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PHENOLIC CONSTITUENTS FROM PROPOLIS OF STINGLESS BEE *TETRIGONILLA COLLINA*

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Summary

Phenolic Constituents from Propolis of Stingless Bee *Tetrigonilla collina*

Phytochemical study of propolis of stingless bee *Tetrigonilla collina* collected in Kon Ka Kinh forest led to the isolation of six phenolic compounds: siamyl benzoate (**1**), (+)-pinoresinol (**2**), coniferyl aldehyde (**3**), *p*-coumaraldehyde (**4**), *trans*-cinnamic acid (**5**), and vanillin (**6**). The structures of isolated compounds **1-6** were elucidated by spectroscopic (NMR and MS) analysis and comparison with the previous data. This is the first chemical investigation of *T. collina* propolis.

Keywords: *Tetrigonilla collina, Lignan, Benzoate, Phenylpropanoid, Phenolic.*

1. Introduction

Stingless bees that belong to the Meliponini tribe are predominantly found in tropical regions, including South America, Africa, Australia, India, and Southeast Asia [1]. Similar to honey bees (*Apis mellifera*), stingless bees gather plant resin, a substance known as propolis, which they use to construct and protect their hives. In many countries, propolis has been utilized as a traditional remedy for various ailments and to enhance immunity. Nowadays, propolis is incorporated into numerous cosmetic products and functional foods. As a natural product derived from plants, the chemical constituents of propolis vary based on the local flora, resulting in significant chemical diversity [2]. Previous phytochemical studies have revealed that stingless bee propolis contains various natural compounds, including flavonoids, coumarins, xanthones, phenolic acids, and triterpenes. Extracts of propolis, and its isolated compounds, exhibit a range of pharmacological activities such as antibacterial, anticancer, antioxidant, and anti-inflammatory effects [2],[3],[4],[5],[6].

Tetrigonilla collina (Smith, 1857) is a species of stingless bee found in Southeast Asian countries, including Indonesia, Laos, Malaysia, Myanmar, and Vietnam. This bee species typically constructs its nests underground, often within termite colonies [7]. To date, there have been no chemical or biological studies published on the propolis derived from the stingless bee *T. collina*. In this report, we aim to outline the phenolic constituents isolated from *T. collina* propolis collected in the Kon Ka Kinh forest, Gia Lai province.

2. Materials and methods

2.1. Material

The propolis sample was collected from a stingless bees' hive in Kon Ka Kinh forest, Gia Lai province, Vietnam, in 2023. The stingless bee species was identified as *Tetrigonilla collina* (Smith, 1857) by Prof. Nguyen Thi Phuong Lien and Dr. Tran Thi Ngat, Institute of Ecology and Biological Resources, Vietnam Academy of Science and Technology. The specimen of the stingless bee is preserved at the Institute of Ecology and Biological Resources, bearing the voucher specimen designation SB-GL-01.

2.2. General experimental procedures

NMR spectra were measured on Bruker AVANCE III HD 500 MHz or Bruker AVANCE NEO 600 MHz spectrometers with tetramethylsilane as the internal standard. ESI-MS analyses were conducted on an Agilent 1260 series single quadrupole LC/MS system. Column chromatography (CC) was carried out on *silica gel* 60 (230-400 mesh, Merck), Sephadex® LH20, and YMC RP-18 (120 Å, 150 µm). Thin layer chromatography was performed on *silica gel* 60

GF_{254} and RP-18 GF_{254s}. Compounds were visualized by spraying with 10% sulfuric acid and heating. All solvents used were laboratory grade reagents and were distilled before use.

2.3. Extraction and isolation

The propolis of *T. collina* (410 g) was extracted with EtOH (4 × 5 L for 48 h) at room temperature, and the EtOH extracts were filtered and concentrated under reduced pressure. The crude extract (304 g) was suspended in distilled water (1 L) and extracted with EtOAc. After evaporation of the solvent *in vacuo*, the EtOAc extract (250 g) was obtained. The EtOAc residue was applied to *silica gel* CC, eluted with a solvent gradient of *n*-hexane/EtOAc (100/1 to 0/100, v/v) to yield 15 fractions (E1-E15).

Fraction E7 (5.4 g) was chromatographed by *silica gel* CC (*n*-hexane/EtOAc v/v, 19:1) to give 11 fractions E7A-E7K. Fraction E7I (1.24 g) was separated by *silica gel* CC (*n*-hexane/EtOAc v/v, 9:1) to yield ten fractions E7I1-E7I10. Fraction E7I10 (454 mg) was fractionated by *silica gel* CC (*n*-hexane/EtOAc v/v, 8:2) to give compound **3** (5.5 mg).

Fraction F9 (5.0 g) was separated by *silica gel* CC (CH_2Cl_2/EtOAc v/v, 50:1) to afford 8 fractions E9A-E9H. Fraction E9C (1.31 g) was chromatographed by *silica gel* CC (*n*-hexane/EtOAc v/v, 9:1) to give 10 fractions E9C1-E9C10. Fraction E9C6 (104 mg) was further purified by *silica gel* CC (*n*-hexane/acetone v/v, 9:1) to obtain 2 fractions E9C6A-E9C6B (2.8 mg). Fraction E9C6A (50 mg) was purified by reversed-phase *silica gel* CC (acetone/H_2O v/v, 1:1) to give compound **4** (3.2 mg). Fraction E9C6B (52 mg) was fractionated by reversed-phase *silica gel* CC (acetone/H_2O v/v, 2:1) to give compound **5** (5.1 mg).

Fraction F11 (8.2 g) was subjected to *silica gel* CC (*n*-hexane/acetone v/v, 9:1) to afford 9 fractions E11A-E11I. Fraction E11G (0.545 g) was chromatographed by *silica gel* CC (CH_2Cl_2/EtOAc v/v, 20:1) to give 4 fractions E11D1-E11D4. Fraction E11D4 (150 mg) was purified by reversed-phase *silica gel* CC (acetone/H_2O v/v, 2:1) to yield compound **1** (2.8 mg). Fraction E11G (0.545 g) was purified by *silica gel* CC (CH_2Cl_2/EtOAc v/v, 20:1) to obtain 3 fractions E11G1-E11G3. Fraction E11G1 (120 mg) was further purified by reversed-phase *silica gel* CC (acetone/H_2O v/v, 2:1) to give compound **6** (10 mg). Fraction E11H (1.4 g) was fractionated by reversed-phase *silica gel* CC (acetone/H_2O v/v, 2:1) to afford 7 fractions E11H1-E11H7. Fraction E11H1 (30 mg) was purified by Sephadex® LH20 CC and eluted with MeOH to yield compound **2** (10 mg).

Siamyl benzoate (**1**): pale yellow solid; ESI-MS: *m/z* 319 $[M+H]^+$. ^{1}H-NMR (500 MHz, $CDCl_3$) δ (ppm): 8.00 (2H, d, J = 8.0 Hz, H-2', H-6'), 7.57 (1H, t, J = 8.0 Hz, H-4'), 7.44 (2H, t, J = 8.0 Hz, H-3', H-5'), 6.92 (1H, d, J = 1.5 Hz, H-2), 6.89 (1H, d, J = 8.0 Hz, H-5), 6.87 (1H, d, J = 8.0, 1.5 Hz. H-6), 4.67 (1H, d, J = 7.0 Hz, H-7), 4.05 (1H, m, H-8), 4.37 (1H, dd, J = 3.5, 17.0 Hz, H-9a), 4.22 (1H, dd, J = 5.5, 17.0 Hz, H-9b), 3.86 (3H, s, OCH_3). ^{13}C-NMR: See **Table 1**.

(+)-Pinoresinol (**2**): white solid, $[\alpha]_D^{25}$: +67 (*c* 0.2, $CHCl_3$). ESI-MS *m/z* 359 $[M+H]^+$. ^{1}H-NMR (500 MHz, $CDCl_3$), δ (ppm): 6.89 (2H, d, J = 2.0 Hz, H-2, H-2′), 6.88 (2H, d, J = 8.0 Hz, H-5, H-5′), 6.82 (2H, dd, J = 1.5, 8.0 Hz, H-6, H-6′), 5.64 (2H, s, OH), 4.73 (2H, d, J = 4.5 Hz, H-7, H-7′), 4.25 (2H, dd, J = 7.0, 9.0 Hz, H-9β, H-9′β), 3.89 (6H, s, OMe), 3.89 (2H, dd, J = 4.0, 9.5 Hz, H-9α, H-9′α), 3.10 (2H, m, H-8, H-8′). ^{13}C-NMR: See **Table 1**.

Coniferyl aldehyde (**3**): yellow solid, ESI-MS: *m/z* 179 $[M+H]^+$. ^{1}H-NMR (500 MHz, $CDCl_3$) δ (ppm): 9.65 (1H, d, J = 8.5 Hz, H-9), 8.40 (1H, d, J = 15.5 Hz, H-7), 7.12 (1H, dd, J = 2.0, 8.0 Hz, H-6), 7.07 (1H, d, J = 2.0 Hz, H-2), 6.96 (1H, d, J = 8.0 Hz, H-5), 6.59 (1H, dd, J = 8.5, 15.5 Hz, H-8), 3.95 (3H, s, OMe). ^{13}C-NMR: See **Table 2**.

***p*-Coumaraldehyde** (**4**): yellow solid, ESI-MS: *m/z* 149 $[M+H]^+$. ^{1}H-NMR (600 MHz, $CDCl_3$) δ (ppm): 9.65 (1H, d, J = 7.8 Hz, H-9), 7.48 (2H, d, J = 8.4 Hz, H-2 and H-6), 7.41 (1H, d, J = 15.6 Hz, H-7), 6.89 (2H, d, J = 8.4 Hz, H-3 and H-5), 6.60 (1H, dd, J = 7.8, 15.6 Hz, H-8). ^{13}C-NMR: See **Table 2**.

***trans*-Cinnamic acid** (**5**): yellow solid, ESI-MS: *m/z* 181 $[M+H]^+$. ^{1}H-NMR (600 MHz, CD_3OD) δ (ppm): 7.69 (1H, d, J = 15.6 Hz, H-7), 7.62 (2H, m, H-2 and H-6), 7.42 (1H, m, H-5), 6.50 (1H, dd, J = 15.6 Hz, H-8). ^{13}C-NMR: See **Table 2**.

Vanillin (**6**) pale yellow solid, ESI-MS: *m/z* 153 $[M+H]^+$. ^{1}H-NMR (500 MHz, $CDCl_3$) δ (ppm): 9.82 (1H, s, CHO), 7.43 (2H, m, H-2 and H-6), 7.04 (1H, d, J = 8.5 Hz, H-5), 6.26 (1H, s, OH), 3.96 (3H, s, OMe). ^{13}C-NMR (500 MHz, $CDCl_3$) δ (ppm): 190.8 (CHO), 151.7 (C-3), 147.1 (C-4), 129.9 (C-1), 127.4 (C-6), 114.4 (C-5), 108.8 (C-2), 56.1 (OMe).

3. Results and discussion

The ethyl acetate extract of *T. collina* propolis was repeatedly fractionated by chromatographic methods to obtain six phenolic compounds. The

structures of compounds **1-6** were elucidated as siamyl benzoate (**1**), pinoresinol (**2**), coniferyl aldehyde (**3**), *p*-coumaraldehyde (**4**), *trans*-cinnamic acid (**5**), and vanillin (**6**) by NMR and ESI-MS data analyses and comparison with the published results in the literature (Fig. 1).

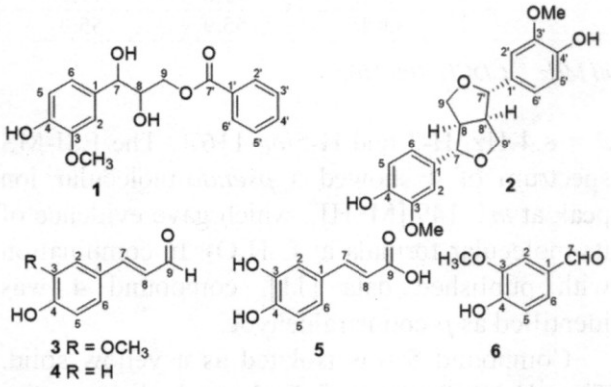

Fig. 1. Chemical structures of compounds **1-6** from *T. collina* propolis

Compound **1** was isolated as a pale yellow solid. The ^{1}H-NMR spectrum of **1** exhibited signals of a phenyl moiety at δ_H 8.00 (2H, d, J = 8.0 Hz, H-2', H-6'), 7.57 (1H, t, J = 8.0 Hz, H-4'), and 7.44 (2H, t, J = 8.0 Hz, H-3', H-5'). Another 1,3,4-substituted phenyl ring was detected by the presence of an ABX spin system at δ_H 6.92 (1H, d, J = 1.5 Hz, H-2), 6.89 (1H, d, J = 8.0 Hz, H-5) and 6.87 (1H, dd, J = 8.0, 1.5 Hz, H-6). In addition, two oxymethine protons appeared at δ_H 4.67 (1H, d, J = 7.0 Hz, H-7) and 4.05 (1H, m, H-8), two oxymethylene protons appeared at δ_H 4.37 (1H, dd, J = 3.5, 17.0 Hz, H-9a) and 4.22 (1H, dd, J = 5.5, 17.0 Hz, H-9b), and a methoxy group was detected at δ_H 3.86 (3H, s, OMe). The ^{13}C-NMR spectrum of **1** showed 17 carbons, including one downfield carbonyl carbon at δ_C 166.8 (C-7'); 12 aromatic ring carbons ranging from δ_C 146.8 to 109.0; 2 oxymethine carbons at δ_C 74.7 (C-7) and 74.5 (C-8); one oxymethylene carbon at δ_C 65.8 (C-9); and one methoxy group at δ_C 55.9. The ESI-MS spectrum of **1** showed a *pseudo*-molecular ion peak at *m/z* 319 $[M+H]^+$, giving evidence of its molecular formula as $C_{17}H_{18}O_6$. The NMR and MS spectra suggested that **1** was the benzoate ester of a phenylpropanoid.

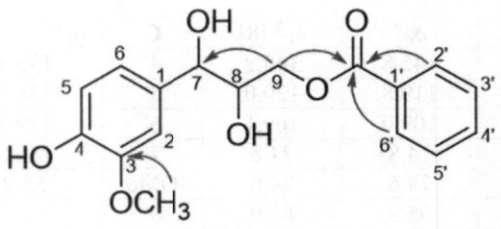

Fig. 2. Key HMBC correlations of compound **1**

The location of the benzoate moiety at C-9 was confirmed by the HMBC correlations of H-9 (δ_H 4.37 and 4.22) to C-7' (δ_C 166.8). In addition, the position of the methoxy group at C-3 was determined by the HMBC correlation of the methoxy group (δ_H 3.86) and C-3 (δ_C 146.9) (Fig. 2). Compound **1** was identified as siamyl benzoate by the comparison of NMR data with the previous literature [8].

Compound **2** was isolated as a white solid. The molecular formula of **2** was assigned as $C_{20}H_{22}O_6$ based on a *pseudo*-molecular ion peak at *m/z* 359 $[M+H]^+$ in the ESI-MS spectrum and NMR data. The ^{1}H and ^{13}C NMR spectra revealed signals of eleven protons and ten carbons, respectively, hence, these signals are doubling peaks. In the ^{1}H-NMR spectrum, signals of two identical ABX systems were observed at δ_H 6.89 (d, J = 2.0 Hz, H-2, H-2′), 6.88 (d, J = 8.0 Hz, H-5, H-5′), 6.82 (dd, J = 1.5, 8.0 Hz, H-6, H-6′). In addition, two oxymethine protons at δ_H 4.73 (d, J = 4.5 Hz, H-7, H-7′); four oxymethylene protons at δ_H 4.25 and 3.89 (H-9, H-9′), two methoxy groups at δ_H 3.89 and two methine protons at δ_H 3.10 (H-8, H-8′) were detected. The ^{13}C-NMR spectrum of **2** showed 20 carbons, including 12 aromatic ring carbons at δ_C 146.7 (C-4, C-4′), 145.2 (C-3, C-3′), 132.9 (C-1, C-1′), 118.9 (C-6, C-6′), 114.2 (C-5, C-5′), 108.6 (C-2, C-2′); two oxymethine carbons at δ_C 85.8 (C-7); two oxymethylene carbon at δ_C 71.6 (C-9); two methoxy groups at δ_C 55.9; and two methine carbons at δ_C 54.1. Comparing spectral and optical rotation data [9], compound **2** was assigned as (+)-pinoresinol.

Table 1. ^{13}C-NMR spectroscopic data of **1** and **2**

	1					**2**		
C	δ_C[a]	δ_C[b] [8]	**C**	δ_C[a]	δ_C[b] [8]	C	δ_C[a]	δ_C[c] **[9]**
1	131.9	132.0	1′	129.6	129.7	1,1′	132.8	132.9
2	114.5	114.6	2′	129.7	129.8	2,2′	108.6	108.6
3	146.8	146.9	3′	128.4	128.6	3,3′	146.5	146.7

	1					2		
C	δ_C[a]	δ_C[b] [8]	C	δ_C[a]	δ_C[b] [8]	C	δ_C[a]	δ_C[c] [9]
4	145.8	145.9	4′	133.3	133.4	4,4′	145.1	145.3
5	119.8	120.0	5′	128.5	128.6	5,5′	114.2	114.3
6	109.0	109.1	6′	129.7	129.8	6,6′	118.9	120.7
7	74.8	74.8	7′	166.8	167.0	7,7′	85.8	85.9
8	74.6	74.6	OMe	55.9	55.9	8,8′	54.2	54.2
9	65.9	65.9				9,9′	71.6	71.7
						OMe	55.9	55.9

[a] $CDCl_3$, 125 MHz, [b] $CDCl_3$, 100 MHz. [c] $CDCl_3$, 100 MHz.

Compound **3** was isolated as a yellow solid. The ^{1}H-NMR data of **3** showed signals for an ABX system at δ_H 7.12 (1H, dd, J = 2.0, 8.0 Hz, H-6), 7.07 (1H, d, J = 2.0 Hz, H-2), 6.96 (1H, d, J = 8.0 Hz, H-5), two protons of a *trans*-double bond at δ_H 8.40 (1H, d, J= 15.5 Hz, H-7), and 6.59 (1H, dd, J = 8.5, 15.5 Hz, H-8), an aldehyde group at δ_H 9.65 (1H, d, J = 8.5 Hz, H-9) and a methoxy group at δ_H 3.95 (3H, s, OMe). The ^{13}C-NMR revealed signals of nine carbons, including a carbonyl at δ_C 193.5, six aromatic carbons and two *trans*-olefinic carbons in the δ_C range of 152.9-109.4, and a methoxy group at δ_C 56.0 (Table 2). The ESI-MS spectrum of **3** showed a *pseudo*-molecular ion peak at *m/z* 179 $[M+H]^+$, which indicated its molecular formula as $C_{10}H_{10}O_3$. The NMR data of **3** were consistent with those of coniferyl aldehyde [10]. Thus, compound **3** was determined as coniferyl aldehyde.

Compound **4** was isolated as a yellow solid. Its ^{1}H- and ^{13}C-NMR spectra were very similar to those of **3**, except that the 1,3,4-substituted phenyl moiety was replaced by a 1,4-substituted phenyl group. The signals of the 1,4-substituted phenyl group were observed at δ_H 7.48 (2H, d, J = 8.4 Hz, H-2 and H-6)/δ_C 130.8 and 6.89 (2H, d, J = 8.4 Hz, H-3 and H-5/δ_C 116.1. The ESI-MS spectrum of **4** showed a *pseudo*-molecular ion peak at *m/z* 149 $[M+H]^+$, which gave evidence of its molecular formula as $C_9H_8O_2$. In combination with published data [11], compound **4** was identified as *p*-coumaraldehyde.

Compound **5** was isolated as a yellow solid. The ^{1}H-NMR data of **5** showed signals of a phenyl group at δ_H 7.62 (2H, m, H-2 and H-6) and 7.42 (3H, m, H-3, H-4, and H-5), two protons of a *trans*-double bond at δ_H 7.69 (1H, d, J = 15.6 Hz, H-7) and 6.50 (1H, dd, J = 15.6 Hz, H-8). The ^{13}C-NMR revealed signals of nine carbons, including a carboxylic acid group at δ_C 170.5 (C-9), six carbons of a phenyl group at δ_C 135.8 (C-1), 129.2 (C-2, C-6), 130.0 (C-3, C-5), 131.4 (C-4), and two *trans*-olefinic carbons at δ_C 146.2 (C-7) and 119.4 (C-8). The ESI-MS spectrum of **5** revealed a *pseudo*-molecular ion peak at *m/z* 181 $[M+H]^+$ suggesting its molecular formula as $C_9H_8O_4$. In comparison with the previously reported data [12], compound **5** was assigned as *trans*-cinnamic acid.

Compound **6** was determined as vanillin based on the agreement of NMR data with those in the previous report [13].

Table 2. ^{13}C-NMR spectroscopic data of **3-5**

	3		4		5	
C	δ_C[a]	δ_C[b] [10]	#δ_C[a]	δ_C[c] [11]	#δ_C[d]	δ_C[e] [12]
1	126.7	126.7	126.9	126.8	135.8	135.8
2	109.4	109.3	130.6	130.8	129.1	129.2
3	146.9	147.0	116.1	116.1	130.0	130.0
4	148.9	148.8	158.8	158.6	131.4	131.4
5	114.9	114.9	116.1	116.1	130.0	130.0
6	124.0	124.1	130.6	130.8	129.1	129.2
7	152.9	153.1	152.6	152.9	146.2	146.3
8	126.5	126.4	126.5	126.4	119.4	119.4
9	193.5	193.6	193.7	193.9	170.5	170.4
OMe	56.0	56.0				

[a] $CDCl_3$, 125 MHz, [b] $CDCl_3$, 75 MHz, [c] $CDCl_3$, 100 MHz, [d] CD_3OD, 125 MHz, [e] CD_3OD, 100 MHz.

This is the first chemical investigation of the propolis of the stingless bee *T. collina*. Compounds **2** and **6** were also isolated from stingless bee *Lepidotrigona ventralis* propolis collected in Tuyen Quang province [14]. *trans*-Cinnamic acid (**5**) was found in the stingless bee *Trigona sirindhornae* propolis in Thailand [15]. To the best of our knowledge, compounds **1**, **3**, and **4** were first reported from stingless bee propolis. Notably, siamyl benzoate (**1**) has only been found in the plant *Styrax tonkinensis* [8]. In addition, compounds **2**, **3**, **5**, and **6** were also isolated from *S. tonkinensis* and other *Styrax sp.* [16],[17]. A study by Athikomkulchai *et al.* has suggested that the *Styrax* tree was a plant source for propolis of honeybee *Apis melifera* in Thailand [18]. *Styrax tonkinensis* is widely available in Thailand as well as in Vietnam [18],[19]. Based on our chemical results and the wide distribution of *Styrax* trees in Vietnam, *S. tonkinensis* was proposed as one of the botanical sources of *T. collina* propolis.

4. Conclusion

A phytochemical study of *T. collina* stingless bee propolis has resulted in the isolation of six phenolic compounds, including siamyl benzoate (**1**), (+)-pinoresinol (**2**), coniferyl aldehyde (**3**), *p*-coumaric aldehyde (**4**), *trans*-cinnamic acid (**5**), and vanillin (**6**). Compounds **1**, **3**, and **4** were isolated for the first time from stingless bee propolis. Plant *Styrax tonkinensis* was proposed as one of the resin sources for *T. collina* propolis.

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