Discolored Components from the Black-streaked Heartwood Extracts of Teak

Ganis Lukmandaru, Tatsuya Ashitani, and Koetsu Takahashi

Abstract

With regard to black-streaked discoloration of teak wood, the ethyl acetate extract from successive extraction was examined. By column chromatography, tectol and and two unknown compounds i.e. C-1 (molecular weight of 240) and C-2 (molecular weight of 210) were isolated. Discoloration tests, i.e. air oxidation, acidic (acetic acid), and alkaline (potassiun hydrogen carbonate) treatments were conducted to the isolated and other standard quinoine compounds (tectoquinone, lapachol, 2-tert-buthyl- anthraqunone, 2-hydroxy-methyl-anthraqunone). The results showed that tectol changes its color by the considerable decreasing in brightness whereas C-1 showed huge decrease in yellowness by air oxydation. Tectol did not change its color in pH value of 2.9 to 8.3 but C-1, C-2, and lapachol did change. Tectoquinone was hardly change color under alkaline or acidic treatment. The difference in quinone coloration might be due the structural features of a hydroxyl group and a double bond conjugated.

Keywords : darkening wood, air oxydation, extractives, qunoines, tectol.

Introduction

Teak is an economically important tree species indigenous to Java. One of the values of teak wood particularly depends on its aesthetic properties. Coloured wood is in great demand for sawntimber, furniture, and handy craft products. Heartwood color of normal teak is dark brown, but abnormal color due to blackness is not desirable by the people. This discoloration is serious problem that decreases the value of teak products. This natural defect is called 'doreng' in Indonesia and is classified as a cross-sectional defect (BSN 1999).

Color and extractve content of black-streaked heartwood in teak have been investigated (Lukmandaru 2009; Lukmandaru *et al*. 2009). It is commonly known that the phenolic compounds of wood are closely related to the coloration (Takahashi 1996; Paques *et al*. 2013). Unfortunately, the phenols of teak related to the heartwood color are remained unknown. In order to investigate the cause of discoloration, it is expected to identify the extractives contributing to the discoloration and to understand their conversion mechanism. Technically, the causes of discoloration in the wood are various e.g. enzymatic reactions, iron staining, microorganisms, basic and acid conditions of wood etc. (Hon and Shiraishi 2001; Koch *et al*. 2003).

Previous study showed that ethyl acetate (EtOAc) fraction of teak extractives from successively extraction highly correlated with the darkness of heartwood (Lukmandaru *et al*. 2014). Furthermore, it was also found that the lower brightness values correlated with higher pH values. It is thought that some colored components (pigments) were extracted and further examination was conducted with previous methods (Burtin *et al*. 1998; Takahashi and Mori 2006). In this paper, the compounds were isolated and the significance of coloration was described.

Materials and Methods

Chromatography and Physical Property Determinations

Si-gel 20-40 mesh (Wako) and 63-210 mesh spherical neutral (Kanto chemical) were used for flash and column chromatography, respectively. Precoated aluminum sheets silica gel 60 F254 (Merck) were used for thin layer chromatography (TLC). Spots were visualized by UV light irradiation (λ254 nm and λ 360 nm) and by spraying with vaniline-sulfuric acid (for color test) followed by heating at 110 °C for 10 minutes. Developing solvents used for TLC were hexane/acetone (1:5, v/v). Melting points were determined on a YANACO Micro Melting Point Apparatus.

Extraction and Isolation

Black streaked wood samples from a teak tree (35 years) were collected from Randublatung, Central Java Province. Samples from the tree disc (Fig. 2) were ground in a blender. Further, the ground samples (20-40 mesh, 200 g) were extracted by refluxing with n-hexane, EtOAc, and methanol, successively while heated for 6 hours each. The extracts were rotary-evaporated to yield 7.62 g (38.1 %), 6.70 g (33.5 %) and 5.63 g (28.1%), respectively. The EtOAc extract (4.28 g) were separated into low molecular weight and and polymeric fractions by column chromatography. The extract was chromatographed on a silica gel column using n-hexane and acetone as eluents of increasing polarity. The scheme of separation is displayed in Fig. 1. Fractions 2 to 5 contain an unknown compound (C-1) and tectoquinone, respectively. From fraction 8 to 10 (combination), tectol was isolated, whereas from fraction 11 and 12 (combination), another unknown compound (C-2) was isolated from the combination of fraction 8~10.

Figure 1. Teak wood with black streak.

Spectrum Determinations

The ^{13}C (in in 400 MHz), ^{1}H NMR (in 100 MHz) and COSY spectra were determined by a JEOL JNX-400 spectrometer. Chemical shifts are given as δ (ppm) values with TMS as internal standard. Coupling constants (J) are given in [Hz]. The UV-VIS spectral data were determined with SHIMADZU UV-1600 PC in an acetone solution. The amount of 0.1 mg of tested sample was dissolved in 20 ml acetone (reagent grade). GC and GC-MS analysis were using a Hitachi G-3500 GC equipped with FID and NB-1 capillary 30-m column. Operation temperature was 120 - 300 ºC with a heating rate of 4 C/min and held at 300 ºC for 15 minutes. Injector and detector temperatures were set at 250 ºC. Helium was used as the carrier gas, the split ratio was 80:1, and the injected volume was 1.0 µL Mass spectrometry measurements were obtained from GC-MS analysis on a Shimadzu QP-5000 with operation conditions similar to GC analysis. The MS operating parameters were temperature ionization voltage of 70 eV, transfer line temperature at 250 ºC, and scan range of 50 – 500 atomic mass unit.

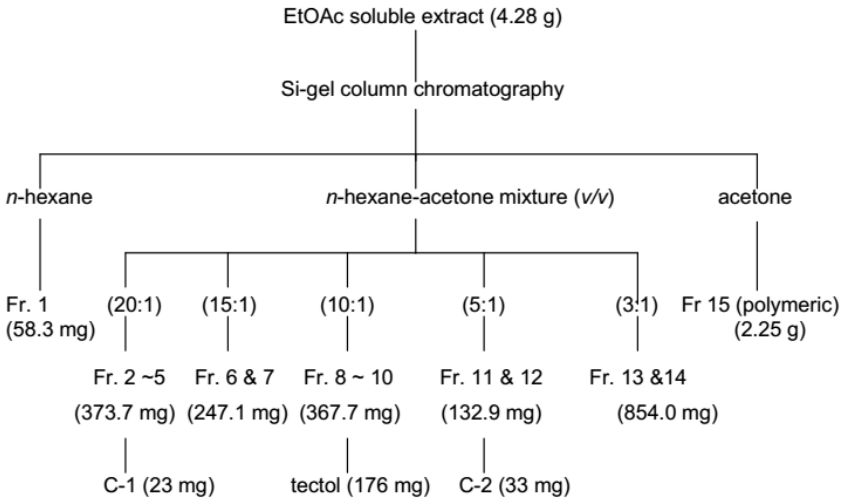

Figure 2. Scheme of compounds isolation from ethyl acetate soluble extract.

Properties of Tectol

Tectol (Fig. 4) was crystallized as a white powder from an n-hexane and acetone solution. Yield : 176 mg; Rf value (solvent n-hexane-acetone = 5 : 1) : 0.32; color reaction (vanillin-sulfuruic acid reagent) : blue; mp: 214-216 0C. GC-MS m/z (rel. int.) 450 (98) (M+), 435 (100), 211 (77), and 210 (54). UV-Vis spectrum λ max acetone : 364 nm. NMR (acetone d1): δ 13C 27.4 (C-15, C-15'), 27.6 (C-14, C-14'), 76.3 (C-13, C-13'), 111.6 (C-3, C-3'), 116.8 (C-2, C-2'), 121.7(C-11, C-11'), 122.3(C-8, C-8'), 123.5(C-5, C-5'), 126.0(C-10, C-10'), 126.2(C-7, C-7'), 126.6(C-10, C-10'), 126.8(C-9, C-9'), 130.9(C-12, C-12'), 142.3(C-1, C-1'), 145.7(C-4, C-4'). 1H 1.47(s, H-15, H-15'), 1.52(s, H-14, H-14'), 5.61 (d, J = 9.8, H-12, H-12'), 5.90 (d, J = 9.9, H-11, H-11'), 7.51(m, H-7, H-7', H-6, H-6'), 7.71 (s, OH), 8.15 (d, J = 8.1, H-8, H-8'), 8.21(d, J = 8.4, H-5, H-5').

Properties of C-1

C-1 was isolated as a reddish crystal from repeated column chromatography. Yield : 23 mg; Rf value (solvent n-hexane-acetone = 5 : 1) : 0.40; mp: 98-100 0C. GC-MS m/z (rel. int.) 240 (16) (M+), 225 (100), 211 (5), and 197 (34). UV-Vis spectrum λ max acetone : 416 nm. NMR (chloroform-d1) : δ 13C 19.6, 74.6, 77.2, 80.9, 122.4, 126.1, 126.2, 129.9, 131.7, 133.1, 137.5, 140.9, 149.4.1H 1.22 (s, 2H), 1.53 (s, 3H), 2.15 (s, 3H), 3.44(s, 4H), 5.70 (d, J = 10.1, 1H), 6.63 (d, J = 9.9, 1H), 7.68 (m, 1H), 8.06 (m, 1H).

Properties of C-2

C-2 was isolated as dark powder from repeated column chromatography. Yield : 33 mg; Rf value (solvent n-hexane-acetone = 5 : 1) : 0.22; mp: 85-87 0C. GC-MS m/z

(rel. int.) 210 (100) (M+), 198 (10), and 182 (55). NMR (chloroform-d1) 13C 19.64, 74.63, 77.23, 80.98, 122.39, 126.14, 126.26, 129.99, 131.77, 133.14, 137.57, 140.99, 149.43, 184.46. 1H 1.23 (s, 1H), 1.69 (s, 3H), 3.81(d, J = 6.3, 1H), 3.97 (t, 1H), 6.12 (s, 2H), 6.44 (d, J = 5.5, 2H), 7.52 (m, 1H), 7.63 (m, 1H), 7.74 (d, J =7.7, 1H), 8.07 (d, J =7.8, 1H).

Air Oxidation Test

The three samples prepared as mentioned above were dissolved in acetone. The solution was then applied to a TLC plate and left in the laboratory desk for 5 days (ambient temperature). The color of TLC before and after exposure was measured with a CIEL*a*b* system, described in using a colorimeter (NF777). L*a*b* system gives the brightness (L*), redness (a*), and yellowness (b*). The resulting total color difference (ΔE*ab) was evaluated using the following equation : $(\Delta L^{*\,2} + \Delta a^{*\,2} + \Delta\, b^{*\,2})^{1/2}$. Tectoquinone (2-methyl anthraquinone, 25753-31 Kanto Chemical), along with several commercially available compounds, which previously reported to occur in the teak extract (Sandermann and Simatupang 1966; Windeisen *et al*. 2003; Lukmandaru and Takahashi 2009), i.e. lapachol 142905 Sigma-Aldrich), 2-hydroxymethyl anthraquinone (17241-59-7 Acros Organics) were also measured.

Alkaline and Acidic Treatment of Quinones

The 0.5 ml solution of each compound mentioned in discoloration test (1 mg/ml) was placed in a test tube. 100 µl of potassium hydrogencarbonate ($KHCO_3$) and acetic acid (CH_3COOH) solution at different concentrations (0.01, 0.1, and 1 %), correspond to pH of 3~9, was added to each sample. The color change of each solution was observed after 4 h.

Results and Discussion

Identification of Components

Teak heartwood meals were extracted with EOAc and treated successively with n-hexane and acetone. Polymeric parts were fairly large in the EtOAc extract (52 %). In the low molecular weight parts, a combination of fractional crystallization and chromatographic methods led to the separation of tectol and 2 other unknown compounds. Tectol was also isolated in considerable amount. The identification of tectol was confirmed by comparing the NMR spectra data reported by Lemos *et al.* 1999). Tectoquinone was detected by GC-MS as one of major compounds. C-1 was separated as orange crystals and as determined by analyzing its GCMS molecular ion at m/z 240.

^{13}C-NMR spectrum of C-1 showed 13 carbon signals, including three methylene carbons (δC 74.6, 77.2, 80.9), three olefinic carbons (δC 137.5, 140.9, 149.4), one methyl carbon (δC 19.6) and six aromatic carbons (δC 122.4, 126.1, 126.2, 129.9, 131.7, 133.1). In the ^{1}H-NMR spectrum, the resences of methylene proton (δH 3.4), aromatic proton (δH 6.6), methylenedioxy protons (δH 5.6, 5.7), and vinyl protons (δH 7.68, 8.0) were found.

C-2 was separated as dark powders, and was determined by analyzing its GC-MS molecular ion at m/z 210. 13C-NMR spectrum of C-2 showed 13 carbon signals, including carbonyl carbon (δC 184.4), two methylene carbons (δC 77.2; 80.9), three olefinic carbons (δC 133.1; 137.5; 149.4), two methyl carbons (δC 19.6; 74.6) and four aromatic carbons (δC 122.3; 126.1; 126.2; 131.7). In the ^{1}H-NMR spectrum, it revealed methylene proton (δH 3.8; 3.9), aromatic protons (δH 6.1; 6.4), and vinyl protons (δH 7.5; 7.6; 7.7; 8.0). Due to the presence of aromatic rings and carbonyl carbons, this compound is suggested to be a naphtaquinone compound.

Air Oxidation Test

The results of air oxidation of the compounds are shown in Table 1. It was revealed that tectol changes its color (ΔE*ab = 27.2) as indicated by the considerable decreasing in brightness and increasing in yellowness index (Fig. 3). Lapachol and C-1 were moderately changed, whereas tectoquinone was slightly changed. The color change in C-1 was mostly from its yellowness. The changes of the color in other compounds were small.

Table 1. Color changes of compounds after 5-day exposure

Compounds	Color measurement			Color changes after exposure			
	L*	a*	b*	ΔL*	Δa*	Δb*	ΔEab*
Tectol	91.4	0.6	2.7	-13.1	5.1	23.3	27.2
C-1	85.3	4.0	24.7	3.1	-2.5	-10.3	11.0
C-2	84.3	0.1	15.7	1.5	1.3	1.6	2.5
Tectoquinone	91.7	-1.3	6.4	-0.4	-6.0	4.3	7.3
Lapachol	83.0	9.0	17.6	6.9	-7.9	8.6	13.5
2-tert-buthyl-AQ	92.1	0.9	-0.1	-0.3	-0.3	2.1	2.1
2-hydroxy-methyl-AQ	92.1	1.0	-0.1	-0.5	-1.1	3.7	3.8

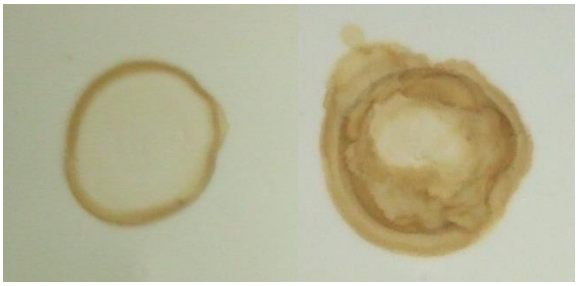

Figure 3. Air oxidation of tectol after 1-day (left) and 3-day exposures (right).

Alkaline and Acidic Treatment of Quinones

Compounds were treated in acetic acid and potassium hydrogencarbonate in the pH range of 3-9 to investigate the influence of pH values. After the treatment, changes in color and UV-VIS absorption were examined. The results (Table 2, Fig. 4) revealed that C-1, C-2, and lapachol changed their color after alkaline treatment at 0.1 % $KHCO_3$ (pH 8.3), whereas tectol changed its color at 1 % $KHCO_3$ (pH 9.3). Tectol showed no clear absorption spectrum, although tectol changed color to dark green. In acidic condition, only C-1 and lapachol changed their color at acetic acid 0.1 % solution (pH 3.9). C-1 showed absorption maxima at 416 (at pH 3.9 and 8.3) and 520 nm (at pH 9.5). It was concluded that C-1, C-2, and tectol is sensitive to pH changes, while other quinones hardly change color even at high pH. The pH changes does not correspond to the changes in the color of the heartwood that were caused by acidic or alkaline treatment since the pH of black streak part ranges from 5.5 to 7.0 as described in the previous report (Lukmandaru *et al.* 2009; 2014). Furthermore, the cause of pH change in the discolored part is remained unknown. This trend was also differed in another species related to pH values (Starck *et al.* 1984; Takahashi and Mori 2006). Thus, the question about the reason of color changes of woods upon pH change cannot be answered in a definite way.

Table 2. Color changes and UV-VIS measurement of compounds after 4-hr acidic and alkaline treatment.

Compounds	Acetic acid			Potassiun hydrogen carbonate		
	pH 2.9	pH 3.9	pH 5.9	pH 6.9	pH 8.3	pH 9.5
Tectol	x	x	x	x	x	Deep green (362 nm)
C-1	x	Weak reddish (416 nm)	x	x	Weak reddish (416 nm)	Reddish (520 nm)
C-2	x	x	x	x	Weak brownish	Weak brownish
Tectoquinone	x	x	x	x	x	x
Lapachol	x	Weak reddish (378 nm)	x	x	Weak reddish (368 nm)	Deep reddish (484 nm)
2-tert-buthyl-AQ	x	x	x	x	x	x
2-hydroxy-methyl-AQ	x	x	x	x	x	x

Remark : x = no color change AQ - anthraquinone

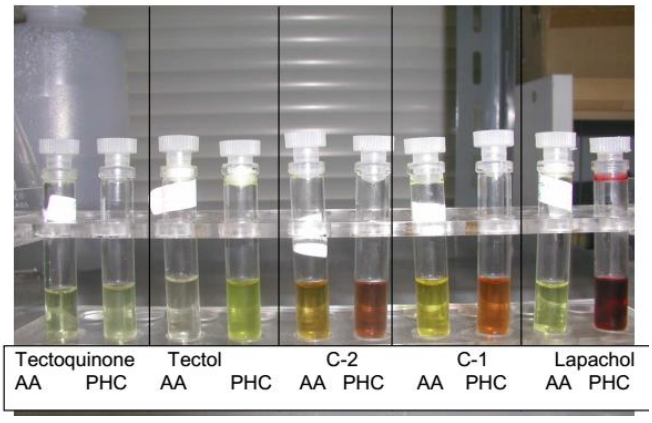

Figure 4. The color changes of compounds after the treatment of acidic and alkaline. (AA = acetic acid 1 %; PHC = potassium hydrogen carbonate 1 %).

Relationship between Structure and Coloration of Quinone

Previously, the discoloration behaviour of teak during irradiation was observed which the concentration tectoquinone and phenol decreased substantially (Li *et al*. 2019). Furthermore, darkened wood samples by heat treatment showed the total extractives content were higher in the treated teakwood (Gasparik *et al*. 2019; Lengowski *et al.* 2021). Table 1 and 2 shows how quinones are colored with air oxidation, acidic, and alkaline treatment. Fig. 5 displayed the chemical structure of the treated compounds.Tectoquinone and other two anthraquinones are remained stable, whereas tectol, C-1, C-2 and lapachol changed their color. It is thought that the color changes are due to the structural differences among those compounds. The structural differences are the structure of double bond conjugated and hydroxyl groups. The structure of tectoquinone, 2-tert-butyl-anthraquinone, 2-hidroxy-methyl-anthraquinone which is not colored, are lack of hydroxyl group and double bond conjugated (Table 3). Thus, it is reasonable to assume that the structural features of a hydroxyl group and a double bond conjugated have a significant effect on quinone coloration (Takahashi and Mori 2006). Tectol with two hydroxyl groups and a possibility to extend its double bonds conjugated system, changes its color through air oxidation and through treatment at pH 9.5. Lapachol with one hydroxyl group and a possibility to extend its double bonds conjugated system changes to weak reddish at pH 3.9 and 8.3 as well as moderately changed in color through air oxidation.

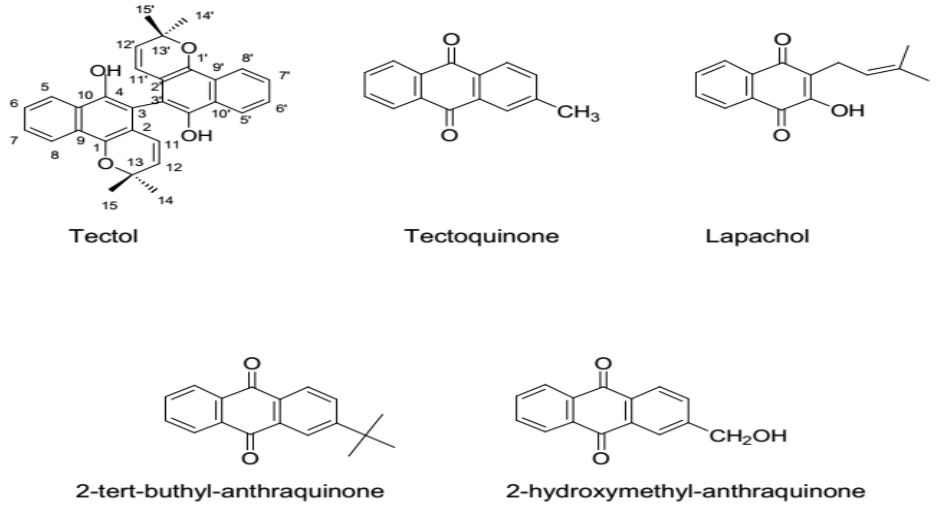

Figure 5. Compounds from the extractives of teak heartwood.

Table 3. Structural differences among the compounds from teak extractives.

Compounds	Structure	
	Hydroxyl group	Extension of double bond conjugated
Tectol	2	2
Tectoquinone	0	0
Lapachol	1	1
2-tert-buthyl-AQ	0	0
2-hydroxy-methyl-AQ	1	0

The Contribution of Components to the Discoloration of Teak Wood

As tectol providing the most intense color on TLC plates and showing the most abrupt decrease in brightness, it could be regarded as a major heartwood color precursor or pigment. Tectol can easily be oxidized, therefore, this could be the first stage of tectol before being polymerized by enzymatic reaction to produce special colored compounds. This similar phenomenon was also observed in a naphtaquinone, hydrojuglone glucoside, in *Juglans* sp wood (Burtin *et al*. 1998). The yellowness value of C-1 was decreased considerably after air exposure. This fact is in line with characteristic of black streak part of teak in the previous study (Lukmandaru 2009; Lukmandaru *et al*. 2009), which has lesser value in yellowness. In an earlier report, lapachol was also detected in *Tabebuia serratifolia* wood extract which it contribute in yellow color of wood (Romagnoli *et al*. 2013).

Heartwood and sapwood of teak were measured to be weakly acidic (Qiu *et al.* 2019). In addition, the main

different substances between heartwood and sapwood were 4-tert-butyl-2-phenyl-phenol, tectoquinone, and 2,3-dimethyl-1,4,4a, 9a-tetrahydro-9,10-anthracenedione#. The sensitivity to pH treatment also suggests that C-1 and C-2 could be the one of precursors of colored substances although the changes did not occur in the weakly acid range. It is assumed that a structure changed to a p-quinone or o-quinone structure after acidic/alkaline treatments due to the formation of a large conjugated system (Takahashi and Mori 2006). The exact structure of and C-1 and C-2 will be elucidated after finishing the measurements COSY CH, HMBC, and HMQC- NMR and will be reported in the future. This study proved that tectoquinone is not easily to change its color although this compound was observed in comparatively higher amount (Lukmandaru *et al*. 2009; 2014). Various constituents other than quinones are found in the teak heartwood, therefore, more detailed investigations are needed.

The higher extractive contents of discolored heartwood to those of normal wood mean that both high and low molecular weight substances could be studied using HPLC and GC-MS analysis. The significantly high *n*-hexane and EtOAc extractive content in the black streak part (Lukmandaru *et al*. 2009; 2014) indicated that the intense production of pigmented heartwood substances. It was also observed that the final color of the wood after extraction was still darker than that of the normal heartwood. Above a certain degree a polymerization, some compounds remain bound to the matrix. To understand more about the background of the blackening, the acidolysis treatment (Kai and Swan 1990) might be necessary to characterize the polymeric parts, as well as, enzymatic or derivation treatment to the low molecular weight substances.The blackening process was assumed to be a kind of protective function against biological origins. This hypothesis was supported by the comparatively high level of tectoquinone in that region (Lukmandaru *et al*. 2009; 2014). However, tectoquinone was relatively stable at low and high pH, as well as in air oxidation. In studying the blackening *Diospyros kaki*, Yasue *et al*. (1975) observed that a quinone compound is indicated as a precursor of polymeric substances. On the other hand, tectol, a dimeric naphtaquinone, provided dark colored after air oxidation. Thus, it is proved that the structural of quinone will affect the ability to change its color. Unfortunately, two isolated compounds remain unidentified, as well as, another dimeric quinone as not isolated, therefore, the conclusions with regard to structural differences of quinone could not be drawn more thoroughly. The structure of tectoquinone, 2-tert-butyl-anthraquinone, 2-hidroxy-methyl-anthraquinone which is not colored, are lack of hydroxyl group and double bond conjugated.

Conclusions

By column chromatography, tectol and two unknown components (C-1 and C-2) were isolated from the low molecular weight parts of ethyl acetate extract. By air oxidation test, tectol, a dimeric naphtaquinone, showed an abrupt decrease in brightness. Therefore, tectol could be a precursor of colored compounds by enzymatic reactions. Tectoquinone was relatively stable at low and high pH, as well as in air oxidation.The discoloration sensitivity might be due the structural differences in double bond conjugated and hydroxyl groups among the tested quinones. All tested components did not change their color after weak acidic treatment which suggest the other unisolated colored substances and high molecular parts from ethyl acetate extract might be involved in discoloration process under the weakly acid range.

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