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Stilbenes from the roots of *Pleuropterus ciliinervis* and their antioxidant activities

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Abstract

Two stilbene glycosides, pieceid-2″-*O*-gallate and pieceid-2″-*O*-coumarate, were isolated from the MeOH extract of the roots of *Pleuropterus ciliinervis* Nakai (Polygonaceae), together with two known compounds, resveratrol and pieceid. Their structures were determined spectroscopically, particularly by 2D NMR spectroscopic analysis. The antioxidant activities of stilbenes isolated were determined in vitro against 1,1-diphenyl-2-picrylhydrazyl (DPPH) radicals, superoxide radicals and by determining their lipid peroxidation inhibitory activities. Among the compounds isolated, pieceid-2″-*O*-gallate had the most potent inhibitory scavenging effect on DPPH, superoxide radicals and upon lipid peroxidation inhibition with IC_{50} values of 16.5, 23.9 and 5.1 μM, respectively.

Keywords: *Pleuropterus ciliinervis*; Polygonaceae; Pieceid-2″-*O*-gallate; Pieceid-2″-*O*-coumarate; DPPH radical scavenging activity; Superoxide radical scavenging activity; Lipid peroxidation activity

1. Introduction

Stilbenes have been found in many families of higher plants, such as Vitaceae, Gnetaceae, Polygonaceae, Liliaceae, Moraceae and Cyperaceae (Teguo et al., 1998; Li et al., 2001; Meng et al., 2001; Iliya et al., 2002; Kanchanapoom et al., 2002; Su et al., 2002; Xiao et al., 2002). These compounds have attracted much attention for their biological effects, which include antioxidant (Fauconneau et al., 1997; Teguo et al., 1998; Ryu et al., 2002), cyclooxygenase-I and-II-inhibitory (Cichewicz et al., 2000), antiplatelet-aggregation (Orsini et al., 1997), antifungal (Pacher et al., 2002), tyrosinase-inhibitory (Likhitwitayawuid and Sritularak, 2001) and anti-HIV-1 and cytotoxic effects (Dai et al., 1998).

As a part of our continuing studies to identify novel antioxidant agents from natural sources, the MeOH extracts from approximately 200 Korean plants were screened for 1,1-diphenyl-2-picrylhydrazyl (DPPH) radical-scavenging activity (Na et al., 2002) and the root of *Pleuropterus ciliinervis* Nakai (Polygonaceae) was found to have the greatest activity. This plant is used in a traditional China medicine ''Hasuo'', which is used to treat inflammation and bacterial infections (Namba, 1993). Stilbenes, anthraquinones and flavonoids have been isolated from the genus *Pleuropterus* (Han and Cho, 1981; Yoshizaki et al., 1987; Tang and Eisenbrand, 1992). In this paper, we report the isolation, structure elucidation and antioxidant activities of four stilbenes, which include two new stilbene natural products (**1** and **2**).

2. Results and discussion

Silica gel and Sephadex LH-20 column chromatography and preparative HPLC of the EtOAc-soluble fraction of the MeOH extract of *P. ciliivervis* (root) led

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to the isolation of four stilbenes (**1**–**4**). Two were known and were identified as resveratrol (**3**, 3,4′,5-trihydroxystilbene) and pieceid (**4**), (resveratrol-3-*O*-*β*-D-glucopyranoside) (Teguo et al., 1996) by comparing their spectral data with those previously reported (Fig. 1).

Compound **1** was obtained as a white amorphous powder. FABMS showed a quasi-molecular ion peak at *m*/*z* 543 $[M+H]^+$, with a molecular formula $C_{27}H_{26}O_{12}$, which was supported by the ^{13}C NMR spectrum. Its UV spectrum exhibited absorption maxima at 216 and 300 nm, suggesting the presence of a stilbene group (Ioset et al., 2001). The ^1H NMR spectrum of **1** showed a combined total of 11 aromatic and olefinic protons, which produced A_2X, A_2B_2 and AB spin patterns, as follows: an A_2X spin system of a 3,5-dihydroxphenyl group at *δ* 6.70 (1H, *brs*, H-2), 6.36 (1H, *brs*, H-4), and 6.63 (1H, *brs*, H-6); an A_2B_2 spin system assigned to a 4-hydroxyphenyl group at *δ* 7.35 (2H, *d*, *J* = 8.6 Hz, H-2′, 6′) and 6.79 (2H, *d*, *J* = 8.6 Hz, H-3′, 5′); AB spin system protons on a 1,3,4,5-tetrasubstituted benzene ring at *δ* 7.18 (2H, *s*, H-2‴, 6‴); two olefinic protons at *δ* 6.81 (1H, *d*, *J* = 16.3 Hz, H-7) and 6.94 (1H, *d*, *J* = 16.3 Hz, H-8) (Table 1). Furthermore, the ^1H NMR spectrum showed signals for a sugar at *δ* 5.14 (1H, *m*, H-2″), 5.12 (1H, *d*, *J* = 7.9 Hz, H-1″), 4.00 (1H, *d*, *J* = 11.4 Hz, H-6″), 3.66 (2H, *m*, H-3″, 6″), 3.45 (2H, *m*, Glu H-4″, 5″), indicating the presence of a glucose moiety. The ^{13}C NMR spectrum of **1**, in combination with DEPT and HMQC experiments, showed signals for six sugar carbons including an anomeric carbon at *δ* 101.5, a carbonyl carbon at *δ* 167.8 and 20 aromatic and olefinic carbons at *δ* 104.4–160.4 (Table 1). Comparing these with the ^1H and ^{13}C NMR spectra of resveratrol (**3**) and pieceid (**4**), it appeared that **1** was composed of a resveratrol unit, gallic acid and glucose groups. This finding was further supported by the presence of prominent fragment ion peaks at *m*/*z* 315 $[M-\text{resveratrol}+H]^+$, 228 $[\text{resveratrol}]^+$ and 154 $[\text{gallic acid}+H]^+$.

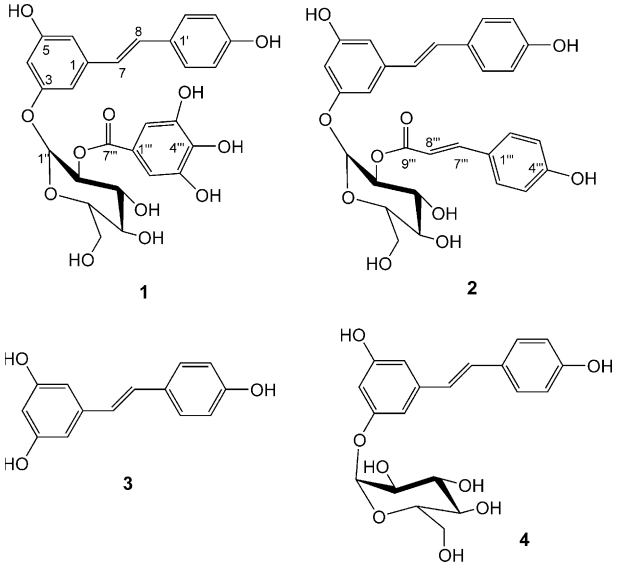

Fig. 1. Structures of compounds **1**–**4** isolated from *P. ciliinervis*.

The connectivites of resveratrol, sugar and gallic acid units in **1** were confirmed by HMBC, which showed correlations between signals at δ_H 5.12 (Glc H-1″) and δ_C 160.4 (resveratrol C-3), and δ_H 5.14 (Glc H-2) and δ_C 167.8 (gallic acid C-7‴), indicating glycosylation at C-3 of resveratrol with a gallic acid (7‴→2″) glucose moiety (Min et al., 2000) (Fig. 2). Acid hydrolysis of **1** afforded the monosaccharide glucose, which was identified on TLC by reference to an authentic sample. Furthermore, this sugar was determined to be D-glucose by the GLC

Table 1
^1H and ^{13}C NMR (400 MHz, CD_3OD) spectroscopic data for **1** and **2** (*δ* values, *J* in Hz)

	δH		δC	
	1	**2**	**1**	**2**
1			139.9	140.1
2	6.70 *brs*	6.59 *brs*	109.0	105.8
3			160.4	158.1
4	6.36 *brs*	6.28 *t* (2.2)	104.4	103.0
5			159.5	158.9
6	6.63 *brs*	6.51 *brs*	107.2	105.8
7	6.81 *d* (16.3)	6.66 *d* (16.3)	126.4	125.0
8	6.94 *d* (16.3)	6.82 *d* (16.3)	130.1	128.7
1′			130.2	128.8
2′	7.35 *d* (8.6)	7.14 *d* (8.6)	129.0	127.6
3′	6.79 *d* (8.6)	6.63 *d* (8.6)	116.5	115.1
4′			158.4	156.9
5′	6.79 *d* (8.6)	6.63 *d* (8.6)	116.5	115.1
6′	7.35 *d* (8.6)	7.14 *d* (8.6)	129.0	127.6
1″	5.12 *d* (7.9)	4.99 *d* (8.0)	101.5	99.8
2″	5.14 *m*	5.00 *m*	78.4	77.0
3″	3.66 *m*	3.71 *m*	76.2	74.7
4″	3.45 *m*	3.45 *m*	71.6	70.1
5″	3.45 *m*	3.45 *m*	75.4	73.8
6″	4.00 *d* (11.4), 3.66 *m*	3.84 *m*, 3.71 *m*	62.5	61.1
1‴			121.5	159.8
2‴	7.18 *brs*	6.68 *d* (8.6)	110.4	115.5
3‴		7.33 *d* (8.6)	146.5	129.9
4‴			141.5	125.8
5‴		7.33 *d* (8.6)	146.5	129.9
6‴	7.18 *brs*	6.68 *d* (8.6)	110.4	115.5
7‴		6.28 *d* (15.9)	167.8	113.6
8‴		7.59 *d* (15.9)		145.8
9‴				167.2

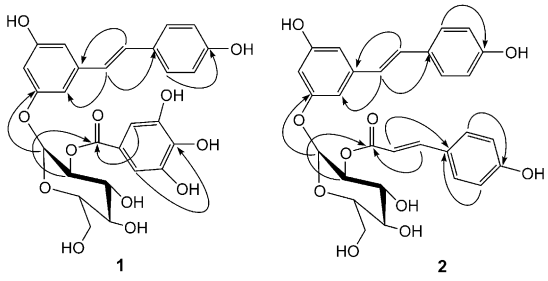

Fig. 2. Important ^1H-^{13}C long-range correlations for compounds **1** and **2**.

of its pertrimethylsilylated L-cysteine methyl ester derivative (Min et al., 2001). The configuration of the glycosidic linkage of the glucopyranoside moiety in **1** was determined to be β on the basis of the $J_{1''', 2'''}$ value (7.9 Hz) of the anomeric proton. On the other hand, the two olefinic protons at δ 6.81 and 6.94 (*d*, $J = 16.3$ Hz) indicated a *trans* geometry owing to the large coupling constant. Thus, the structure of compound **1** was established as pieceid-2″-*O*-gallate.

Compound **2** was obtained as a white amorphous powder and exhibited absorption maxima at 215 and 305 nm in the UV spectrum. Its positive-ion-mode FABMS spectrum produced a quasi-molecular ion peak at m/z 537 $[M+H]^+$ compatible with a molecular formula $C_{29}H_{28}O_{10}$, which was supported by the ^{13}C NMR spectra. The ^1H NMR spectrum of **2** showed similar patterns to that of **1**. However, the ^1H NMR spectrum of **2** exhibited a combination of 15 aromatic and olefinic protons, resembling that of a 3,5-dihydroxyphenyl group in an A_2X spin system at δ 6.59 (1H, *brs*, H-2), 6.51 (1H, *brs*, H-6), and 6.28 (1H, *t*, $J = 2.2$ Hz, H-4); two sets of A_2B_2 spin systems assigned to *ortho*-coupled aromatic protons at δ 7.14 (2H, *d*, $J = 8.6$ Hz, H-2′, 6′) and 6.63 (2H, *d*, $J = 8.6$ Hz, H-3′, 5′), and 6.68 (2H, *d*, $J = 8.6$ Hz, H-2‴, 6‴) and 7.33 (2H, *d*, $J = 8.6$ Hz, H-3‴, 5‴); and four olefinic protons were observed at δ 7.59 (1H, *d*, $J = 15.9$ Hz, H-8‴), 6.82 (1H, *d*, $J = 16.3$ Hz, H-8), 6.66 (1H, *d*, $J = 16.3$ Hz, H-7), and 6.28 (1H, *d*, $J = 15.9$ Hz, H-7‴). The spectrum also exhibited a set of sugar protons at δ 5.00 (1H, *m*, H-2″), 4.99 (1H, *d*, $J = 8.0$ Hz, H-1″), 3.84 (1H, *m*, H-6″), 3.71 (2H, *m*, H-3″, 6″), and 3.45 (2H, *m*, H-4″, 5″). The ^{13}C NMR spectrum of **2**, in combination with DEPT and HMQC, revealed the presence of a carbonyl carbon at δ 167.2 and of an anomeric carbon at δ 99.8, and included five sugar carbons at δ 77.0–61.1, in addition to 22 aromatic and olefinic carbons at δ 103.0–159.8. Therefore, **2** was similar to **1**, the structure was composed of resveratrol, glucose, and coumaric acid units.

The connectivities of the three units were established by interpreting the significant HMBC signals, which exhibited long-range signals at δ_H 4.99 (Glc H-1″) and δ_C 158.1 (resveratrol C-3), and between δ_H 5.00 (Glc H-2″) and δ_C 167.2 (coumaric acid C-9‴), indicating glycosylation at C-3 of resveratrol by a coumaric acid (9‴→2″) glucose moiety (Fig. 2). Glucose was identified by GLC of its pertrimethylsilylated L-cysteine methyl ester derivative after acid hydrolysis. The configuration of the glucosidic linkage was assigned as β based on its coupling constant, which was similar to that in **1**. The structure of **2** was thus determined to be (*E*)-pieceid-2″-*O*-coumarate.

The compounds (**1**–**4**), which are all above 95% grade of purity, were tested for their antioxidant scavenging effects on DPPH and superoxide radicals and in terms of their abilities to inhibit lipid peroxidation. All compounds tested showed antioxidant activity and the results (IC_{50} values) are summarized in Table 2. Of these compounds, **1** exhibited potent antioxidant scavenging activity against DPPH and superoxide radicals, and inhibited lipid peroxidation with IC_{50} values of 16.5, 23.9 and 4.3 μM; α-tocopherol and BHT were used as positive controls. This result shows that the galloyl group in stilbene might play an essential role in antioxidant activity. On comparing resveratrol (**3**) with pieceid (**4**), it appears that the glycosylation of stilbenes reduces their antioxidant activity (Ryu et al., 2002).

3. Experimental

3.1. General

UV: UV-2200 UV–VIS recording spectrophotometer (Shimadzu, Japan); IR: Jasco Report-100 spectrophotometer; NMR: Bruker AMX 400 spectrometer (Bruker, USA), the chemical shifts being represented as ppm with tetramethylsilane as a internal standard; positive-ion FABMS: JMS-AX 110/110A (Jeol, Japan); preparative HPLC was carried out on a Shimadzu system: LC10AD pump, SPD-10AV UV detector; GC-MASS (HP 5890 series II plus GC, HP 5972 series mass selective Detector, Column: HP-1MS); cc: silica gel 60 (70∼230 and 230∼400 mesh, Merck), Sephadex LH-20 (Pharmacia, Sweden), Amberlite MB-3 (Rohm and Haas Co., USA), and YMC-GEL ODS-A (12 nm, S-75 μm, YMC); TLC and preparative TLC: pre-coated silica gel 60 F_{254} and RP-18 F_{254s} (Merck).

3.2. Plant material

P. ciliinervis Nakai was collected at Whachen, Korea, in August 1998. A voucher specimen (No. CNU477) is deposited at the Laboratory of Pharmacognosy, College of Pharmacy, Chungnam National University, Korea.

Table 2
Antioxidant activities of compounds (**1**–**4**) from *P. ciliinervis*

	DPPH radical Scavenging activity	Superoxide radical Scavenging activity	Inhibitory activity of lipid peroxidation
1	16.5	23.9	4.3
2	84.3	74.6	5.1
3	38.9[c]	51.1	3.3
4	82.4	N.T.[d]	67.2
α-tocopherol[a]	20.7	N.T.	5.3
BHT[b]	12.6	24.6	1.0

[a] Positive control.
[b] Butylated hydroxytoluene (positive control).
[c] IC_{50} (μM) values were calculated from regression lines using six different concentrations in triplicate.
[d] N.T.: 50% inhibition was not reached at concentrations of 100 μM.

3.3. Extraction and isolation

The air-dried roots of *P. ciliinervis* Nakai (3.0 kg) were extracted with MeOH, and the MeOH solution was then evaporated to dryness. The residue (315.6 g) was diluted with H_2O and partitioned against hexane, EtOAc, and *n*-BuOH. The EtOAc-soluble fraction (120.0 g) was subsequently fractionated on silica gel (60–230 mesh; 1.5 kg) cc and eluted using a gradient of 2–50% EtOAc/hexane (2, 10, 15, 20, 30, 40, 50%, each 1000 ml) and EtOAc–MeOH (5:1, 1000 ml), to afford six fractions (Fr. 1–6; 5.9, 36.4, 27.2, 19.0, 15.3 and 15.4 g, respectively). Cc on silica gel (230–400 mesh, 400 g; hexane–EtOAc, 6:1, 3:1, 2:1, 1:1, 1:2, each 200 ml) of fr. 4 (19.0 g) furnished the stilbene, resveratrol (**3**, 1238.0 mg). Fraction 5 (15.3 g) was further subjected for Sephadex LH-20 cc and eluted with H_2O–MeOH (1:0→0:1) and then preparative HPLC (C_{18}-column; MeOH–H_2O, 4:6) to yield the stilbenes, pieceid-2″-*O*-gallate (**1**, 538.6 mg, R_t 46.8 min) and pieceid-2″-*O*-coumarate (**2**, 650.8 mg, R_t 58.5 min). On the other hand, cc of Fr. 6 (15.4 g) on silica gel (230∼400 mesh, 400 g; EtOAc–MeOH, 6:1) yielded **4** (1029 mg).

3.4. Pieceid-2″-O-gallate (1)

White amorphous powder (MeOH–H_2O). $FeCl_3$ reaction: positive (green); $[\alpha]_D$−37.0° (MeOH, *c* 0.22); UV λ_{max} nm: 216, 300; IR υ_{max} cm^{-1}: 3310(OH), 1588, 1513 (aromatic ring), 1443, 1250, 1150 (CO), 965 (*trans* C=C), 831 (trisubstituted aromatic ring); for ^1H and ^{13}C NMR, see Table 1; Positive-ion FABMS *m/z*: 543 $[M+H]^+$, 315 $[M-\text{resveratrol}+H]^+$, 228 $[\text{resveratrol}]^+$; HR-FABMS *m/z*: 565.1322 ($[M+Na]^+$, calc. for $C_{27}H_{26}O_{12}Na$ 565.1322).

3.5. Pieceid-2″-O-coumarate (2)

White amorphous powder (MeOH). $FeCl_3$ reaction: positive (green); $[\alpha]_D$ 2.5° (MeOH, *c* 0.22); UV λ_{max} nm:215, 305; IR υ_{max} cm^{-1}: 3310 (OH), 1588, 1513 (aromatic ring), 1443, 1250, 1150 (CO), 965, 831 (trisubstituted aromatic ring); for ^1H and ^{13}C NMR, see Table 1; Positive-ion FABMS *m/z*: 537 $[M+H]^+$, 309.0 $[M-\text{resveratrol}+H]^+$, 228 $[\text{resveratrol}]^+$; HR-FABMS *m/z*: 537.1763 ($[M+H]^+$, calc. for $C_{29}H_{29}O_{10}$ 537.1761).

3.6. Determination of sugars in 1 and 2

Each sample (2 mg) was refluxed with 4-N HCl-dioxane (1:1, 2 ml) for 2 h. The mixture was then extracted with EtOAc (5 ml-×-3). The residual water layer was neutralized with Amberlite MB-3 and dried to give a residue, which was dissolved in pyridine (1 ml), to which 0.1 M L-cysteine methyl ester hydrochloride in pyridine (2 ml) was added. The mixture was kept at 60°C for 1.5 h, dried in vacuo, and the residue trimethylsilylated with hexamethyldisilazane-trimethylchlorosilane (0.1 ml) at 60 °C for 1 h. The mixture obtained was partitioned between hexane and H_2O (0.3 ml each) and the hexane extract was analyzed by GC-MS. In the acid hydrolysate of **1** and **2**, D-glucose was confirmed by comparing the retention times of the derivatives with those of D-glucose and L-glucose, and derivatives prepared in a similar way, which showed retention times of 21.30 and 22.00 min, respectively (Park et al., 2002).

The sugars obtained by the acid hydrolysis of **1** and **2** were identified by TLC on silica gel using EtOAc–MeOH–H_2O–AcOH (65:20:15:15) as a solvent system. The spots on the plate were visualized by spraying with an anisaldehyde–H_2SO_4 solution.

3.7. DPPH radical scavenging activity

DPPH radical scavenging activity was measured according to the procedure of Takao et al. (1994). The purities of compounds used for the assay were above 95% checked by HPLC.

3.8. Superoxide (O_2^-) radical scavenging activity

Superoxide was generated by xanthine/xanthine oxidase and measured by the nitroblue tetrazolium (NBT) reduction method (Nishikimi et al., 1972; Cheng et al., 1998).

3.9. Preparation of rat brain homogenate

The rat brain homogenate was prepared as previously described, with some modification (Huong et al., 1998). Rat brains were removed, washed with ice-cold saline, homogenized in 9 volumes of ice-cold 5 mM phosphate buffer (pH 7.4) using a glass homogenizer and then centrifuged at 1000 rpm for 10 min. The supernatant was stored at −70 °C until required for the lipid peroxidation determination.

3.10. Lipid peroxidation inhibitory activity

Lipid peroxidation inhibitory activity in rat brain homogenate was evaluated by the thiobarbituric acid (TBA) method with some modification (Huong et al., 1998).

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