A New Flavonoid from Malaysian *Dipterocarpus cornutus*

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Abstract: Dipterocarpus cornutus *Dyer is commonly known as 'keruing'. It belongs to the family of Dipterocarpaceae, an important timber family in South East Asia.* D. cornutus *is listed as critically endangered on IUCN Red List. Since no comprehensive study has been documented on the chemical constituents of* D. cornutus*, there is an urgent need to study this plant comprehensively. Phytochemical study of the stem bark of* D. cornutus *afforded a new flavonoid (**1**) and nine known compounds, which consist of flavonoids (**2**, **3**), oligostilbenoids (**4**, **5**, **7**, **8**, **9**, **10**), and coumarin (**6**). The finding of the study contributes to the chemotaxonomic differentiation in the plants of the tribe Dipterocarpae.*

Keywords: Dipterocarpus cornutus*; Dipterocarpaceae; flavonoid; oligostilbenoids*

■ INTRODUCTION

Dipterocarpus, commonly known as 'keruing', is a plant genus that belongs to the family Dipterocarpaceae, which consists of approximately 75 species distributed in tropical regions [1-2]. This family of plant is known to contain sesquiterpenes [3], triterpenes [4-7], flavonoids [8], and resveratrol oligomers [1-2,9-14] and possesses diverse biological activities, such as anti-inflammatory [15-16], antioxidant [17], anticancer [1,4], anti-human immunodeficiency virus (HIV) [1,4,7,18], and antibacterial [19-21] activities. As part of our ongoing project searching for resveratrol oligomers in Malaysia Dipterocarpace [12,17,19-20,22-23], *Dipterocarpus cornutus* was selected for phytochemical investigation. *Dipterocarpus cornutus* is a species of tree in the family Dipterocarpaceae native to peninsular Malaysia, Singapore, Sumatra, and Kalimantan. It is known for having large leaves due to reaching heights of up to 50 meters tall. Its flowers are around 4 cm in diameter and of a pale yellow coloration [24]. Traditionally, this genus has been prescribed to cure skin inflammation, bronchial infection, colitis and anxiety, gonorrhea, gleets, ulcer, rheumatism, and liver diseases in Thailand, Cambodia, Laos, Vietnam, and the Philippines [25]. The Malaysian Red List of Peninsular Malaysia has reported that *Dipterocarpus cornutus* is critically endangered. To the best of our knowledge, there has been no report of pharmacological and phytochemical investigation for *D. cornutus* so far. Hence, the present study described the structural characterization of the new flavonoid derivative, 4-methoxy epigallocatechin-3-*O*-(3-methyl) gallate (**1**), based on spectroscopic data including ultraviolet (UV),

infrared (IR), mass spectrometry (MS), 1D and 2D nuclear magnetic resonance (NMR).

■ EXPERIMENTAL SECTION

Materials

Samples of the stem bark of *Dipterocarpus cornutus* was collected from Universiti Teknologi MARA Pahang Forest Reserve, Pahang, Malaysia. The voucher specimen SKD 2/6 was deposited at the Herbarium of Universiti Teknologi MARA Pahang, Malaysia, and identified by the wood lecturer, Tuan Sheikh Abdul Karim bin Tuan Yamani and with help from a botanist. The identification of species was carried out by comparing with the existing specimen in the herbarium by using the existing taxonomic keys of Symington [24]. The following adsorbents were used for purification: vacuum liquid chromatography with Merck Si-gel 60 (5–40 μm, cat.no. 1.07747), radial chromatography with Merck Si-gel 60 GF254 containing gypsum (5–40 μm, cat.no. 1.07749), and TLC analysis with Merck Kieselgel 60 F254 0.25 mm (cat.no. 1.05554). The silica gel and TLC were purchased from Merck (Germany). Solvents used for purification compounds were analytical grade (RCL Labscan), while extraction of sample industrial grade (Fisher chemical) is used in this study. The industrial grade was distilled before being used.

Instrumentation

UV and IR spectra were measured with Varian Conc. 100 instruments and a Perkin Elmer Spectrum One FTIR spectrometer (Perkin Elmer, USA), respectively. LC-MS/MS was determined on Agilent 6224 TOF-LC/MS using positive and negative mode (Agilent, Santa Clara, USA). The ^{1}H and ^{13}C APT NMR spectra were recorded using Bruker Advance Model (500 MHz, 300 MHz for ^{1}H and 125 MHz, 75 MHz for ^{13}C, respectively) (Switzerland). The melting points were measured using Melting-Point Apparatus with microscope JM628.

Procedure

Extraction and isolation

The dried powder of the stem bark of *D. cornutus* (5 kg) was macerated with acetone (3 × 10 L) and evaporated under reduced pressure to give a dark brown residue (300 g). The dried acetone extract was dissolved in a small volume of MeOH (300 mL), then added with diethyl ether to a volume ± 2 L to give a MeOH-diethyl ether soluble fraction (50 g) after decantation and evaporation. Further fractionation using various chromatography techniques was carried out consecutively [26]. Part of the fraction (2 × 20 g) was subjected to vacuum liquid chromatography (VLC), (diameter; 10 cm, silica gel: 250 g) with mixtures of *n*-hexane/EtOAc and MeOH to give four major fractions (DC_2–DC_5). DC_5 was purified yielded compound **1** (10 mg), compound **2** (15 mg), compound **3** (12 mg), and compound **7** (8 mg). Using the same methodology, purification of fraction DC_2 gave four major fractions ($DC_{2.1}$–$DC_{2.4}$). Purification of each fraction manages to isolate compound **4** (15 mg), compound **5** (15 mg), compound **6** (17 mg), and compound **9** (40 mg). DC_3 (7.5 g) was subjected to VLC (diameter: 10 cm and silica gel 250 g), which was performed with Hex:EtOAc:MeOH to give $DC_{3.1}$–$DC_{3.4}$. Purification of fraction $DC_{3.2}$ (245 mg) with radial chromatography (plate 1 mm, $CHCl_3$:MeOH (9.5:0.5) yielded compound **8** (8 mg). DC_4 (5.4 g) was selected to further purification using VLC with Hex:EtOAc:MeOH and yielded four fractions $DC4_{.1}$–$DC4_{.4}$. Subfraction $D_{4.2}$ (600 mg) chromatographed with RC and eluent system $CHCl_3$:EtOAc:MeOH (7.0:2.5:0.5 to 5.0:4.5:0.5) yielded compound labelled as compound **10** (7 mg).

4-methoxy-epigallocatechin-3-*O*-(4-methyl)gallate (1). Mp.: 192–195 °C. UV (MeOH) χmax (Log$_\epsilon$): 284 nm. IR spectrum (KBr) υ max (cm^{-1}): 3423 (OH), 2935 (C–H stretching), 1054 (C-O stretching). The MS *m/z* at [M-H]$^-$: 485. ^{1}H-NMR (methanol-d_4, 300 MHz) δ_H: 5.10 (1H, *s*, H-2), 5.50 (1H, *m*, H-3), 2.98 (1H, *dd*, *J* = 17.1, 4.5 Hz, H-4α), 3.06 (1H, *dd*, *J* = 17.1, 4.5 Hz, H-4β), 6.03 (1H, *d*, *J* = 2.1 Hz, H-6), 5.99 (1H, *d*, *J* = 2.4 Hz, H-8), 6.56 (1H, *s*, H-2′/6′), 3.76 (3H, *s*, OMe), 7.16 (1H, *s*, H-2′/6″), 7.16 (1H, *s*, H-6″) , 3.87 (1H, *s*, OMe). ^{13}C-NMR (methanol-d_4, 75 MHz) δ_C: 76.8 (C-2), 69.4 (C-2), 26.7 (C-4α/4β), 97.9 (C-4a), 155.6 (C-5/7), 95.3 (C-6), 94.8 (C-8), 151.0 (C-8a), 127.6 (C-1′), 105.6 (C-2′/6′), 147.4 (C-3′/5′), 134.5 (C-4′), 59.5 (OMe), 120.0 (C-1″), 106.2 (C-2″), 147.4 (C-3″), 143.0 (C-4″), 144.4 (C-5″), 106.2 (C-6″), 166.2 (CO), 55.4 (OMe).

■ RESULTS AND DISCUSSION

The phytochemical study on the acetone extract of the stem barks of *D. cornutus* yielded a new compound **1** together with nine known compounds known as davidiol A (**4**) [27], stenophyllol B (**5**) [28], hemsleyanol D (**7**) [29], ε-viniferin (**8**) [30], laevifonol (**9**) [31], ampelopsin F (**10**) [32], one coumarins: scopoletin (**6**) [33], two flavonoids: 4-*O*'-metylgallocatechin (**2**), and 4-*O*'-methylepigallocatechin (**3**) [34]. (Table 1, Fig. 1). Their chemical structures were established based on their spectroscopic evidence and comparison with the published data (Table 2, 3, and Fig. 2).

Compound **1** was obtained as an amorphous light-yellow solid and was predicted as a flavonoid compound based on TLC analysis. The sulphuric acid-vanillin spraying reagent was applied, and the yellow spot was formed. The UV spectrum showed maximum absorption signals at 286 and 320 nm in MeOH, which is typical for flavan-3-ol derivatives. The IR spectrum of compound **1** showed an absorption band for the hydroxyl group at 3518 cm^{-1}, 1602, and 1461 cm^{-1} for C=C aromatic and 1713 cm^{-1} for carbonyl (C=O). The melting point was determined between 192–195 °C. The molecular formula of $C_{24}H_{22}O_{11}$ was deduced from the molecular ion peak observed at m/z 485.1141 $[M-H]^-$ in the LC mass spectrum with calculated 14 DBE. The ^{1}H-NMR spectrum (Table 1) of compound **1** displayed its catechin

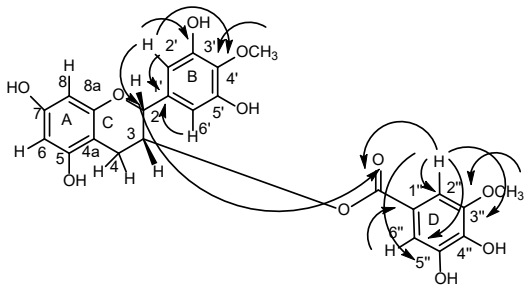

Fig 1. HMBC correlations of Compound **1**

Table 1. ^{1}H-NMR spectroscopy data of compound **1**

No C	δH (mul., *J* in Hz)	δC	HMBC (^{1}H⇔^{13}C)
2	5.10 (*d*, 1.5)	76.8	H-2-CO
3	5.50 (*m*)	69.4	-
4ax	2.98 (*dd*, 17.1, 4.5)	26.7	-
4eq	3.06 (*dd*, 17.1, 4.5)	26.7	-
4a		97.9	-
5/7		155.6	-
6	6.03 (*d*, 2.1)	95.3	-
8	5.99 (*d*, 2.1)	94.8	-
8a		151.0	--
1'		127.6	-
2'/6'	6.56 (*s*)	105.6	H-2'/6'-C-2,C-2'/6',C-4',C-3'/5'
3'/5'		147.4	-
4'		134.5	-
OMe	3.76 (*s*)	59.5	OMe-C-4'
1"		120.0	-
2"	7.16 (*s*)	106.2	H-2"-C-2"/6",C-1",C-5",C-3", CO
3"		147.4	-
4"		143.0	-
5"		144.4	-
6"	7.16 (*s*)	106.2	H-6" -C-2"/6",C-1",C-5",C-3", CO
CO		166.2	H-2/CO
OMe	3.87 (*s*)	55.4	C-3"

Measured in methanol, d_4 at 300 MHz (^{1}H) and 75 MHz (^{13}C APT)

Table 2. [1]H-NMR spectroscopy data of for oligomeric resveratrol

No H	tetramer	trimer		dimer		
	7	4	5	8	9	10
2/6a	7.22 (d, 8.7)	7.21 (d, 8.7)	6.88 (d, 8.7)	7.18 (d, 8.7)	6.76 (d, 8.1)	7.09 (d, 8.4)
3/5a	6.78 (d, 8.7)	6.80 (d, 8.7)	6.77 (d, 8.7)	6.81 (d, 8.7)	6.76 (d, 8.1)	6.76 (d, 8.4)
7a	5.77 (d, 11.7)	6.09 (d, 3.3)	5.84 (d, 3.3)	5.39 (d, 6.6)	5.06 (d, 7.5)	4.18 (d, 1.5)
8a	4.41 (d, 11.7)	4.42 (d, 9.6)	5.07 (d, 3.3)	4.35 (d, 6.6)	3.29 (d, 7.5)	3.35 (br s)
10a	-	-	-	6.18 (d, 1.8)	5.93 (d, 2.1)	-
12a	6.36 (d, 2.4)	6.44 (d, 2.1)	6.31 (d, 2.1)	6.20 (d, 2.1)	6.17 (t, 2.1)	6.06 (d, 2.4)
14a	6.12 (d, 1.8)	6.57 (d, 2.4)	6.25 (d, 2.4)	6.18 (d, 1.8)	5.93 (d, 2.1)	6.53 (d, 2.4)
2/6b	6.94 (d, 8.7)	7.02 (d, 8.7)	7.20 (d, 8.4)	7.07 (d, 8.7)	6.97 (d, 8.1)	6.78 (d, 8.4)
3/5b	6.48 (d, 8.7)	6.60 (d, 8.7)		6.68 (d, 8.7)	6.76 (d, 8.1)	6.58 (d, 8.5)
7b	5.29 (d, 3.4)	5.28 (br s)		6.87 (d, 16.2)	5.29 (d, 10.8)	3.64 (br s)
8b	3.38 (d, 10.9)	4.24 (d, 11.4)		6.61 (d, 16.2)	3.27 (d, 10.8)	4.12 (br s)
12b	6.02 (s)	6.04 (s)		6.27 (d, 1.8)	6.20 (d, 2.0)	6.15 (d, 2.4)
14b	-	-		6.65 (d, 1.8)	7.14 (br s)	6.45 (d, 2.4)
Ascorbic acid					4.42(br s), 4.22 (m), 3.96 (dd, 4.4.10)	
2/6c	6.72 (d, 8.7)	6.74 (d, 8.7)	7.29 (d, 8.1)			
3/5c	6.52 (d, 8.7	6.61 (d, 8.7)	6.68 (d, 8.1)			
7c	4.55(d, 1.9)	4.39 (d, 9.3)				
8c	3.89 (dd, 11.7, 11.2)	2.97 (dd, 11.7, 9.9)				
10c	-	6.43 (d, 2.4)				
12c	6.23 (d, 2.0)	6.19 (t, 2.1)				
14c	6.79 (s)	6.43 (d, 2.4)				
2/6d	7.06 (d, 8.4)					
3/5d	6.82 (d, 8.4)					
7d	4.92(d, 1.5)					
8d	3.50 (br s)					
10d	5.34 (br s)					
12d	6.07 (t, 2.1)					
14d	5.34 (br s)					

The [1]H were measured with 500 MHz in acetone, d_6

Table 3. [1]H-NMR spectroscopy data of for non-oligomeric resveratrol

No H	2	3	6
2	4.59 (d, 7.5)	4.822 (s)	-
3	3.99 (m)	4.20 (m)	6.20 (d, 9.3)
4a	2.84 (dd, 16.2, 5.1)	2.73 (dd, 16.5, 3.0)	7.84 (d, 9.3)
4b	2.56 (dd, 16.2, 7.8)	2.83 (dd, 16. 5,3.0)	
5		-	7.12 (s)
6	5.95 (d, 2.1)	6.01 (d, 2.0)	
8	5.86 (d, 2.1)	5.91 (d, 2.0)	6.78 (s)
OMe	3.80 (s)	3.78 (s)	3.92 (s)
2'/6'	6.42(s)	6.58 (s)	

The [1]H was measured with 300 MHz in methanol, d_4

skeleton through resonances displayed in the ring A at the downfield region at δ_H 6.03 (*d*, *J* = 2.1 Hz) and 5.99 (*d*, *J* = 2.1 Hz), which were assigned to proton H-6 and H-8, respectively. An AB spin system signal was also observed in the downfield region at δ_H 6.56 as singlet signals each for H-2′/6′ indicated there were trisubstituted at H-3′,4′ and 5′. Thus, the signals revealed the presence of ring B for this catechin derivative. The presence of ring C was identified upfield region for the signal of proton aliphatic at δ_H 5.10 (H-2), 5.50 (H-3), 2.98 (H-4_{ax}), and 3.33 (H-4_{eq}), which revealed the occurrence of dihydroxypyran heterocyclic compound

with a molecular formula of C_5H_8O with the attachment of hydroxyl group on C-3. Thus, establishing this compound as a flavan-3-ol skeleton.

Basically, the ^{1}H and ^{13}C APT NMR of compound **1** closely resembled those of compound **3** as 4-*O'*-methylepigallocatechin [34]. However, from the HMBC spectrum correlation (Fig. 2), it showed the occurrence of a cross peak at ^3J HMBC for H-2 and carbon carbonyl, which revealed that this catechin was attached to GMe (methyl gallate) established for the ring D. The addition of GMe, which is proposed to be attached at C-3 establishing from compound **3** (4-*O'*-methylepigallocatechin) and was named as 4-methoxyepigallocatechin-3-*O*-(3-methyl) gallate.

The remaining ^{1}H-NMR for *ortho* coupling H-2″/6″ at δ_H 7.16 (*s*) revealed the occurrence of GMe, which attached to the methoxy group at C-3″. The important peak for this methoxy at ring D can be seen at δ_H 3.87, and another methoxy at δ_H 3.76 was attributable for C-4′ at ring B. HMBC spectrum revealed the occurrence for proton methoxy of GMe (ring D) correlate with C-3″ at ^3J while proton methoxy (ring B) correlate with C-4′ at ^3J (Fig. 1). The ^{1}H and ^{13}C-NMR assignments obtained in this work were achieved primarily using proton–carbon correlation methods, specifically HMQC and HMBC experiments for long-range correlations. Basically, the ^{13}C APT NMR indicated the presence of eight quaternary δ_C 97.9 (C-4a), 97.9 (C-5/7), 151.0 (C-8a), 151.0 (C-1′), 147.5 (C-3′/5′), 134.5 (C-4′), six methine carbon at δ_C 95.3 (C-6), 94.8 (C-8), 69.4 (C-3), 76.8 (C-2), 106.7 (C-2′/C-6′), one methylene carbon at δ_C 26.7 (C-4) and one methoxy at δ_C 3.871. The ^{13}C APT NMR also indicated there were six additional signals from spectrum compound **3** which have four quaternary carbons at δ_C 127.0 (C1″), 147.4 (C-3″), 143.0 (C-4″), 144.4 (C-5″), two methane at δ_C 105.6 (C-2″), 120 (C-6″), the signal for carbonyl (C=O) at δ_C 166.24 and also methoxy carbon at δ 3.753.

In our present study, the discovery of six oligomeric which consist of dimer (**8**, **9**, **10**), trimer (**4**, **5**), tetramer (**7**), and also non-oligomeric compounds (**6**, **1**, **2,** and **3**), indicated the variations in their chemical

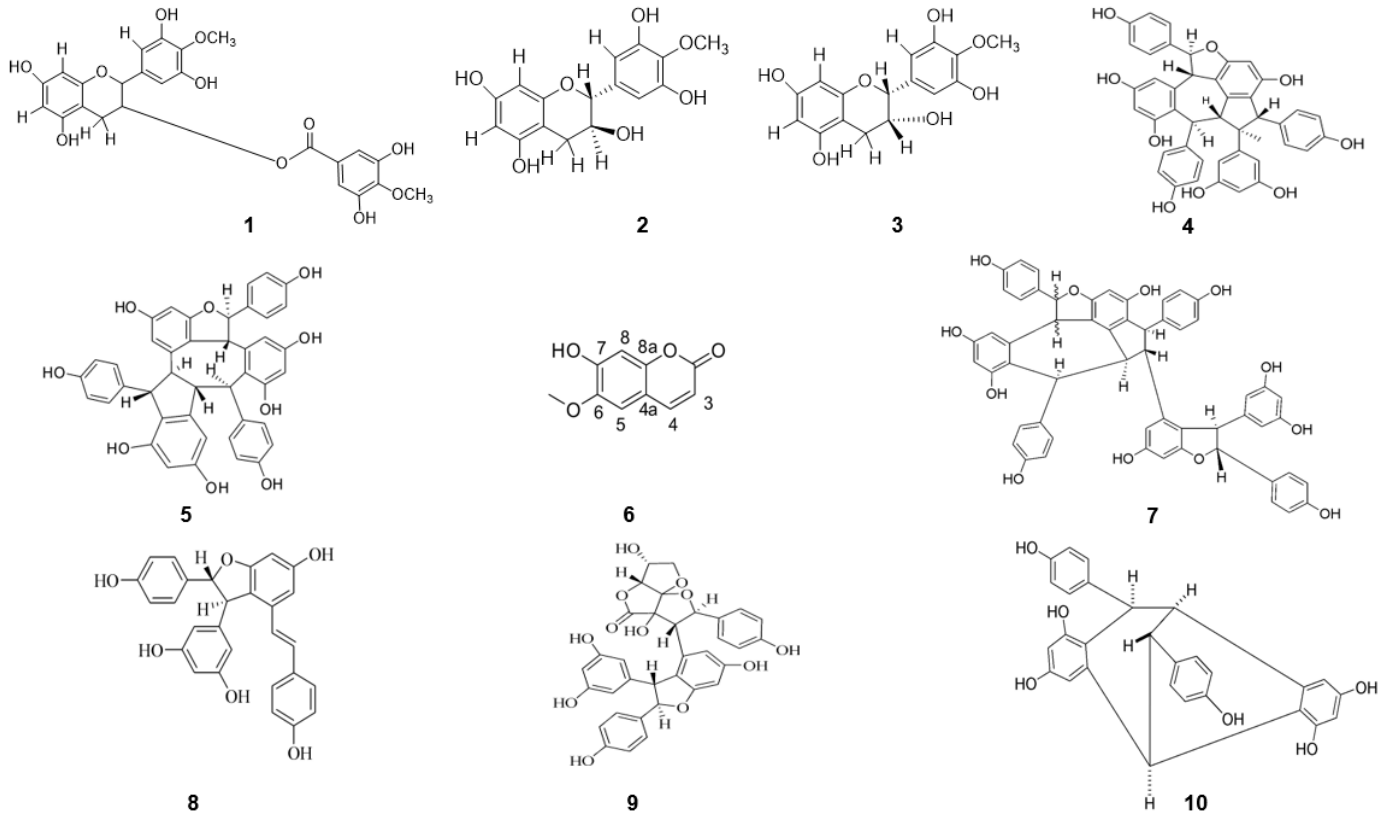

Fig 2. Compounds isolated from *D. cornutus*

constituents from *D. cornutus*. The six oligometric are commonly found in Dipterocarpaceae [14]. The presence of (**8**) has no chemotaxonomic significance as it is regarded as the general precursor for oligostilbenoids [14,26]. Compound (**9**) is a unique oligostilbenoid formed from the condensation of (**8**) and ascorbic acid. Another dimer resveratrol, (**4**) with the skeleton bicycle[3.2.1] octane found in *Dipterocarpus grandiflorus,* indicated that these metabolites have a significant relationship with these species [9]. The significant findings of these resveratrol oligomers are compound (**4**) and (**5**), which is the first time reported in *Dipterocarpus*. The presence of compound (**7**), tetramer resveratrol, also revealed the relationship of chemotaxonomy characteristics between *D. cornutus* and *D. grandiflorus*.

This study also discovered non-oligomeric resveratrol, which is (**6**, **2,** and **3**). Compound **6** can be classified as a significant compound in Dipterocarpaceae, which can be found abundantly. However, the presence of two flavonoids: 4-*O*′-methylgallocatechin (**2**) and 4-*O*′-methylepigallocatechin (**3**) are only reported in the family other than Dipterocarpaceae. To the best of our knowledge, 4-methoxy epigallocathechin-3-*O*-(3-methyl) gallate (**1**), flavan-3-ol derivative, was isolated for the first time in the plant. Catechin, epicatechin, gallocatechin, and epigallocatechin are the flavanol units, except that the two latter compounds have hydroxyl units. This methoxylated analog in the catechin or gallocatechin series has not yet been reported in Dipterocarpaceae family. We now describe the isolation and structural elucidation of compound **2** as an isomer mixture of compound **3**.

Compound **6** is not in the same class as stilbenoid, but it's derived from the same route called the shikimic acid pathway, which replaces most plant phenolic biosynthesis. The coumarin nucleus (benzo-2-pyrone) is derived from cinnamic acid (phenyl acrylic skeleton) in its biosynthesis [35]. Basically, all the compounds originate from the same route alongside coumarin and flavonoid. It starts from phenylalanine via the shikimate pathway, where it branches off to different biosynthetic routes in order to synthesize oligomeric and non-oligomeric compounds [35]. Tables 2 and 3 showed the ^{1}H-NMR spectroscopy for oligomeric resveratrol and non-oligomeric resveratrol, respectively. The identification of the flavonoids in this study contributes significantly towards the diversity of secondary metabolites in the tribe Dipterocarpae. Flavonoids are potential taxonomic markers to distinguish Dipterocarp with closely anatomical features such as the Balau Group in the genus *Shorea* [35]. Flavonoids are important chemical markers due to their characteristics such as structural variability, chemical stability, ubiquitous occurrence, easy and rapid identification. Moreover, flavonoids are also used to solve the problems of plant identification where flowering and fruit development do not frequently occur [36].

■ CONCLUSION

This study found that *Dipterocarpus cornutus* possesses the ability to synthesize oligomeric and non-oligomeric compounds *via* different biosynthetic routes. This finding was rather interesting, as this can be used to investigate the relationship between species and genera in Dipterocarpaceae.

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