27.58
			Average	55.71 c	6.67	25.91 e	75.51 g	26.69 i
		Std. dev.	3.97	1.55	1.59	3.53	1.34	

Remarks: Std. dev. = standard deviation; the same letters on the same column are not statistically different at $P < 0.05$ by Duncan's test.

Extractive Content

Darker heartwood is generally assumed to contain more extractives but the values obtained are presented in Table 2. The average extractive contents through successive extraction showed *n*-hexane, EtOAc, and TEC levels were clearly different between thick black-streaked and normal wood groups while slight differences were found between the normal and discolored portion in cold-water and hot-water extractive contents. These results indicate black-streaked heartwood has higher apolar compounds than the normal tissues while only *n*-hexane and total extractive contents were observed to have varied

significantly between its thick and thin samples using analysis of variance. Meanwhile, the Duncan test showed that only the total extractive content followed the tendency of the brightness index and it seems reasonable to assume the increases in its value by 14–19% were due to the production of darkened heartwood. This is, however, more pronounced in the EtOAc extractive content levels which streaks and normal heartwood areas ranged from 4.38 to 7.42% and 0.75 to 3.86%, respectively.

The GC analysis of ethanol-benzene extract for both black-streaked and normal heartwood is shown in Figure 2. The major components identified were lapachol,

tectoquinone, squalene, tectol, and desoxylapachol or its isomer (Lukmandaru and Takahashi 2009) and their quantification is summarized in Table 3. ANOVA, however, showed significant differences exist only in tectoquinone and squalene contents among the groups. The tectoquinone content was observed to have varied from 0.72 to 4.20% in thick black streaks part and 0.29 to 2.54% in normal part but a contrasting result was recorded for squalene with the value found to be significantly lower in the discolored part which was 0.26 to 2.88% compared to the normal heartwood at 0.26 to 1.24%. Meanwhile, the tectoquinone contents recorded in previous studies were reported to have varied from 0.3 to 2% while squalene was 1 to 2% in teak wood (Sandermann and Simatupang 1966; Windeisen *et al.* 2003; Lukmandaru and Takahashi 2009).

The desoxylapachol concentration was found to be lower in discolored areas than normal heartwood but the ANOVA showed the difference between the groups is not statistically significant. A similar tendency was also measured in an unknown compound (UN1) which showed a comparatively higher concentration in the black-streaked region while another unknown component, UN2, was not detected in the black-streaked area but in three individuals containing normal heartwood. The results also showed no

considerable differences between the groups based on the amounts of other constituents due to wide variations by examining their standard deviations.

The higher extractives and tectoquinone contents as well as the lower squalene content found in the black streak parts have been reported in a previous study (Lukmandaru *et al.* 2009). The phenolic compounds of wood are commonly closely related to coloration (Moya *et al.* 2012; Romagnoli *et al.* 2013; Paques *et al.* 2013) but some phenolics have already been identified in teak while those in the wood color of this species are unknown. Meanwhile, the discolorations in living trees are initiated predominantly through wounds, dying branches, and roots (Shigo 1976). Moreover, the role of tectoquinone in natural durability of teak has been recognized in several reports (Rudman *et al.* 1958; Sandermann and Simatupang 1966; Haupt *et al.* 2003) and even though a direct bioassay test was not attempted and the mechanism of resistance associated with the discolored tissue was not determined in this study; the blackening processes was suggested to be related to some protective functions against biological origin. This may have much in common with *Diospyros kaki* where the black portion is more resistant than the adjacent normal heartwood (Noda *et al.* 2002).

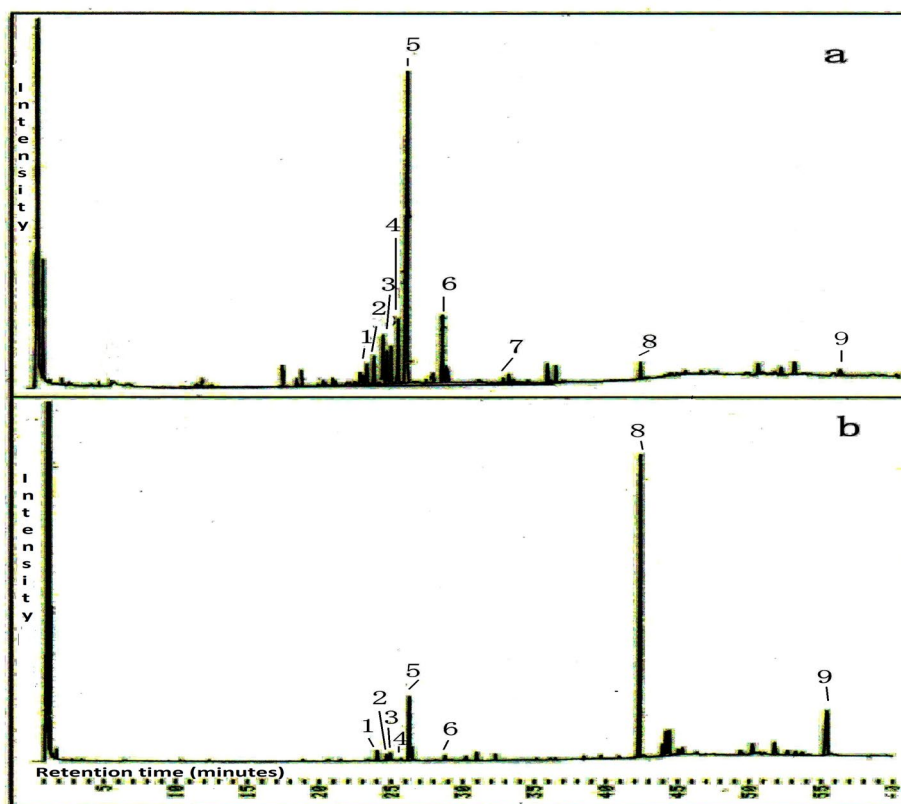


Figure 2. Gas chromatogram of ethanol-benzene extracts of a) black-streaked heartwood and b) normal heartwood of teak. Nine main components are detected: 1. desoxylapachol, 2. palmitic acid, 3. lapachol, 4. isodesoxylapachol, 5. tectoquinone, 6. Unknown compound (UN3), 7. 2-hydroxymethyl-anthraquinone, 8. squalene, 9. tectol.

Table 2. The extractive contents (% based on dry wood) in teak heartwood of black-streak and normal

Extract	Thick black streaked			Thin black streaked			Normal			F-ratio
	Min.	Max.	Average	Min.	Max.	Average	Min.	Max.	Average	
n-hexane	6.35	9.17	8.06 (1.19) b	5.50	6.92	6.33(0.62) a	3.88	6.37	5.15(0.95) a	14.60**
Ethyl acetate	4.38	7.42	5.99 (2.2) d	4.21	5.60	4.90(0.56) d	0.75	3.86	2.09(1.17) c	23.22**
Methanol	2.77	5.61	4.54 (0.95) fg	4.39	5.05	4.95(0.34) g	2.04	5.26	3.42(1.26) f	3.97*
Total extractive	17.11	19.68	18.61 (0.97) k	15.14	17.30	16.19(0.78) j	8.20	14.77	10.66(2.74) i	30.95**
Cold-water	1.19	2.21	1.91(0.38) x	1.62	3.58	2.76(0.75) y	1.33	3.16	2.11(0.64) xy	2.89*
Hot-water	3.60	5.18	4.41(0.56) lm	3.65	5.59	4.73(0.74) m	2.99	4.68	3.69(0.68) l	7.89**

Remarks: min. = minimum values, max. = maximum values, ** significant at 1 % level, * significant at 5 % level. Total extractive content is the sum n-hexane, ethyl acetate, and methanol extractive contents (successive extraction). Cold-water and hot-water solubles were extracted separately. Average of 6 trees (thick black streaked), 5 trees (thin black streaked), and 7 trees (normal) with the standard deviation in parentheses. The same letters on the same row are not statistically different at $P < 0.05$ by Duncan's test.

Table 3. The amount of major components (% based on dry wood) in the ethanol-benzene extracts in teak heartwood of black-streak and normal

Components	Thick black streaked			Thin black streaked			Normal			F-ratio
	Min.	Max.	Average	Min.	Max.	Average	Min.	Max.	Average	
Desoxylapachol	0.10	0.35	0.21(0.11)	0.04	0.78	0.46(0.28)	0.15	2.38	0.79(0.86)	1.66
Palmitic acid	0.03	0.10	0.06(0.02)	0.03	0.14	0.08(0.04)	0.03	0.70	0.20(0.22)	1.85
Lapachol	0.00	0.15	0.05(0.05)	0.00	0.86	0.29(0.35)	0.00	0.48	0.24(0.16)	2.06
Isodesoxylapachol	0.58	2.24	1.20(0.58)	0.31	2.45	1.26(0.77)	0.10	3.54	1.11(1.22)	0.03
Unknown 1	0.00	1.13	0.06(0.04)	0.03	0.12	0.06(0.04)	0.00	0.09	0.03(0.03)	1.05
Tectoquinone	0.72	4.20	2.42(1.26) b	0.95	2.78	1.71(0.92) ab	0.27	2.54	0.94(0.93) a	3.20*
Unknown 2	0.00	0.00	0.0 (0.0)	0.00	0.00	0.0+0.0	0.00	0.60	0.10(0.22)	1.17
Unknown 3	0.04	0.20	0.12(0.05)	0.02	0.29	0.11(0.12)	0.02	0.19	0.12(0.05)	0.02
HMAQ	0.00	0.80	0.16(0.31)	0.00	0.18	0.05(0.08)	0.00	0.43	0.08(0.15)	0.41
Squalene	0.43	0.68	0.55(0.09)c	0.26	1.24	0.65(0.37) c	0.23	2.88	1.66(0.89) d	6.80**
Tectol	0.46	1.45	0.85(0.35)	0.20	1.20	0.64(0.38)	0.24	1.21	0.84(0.31)	0.68

Remarks: min. = minimum values, max. = maximum values, ** significant at 1 % level, * significant at 5 % level

HMAQ = 2-hydroxymethyl-anthraquinone. Average of 6 trees (thick black streaked), 5 trees (thin black streaked), and 7 trees (normal) with the standard deviation in parentheses. The same letters on the same row are not statistically different at $P < 0.05$ by Duncan's test.

Inorganic Contents and pH Value

Table 4 shows the amounts of inorganic materials as well as the pH value in the outer heartwood and the ash and silica levels of discolored parts are observed to be comparatively higher than in normal ones but ANOVA indicated no significant differences in the brightness level factor. Meanwhile, the values for Ca and K contents were observed to be significant as observed with the 2190 to 4760 ppm variation reported for Ca in the discolored portion compared to 1140 to 3090 ppm in normal part. This is in line with the report of the previous study (Lukmandaru *et al.* 2009) which showed Ca values to be slightly higher in the partially black-streaked heartwood compared to the adjacent

normal ones. The Duncan test, however, reported no statistically significant difference between thick black-streaked and normal parts. The same pattern was also observed in K contents.

All the pH values were in the weakly acidic range, except for sample no. 8 with pH 7.07. A comparison of these values showed the discolored wood to have higher levels as observed with the 5.66 to 7.07 recorded for black streak portion while the normal heartwood ranged from 4.96 to 6.35. The ANOVA showed a significant brightness level factor and Duncan test also confirmed a significant difference between the black-streaked and normal heartwood parts.

Table 4. The amount of inorganic elements (% based on dry wood) and pH values in teak heartwood of black-streak and normal

	Thick black streaked			Thin black streaked			Normal			F-ratio
	Min.	Max.	Average	Min.	Max.	Average	Min.	Max.	Average	
Ash	1.40	2.90	2.11 (0.57)	1.80	3.09	2.39 (0.48)	0.85	3.05	1.64 (0.93)	1.65
Silica	7500	16000	10500 (3446)	7500	21000	11800 (5472)	500	12500	7500 (4387)	1.46
Potassium	330	1040	551 (264) a	480	1860	1336 (532) b	340	1680	758(517) a	4.30*
Magnesium	310	480	398 (60)	310	630	454 (116)	230	640	445 (174)	0.31
Calcium	2190	2840	2535 (241) c	2740	4760	3458 (819) d	1140	3090	2031(849) c	6.10*
Iron	37	118	67 (27)	42	60	55 (7)	32	75	56 (14)	0.72
Phosphorous	62	155	115 (40) ef	93	383	184 (117) e	24	180	77 (50) f	3.23*
pH values	5.66	6.97	6.39 (0.46) h	5.74	7.07	6.20 (0.50) h	4.96	6.35	5.39(0.48) g	7.89**

Remarks: min. = minimum values, max. = maximum values, ** significant at 1 % level, * significant at 5 % level. Average of 6 trees (thick black streaked), 5 trees (thin black streaked), and 7 trees (normal) with the standard deviation in parentheses. The same letters on the same row are not statistically different at $P < 0.05$ by Duncan's test.

Relationship between the Brightness Degree and Chemical Properties

The correlation analysis in Table 5 confirmed there is a highly significant negative correlation between the brightness and extractive contents with the strong negative linear relationship measured using ethyl acetate soluble content ($r = -0.94$) or total extractive content ($r = -0.92$). The high correlation coefficient between EtOAc extractive content and brightness is particularly emphasized in the scatter diagram in Figure 3 and an increase in EtOAc soluble extractives were observed to have corresponded thoroughly with the degree of blackening. This is established with its value recorded to be four times in the darkest streak heartwood while the total extractive content was twice higher compared to normal heartwood. This, therefore, means EtOAc soluble extractives have considerable roles in the formation of the darkened parts. A previous study conducted on *Tabebuia serratifolia* wood showed large amounts of polyphenols were found in the black parts, together with other quinone compounds, but low in lapachol concentrations (Romagnoli *et al.* 2013). The EtOAc soluble extracts are, however, believed to contain polymeric quinones which cause wood darkening.

In the component levels, the tectoquinone content was found to be negatively correlated ($r = -0.62^{**}$) with the brightness value while the squalene ($r = 0.56^{**}$) and palmitic acid concentration ($r = 0.49^{*}$) were positively correlated as shown in Figure 4. This means the teak is darker when it contains higher amounts of tectoquinone and lower amounts of squalene and palmitic acid. The reason for this relationship does not necessarily have to be related to the chemical nature of tectoquinone but a specific feature in the responsible compounds correlated with anthraquinones concentration. Moreover, lapachol and dehydro- α -lapachone from the naphthoquinone group were also detected in *Tabebuia serratifolia* wood extract to have contributed to the yellow and pink color of the wood (Romagnoli *et al.* 2013). The enzymatic oxidation of anthraquinones and naphthoquinones was also assumed to be the first step of darkening.

The cause of lower squalene production in the discolored wood and its relationship with the degree of

blackening has not been ascertained. Squalene, a triterpene, as well as palmitic acid from fatty acid family are both not a coloring matter. The tectoquinone, however, might have an indirect role through the formation of polymeric compounds responsible for discoloration which further reduces the production of terpenes such as squalene. Furthermore, the higher content of UN1 with 25 mins retention time and the absence of UN2 with 27.2 min in the discolored parts is also possibly related to the blackening phenomenon.

Previous reports on other species showed inorganic elements in wood to be correlated with the blackening process in heartwood (Kubo and Ataka 1998; Minato and Morita 2005). Even though iron technically forms a complex of dark color due to its reaction with phenolic compounds, no considerable difference was found in its content for both normal and black-streaked woods studied. Moreover, the Ca, K, and silica contents were observed to be higher in the black streak parts compared to the normal ones but there was no significant correlation between them and the brightness index level. This means the formation of blackened heartwood is not directly related to these metals and they are hardly involved in the degree of blackening.

pH value also has the ability to play a role in the discoloration of some wood species (Sanderemann and Dietrichs 1959) such as *Cryptomeria japonica* (Takahashi 1996) and *Pycnanthus angolensis* (Starck *et al.* 1984) which were found to be discolored due to a comparatively huge gradient of pH. Meanwhile, the brightness and pH levels were moderately inversely related according to Pearson's correlation ($r = -0.75$) while the relationships between the degree of blackening and pH values are illustrated in Figure 5. This, therefore, means it is possible to attribute teak wood discoloration from black to pH change even though the value is still in the weakly acidic range. Moreover, the degrees of correlation between pH values and Ca or K content were low (data not shown). Another alternative reason or the blackening phenomenon is attached to the polymerization of phenolic compounds contained in the heartwood under a more weakly acidic condition. It is, therefore, necessary to determine the cause of higher pH in the black streak regions and relate it with the blackening process.

Table 5. Correlation coefficient (*r*) between L* brightness and chemical properties in heartwood of black-streak and normal

Chemical properties	L*	Inorganic contents	L*
Extractive contents		Ash	-0.39
n-hexane	-0.69**	Silica	-0.24
Ethyl acetate	-0.94**	Calcium	-0.39
Methanol	-0.40	Magnesium	0.14
Total extractive	-0.92**	Potassium	-0.06
Cold-water	-0.05	Iron	-0.19
Hot water	-0.55*	Phosphorous	-0.43
		pH values	-0.75**
Extractive component			
Desoxylapachol	0.38		
Palmitic acid	0.49*		
Lapachol	0.25		
Isodesoxylapachol	-0.19		
Unknown 1	-0.45		
Tectoquinone	-0.62**		
Unknown 2	0.64*		
Unknown 3	0.03		
2-hydroxymethyl-anthraquinone	-0.02		
Squalene	0.56**		
Tectol	-0.03		

Remarks: ** significant at 1 % level, * significant at 5 % level

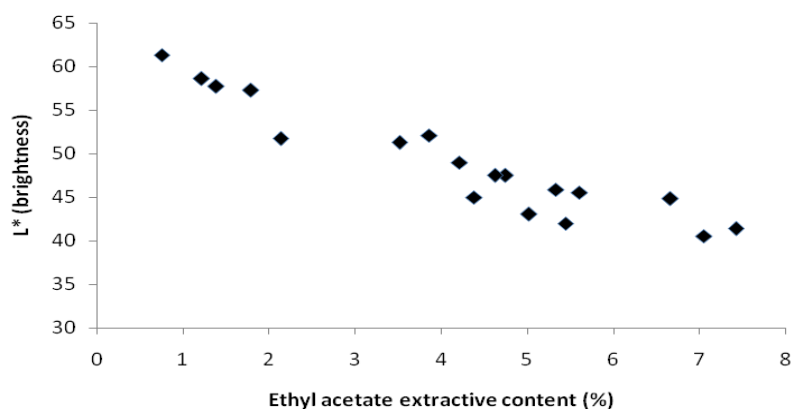


Figure 3. Relation between ethyl acetate extractive content and L* (brightness) in teak heartwood of black-streak and normal.

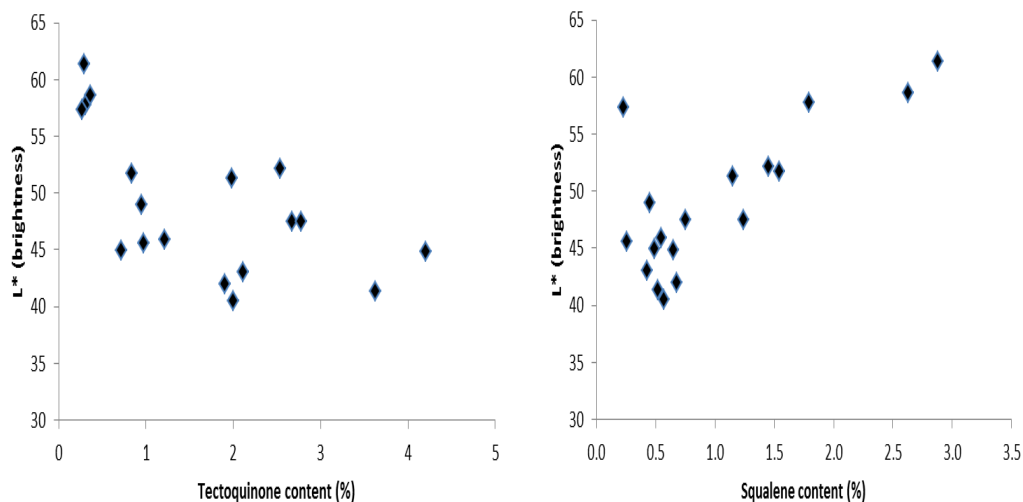


Figure 4. Relation between ethyl acetate extractive content and L* (brightness) in teak heartwood of black-streak and normal.

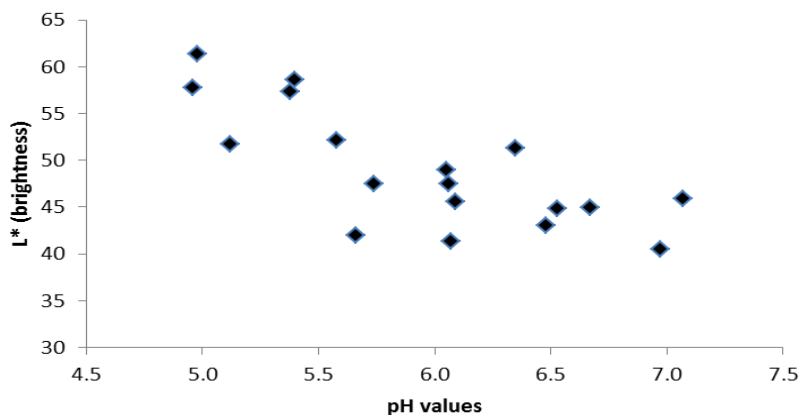


Figure 5. Relation between pH values and L* (brightness) in teak heartwood of black-streak and normal.

Conclusions

The black streak defect of teak wood causes serious losses to the wood industry and this led to the analysis of samples containing this defect in the outer heartwood parts after which they were compared to the normal ones. The black streak area was found to have lower brightness (L*) values up to 10 units, lower levels of yellowness (b*), hue, and chroma in comparison with the normal heartwood. Moreover, the extractive content was discovered to be considerably higher in the blackened portion, particularly in the apolar fraction while the inverse relationship ($r = -0.94$) between the ethyl acetate soluble extractive content levels and brightness values (L*) was also measured. The thick black-streaked heartwood significantly contained more tectoquinone but less squalene while the ash, potassium, calcium, and silica concentration levels were not significantly different among the groups. Furthermore, the variation in the ash contents and individual inorganic

elements were unable to adequately explain the differences in the degree of blackening. The increase in pH values was also observed to be corresponding with the blackening of heartwood ($r = -0.75$). Therefore, further research is necessary to elucidate the chemical structures of the ethyl acetate soluble material relating to the formation of the dark-colored compounds in weakly acidic conditions.

Acknowledgement

This research was financed by MONBUKAGAKUSHO Scholarship. The authors thank to Prof. Tadao Wagatsuma (Faculty of Agriculture, Yamagata University, Japan) for conducting inorganic elements analysis. The authors is also indebted to Mr. Trisno Aji (Perhutani Estate) and Mr. Mufti Wibowo (Brebes Forest Service) for providing the teak samples.

References

- ASTM D1110. 1984. Standard Test Methods for Water Solubility of Wood. Annual Book of ASTM Standards. 1916 race St., Philadelphia, Pa. 19103 : American Society for Testing and Materials.
- ASTM D1102. 1984. Standard Test Methods for Ash in Wood. Annual Book of ASTM Standards. 1916 race St., Philadelphia, Pa. 19103 : American Society for Testing and Materials.
- Bhat, K.M.; P.K. Thulasidas; E.J.M. Florence; K. Jayaraman 2005. Wood durability of home-garden teak against brown-rot and white-rot fungi. *Trees* 19:654–660.
- Haupt, M.; H. Leithoff; D. Meier; J. Puls; H.G. Richter; O. Faix. 2003. Heartwood extractives and natural durability of plantation-grown teakwood (*Tectona grandis* L.)—A Case Study. *Holz als Roh-und Werkstoff* 61:473-474.
- Hillis, W.E. 1987. Heartwood and Tree Exudates. Springer-Verlag, Berlin, Germany.
- Kokutse, A.D.; A. Stokes; H. Bailleres, K. Kokou, C. Baudasse. 2006. Decay resistance of Togolese Teak (*Tectona grandis* L.) heartwood and relationship with colour. *Trees* 20:219-223
- Kubo, T.; S. Ataka. 1998. Blackening of Sugi (*Cryptomeria japonica* D. Don) heartwood in relation to metal content and moisture content. *Journal of Wood Science* 44:137-141.
- Lemos, T.G.; S.M. Costa; O.L. Pessoa; R. Braz-Filho. 1999. Total assignment of ^1H and ^{13}C NMR spectra of tectol and tecomaquinone. *Magnetic Resonance in Chemistry* 37:908-911.
- Lukmandaru, G.; K. Takahashi. 2009. Radial distribution of quinones in plantation teak (*Tectona grandis* L.f.). *Annals of Forest Science* 66(605): 1-9.
- Lukmandaru, G.; T. Ashitani; K. Takahashi. 2009. Characterization of partially black streaked heartwood in plantation teak. *Journal of Forestry Research* 20: 377-380.
- Minato, K.; T. Morita. 2005. Blackening of *Diospyros* genus xylem in connection with boron content. *Journal of Wood Science* 51: 659-662.
- Moya, R.; A. Berrocal. 2010. Wood colour variation in sapwood and heartwood of young trees of *Tectona grandis* and its relationship with plantation characteristics, site, and decay resistance. *Annals of Forest Science* 67: 109.
- Moya, R.; R. Soto Fallas; P. Jiménez Bonilla; C. Tenorio. 2012. Relationship between wood color parameters measured by the CIELab System and extractive and phenol content in *Acacia mangium* and *Vochysia guatemalensis* from fast-growth plantations. *Molecules* 17:3639-3652.
- Noda E., T. Aoki; K. Minato. 2002. Physical and chemical characteristics of the blackened portion in Japanese Persimmon (*Diospyros kaki*). *Journal of Wood Science* 48: 245–249.
- Paques, L.E.; M.D.C. Garcia-Casas; J.P. Charpentier. 2013. Distribution of heartwood extractives in hybrid larches and in their related European and Japanese Larch Parents: Relationship with wood colour parameters. *European Journal of Forest Research* 132:61–69.
- Perry, N.B.; J.W. Blunt; M.H.G. Munro. 1991. A Cytotoxic and antifungal 1,4 Naphtquinone and related compounds from a New Zealand brown alga, *Landsburgia quercifolia*. *Journal of Natural Product* 54:978-985.
- Romagnoli, M.; E. Segoloni; M. Luna; A. Margaritelli; M. Gatti; U. Santamaria; V. Vinciguerra. 2013. Wood colour in Lapacho (*Tabebuia serratifolia*): Chemical composition and industrial implications. *Wood Science and Technology* 47:701–716.
- Rudman, P.; E.W.B. Da Costa; F.J. Gay; A.H. Wetherly. 1958. Relationship of Tectoquinone to durability in *Tectona grandis*. *Nature* 181:721–722.
- Sandermann, W.; H.H. Dietrichs. 1959. Chemische Untersuchungen an Teakholz. *Holzforschung* 13:137-148.
- Sandermann, W.; M.H. Simatupang. 1966. On the chemistry and biochemistry of Teakwood (*Tectona grandis* L. fil). *Holz als Roh-und Werkstoff* 24:190-204.
- Shigo, A.L. 1976. Compartmentalization of discolored and decayed wood in trees. *Material and Organismen* 3: 221–226.
- Starck, M.; J. Bauch; M.H. Simatupang. 1984. Characteristics of normal and discolored wood of Ilomba (*Pycnanthus angolensis* Exell). *Wood Science and Technology* 18:243-253.
- Suhaendi, H. 1998. Teak improvement in Indonesia. In : Teak for the Future, Proceedings of the Second Regional Seminar on Teak. M Kashio and K White (Ed.). RAP Publication 1998/5 TEAKNET Publication: No. 1. Yangon, Myanmar. FAO Regional Office for Asia and the Pacific Bangkok, Thailand. p. 179-188.
- Takahashi, K. 1996. Relationships between the blackening phenomenon and norlignans of Sugi (*Cryptomeria japonica* D. Don) Heartwood. I. A case of partially black-heartwood. *Mokuzai Gakkaishi* 42:1119-1125.
- Windeisen, E.; A. Klassen; G. Wegener 2003. On the chemical characterization of plantation teakwood (*Tectona grandis* L.) from Panama. *Holz als Roh-und Werkstoff* 61:416-418.

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