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Hopeanolin and other Resveratrol Oligomers from the Twigs of *Shorea acuminata*: Antioxidant Propertiaes and Chemotaxonomic Significance

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Abstract: (-)-Hopeanolin (**1**) was isolated from the acetone extract of the twigs of *Shorea acuminata* (Dipterocarpaceae), together with four resveratrol oligomers namely (-)-laevifonol, (-)-α-viniferin, (-)-vaticanol B and (-)-hopeaphenol. The structures of these compounds were established based on spectroscopic evidence, including UV, IR, NMR and mass spectra. Compound **1** showed the potent ability to protect β-carotene bleaching by linoleic acid and also to scavenge DPPH radicals with IC_{50}s 0.18 and 6.58 mM respectively. The presence of compound **1** and the other four resveratrol oligomers in this species have great chemotaxonomic significance on the relationship between *Shorea* and other genera of Dipterocarpaceae especially *Hopea.*

Keywords: β-carotene bleaching, Dipterocarpaceae, DPPH, (-)-Hopeanolin, *Shorea acuminata*.

INTRODUCTION

Resveratrol oligomer has been reported first discovered by Coggon *et al.* in 1965 [1]. Since then, studies have been conducted to isolate resveratrol oligomer from different genus and plant families including the Dipterocarpaceae. Resveratrol oligomers have been classified into monomers, dimers, trimers, tetramers, hexamers and octamer. Some of the compounds obtained were found to show biological activity such as antioxidants [2], anti-bacteria [3], anti-virus and cytotoxic effects [4].

Shorea which is the largest genus in the family Dipterocarpaceae is widely distributed in Malaysia. Symington (1943) [5] has classified the genus *Shorea* into four groups that is Balau (15 species), Meranti Pa'ang (11 species), Meranti Damar Hitam (10 species) and Red Meranti (23 species). To date, about 26 resveratrol oligomers were successfully isolated from this genus [6]. So, in our continuing investigation on the chemical constituents of *Shorea*, *S. acuminata* Dyer who belong to the Red Meranti group of *Shorea* better known by locals as 'Meranti Rambai Daun', was selected as the study species. This paper will report the isolation and structure elucidation of (-)-hopeanolin (**1**) [7], (-)-laevifonol (**2**) [8], (-)-α-viniferin (**3**) [9], (-)-vaticanol B (**4**) [10] and (-)-hopeaphenol (**5**) [11] (Fig. **1**), isolated from the twigs of *S. acuminata*, as well as their antioxidant activity towards DPPH radicals and β-carotene bleaching by linoleic acid.

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MATERIALS AND METHODOLOGY

General Procedure

An IR spectra was recorded on a Perkin Elmer GX FT-IR spectrophotometer. An UV spectra was recorded on Shimadzu UV-160 (200-400 nm). ^{1}H and ^{13}C-APT NMR were recorded in Acetone-d_6 using JEOL ECP400 spectrometer (400 MHz for ^{1}H and 100 MHz for ^{13}C). A mass spectra was detected by LC-MS (ToF) spectrometer. Melting points was measured by Stuart SMP10 melting point apparatus and is uncorrected. Optical rotation was recorded on JASCO DIP-370 Digital polarimeter in MeOH. Vacuum liquid (VLC) chromatography was carried out on Si-gel 60 GF254 (Merck), radial chromatography was carried out on Si-gel 60 PF254 (Merck), gel chromatography was carried out on Sephadex LH-20 (Merck) and TLC was performed on pre-coated silica gel (Merck, Kieselgel 60 F_{254} 0.25 mm) and detected by UV light (254 nm) or by $CeSO_4$ spraying reagent followed by heating. All solvents used were analytical grades.

Plant Material

The twigs of *S. acuminata* were collected from UKM Forest Reserve, Bangi, in September 2009. A voucher specimen (NYM 001) has been deposited in the Universiti Kebangsaan Malaysia Herbarium.

Extraction and Isolation

The dried powdered of twigs of *S. acuminata* (3200 g) was macerated with acetone at room temperature. The

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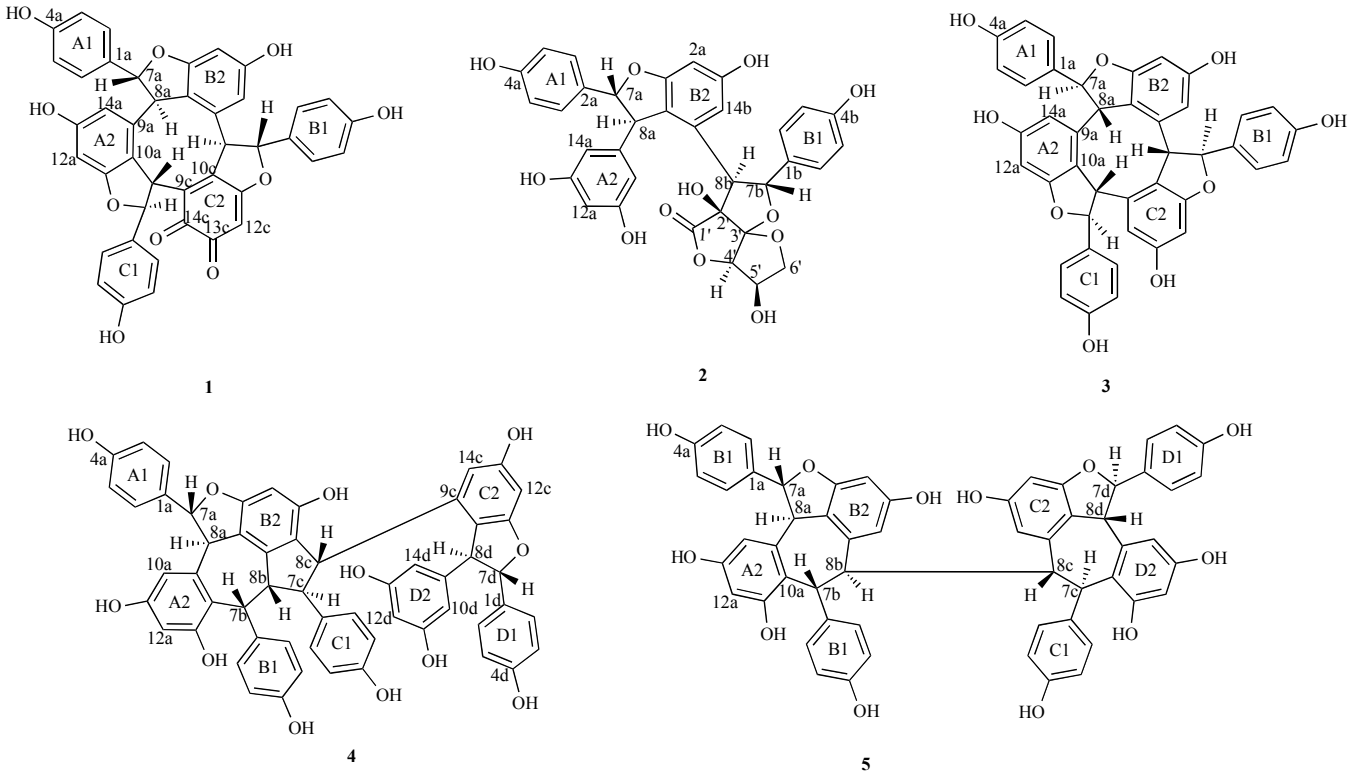

Fig. (1). Structures of compounds **1**- **5**.

extract solution was concentrated using rotary evaporator to yield a brownish acetone extract (180.0 g, 5.6 %). The acetone extract was then fractionated by vacuum liquid chromatography (VLC) eluted with *n*-hexane:EtOAc (increasing polarity of EtOAc). The eluates that showed the same profile on TLC chromatogram were combined to give four fractions (A-D). Further purification of fraction A (6200 mg) by radial chromatography using $CHCl_3$:MeOH (8.5:1.5) solvent system afforded compound **3** (180.0 mg). Refraction of fraction A with column chromatography ($CHCl_3$:MeOH, 8.0:2.0) followed by preparative TLC ($CHCl_3$:MeOH, 9.0:1.0) and gel chromatography (MeOH) gave compound **1** (1.4 mg). Purification of fraction C (2000 mg) by radial chromatography using $CHCl_3$:MeOH (8.5:1.5) solvent system followed by preparative TLC afforded compound **2** (2.3 mg) and **4** (7.3 mg). Compound **5** (4.1 mg) was obtained from fraction D by column chromatography followed by preparative TLC ($CHCl_3$:MeOH, 8:2).

(-)-Hopeanolin (**1**), obtained as a brownish orange amorphous solid, ESIMS $[M-H]^+$ m/z 691.1525, optical rotation, $|\alpha|_D^{122}$ = -14°(c 0.00014 MeOH). UV absorption λ_{max} (MeOH) 282 nm. IR (KBr) γ_{max} 3382 cm^{-1} (OH), 2925 cm^{-1} (C-H aliphatic), 1700 cm^{-1} (C=O), 1615 cm^{-1}, 1519 cm^{-1} and 1451 cm^{-1} (C=C aromatic), 1213 cm^{-1} and 1171 cm^{-1} (C-O oxyaryl) and 835 cm^{-1} (*p*-disubstitutedbenzene). ^1H and ^{13}C NMR spectrum data (see Table **1**).

(-)-Laevifonol (**2**), optical rotation, $|\alpha|_D^{19}$= -117º (*c* 0.0023, MeOH). UV absorption λ_{max} (MeOH) 283 nm. IR (KBr) γ_{max} 3402 cm^{-1} (OH), 1789 cm^{-1} (C=O), 1614 cm^{-1}, 1517 cm^{-1} and 1454 cm^{-1} (C=C aromatic) and 1267 cm^{-1} (C-O oxyaryl) and 835 cm^{-1} (*p*-disubstitutedbenzene). ^1H NMR spectrum data (asetone-*d6*, 400 MHz) δ_H (ppm): 6.98 (d, *J* = 8.4 Hz, 2H, H-2b,6b), 6.76 (m; 6H; H-2a,6a; H-3a,5a and H-3b,5b), 6.17 (br s, H-12a), 5.92 (br s, 2H, H-10,14a), 6.19 (d, *J*=1.8, H-12b), 7.17 (br s, H-14b), 5.05 (d, *J*=7.3, H-7a), 3.30 (d, *J*=10.6, H-8a), 5.29 (d, *J*=10.6, H-7b), 3.31 (d, *J*=10.6, H-8b), 4.42 (br s, H-4'), 4.23 (m, H-5'), 3.99 (dd, *J*=10.2, 4.8 Hz, H-6'a) and 4.07 (dd, *J*=9.9, 2.2 Hz, H-6'b). RMN ^{13}C (asetone d_6 400 MHz), δ_C ppm: 171.2 (C-1'), 160.3 (C-11b), 159.0 (C-11a,13a), 158.3 (C-4b), 157.9 (C-13b), 157.5 (C-4a); 145.1 (C-9a), 131.6 (C-9b), 131.1 (C-1a), 129.2 (C-1b), 128.4 (C-2a,6a), 127.5 (C-2b,6b), 122.7 (C-10b), 118.0 (C-3'), 115.3 (3a,5a), 115.0 (3b,5b), 110.0 (C-14b), 101.6 (C-12a), 96.2 (C-12b), 93.5 (C-7a), 89.3 (C-7b), 88.3 (C-4'), 80.3 (C-2'), 74.9 (C-6'), 73.9 (C-5') and 55.4 (C-8a,8b).

(-)-α-Viniferin (**3**), obtained as brown powder, optical rotation, $|\alpha|_D^{19}$: –34º (*c* 0.0003, MeOH). ESIMS $[M-H]^-$, *m/z*: 677.0315 ($C_{42}H_{29}O_9$). UV absorption λ_{max} (MeOH) 284 nm. IR (KBr) γ_{max} 3367 cm^{-1} (OH), 2974 cm^{-1} (C-H aliphatic), 1614 cm^{-1}, 1514 cm^{-1} and 1441 cm^{-1} (C=C aromatic), 1244 cm^{-1} and 1171 cm^{-1} (C-O oxyaryl) and 831 cm^{-1} (*p*-disubstitutedbenzene). ^1H NMR spectrum data (asetone-*d6*, 400 MHz) δ_H (ppm): 7.02 (2H; d, *J*=9.0 Hz; 2a,6a), 6.71 (2H; d, *J*=7.8 Hz; 3a,5a); 7.04 (2H; d, *J*=9.0 Hz; 2b,6b), 6.76 (2H; d, *J*=7.8 Hz; 3b,5b), 7.22 (2H; d, *J*=8.4 Hz; 2c,6c), 6.77 (2H; d, *J*=7.8 Hz; 3c,5c), 6.22 (1H; *J*=1.2 Hz; 12a), 6.71 (1H; br s; 14a), 5.98 (1H; *J*=1.8 Hz; 14b), 6.20 (1H; *J*=1.2 Hz; 12b), 6.23 (1H; *J*=1.2 Hz; 12c), 6.59 (1H; *J*=2.0 Hz; 14c), 6.06 (1H; br s; 7a), 3.94 (1H; br s; 8a); 4.89 (1H; *J*=6.6 Hz; 7b), 4.61 (1H; *J*=6.6 Hz; 8b), 5.94 (1H; *J*=10.2 Hz; 7c)

Table 1. Comparison ^{1}H and ^{13}C-APT NMR (600 and 150 MHz, asetone d_6) Data of 1 with (-)-hopeanolin from *Hopea exalata* (300 and 75 MHz)

Position	δH (mult., *J* in Hz)		δC (mult., *J* in Hz)	
	1	(-)-Hopeanolin*	1	(-)-Hopeanolin*
1a	-	-	129.7	130.6
2a(6a)	7.30 (d, 8.4)	7.32 (d,8.6)	128.3	128.9
3a(5a)	6.74 (d, 8.4)	6.75 (d,8.6)	115.2	116.1
4a	-	-	157.7	158.6
7a	5.97 (d, 12.0)	5.98 (d, 12.0)	89.1	90.0
8a	4.74 (d, 12.0)	4.75 (d,12.0)	51.9	52.8
9a	-	-	136.7	137.6
10a	-	-	117.1	118.0
11a	-	-	161.9	162.8
12a	6.27 (d, 1.8)	6.29 (d, 1.8)	96.7	97.6
13a	-	-	160.7	161.6
14a	6.67 (d, 1.8)	6.69 (d, 1.8)	105.3	106.2
1b	-	-	133.7	134.6
2b(6b)	7.17 (d, 9.0)	7.19 (d, 8.5)	128.3	129.2
3b(5b)	6.80 (d, 8.4)	6.81 (d, 8.5)	115.0	116.4
4b	-	-	158.1	159.0
7b	6.19 (br s)	6.20 (br s)	88.0	88.9
8b	3.96 (br s)	3.97 (br s)	43.8	44.7
9b	-	-	139.0.	139.8
10b	-	-	121.3	122.1
11b	-	-	160.2	161.0
12b	6.30 (d, 1.8)	6.32 (d, 1.8)	96.9	97.8
13b	-	-	159.0	159.9
14b	6.52 (d, 1.2)	6.54 (d, 1.8)	105.7	106.5
1c	-	-	133.0	133.8
2c(6c)	7.25 (d, 8.4)	7.26 (d, 8.5)	127.2	128.0
3c(5c)	6.75 (d, 8.4)	6.75 (d, 8.5)	115.5	115.9
4c	-	-	157.3	158.2
7c	5.56 (d, 2.4)	5.56 (d, 2.2)	90.3	91.2
8c	4.67 (d, 4.2)	4.68 (d, 2.2)	49.3	50.2
9c	-	-	128.6	129.5
10c	-	-	147.2	148.1
11c	-	-	170.5	171.4
12c	5.78 (s)	5.80 (s)	100.8	101.7
13c	-	-	176.5	177.4
14c	-	-	179.5	180.4

Table 1. contd…

Position	δH (mult., *J* in Hz)		δC (mult., *J* in Hz)	
	1	(-)-Hopeanolin*	1	(-)-Hopeanolin*
OH-4a	8.52 (br s)	8.52 (br s)		
OH-13a	8.79 (br s)	8.78 (br s)		
OH-4b	8.71 (br s)	8.71 (br s)		
OH-13b	8.61 (br s)	8.62 (br s)		
OH-4c	8.45 (br s)	8.44 (br s)		

*Source: Ge *et al*. 2006 [7]

and 4.69 (1H; *J*=10.2 Hz; 8c). ^{13}C NMR spectrum data (asetone-*d6*, 150 MHz) δ_C (ppm): 95.7 (C-12c), 96.0 (C-12b), 97.1 (C-12a), 104.9 (C-14c), 105.3 (C-14a), 107.7 (C-14b), 114.8 (C-3a,5a), 115.2 (C-3b,5b), 115.2 (C-3c,5c), 118.0 (C10b), 118.9 (C-10c), 120.0 (C-10a), 127.5 (C-2a,6a), 127.4 (C-2c,6c), 127.8 (C-2b,6b), 131.1 (C-1c), 131.3 (C-1a), 131.7 (C-1b), 137.8 (C-9c), 138.9 (C-9b), 140.4 (C-9a), 157.4 (C-4a), 158.6 (C-11a), 160.7 (C-13a); 157.6 (C-4b), 158.5 (C-11b) , 160.0 (C-13b), 157.0 (C-4c), 159.8 (C-11c), 160.9 (C-13c), 45.4 (C-8a), 85.5 (C-7a); 54.8 (C-8b), 94.8 (C-7b), 52.0 (C-8c) and 89.2 (C-7c).

(-)-Vaticanol B (**4**), obtained as a dark yellow amorphous solid, exhibiting molecular ion peak in the negative-ion ESIMS spectrum at [M-H]$^-$ *m/z* 905 corresponding to molecular formula $C_{56}H_{42}O_{12}$, $\left|\alpha\right|_D^{20}$: -29º (c : 0.0005 MeOH). UV absorption λ_{maks} (MeOH) 283 nm. IR spectrum (KBr) γ_{max} 3400 cm^{-1} (OH), 2920 cm^{-1} (C-H aliphatic), 1614 cm^{-1}, 1514 cm^{-1} and 1450 cm^{-1} (C=C aromatic), 1242 cm^{-1} and 1159 cm^{-1} (C-O oxyaryl) and 832 cm^{-1} (*p*-disubstitutedbenzene). ^{1}H NMR spectrum data (asetone-*d6*, 400 MHz) δ_H (ppm): 7.21 (2H; d, *J*=8.4 Hz; 2a,6a), 6.77 (2H; d, *J*=8.4 Hz; 3a,5a), 7.15 (2H; d, *J*=8.4 Hz; 2b,6b), 6.69 (2H; d, *J*=8.4 Hz; 3b,5b), 6.39 (2H; d, *J*=8.4 Hz; 2c,6c), 6.50 (2H; d, *J*=8.4 Hz; 3c,5c), 7.18 (2H; d, *J*=8.8 Hz; 2d,6d), 6.77 (2H; d, *J*=8.4 Hz; 3d,5d), 6.28 (1H; d, *J*=2.2 Hz; 12a), 6.11 (1H; d, *J*=2.6 Hz; 14a)], 6.18 (1H; *J*=2.2 Hz; 12c), 6.47 (1H; *J*=2.2 Hz; 14c), 6.09 (2H; d, br s; 10d,14d), 6.27 (1H; t, *J*=2.2 Hz; 12d), 6.04 (1H; 12b), 5.76 (1H; *J*=12.1 Hz; 7a), 4.43 (1H; *J*=11.0 Hz; 8a), 5.36 (1H; *J*=5.1 Hz; 7d), 4.67 (1H; *J*=5.1 Hz; 8d), 5.20 (1H; *J*=3.7 Hz; 7b), 3.11 (1H; br d; 8b) 4.09 (1H; t; *J*=11.4 Hz; 7c) and 4.54 (1H; *J*=10.6 Hz; 8c). ^{13}C NMR spectrum data (aseton d_6 400 MHz), δ_C ppm: 94.8 (C-12c), 95.6 (C-12b), 100.8 (C-12a), 101.4 (C-12d), 104.9 (C-14a), 106.2 (C-14c), 106.7 (C-10d,14d), 114.7 (C-3b,5b), 115.1 (C-3c,5c), 115.1 (C-3d,5d), 115.2 (C-3a,5a), 127.4 (C-2d,6d), 128.4 (C-2c,6c), 129.4 (C-2a,6a), 130.0 (C-2b,6b), 114.9 (C10b), 121.3 (C-14b), 122.6 (C-10c), 123.7 (C-10a), 130.0 (C-1a), 130.6 (C-1c), 132.7 (C-1b), 133.9 (C-1d), 141.0 (C-9a), 140.9 (C-9c), 142.4 (C-9b), 147.2 (C-9d), 154.1 (C-13b), 154.9 (C-11a), 155.1 (C-4b), 155.6 (C-4c), 156.0 (C-13a), 157.8 (C-4a), 157.2 (C-4d), 158.0 (C-11b), 158.6 (C-13c), 159.0 (C-11d,13d), 160.9 (C-11c), 36.2 (C-7b), 48.1 (C-8a), 48.5 (C-8c), 52.3 (C-8b), 56.8 (C-7c), 56.8 (C-8d), 89.6 (C-7a) and 93.8 (C-7d).

(-)-Hopeaphenol (**5**), obtained as a dark yellow amorphous solid, exhibiting molecular ion peak in the negative-ion ESIMS spectrum at [M-H]$^-$ *m/z* 905.2696 corresponding to molecular formula $C_{56}H_{42}O_{12}$, which is classified to a resveratrol tetramer. $\left|\alpha\right|_D^{20}$: -396º (c : 0.0005 MeOH). UV absorption λ_{maks} (MeOH) 277 nm. IR spectrum (KBr) γ_{max} 3412 cm^{-1} (OH), 2950 cm^{-1} (CH-aliphatic), 1644 cm^{-1} and 1453 cm^{-1} (C=C aromatic) and 1111 cm^{-1} and 1032 cm^{-1} (C-O oxyaryl). ^{1}H NMR spectrum data (asetone-*d6*, 400 MHz) δ_H (ppm): 7.12 (2H; d, *J*=8.4 Hz; 2a,6a), 6.77 (2H; d, *J*=8.4 Hz; 3a,5a), 6.89 (2H; d, *J*=8.1 Hz; 2b,6b), 6.54 (2H; d, *J*=8.8 Hz; 3b,5b), 6.28 (1H; br s; 14a), 6.53 (1H; br s; 12a), 5.15 (1H; *J*=2.2 Hz; 14b), 5.71 (1H; *J*=2.2 Hz; 12b), 5.74 (1H; *J*=12.1 Hz; H-7a), 4.22 (1H; *J*=12.1 Hz; H-8a), 5.77 (1H; br s; H-7b) and 3.93 (1H; s; H-8b). ^{13}C NMR (aseton d_6 400 MHz), δ_C (ppm): 95.4 (C-12b), 101.3 (C-12a), 106.5 (C-14a), 111.4 (C-14b), 115.4 (C-3a,5a), 116.2 (C-3b,5b), 130.4 (C-2a,6a), 118.7 (C10b), 121.3 (C-10a), 131.1 (C-1a), 135.4 (C-1b), 140.7 (C-9b), 142.6 (C-9a) 155.8 (C-4b), 157.3 (C-13b), 157.4 (C-13a), 158.7 (C-4a), 159.0 (C-11a), 159.5 (C-11b) 41.4 (C-7b), 48.4 (C-8b), 49.9 (C-8a) and 88.4 (C-7a).

Antioxidant Activity

The antioxidant activities of compound 1 was screened by β-carotene bleaching and DPPH assay as previously described [12]. Butylatedhydroxytoluene (BHT) was used as a positive control.

Statistical Analysis

Values expressed are means of the three replicate determination ± standard deviation. All statistical analyses were carried out using SPSS 16.00 for Windows. To determine whether there were any differences between activities of samples, variance analysis (one-way ANOVA) was applied to the results. Values of $p < 0.05$ were considered as significant difference ($\alpha = 0.05$).

RESULTS AND DISCUSSION

Compound **1**, obtained as a brownish orange amorphous solid, exhibiting molecular ion peak in the positive-ion ESIMS spectrum at [M-H]$^+$ m/z 691.1525 corresponding to molecular formula $C_{42}H_{27}O_{10}$, which is classified to a resveratrol trimer. $\left|\alpha\right|_D^{122}$ = -14°(c 0.00014 MeOH). UV

Table 2. Antioxidant Activities of Compounds 1-5 from *S. acuminata*

Compounds	β-Carotene Bleaching (IC_{50} mM)	DPPH Radical (IC_{50} mM)
(-)-Hopeanolin	0.18 ± 0.01^{a}	6.58 ± 0.02^{d}
(-)-Laevifonol	0.22 ± 0.02^{a} [12]	4.21 ± 0.23^{c} [12]
(-)-α-Viniferin	0.18 ± 0.00^{a} [12]	6.29 ± 0.05^{d} [12]
(-)-Vaticanol B	0.10 ± 0.01^{a} [12]	0.84 ± 0.02^{a} [12]
(-)-Hopeaphenol	0.10 ± 0.01^{a} [12]	2.78 ± 0.16^{b} [12]
BHT*	0.09 ± 0.00^{a} [12]	0.95 ± 0.05^{a} [12]

[a-d] Mean within each column with different letters differ significantly ($p < 0.05$). Each value is presented as mean ± SD ($n = 3$). *Positive reference standards. IC_{50}, 50 % inhibition concentration.

absorption λ_{max} (MeOH) 282 nm. IR (KBr) γ_{max} 3382 cm^{-1} (OH), 2925 cm^{-1} (C-H aliphatic), 1700 cm^{-1} (C=O), 1615 cm^{-1}, 1519 cm^{-1} and 1451 cm^{-1} (C=C aromatic), 1213 cm^{-1} and 1171 cm^{-1} (C-O oxyaryl) and 835 cm^{-1} (*p*-disubstitutedbenzene). ^{1}H- and ^{13}C- NMR spectra closely resemble those of α-viniferin (**3**) [9], however, **1** showed two additional carbonyl signals (δ_C 176.5 and 179.5) and the disappearance of *meta*-coupled aromatic protons on the 3,5-dioxygenated phenyl group (ring C2) in the NMR spectra of **1**. This evidence strongly implies that **1** is an oxidation product of α-Viniferin (**3**). This inference is further strengthened by the observation of strong IR absorption bands recorded in ν_{max} 1700 cm^{-1} (C = O) and different *m/z* of 14 which corresponds to the addition of one oxygen atom and two hydrogen atoms decrease compared to α-viniferin [9].

^{1}H NMR spectrum of **1** recorded three pairs of signals of ortho coupled aromatic protons assignable to three units of 1.4-disubstituted benzene rings (rings A1, B1 and C1), which at δ_H 7.30 (2H, d, J = 8.4 Hz; 2a, 6a), 6.74 (2H, d, J = 8.4 Hz; 3a, 5a), 7.17 (2H, d, J = 9.0 Hz; 2b, 6b), 6.80 (2H, d, J = 8.4 Hz; 3b, 5b) and 7.25 (2H, d, J = 8.4 Hz; 2c, 6c), 6.75 (2H, d, J = 8.4 Hz; 3c, 5c). However, there are two pairs of doublet signals for meta coupled aromatic protons suggested there are only two units of 1,2,3,5-tetrasubstituted of the ring A2 and B2 [δ_H 6.27 (1H; J = 1.8 Hz; 12a), 6.67 (1H; J = 1.8 Hz; 14a) and 6.30 (1H; J = 1.8 Hz; 12b), 6:52 (1H; J = 1.2 Hz; 14b)]. Aliphatic proton signals of dihydrobenzofuran units were recorded at δ_H 5.97 (1H; J = 12.0 Hz; 7a), 4.74 (1H; J = 12.0 Hz; 8a) 6.19 (1H; br s; 7b), 3.96 (1H; br s; 8b) and 5.56 (1H; J = 2.4 Hz; 7c), 4.67 (1H; J = 4.2 Hz; 8c). In addition, there is an olefinic proton signals were recorded at δ_H 5.78 (1H; s; 12c).

APT ^{13}C-NMR spectrum recorded thirty six signals representing forty-two carbon atoms as α-viniferin [9] although, the signal recorded for aromatic carbon consists of twenty-four signals compare to α-viniferin [9], at δ_C 96.9 (C-12b), 96.7 (C-12a), 105.3 (C-14a), 105.7 (C-14b), 115.2 (C-3a, 5a), 115.0 (C-3b, 5b), 115.5 (C-3c, 5c), 121.3 (C10b), 117.1 (C-10a), 128.3 (C-2a, 6a), 127.2 (C-2c, 6c), 128.3 (C-2b, 6b) , 133.0 (C-1C), 129.7 (C-1a), 133.7 (C-1b), 139.0 (C-9b) and 136.7 (C-9a); seven carbon oxyaryl with each of the three signals on the A and B rings , while ring C only one signal was recorded, δ_C 157.7 (C-4a), 161.9 (C-11a), 160.7 (C-13a), 158.1 (C-4b), 160.2 (C-11b), 159.0 (C -13b) and 157.3 (C-4c). Signals for the three pairs of aliphatic carbon was recorded at δ_C 51.9 (C-8a), 89.1 (C-7a), 43.8 (C-8b), 88.0 (C-7b) and 49.3 (C-8c), 90.3 (C-7c)]. In addition, there are four carbon signals were recorded on a cyclic olefinic C2 that is δ_C 100.8 (C-12c), 147.2 (C-10c), 128.6 (C-9c) and 170.5 (C-11c) and two carbonyl carbon also been recorded at δ_C 176.5 (C-13c) and 179.5 (C-14c).

Comparison of spectroscopic data (Table **1**) with literature by Ge *et al.* (2006) [7] reinforce that the compound **1** is trimer resveratrol (-)-hopeanolin. Using the same methodology as that of **1**, it can be concluded that **2** was (-)-laevifonol [8], **3** was (-)-α-viniferin [9], **4** was (-)-vaticanol B [10] and **5** was (-)-hopeaphenol [11].

Compound **1**-**5** were screened for its antioxidant activities by β-carotene bleaching and DPPH assay. The results are summarized in Table **2**. Compound **1**-**5** showed potent ability to protect β-carotene bleaching by linoleic acid and also to scavenge DPPH radicals, which was comparable with that of BHT. Compound **1** exhibited antioxidant activity with no significant difference than the positive control BHT in the ability to protect β-carotene bleaching by linoleic acid and moderate antioxidant activity to scavenge DPPH radicals.

From literature reports, compound **1** which was first isolated from *H. exalata* [7] has been successfully isolated for the first time from the genus *Shorea* in this study, whereas (-)-laevifonol which commonly had been isolated from *Shorea* species that is *S. laeviforia* [8], *S. pinanga* [13], *S. balangeran* [14], *S. parvifolia* [15], *S. macroptera* [16] and *S. selanica* [17] also found in *H. nutans* [18]. Instead, vaticanol B which had been isolated more frequently in *Hopea* that is *H. utilis* [19], *H. sangal* [20], *H. drybalanoides* [21], *H. gregaria* [22], *H. mengarawan* [23] and *H. nutans* [18] also found in a few species of *Shorea* that is *S. selanica* [14] and *S. assamica* [24]. α-Viniferin and hopeaphenol which had been isolated from almost all of Dipterocarpacae plants [6, 21] can be regarded as a "chemical marker" of Dipterocarpaceous plants. The presence of compound **1** and the other four resveratrol oligomers in this species have great chemotaxonomic significance on the relationship between *Shorea* and *Hopea*.

CONCLUSION

Five resveratrol oligomers, namely (-)-hopeanolin, (-)-laevifonol, (-)-α-viniferin (-)-vaticanol B and (-)-

hopeaphenol, have been successfully isolated from the acetone extract of the twigs of *Shorea acuminata* (Dipterocarpaceae). The structures of these compounds were determined based on spectral analysis and comparison with literature reports. Compound **1** showed good antioxidant activity in β-carotene bleaching and moderate ability to scavenge DPPH radicals. The presence of these compounds in this species has great chemotaxonomic significance on the relationship between *Shorea* and *Hopea.*

CONFLICT OF INTEREST

The authors confirm that this article content has no conflicts of interest.

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