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Antitumor and Antiinflammatory Constituents from *Celtis sinensis*

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Eight compounds were isolated from the methanolic extract of the twigs of *Celtis sinensis* through repeated silica gel and Sephadex LH-20 column chromatography. Their chemical structures were elucidated as two triterpenoids, germanicol and epifriedelanol, two amide compounds, *trans-N*-caffeoyltyramine and *cis-N*-coumaroyltyramine, two lignan glycoside, pinoresinol glycoside and pinoresinol rutinoside, and two steroids by spectroscopic analysis.

Key words: *Celtis sinensis*, Germanicol, Epifriedelanol, Amide compounds, Pinoresinol glycoside, Pinoresinol rutinoside, Antitumor, Antiinflammatory

INTRODUCTION

Seltis sinensis Persoon (Ulmaceae) is widely distributed in Korea and has been often used as folk medicine in lumbago, gastric disease, abdominal pain and urticaria eczema (But *et al.*, 1997). Previously, we reported the isolation of *trans-N-p*-coumaroyltyramine as an acetylcholinesterase inhibitor (Kim and Lee, 2003). In the course of our continuing work, two triterpenoids, germanicol and epifriedelanol (antitumor principle, Kundu *et al.*, 2000), two amide compounds, *trans-N*-caffeoyltyramine and *cis-N*-coumaroyltyramine, two lignan glycoside, pinoresinol glycoside and pinoresinol rutinoside (anti-inflammatory principle, Cho *et al.*, 2001), and two steroids were isolated. This paper describes the isolation and structural characterization of these compounds.

MATERIALS AND METHODS

General experimental procedures

^{1}H- and ^{13}C-NMR spectra were determined on a JEOL JMN-EX 400 spectrometer in $CDCl_3$ and CD_3OD. IR spectra were obtained on a JASCO FT/IR 410 spectrometer and UV spectra were recorded on Shimadzu UV-1601 UV-Visible spectrophotometer. TLC work was carried out using plates coated with silica gel 60 F_{254} (Merck Co.).

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Silica gel column chromatography was performed on Merck silica gel 60 (230-400 mesh). Sephadex LH-20 was used for the column chromatography (Pharmacia, 25-100 μm). The column used for LPLC was Lobar-A (Merck Lichroprep Si 60, 240-10 mm). All solvents were routinely distilled prior to use. Other chemicals were commercial grade without purification.

Plant materials

The twigs of *C. sinensis* were collected and air-dried in June 2001 at Bongdong, Chonbuk, Korea. A voucher specimen was deposited in the herbarium of the college of pharmacy, Woosuk University (WSU-01-020).

Extraction and isolation

The shade dried plant material (500 g) was extracted (three times with MeOH at room temperature) and filtered. The filtrate was evaporated *in vacuo* to give a dark brownish residue. The resultant methanolic extract (95 g) was followed by successive solvent partitioning to give *n*-hexane (7 g), $CHCl_3$ (15 g), EtOAc (5 g), *n*-BuOH (30 g) and H_2O soluble fractions. *n*-Hexane soluble fraction was chromatographed on silica gel column (*n*-hexane-$CHCl_3$-EtOAc, 12:1:1) to give four fractions (H1-H4). Fraction H3 was was purified by LPLC (Lobar A column, *n*-hexane-$CHCl_3$-EtOAc, 10:1:1) to give **1** (8 mg). Silica gel column chromatography of the $CHCl_3$ soluble fraction with $CHCl_3$-EtOAc-MeOH (10:1:1) gave six fractions (C1-C6). The major fraction C1 was chromatographed on silica gel column using $CHCl_3$-EtOAc (40:1) to yield five fractions

(C11-C15). Fraction C11 was purified by Sephadex LH-20 column with MeOH to give **2** (10 mg). Recrystallization of the fraction C15 with *n*-hexane-acetone (10:1) gave **3** (15 mg). Silica gel column chromatography of the EtOAc soluble fraction with $CHCl_3$-EtOAc-MeOH (10:1:1) gave five fractions (E1-E5). The major fraction E2 was rechromatographed on the Sephadex LH-20 column (MeOH) and purified by Lobar-A column chromatography ($CHCl_3$-MeOH, 7:1) to yield compound **4** (7 mg), and **5** (9 mg). Recrystallization of the fraction E4 with MeOH gave **6** (15 mg). 15 g of *n*-BuOH soluble fraction was chromatographed over Sephadex LH-20 column with MeOH to give four fractions (B1-B4). The major fraction B2 was chromatographed on silica gel column using $CHCl_3$-MeOH-H_2O (50:10:1) to yield four fractions (B21-B24). Fraction B22 was purified by Lobar-A column ($CHCl_3$-MeOH, 5:1) to give **7** (11 mg). Fraction B24 was purified by Lobar-A column (MeOH) to give **8** (11 mg).

Germanicol (1) (Koch *et al.*, 2005)

mp 173-174°C; ^{1}H-NMR (400 MHz, DMSO-d_6) δ: 4.85 (1H, s, H-19), 3.20 (1H, dd, *J*=11.2, 4.8 Hz, H-3α), 1.08, 1.02, 0.97, 1.88, 0.77, 0.73 (each 3H, s, CH_3), 0.94 (6H, s, 2×CH_3). ^{13}C-NMR (100 MHz, DMSO-d_6) δ: 142.7 (C-18), 129.7 (C-19), 79.0 (C-3), 55.5 (C-5), 51.2 (C-9), 43.3 (C-14), 40.7 (C-8), 38.9 (C-4), 38.9 (C-13), 38.4 (C-1), 37.7 (C-16), 37.4 (C-22), 37.2 (C-10), 34.6 (C-7), 34.3 (C-17), 33.3 (C-21), 32.4 (C-20), 31.4 (C-29), 29.2 (C-30), 28.0 (C-23), 27.5 (C-15), 27.4 (C-2), 26.2 (C-12), 25.3 (C-28), 21.1 (C-11), 18.2 (C-6), 16.7 (C-26), 16.1 (C-25), 15.4 (C-24), 14.6 (C-27).

Epifriedelanol (2) (Matsunaga *et al.*, 1993)

mp. 277-278°C; ^{1}H-NMR (400 MHz, $CDCl_3$) δ: 3.74 (1H, m, H-3), 1.17, 1.00, 0.99, 0.98, 0.96, 0.86 (each 3H, s, CH_3), 0.95 (6H, s, 2×CH_3). ^{13}C-NMR (100 MHz, $CDCl_3$) δ: 72.8 (C-3), 61.3 (C-10), 53.2 (C-8), 49.1 (C-4), 42.8 (C-18), 41.7 (C-6), 39.7 (C-13), 39.3 (C-22), 38.4 (C-14), 37.8 (C-9), 37.1 (C-5), 36.1 (C-16), 35.5 (C-11), 35.3 (C-19), 35.2 (C-29), 35.0 (C-2), 32.8 (C-21), 32.0 (C-15), 32.1 (C-28), 31.85 (C-30), 30.6 (C-12), 29.7 (C-17), 28.2 (C-20), 20.1 (C-26), 18.7 (C-27), 18.2 (C-25), 17.5 (C-1), 16.4 (C-24), 15.8 (C-7), 11.6 (C-23).

β-Sitosterol (3)

mp 277-278°C; ^{1}H-NMR (400 MHz, CD_3OD) δ: 3.74 (1H, m, H-3), 1.17, 1.00, 0.99, 0.98, 0.96, 0.86 (each 3H, s, CH_3), 0.95 (6H, s, 2×CH_3). ^{13}C-NMR (100 MHz, CD_3OD) δ: 72.8 (C-3), 61.3 (C-10), 53.2 (C-8), 49.1 (C-4), 42.8 (C-18), 41.7 (C-6), 39.7 (C-13), 39.3 (C-22), 38.4 (C-14), 37.8 (C-9), 37.1 (C-5), 36.1 (C-16), 35.5 (C-11), 35.3 (C-19), 35.2 (C-29), 35.0 (C-2), 32.8 (C-21), 32.0 (C-15), 32.1 (C-28), 31.85 (C-30), 30.6 (C-12), 29.7 (C-17), 28.2 (C-20), 20.1 (C-26), 18.7 (C-27), 18.2 (C-25), 17.5 (C-1), 16.4 (C-24), 15.8 (C-7), 11.6 (C-23).

***trans-N*-Caffeoyltyramine (4)**

mp 216-217°C; ^{1}H-NMR (400 MHz, CD_3OD) δ: 7.33 (1H, d, *J*=15.6 Hz, H-7), 7.02 (1H, d, *J*=1.5 Hz, H-2), 7.00 (2H, d, *J*=8.4 Hz, H-2', 6'), 6.95 (1H, dd, *J*=8.4, 1.5 Hz, H-6), 6.69 (1H, d, *J*=8.4 Hz, H-5), 6.62 (2H, d, *J*=8.4 Hz, H-3', 5'), 6.30 (1H, d, *J*=15.6 Hz, H-8), 3.38 (2H, t, *J*=7.5 Hz, H-8), 2.66 (2H, t, *J*=7.5 Hz, H-7), ^{13}C-NMR (100 MHz, CD_3OD) δ: 167.2 (C-9), 155.6 (C-4'), 147.5 (C-4), 145.5 (C-3), 141.0 (C-7), 131.0 (C-1), 130.7 (C-2', 6'), 127.1 (C-1), 121.0(C-6), 117.3 (C-8), 116.3 (C-5), 116.0 (C-3', 5'), 114.1 (C-2), 42.0 (C-8'), 34.0 (C-7').

***cis-N*-Coumaroyltyramine (5)**

mp 234-235°C; ^{1}H-NMR (400 MHz, CD_3OD) δ: 7.26 (2H, d, *J*=8.8 Hz, H-2', 6'), 6.91 (2H, d, *J*=8.4 Hz, H-2, 6), 6.65 (2H, d, *J*=8.8 Hz, H-3', 5'), 6.62 (2H, d, *J*=8.4 Hz, H-3, 5), 6.51 (1H, d, *J*=12.8 Hz, H-8), 5.69 (1H, d, *J*=12.8 Hz, H-7), 3.29 (2H, t, *J*=7.5 Hz, H-8'), 2.59 (2H, t, *J*=7.5 Hz, H-7'), ^{13}C-NMR (100 MHz, CD_3OD) δ: 170.4 (C-9), 159.4 (C-4), 156.9 (C-4'), 138.1 (C-7), 132.3 (C-2', 6'), 131.2 (C1'), 130.7 (C-2, 6), 127.9 (C-1), 121.4 (C-8), 116.2 (C-3, 5), 116.0 (C-3, 5), 42.3 (C-8'), 35.5 (C-7').

β-Sitosterol-3-*O*-glucoside (6)

^{1}H-NMR (400 MHz, DMSO-d_6) δ: 5.42 (1H, m, H-6), 4.95 (1H, d, *J*=5.0, H-1'), 3.80-3.00 (sugar H), 1.05 (3H, s, 19-CH_3), 1.03-0.85 (12H, 21, 29, 27, 26-CH_3), 0.71 (3H, s, 18-CH_3). ^{13}C-NMR (100 MHz, DMSO-d_6) δ: 140.4 (C-5), 121.3 (C-6), 76.8 (C-3), 56.1 (C-14), 55.3 (C-17), 49.6 (C-9), 45.1 (C-24), 41.8 (C-13), 38.3 (C-12), 36.8 (C-1), 36.2 (C-10), 35.4 (C-20), 33.3 (C-22), 31.4 (C-7), 31.3 (C-8), 29.2 (C-2), 28.7 (C-25), 27.7 (C-16), 25.4 (C-23), 23.7 (C-15), 22.6 (C-28), 20.6 (C-11), 19.7 (C-26), 19.1 (C-19), 18.9 (C-27), 18.6 (C-21), 11.7 (C-29), 11.6 (C-18), 100.7 (C-1'), 76.8 (C-3'), 76.6 (C-5'), 73.4 (C-2'), 70.1 (C-4'), 61.0 (C-6').

Pinoresinol-4-*O*-glucoside (7)

^{1}H-NMR (400 MHz, CD_3OD) δ: 7.13 (1H, d, *J*=8.4 Hz, H-5), 7.01 (1H, d, *J*=1.6 Hz, H-2), 6.92 (1H, d, *J*=1.6 Hz, H-2'), 6.90 (1H, dd, *J*=8.4, 1.6 Hz, H-6), 6.80 (1H, dd, *J*=8.4, 1.6 Hz, H-6'), 6.76 (1H, d, *J*=8.4 Hz, H-5'), 4.87 (1H, d, *J*=7.8 Hz, H-1''), 4.74 (1H, d, *J*=4.0 Hz, H-7), 4.68 (1H, d, *J*=4.4 Hz, H-7'), 4.13 (2H, m, H-9β, 9'β), 3.85 (3H, s, OCH_3), 3.84 (3H, s, OCH_3), 3.82 (2H, m, H-9α, 9'α), 3.67-3.18 (sugar H), 3.11 (2H, m, H-8, 8'). ^{13}C-NMR (100 MHz, CD_3OD) δ: 150.9 (C-4), 149.1 (C-4), 147.5 (C-3'), 147.3 (C-3), 137.4 (C-1), 133.7 (C-1'), 120.0 (C-6'), 119.8 (C-6), 118.0 (C-5), 116.1 (C-5'), 111.6 (C-2), 110.9 (C-2'), 102.8 (C-1''), 87.5 (C-7'), 87.1 (C-7), 78.2 (C-5''), 77.8 (C-3''), 74.9 (C-2''), 72.7 (C-9'), 72.7 (C-9), 71.3 (C-4''), 62.5 (C-6''),

56.7 (C-OCH_3), 56.4 (C-OCH_3), 55.5 (C-8'), 55.3 (C-8).

Pinoresinol-4-*O*-rutinoside (8)

UV λ_{max} (MeOH) nm: 280, 235, IR ν_{max} (KBr) cm^{-1}: 3350, 1630, 1520, FABMS: *m/z* 689.0447 (M+Na), ^{1}H-NMR (400 MHz, CD_3OD) δ: 7.05 (1H, d, *J*=8.4 Hz, H-5), 6.95 (1H, d, *J*=1.6 Hz, H-2), 6.87 (1H, d, *J*=1.6 Hz, H-2'), 6.88 (1H, dd, *J*=8.4, 1.6 Hz, H-6), 6.73 (1H, dd, *J*=8.4, 1.6Hz, H-6'), 6.72 (1H, d, *J*=8.4 Hz, H-5'), 4.80 (1H, d, *J*=7.8Hz, H-1"), 4.68 (1H, d, *J*=4.0 Hz, H-7), 4.66 (1H, s, H-1'''), 4.62 (1H, d, *J*=4.4 Hz, H-7'), 4.14 (3H, m, H-9β, 9'β), 3.78 (3H, s, OCH_3), 3.76 (3H, s, OCH_3), 3.95-3.02 (sugar H), 3.02 (2H, m, H-8, 8'), 1.15 (3H, d, *J*=6.4 Hz, H-6'''). ^{13}C-NMR (100 MHz, CD_3OD) δ: 150.7 (C-4), 148.9 (C-4), 147.0 (C-3'), 147.0 (C-3), 137.2 (C-1), 133.6 (C-1'), 119.9 (C-6'), 119.7 (C-6), 117.9 (C-5), 116.0 (C-5'), 111.5 (C-2), 110.9 (C-2'), 102.4 (C-1"), 101.9 (C-1'''), 87.2 (C-7'), 86.8 (C-7), 77.6 (C-5"), 76.7 (C-3"), 74.7 (C-2"), 73.9 (C-4'''), 72.6 (C-9'), 72.5 (C-9), 72.2 (C-3'''), 72.0 (C-2'''), 71.3 (C-4"), 69.7 (C-5'''), 67.7 (C-6"), 56.7 (C-OCH_3), 56.6 (C-OCH_3), 55.2 (C-8'), 55.1 (C-8), 17.9 (C-6''').

RESULTS AND DISCUSSION

In the course of phytochemical study of the MeOH extract from the twigs of *C. sinensis*, eight compounds were isolated by the chromatographic separation of each fraction. They were identified as two triterpenoids, two amide compounds, two lignan glycoside, and two steroids by spectroscopic analysis.

Compound **1** was obtained as an amorphous powder from MeOH. ^{1}H-NMR spectrum of **1** showed eight methyl singlet signals. The singlet peak at δ4.85 (1H, s) was identified as H-19 proton signal attached on an olefinic double bond. In the ^{13}C-NMR spectrum, 30 carbon signals were observed, which included two olefinic carbons at δ142.7 and 129.7, and an oxygenated carbon at δ79.0. As the result, compound **1** was identified as germanicol, which was reported from *Lactuca indica* and *Schaefferia cuneifolia* (Park *et al.*, 1995; Gonzalez *et al.*, 1989). The NMR spectra of **2** was similar to that of **1** but it did not show the olefinic double bond. The ^{1}H-NMR spectrum of **2** revealed the presence of a hydroxyl group at C-3 with δ3.74 (1H, m). In the ^{13}C-NMR spectrum, 30 carbon signals were observed, which included an oxygenated carbon at δ72.8. From the comparison of all spectral data of compound **2**, they were in good agreement with epifriedelanol, which was reported in the literatures (Hong *et al.*, 1990; Ahmad *et al.*, 1994). Compounds **3** and **6** were identified as β-sitosterol (**3**) and β-sitosterol-3-*O*-glucoside (**6**) by

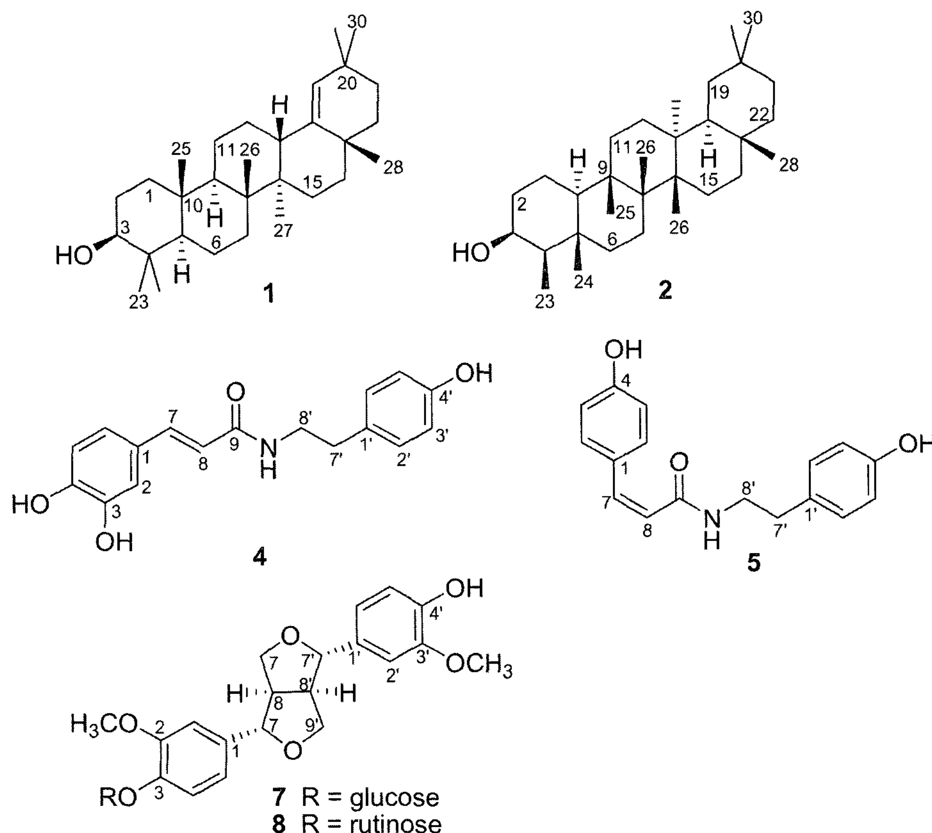

Fig. 1. Isolated compounds from *Celtis sinensis*

comparing physicochemical and spectral data with published literatures (Do *et al.*, 1988; Park *et al.*, 1994).

Compounds **4** and **5** have similar patterns in their NMR spectra. The ^{1}H-NMR spectrum of **4** showed two ortho-coupled doublets each of two protons with a *J* value of 8.4 Hz at β7.00 (2H, d, H-2', 6') and 6.62 (2H, d, H-3', 5'), indicating the presence of a 1,4-disubstituted aromatic ring. In addition, two coupled triplets corresponding to the methylene protons at δ3.38 (2H, t, *J*=7.5 Hz, H-8) and 2.66 (2H, t, *J*=7.5 Hz, H-7) revealed the presence of tyramine moiety in **4**. The signals at δ7.02 (1H, d, *J*=1.5 Hz, H-2), 6.95 (1H, dd, *J*=8.4, 1.5 Hz, H-6) and 6.69 (1H, d, *J*=8.4 Hz, H-5), and two *trans*-coupled vinylic protons at δ7.33 (1H, d, *J*=15.6 Hz, H-7) and 6.30 (1H, d, *J*=15.6 Hz, H-8) revealed the presence of caffeoyl moiety in **4**. The ^{13}C-NMR spectrum of **4** showed a carbonyl carbon, 10 aromatic carbons, two methylene carbons and two vinylic carbons. The structure of **4** was determined to be *trans-N*-caffeoyltyramine on the basis of the above evidences, together with a comparison of the above data with those published in the literature (Lajide *et al.*, 1996; Han *et al.*, 2002).

Compound **5** was obtained as an amorphous powder from MeOH. In the ^{13}C-NMR spectrum of **5**, 13 carbon signals were observed, which included a carbonyl carbon, 8 aromatic carbons, two methylene carbons and two vinylic carbons. The ^{1}H-NMR spectrum of **5** showed two groups of ortho-coupled doublets each of two protons with a *J* value of 8.8 Hz at δ7.26 (2H, d, H-2', 6') and 6.65 (2H, d, H-3', 5'), and a *J* value of 8.4 Hz at δ6.91 (2H, d, H-2, 6) and 6.62 (2H, d, H-3, 5). In addition, two coupled triplets corresponding to the methylene protons at δ3.29 (2H, t, *J*=7.5 Hz, H-8') and 2.59 (2H, t, *J*=7.5 Hz, H-7') and two *cis*-coupled vinylic protons at δ6.51 (1H, d, *J*=12.8 Hz, H-8) and 5.69 (1H, d, *J*=12.8 Hz, H-7) were observed. On the basis of the above evidences, together with a comparison of the above data with those published in the literature, the structure of **5** was determined to be *cis-N*-coumaroyltyramine (Wu *et al.*, 1994).

The ^{1}H-NMR spectrum of **7** showed the typical pattern of two coupling groups of 1,3,4-trisubstituted benzene rings at δ7.13 (1H, d, *J*=8.4 Hz, H-5), 7.01 (1H, d, *J*=1.6 Hz, H-2), 6.92 (1H, d, *J*=1.6 Hz, H-2'), 6.90 (1H, dd, *J*=8.4, 1.6 Hz, H-6), 6.80 (1H, dd, *J*=8.4, 1.6 Hz, H-6') and 6.76 (1H, d, *J*=8.4 Hz, H-5'), two methoxy groups at δ3.85 and 3.84, and an anomeric proton at δ4.87 (1H, d, *J*=7.8 Hz, H-1"). Eight proton signals at δ4.74 (1H, d, *J*=4.0 Hz, H-7), 4.68 (1H, d, *J*=4.4 Hz, H-7'), 4.13 (2H, m, H-9β, 9'β), 3.82 (2H, m, H-9α, 9'α), and 3.11 (2H, m, H-8, 8') were assigned to a *cis*-diequatorial substituted 7,7-diaryldioxabicyclo[3,3,0]octane (Pérez *et al.*, 1995). On the basis of these data, the structure of **7** was elucidated as pinoresinol-4-*O*-glucopyranoside (Tsukamoto *et al.*, 1984; Chiba *et al.*, 1979).

Compound **8** showed a molecular ion peak at *m/z* 666 in the mass spectrum. The NMR spectra of **8** were similar to those of **7**, suggesting it has similar skeleton. The main difference was sugar moiety of aliphatic region. Another anomeric peak at δ_H4.66 (1H, s, H-1'''), δ_C101.9 (C-1'''), and five rhamnose sugar signal were observed in the ^{13}C-NMR spectrum. In the HMBC spectrum, it was evident that the two methoxy groups are placed at each 3 and 3 position of the two aromatic rings. A correlation peak of the H-1 signal of glucose moiety at δ4.80 (1H, d, *J*=7.8 Hz, H-1") and the C-4 signal of the phenyl group at δ_C150.7 suggested that the glucose moiety is connected to a 4-hydroxy group at the phenyl group. And a correlation peak in the HMBC spectrum of the H-1 signal for rhamnose at δ4.66 (1H, s, H-1'''), the C-6 signal for glucose at δ_C67.7 suggested that these two sugar moieties were connected by a 1→6 linkage between β-glucose and α-rhamnose, consistent with a rutinoside structure (Agrawal, 1989). In conclusion, the structure of **8** was determined to be pinoresinol-4-*O*-[α-rhamnopyranosyl (1→6)-*O*-β-glucopyranoside].

Previously, epipinoresinol-4-*O*-[α-rhamnopyranosyl (1→6)-*O*-β-glucopyranoside] and pinoresinol-4-*O*-[α-rhamnopyranosyl(1→2)-*O*-β-glucopyranoside] reported in the literature (Lee *et al.*, 1999; Batirov *et al.*, 1985), but compound **8** has not been reported in any other plant.

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