Vietnam Journal of Science and Technology ***60*** *(2) (2022) 175-182*

doi:10.15625/2525-2518/16060

NEOLIGNANS AND FLAVONOL GLYCOSIDES FROM THE LEAVES OF *VIBURNUM LUTESCENS* BLUME

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Received: 12 May 2021; Accepted for publication: 10 March 2022

Abstract. *Viburnum lutescens* Blume, known as Vot vang nhat in Vietnamese, is a perennial shrub widely distributed South America, China and Southeast Asia. This plant has been used by folk medicine to treat various diseases, such as, cough, diarrhea, rheumatoid arthritis. *V. lutescens* has not been studied for its chemical composition and biological activity. In our study, we used different chromatographic techniques including column chromatography, HPLC, thin layer chromatography, ion exchange chromatography, and size exclusion chromatography to isolate the components. The chemical structures of the isolates were elucidated by using a combination of spectroscopic techniques such as 1D- and 2D-nuclear magnetic resonance spectra, electron spray ionization mass spectroscopy and by comparison with those reported in the literature. As the results, four neolignans including (7*S*,8*S*)-dihydrodehydrodiconiferyl alcohol 9-*O*-*β*-D-glucopyranoside (**1**), (7*S*,8*S*)-dihydrodehydrodiconiferyl alcohol 9′-*O*-*β*-D-glucopyranoside (**2**), (7*S*,8*R*)-5-methoxydihydrodehydrodiconiferyl alcohol 4-*O*-*β*-D-glucopyranoside (**3**) and (7*S*,8*R*)-5-methoxydihydrodehydrodiconiferyl alcohol 9′-*O*-*β*-D-glucopyranoside (**4**), and five flavonol glycosides including kaempferol-3-*O*-*β*-D-glucopyranoside (**5**), kaempferol 3-*O*-rutinoside (**6**), kaempferol 3-*O*-*α*-L-rhamnopyranosyl(1→6)-*β*-D-galactopyranoside (**7**), kaempferol 3-*O*-*β*-D-galactopyranoside (**8**), and quercetin 3-*O*-*β*-D-glucopyranoside (**9**) were isolated from the methanol extract of *Viburnum lutescens* Blume leaves. Interestingly, compounds **1**, **2**, and **4** were reported from *Viburnum* genus for the first time. This study might be a good start to further investigation on the chemical as well as biological activity of this plant.

Keywords: Viburnum lutescens, Caprifoliaceae, neolignan, flavonolglycoside.

Classification numbers: 1.1.1, 1.1.6.

1. INTRODUCTION

The genus *Viburnum* (Caprifoliaceae) consists of over 200 species (8 *Viburnum* species distribute in Viet Nam). *Viburnum* species are used in the folk medicines to treat cough,

diarrhea, rheumatoid arthritis, and tumefaction [1, 2]. Phytochemical investigations of *Viburnum* species indicated the presence of monoterpenoids, sesquiterpenoids, diterpenoids, triterpenoids, iridoids, flavonoids, and lignans, etc. [1]. *V. lutescens* has been used for the treatment of rheumatism, wounds, and obstructive inflammation of the veins [2]. However, chemical constituents and bioactivity of *V. lutescens* have not been studied. We report here the isolation and elucidation of four neolignans and five flavonol glycosides from the leaves of *V. lutescens*.

2. MATERIALS AND METHODS

2.1. Plant materials

The leaves of *Viburnum lutescens* Blume were collected in Vinh Phuc province, Vietnam on May 15th, 2019 and identified by Dr. Nguyen The Cuong, Institute of Ecology and Biological Resources, VAST. A voucher specimen was deposited at University of Science and Technology Hanoi, VAST.

2.2. General experimental procedures

Optical rotations were determined on a Jasco DIP-370 automatic polarimeter. The NMR spectra were recorded using a Bruker DRX 500 spectrometer (^{1}H, 500 MHz; ^{13}C, 125 MHz). ESI-MS spectra were obtained using an Agilent 1100 LC-MSD Trap system. Column chromatography was performed using silica gel 60 (70 - 230 mesh or 230 - 400 mesh, Merck, Germany) or RP-18 resins (30 - 50 μm, Fujisilisa Chemical Ltd., Japan). Pre-coated silica gel 60 F_{254} (0.25 mm, Merck) and RP-18 F_{254}S plates (0.25 mm, Merck) were used for thin-layer chromatography (TLC).

2.3. Extraction and isolation

The dried powder of *V. lutescens* leaves (7.0 kg) was sonicated 3 times with hot MeOH (15 L each) to give a MeOH extract (VLL, 500 g) after removal of solvent under reduced pressure. VLL extract was suspended with water (4.0 L) then partitioned with dichloromethane and ethyl acetate to give dichloromethane (VLL1, 110.0 g), ethyl acetate (VLL2, 3.5 g), and water layer (VLL3). VLL3 was chromatographed on a Diaion HP-20 column eluting with water to remove sugar components, then with increasing concentration of methanol in water (25 % and 100 %) to obtain two fractions, VLL3A and VLL3B, respectively. VLL3B (32.0 g) was separated on a silica gel column eluting with gradient dichloromethane/methanol (1/0 → 0/1, v/v) to give five fractions, VLL3B1 – VLL3B5. VLL3B3 fraction (0.8 g) was chromatographed on an RP-18 column eluting with methanol/water (1/1, v/v) to give four fractions, VLL3B3A – VLL3B3D. Compound **9** (16.1 mg) was obtained from VLL3B3A by a silica gel column eluting with ethyl acetate/methanol/water (8/1/0.1, v/v/v). VLL3B3C fraction was chromatographed on an HPLC system (J'sphere H-80 250 mm length × 20 mm ID column, eluting with 60 % acetonitrile in water, a flow rate of 3 mL/min) to yield compounds **2** (18.7 mg, t_R 50.1 min) and **1** (9.4 mg, t_R 54.3 min). Compound **4** (3.6 mg, t_R 51.5 min**)** was obtained from VLL3B3D fraction on HPLC system (J'sphere H-80 250 mm length × 20 mm ID column) eluting with 20 % ACN in water, a flow rate of 3 mL/min). VLL3B5 fraction was chromatographed on an RP-18 column eluting with methanol/water (1/3, v/v) to give four fractions, VLL3B5A – VLL3B5D. Compound **3** (12.3 mg, t_R 56.1 min) was yielded from VLL3B2B on HPLC system (J'sphere H-80 250 mm length × 20 mm ID column, eluting with 20 % ACN in water, a flow rate of 3 mL/min).

VLL3B5 was chromatographed on an HPLC system (J'sphere H-80 250 mm × 20 mm column, eluting with 18 % ACN in water, a flow rate of 3 mL/min) to yield compounds **7** (16.6 mg, t_R 40.8 min), **6**(33.9 mg, t_R 46.1 min), **8** (3.1 mg, t_R 53.4 min), and **5** (5.2 mg, t_R 60.6 min).

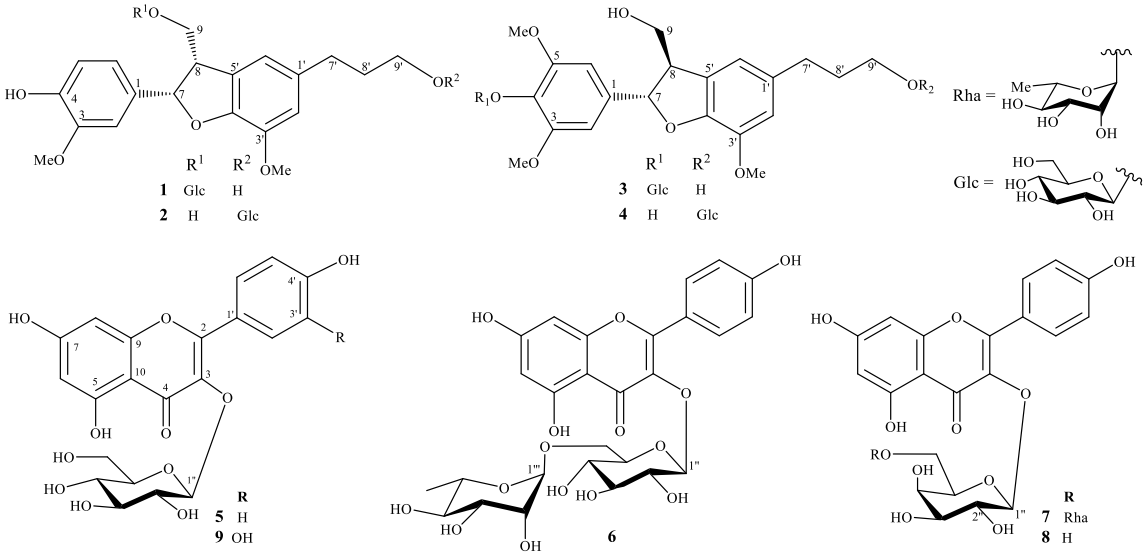

Figure 1. Chemical structures of compounds **1**-**9**.

(7S,8S)-Dihydrodehydrodiconiferyl alcohol 9-O-β-D-glucopyranoside (1)*:* Yellowish powder; ESI-MS: *m/z* 523.49 $[M+H]^+$; $C_{26}H_{34}O_{11}$; $[\alpha]_D^{25}$: -20.7 (*c* = 0.1, MeOH); ^{1}H- and ^{13}C-NMR (CD_3OD): see Table 1.

(7S,8S)-Dihydrodehydrodiconiferyl alcohol 9′-O-β-D-glucopyranoside (2)*:* Yellowishpowder; ESI-MS: *m/z* 523.41 $[M+H]^+$; $C_{26}H_{34}O_{11}$; $[\alpha]_D^{25}$: -18.3 (*c* = 0.1, MeOH);^{1}H-NMR (CD_3OD) δ_H: 6.97 (d, *J* = 2.0 Hz, H-2), 6.78 (d, *J* = 8.0 Hz, H-5), 6.85 (dd, *J* = 2.0, 8.0 Hz, H-6), 5.51 (d, *J* = 6.5 Hz, H-7), 3.49 (m, H-8), 3.78 (dd, *J* = 5.0, 12.0 Hz, H_a-9), 3.86 (dd, *J* = 2.5, 12.0 Hz, H_b-9), 6.77 (s, H-2′), 6.78 (s, H-6′), 2.69 (t, *J* = 7.5 Hz, H-7′), 1.92 (m, H-8′), 3.56 (m, H_a-9′), 3.95 (m, H_a-9′), 4.27 (d, *J* = 7.5 Hz, H-1″), 3.23 (dd, *J* = 7.5, 9.0 Hz, H-2″), 3.39 (t, *J* = 9.0 Hz, H-3″), 3.37 (t, *J* = 9.0 Hz, H-4″), 3.28 (m, H-5″), 3.69 (dd, *J* = 5.5, 11.5 Hz, H_a-6″), 3.88 (dd, *J* = 2.5, 11.5 Hz, H_b-6″), 3.83 (s, 3-OMe), and 3.87 (s, 3′-OMe);^{13}C-NMR (CD_3OD): see Table 1.

(7S,8R)-5-Methoxydihydrodehydrodiconiferylalcohol 4-O-β-D-glucopyranoside (3)*:* Yellowish powder; ESI-MS: *m/z* 553.37 $[M+H]^+$; $C_{27}H_{36}O_{12}$; $[\alpha]_D^{25}$:+26.1 (*c* = 0.1, MeOH); ^{1}H-NMR (CD_3OD) δ_H: 6.65 (s, H-2), 6.65 (s, H-6), 5.47 (d, *J* = 6.0 Hz, H-7), 3.35 (m, H-8), 3.66 (m, H_a-9), 3.78 (m, H_b-9), 6.62 (s, H-2′), 6.59 (s, H-6′), 2.54 (t, *J* = 6.5 Hz, H-7′), 1.73 (m, H-8′), 3.47 (t, *J* = 6.5Hz, H-9′), 4.77 (d, *J* = 7.5Hz, H-1″), 3.39 (dd, *J* = 7.5, 9.0 Hz, H-2″), 3.12 (m, H-3″), 3.30 (t, *J* = 9.0 Hz, H-4″), 3.31 (m , H-5″), 3.57 (m, H_a-6″), 3.70 (m, H_b-6″), 3.78 (s, 3-OMe), 3.78 (s, 5-OMe), and 3.73 (s, 3′-OMe); ^{13}C-NMR (CD_3OD): see Table 1.

(7S,8R)-5-Methoxydihydrodehydrodiconiferyl alcohol 9′-O-β-D-glucopyranoside (4): Yellowish powder; ESI-MS: *m/z* 575.23 $[M+Na]^+$; $C_{27}H_{36}O_{12}$, $[\alpha]_D^{25}$: +21.8 (*c* = 0.1, MeOH)^{1}H-

NMR (CD_3OD) δ_H: 6.97 (d, *J* = 2.0 Hz, H-2), 6.78 (d, *J* = 8.0 Hz, H-5), 6.85 (dd, *J* = 2.0, 8.0 Hz, H-6), 5.51 (d, *J* = 6.5 Hz, H-7), 3.49 (m, H-8), 3.78 (dd, *J* = 5.0, 12.0 Hz, H_a-9), 3.86 (dd, *J* = 2.5, 12.0 Hz, H_b-9), 6.77 (s, H-2′), 6.78 (s, H-6′), 2.69 (t, *J* = 7.5 Hz, H-7′), 1.92 (m, H-8′), 3.56 (m, H_a-9′), 3.95 (m, H_b-9′), 4.27 (d, *J* = 7.5 Hz, H-1″), 3.22 (dd, *J* = 7.5, 9.0 Hz, H-2″), 3.38 (t, *J* = 9.0 Hz, H-3″), 3.37 (t, *J* = 9.0 Hz, H-4″), 3.28 (m, H-5″), 3.69 (dd, *J* = 5.5, 11.5 Hz, H-6″), 3.87 (dd, *J* = 2.5, 11.5 Hz, H-6″), 3.88 (s, 3-OMe), 3.83 (s, 3′-OMe) and 3.88 (s,5-OMe); ^{13}C-NMR (CD_3OD) data: see Table 1.

Kaempferol 3-O-β-D-glucopyranoside (5): Yellow powder; $[\alpha]_D^{25}$: -61.3 (*c* = 0.1, MeOH); ESI-MS *m/z* 448.42 $[M+H]^+$; ^{1}H-and ^{13}C-NMR (CD_3OD) data: see Table 2.

Kaempferol 3-O-rutinoside (6): Yellow powder; ESI-MS: *m/z* 595.19 $[M+H]^+$; $C_{27}H_{30}O_{15}$; $[\alpha]_D^{25}$: -17.8 (*c* = 0.1, MeOH); ^{1}H-NMR (CD_3OD) δ_H: 6.22 (s, H-6), 6.41 (s, H-8), 7.08 (d, *J* = 8.5 Hz, H-2', 6'), 6.91 (d, *J* = 8.5 Hz, H-3', 5'), 5.14 (d, *J* = 7.5 Hz, H-1''), 3.48 (dd, *J* = 7.5, 9.0 Hz, H-2''), 3.45 (t, *J* = 9.0 Hz, H-3''), 3.26 (t, *J* = 9.0 Hz, H-4''), 3.37 (m, H-5''), 3.83 (br d, *J* = 11.5 Hz, H_a-6''), 3.43 (dd, *J* = 4.5, 11.5 Hz, H_b-6''), 4.54 (brs, H-1'''), 3.66 (br d, *J* = 2.5 Hz, H-2'''), 3.49 (dd, *J* = 2.5, 9.0 Hz, H-3'''), 3.27 (t, *J* = 9.0 Hz, H-4'''), 3.43 (m, H-5'''), and 1.14 (d, *J* = 6.0 Hz, H-6'''); ^{13}C-NMR (CD_3OD) data: see Table 2.

Kaempferol 3-O-α-L-rhamnopyranosyl-(1→6)-β-D-galactopyranoside (7): Yellow powder; ESI-MS: *m/z* 615.32 $[M+Na]^+$; $C_{28}H_{32}O_{14}$; $[\alpha]_D^{25}$: -22.7 (*c* = 0.1, MeOH); ^{1}H-NMR (CD_3OD) δ_H: 8.10 (d, *J* = 8.4Hz, H-2′), 6.91 (d, *J* = 8.4 Hz, H-3′), 6.91 (d, *J* = 8.4Hz, H-5′), 8.10 (d, *J* = 8.4 Hz, H-6′), 6.40 (br s, H-6), 6.21 (br s, H-8), 5.03 (d, *J* = 8.0 Hz, H-1″), 4.54 (br s, H-1‴), and 1.16 (d, *J* = 6.4 Hz, H-6‴); ^{13}C-NMR (CD_3OD) data: see Table 2.

Kaempferol 3-O-β-D-galactopyranoside (8): Yellow powder; ESI-MS: *m/z* 471.64 $[M+Na]^+$; $C_{21}H_{20}O_{11}$; $[\alpha]_D^{25}$: +17.9 (*c* = 0.1, MeOH); ^{1}H-NMR (CD_3OD) δ_H: 6.17 (d, *J* = 2.0 Hz, H-6), 6.35 (d, *J* = 2.0 Hz, H-8), 6.90 (d, *J* = 9.0 Hz, H-2′, H-5′), 8.10 (d, *J* = 9.0 Hz, H-3′), 6.90 (d, *J* = 9.0 Hz, H-5′), 8.10 (d, *J* = 9.0 Hz, H-6′), and 5.10 (d, *J* = 8.0 Hz, H-1″); ^{13}C-NMR (CD_3OD) data: see Table 2.

Quercetin 3-O-β-D-glucopyranoside (9): Yellow powder; ESI-MS: *m/z* 465.10 $[M+H]^+$; $C_{21}H_{20}O_{12}$; $[\alpha]_D^{25}$: -66.5 (*c* = 0.1, MeOH); ^{1}H-NMR (CD_3OD) δ_H: 6.22 (d, *J* = 1.5 Hz, H-6), 6.41 (d, *J* = 1.5 Hz, H-8), 7.73 (d, *J* = 1.5 Hz, H-2′), 6.89 (d, *J* = 8.5 Hz, H-5′), 7.63 (dd, *J* = 8.5, 1.5 Hz, H-6′), 5.25 (d, *J* = 7.5 Hz, H-1″), 3.36 (dd, *J* = 9.0, 7.5 Hz, H-2″), 3.47 (t, *J* = 9.0 Hz, H-3″), 3.50 (t, *J* = 9.0 Hz, H-4″), 3.24 (m, H-5″), 3.58 (dd, *J* = 5.0, 12.0 Hz, H_a-6″), and 3.73 (dd, *J* = 2.0, 12.0 Hz, H_b-6″); ^{13}C-NMR (CD_3OD) data: see Table 2.

3. RESULTS AND DISCUSSION

Compound **1** was obtained as a yellowish powder. The ^{1}H-NMR spectrum of **1** showed signals corresponding to an ABX spin system at δ_H 7.01 (1H, d, *J* = 2.0 Hz), 6.88 (1H, dd, *J* = 2.0, 8.0 Hz), 6.79 (1H, d, *J* = 8.0 Hz), two *meta* aromatic protons at δ_H 6.76 (1H, br s) and 6.75 (1H, br s), an anomeric proton at δ_H 4.37 (1H, d, *J* = 8.0 Hz), and two methoxy groups at δ_H 3.83 and 3.88 (each 3H, s). The ^{13}C-NMR and HSQC spectra of **1** showed signals of twenty six carbons including six oxygenated carbons (δ_C 104.6, 78.3, 78.1, 75.2, 71.7, and 62.8) which were assigned to a glucopyranosyl moiety, twelve aromatic carbons at δ_C 149.0, 147.5, 147.5, 145.2, 134.7, 136.9, 129.7, 119.7, 118.2, 116.1, 114.2, and 110.7 assigned to two benzene rings, one methine at δ_C 55.4, one oxymethine at δ_C 88.9, two oxymethylenes at δ_C 72.5 and 62.2, and two methoxycarbons at δ_C 56.8 and 56.4. Analysis of ^{1}H-, ^{13}C-NMR, and HSQC spectra of **1**

indicated the structure of **1** as a dihydrodehydrodiconiferyl alcohol glucoside. Comparing ^{13}C-NMR chemical shifts of two oxygenated methylene groups at C-9 (δ_C 72.5) and C-9′(δ_C 62.2) of **1** with those of (7*S*,8*S)*-dihydrodehydrodiconiferyl alcohol 9-*O*-*β*-D-glucopyranoside [δ_C 72.4 (C-9)/δ_C 62.3 (C-9′)] and (7*S*,8*S)*-dihydrodehydrodiconiferyl alcohol 9′-*O*-*β*-D-glucopyranoside [δ_C 65.1 (C-9)/δ_C 70.0 (C-9′)] confirmed the position of glucopyranosyl unit at C-9 [3]. Consequently, compound **1** was determined as (7*S*,8*S)*-dihydrodehydrodiconiferyl alcohol 9-*O*-*β*-D-glucopyranoside [3], a compound was reported from *Viburnum* genus for the first time.

Table 1. NMR spectral data for compounds **1** – **4** and reference compounds.

C	**1**			**2**		**3**		**4**	
	$\delta_C^{\$,a}$	$\delta_C^{a,b}$	$\delta_H^{a,c}$ (*mult., J* in Hz)	$\delta_C^{\#,a}$	$\delta_C^{a,b}$	$\delta_C^{*,a}$	$\delta_C^{a,b}$	$\delta_C^{@,a}$	$\delta_C^{a,b}$
1	134.8	134.7	-	134.9	134.8	140.3	140.3	134.1	134.1
2	110.9	110.7	7.01 (d, 2.0)	110.7	110.6	104.4	104.4	104.2	104.2
3	149.0	149.0	-	149.1	149.1	154.4	154.4	149.4	149.4
4	147.5	147.5	-	147.5	147.5	135.6	135.6	137.0	136.9
5	116.1	116.1	6.79 (d, 8.0)	116.2	116.2	154.4	154.4	149.4	149.4
6	119.8	119.7	6.88 (dd, 2.0, 8.0)	119.8	119.7	104.4	104.4	104.2	104.2
7	89.0	88.9	5.61 (d, 6.5)	89.0	89.0	88.5	88.5	89.1	89.1
8	55.3	55.4	3.49 (m)	55.4	55.4	55.8	55.8	55.6	55.6
9	72.4	72.5	3.80 (dd, 5.0, 12.0) 4.23 (dd, 2.5, 12.0)	65.1	65.0	65.1	65.1	65.0	65.0
1′	137.0	136.9	-	136.9	136.9	137.2	137.2	136.4	137.0
2′	114.4	114.2	6.75 (s)	114.4	114.3	114.2	114.3	114.3	114.4
3′	145.3	145.2	-	145.2	145.2	145.3	145.3	145.2	145.2
4′	147.5	147.5	-	147.5	147.5	147.4	147.5	147.5	147.5
5′	129.7	129.7	-	130.0	130.0	129.4	129.5	129.8	129.8
6′	118.3	118.2	6.76 (s)	118.1	118.1	117.9	118.0	118.0	118.1
7′	35.8	35.8	1.83 (m)	32.9	32.9	32.9	32.9	32.9	32.9
8′	32.9	32.9	2.64 (t, 7.5)	32.9	32.9	35.8	35.8	32.9	32.9
9′	62.3	62.2	3.58 (t, 6.5)	70.0	70.0	62.2	62.2	69.9	70.0
Glc									
1″	104.6	104.6	4.37 (d, 7.5)	104.5	104.5	105.2	105.2	104.5	104.5
2″	75.2	75.2	3.25 (dd, 7.5, 9.0)	75.2	75.2	75.7	75.7	75.2	75.2
3″	78.3	78.3	3.38 (t, 9.0)	78.2	78.1	78.3	78.3	77.9	78.2
4″	71.7	71.7	3.31 (t, 9.0)	71.8	71.7	71.3	71.3	71.7	71.7
5″	78.0	78.1	3.30 (m)	77.9	77.9	77.8	77.8	77.9	77.9
6″	62.9	62.8	3.68 (dd, 5.5, 11.5) 3.88 (dd, 2.5, 11.5)	62.8	62.8	62.5	62.6	62.8	62.8
3-OMe	56.5	56.4	3.83 (s)	56.4	56.8	57.0	57.0	56.8	56.8
5-OMe						57.0	57.0	56.8	56.8
3′-OMe	56.8	56.8	3.88 (s)			56.9	56.9		

[a] measured in CD_3OD, [b] 500MHz, [c] 125MHz, $\delta_C^{\$}$ of (7S,8S)-dihydrodehydrodiconiferyl alcohol 9-O-β-D-glucopyranoside [3], $\delta_C^{\#}$ of (7S,8S)-dihydrodehydrodiconiferyl alcohol 9′-O-β-D-glucopyranoside [3], δ_C^{*} of (7S,8R)-5-methoxydihydrodehydrodiconiferyl alcohol 4-O-β-D-glucopyranoside [4], $\delta_C^{@}$ of (7S,8R)-5-methoxydihydrodehydrodiconiferyl alcohol 9′-O-β-D-glucopyranoside [5].

Compound **2** was obtained as a yellowish powder. The ^{13}C-NMR data of **2** were found to be very similar to those of **1** except for two oxygenated methylenes [**2**: C-9 (δ_C 65.0) and (C-9′ (δ_C 70.0); **1**: C-9 (δ_C 72.5) and C-9′ (δ_C 62.2)] (Table 1). This suggested the possibility of a

glucopyranosyl location at C-9′. The NMR data of **2** were matched with those of (7*S*,8*S*)-dihydrodehydrodiconiferyl alcohol 9′-*O*-*β*-D-glucopyranoside [3]. Thus, the structure of **2** was identified. This compound was also reported from *Viburnum* genus for the first time.

Table 2. NMR spectral data for compounds **5-9** and reference compounds.

C	5			6		7		8		9	
	δ_C*,[a]	δ_C[a,b]	δ_H[a,c] (*mult., J* in Hz)	δ_C[@,a]	δ_C[a,b]	δ_C[$,a]	δ_C[a,b]	δ_C[&,a]	δ_C[a,b]	δ_C[#,a]	δ_C[a,b]
2	158.5	158.5	-	161.5	161.5	158.7	159.3	158.0	157.3	158.4	159.0
3	135.5	135.5	-	135.5	135.5	135.7	135.7	134.6	134.1	135.6	135.6
4	179.5	179.5	-	179.4	179.4	179.4	179.6	179.8	179.0	179.5	179.5
5	163.1	163.1	-	163.0	163.0	162.9	163.0	162.1	160.2	163.1	163.1
6	99.9	100.0	6.23 (brs)	100.0	100.1	-	100.0	98.8	98.8	99.9	99.9
7	166.0	166.1	-	166.0	166.2	166.0	166.2	165.0	161.0	166.0	166.2
8	94.7	94.8	6.42 (brs)	94.9	95.0	95.2	94.9	93.5	93.5	94.7	94.7
9	159.1	159.1	-	158.6	158.6	159.2	158.5	157.5	155.4	158.5	158.5
10	105.7	105.7	-	105.7	105.6	105.2	105.5	104.6	103.9	105.7	105.7
1′	122.8	122.8	-	122.8	122.8	122.7	122.7	121.5	121.4	123.0	123.1
2′	132.3	132.3	8.07 (d, 8.5)	132.4	132.4	132.4	132.5	131.1	130.9	116.0	116.0
3′	116.1	116.1	6.91 (d, 8.5)	116.1	116.1	116.1	116.1	114.9	114.7	145.9	145.9
4′	161.6	161.6	-	159.4	159.4	161.6	161.6	160.6	160.2	149.8	149.9
5′	116.1	116.1	6.91 (d, 8.5)	116.1	116.1	116.1	116.1	114.9	114.7	117.6	117.6
6′	132.3	132.3	8.07 (d, 8.5)	159.4	132.4	132.4	132.5	131.1	130.9	123.2	123.2
3-*O*-Glc/Gal	**Glc**	**Glc**		**Glc**	**Glc**	**Gal**	**Gal**	**Gal**	**Gal**	**Glc**	**Glc**
1″	104.1	104.2	5.26 (d, 7.0)	104.6	104.6	105.7	105.5	103.7	103.9	104.3	104.4
2″	75.7	75.8	3.46 (dd, 7.0, 9.0)	75.8	75.8	73.0	73.0	71.7	71.7	75.7	75.7
3″	78.1	78.1	3.43 (dd, 9.0, 9.0)	78.2	78.2	75.1	75.0	73.7	73.7	78.1	78.1
4″	71.4	71.4	3.35 (m)	71.4	71.5	70.2	70.1	68.7	68.6	71.2	71.2
5″	78.4	78.4	3.23 (m)	77.3	77.2	75.4	75.3	75.8	75.7	78.4	78.4
6″	62.6	62.7	3.71 (br d, 12.0) 3.55 (dd, 5.0, 12,0)	68.6	68.6	67.4	67.4	60.6	60.6	62.5	62.6
6''-O-Rha											
1‴				102.4	102.4	101.9	101.9				
2‴				72.1	72.1	72.1	72.1				
3‴				72.3	72.3	72.3	72.3				
4‴				73.9	73.9	73.9	73.9				
5‴				69.7	69.7	69.7	69.7				
6‴				17.9	17.9	17.9	18.0				

*[a]measured in CD_3OD, [b]500MHz, [c]125MHz; *δ_C of kaempferol-3-O-β-D-glucopyranoside [6], [@]δ_C of kaempferol 3-O-rutinoside [6], [$]$\delta_C$ of kaempferol 3-O-a-L-rhamnopyranosyl-(1→6)-β-D-galactopyranoside [19], [&]δ_C of kaempferol 3-O-β-D-galactopyranoside [11], [#]δ_C of quercetin 3-O-β-D-glucopyranoside [12].*

Compound **5** was obtained as a yellow amorphous powder. The ^{1}H-NMR spectrum of **5** suggested the presence of a flavonol glycoside with the appearance of two *meta* aromatic protons at δ_H 6.23 (br s) and 6.42 (br s) (ring A) and two *para* aromatic protons at δ_H 6.91 and 8.07 (each 2H, d, J = 8.5 Hz). The ^{13}C-NMR and DEPT spectra of **5** showed signals of 15 carbons of a kaempferol moiety at δ_C 179.5, 166.1, 163.1, 161.6, 159.1, 158.5, 135.5, 132.3×2, 122.8,

116.1×2, 105.7, 100.0, and 94.8. The sugar moiety was determined as glucopyranoside based on the appearance of one anomeric proton at δ_H 5.26 (d, J = 7.0 Hz) along with six carbons at δ_C 104.2, 78.4, 78.1, 75.8, 71.4, and 62.7. In addition, ESI-MS of **5** showed an anion peak at *m/z* 449.42 $[M+H]^+$ corresponding to the molecular formula of $C_{21}H_{20}O_{11}$. Based on above evidence and comparing NMR data of **5** with those reported in the literature (Table 2), the structure of **5** was identified as kaempferol 3-*O*-*β*-D-glucopyranoside [6]. This compound was reported from *Wilkstroemiaindica* [7] and *Clitoriaternatea* [6].

Comparing NMR data of **6** with those of **5** indicated that the structure of **6** was similar to that of **5**, except for the addition of a rhamnopyranosyl unit. This sugar unit was identified based on the chemical shifts at δ_C 102.4 (C-1'''), 72.1 (C-2'''), 72.3 (C-3'''), 73.9 (C-4'''), 69.7 (C-5'''), 17.9 (C-6''') and multiplicity of ananomeric proton [δ_H 4.54 (br s)] and one secondary methyl group [δ_H 1.14 (d, J = 6.0 Hz)] in the ^{1}H-NMR spectrum. Thus, **6** was defined as kaempferol 3-*O*-rutinoside [6]. This compound was reported from *V. plicatum* Thunb. var. *tomemtosum* [8] and found to exhibit antioxidant, antibacterial, and cytotoxic activities [9].

The remaining compounds were elucidated as (7*S*,8*R*)-5-methoxydihydrodehydrodiconiferyl alcohol 4-*O*-*β*-D-glucopyranoside (**3**) [4], (7*S*,8*R*)-5-methoxydihydrodehydrodiconiferyl alcohol 9′-*O*-*β*-D-glucopyranoside (**4**) [5], kaempferol 3-*O*-*α*-L-rhamnopyranosyl-(1→6)-*β*-D-galactopyranoside (**7**) [10], kaempferol-3-*O*-*β*-D-galactopyranoside (**8**) [11], quercetin 3-*O*-*β*-D-glucopyranoside (**9**) [12] by spectroscopic methods and a comparison with those reported in the literature. Among them, **4** was reported from *Viburnum* genus for the first time; **3** was also reported from *V. awabuki* [13] and *V. plicatum* var. *plicatum* f. *plicatum* [14], **7** from *V. punctatum* [15], **8** from *V. tinus* [16], and **9** from *V. dilatatum* [17] and *V. erosum* [18].

Acknowledgment. This research is supported by a grant from Vietnam Academy of Science and Technology under grant code: KHCBHH.02/19-21.

CRediT authorship contribution statement. Thien LD and Dung DT performed the experiments and analysed the data. Minh NP and Dang NH designed the experiments and wrote the article**.**

Declaration of competing interest. All authors declared that they have no conflicts of interest.

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