Jasmultiside, a New Secoiridoid Glucoside from Jasminum multiflorum

Han-Ying Chen, Ya-Ching Shen, and Chung-Hsiung Chen

J. Nat. Prod., **1991**, 54 (4), 1087-1091• DOI:
10.1021/np50076a026 • Publication Date (Web): 01 July 2004

Journal of Natural Products
Vol. 54, No. 4, pp. 1087-1091, Jul-Aug 1991

JASMULTISIDE, A NEW SECOIRIDOID GLUCOSIDE FROM *JASMINUM MULTIFLORUM*

HAN-YING CHEN,

National Taipei College of Nursing, 365 Ming Der Road, Pei-tou 11211, Taipei, Taiwan, Republic of China

YA-CHING SHEN, and CHUNG-HSIUNG CHEN*

School of Pharmacy, National Taiwan University, 1 Jen-Ai Road, Sec. 1, Taipei, Taiwan, Republic of China

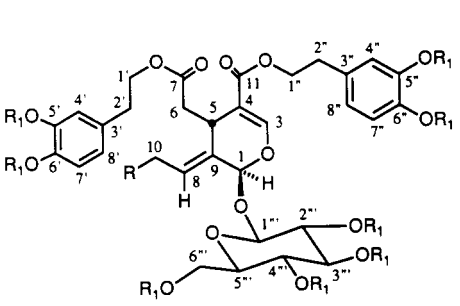
ABSTRACT.—A new secoiridoid glucoside, jasmultiside [**1**], was isolated from the aerial parts of *Jasminum multiflorum*. The structure of the new compound was established on the basis of spectral analysis and chemical correlations.

Jasminum multiflorum (Burm. f.) Andr. (Oleaceae) is an evergreen shrub that is cultivated as an ornamental plant in Taiwan. Previously, in connection with our search of bioactive iridoids, we reported the isolation of novel secoiridoid lactones, jasmolactones A, B, C, and D (1), and secoiridoid glucosides of the oleoside type (2), such as multifloroside, multiroside, 10-hydroxyoleoside 11-methyl ester, and others, from the aerial parts of this plant. Some of these showed coronary vasodilating and cardiotropic activities. In a continuing search along this line, we report here the isolation and structure elucidation of a new secoiridoid glucoside, jasmultiside [**1**], along with 10-hydroxyoleuropein [**2**] from this plant.

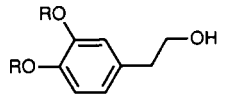
RESULTS AND DISCUSSION

The EtOH extract from the aerial parts of *J. multiflorum* was fractionated by solvent partitions. The *n*-BuOH-soluble fraction was separated by a combination of Sephadex LH-20 and Si gel cc to yield compounds **1** and **2**. Compound **2** was identified to be 10-hydroxyoleuropein (2).

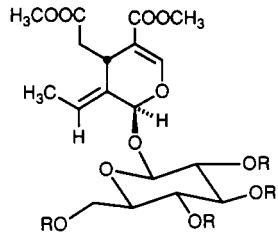
Compound **1**, $[\alpha]_D$ −42.6° (MeOH), was isolated as a pale brown amorphous powder. The fabms of **1** showed a negative quasimolecular ion at *m/z* 661 $[M-H]^-$, consistent with a molecular formula of $C_{32}H_{38}O_{15}$, which was also confirmed by DEPT ^{13}C nmr. The ir and uv spectra of **1** displayed typical absorptions of an enol ether system conjugated with a carbonyl group (1703, 1626 cm^{-1}, 223.6 nm) belonging to a secoiridoid skeleton (3), and, in addition, absorptions due to a catecholic chromophore (1529, 1447 cm^{-1}, 281.6 nm). The ^1H-nmr spectrum of **1** (Table 1) closely resembled those of **2** and multifloroside [**3**] (2), especially those signals arising from the secoiridoid nucleus. In particular, a hemiacetalic proton at δ 5.80 (br s) and a vinylic proton at δ 7.42 (br s) were assignable, respectively, to H-1 and H-3 of the secoiridoid ring. An additional hemiacetalic proton at δ 4.79 (d, $J = 7.8$ Hz) was attributed to the anomeric proton of a β-D-glucopyranosyl moiety. The methylene protons H-6a, -6b consituted part of an AMX spin pattern at δ 2.31 (dd, $J = 13.9, 9.5$ Hz) and δ 2.62 (dd, $J = 13.9, 4.1$ Hz), both of which were shown to be correlated with the methine proton H-5 at δ 3.88 (dd, $J = 9.5, 4.1$ Hz) by inspection of the COSY-45 spectrum. The only feature that is different from **3** is the signal for the vinylic proton H-8, which appeared as a quartet at δ 6.02 ($J = 6.9$ Hz), indicating its coupling with a methyl signal at δ 1.58 (d, $J = 6.9$ Hz, H-10). Both H-8 and H-10 showed long range couplings with H-1 as indicated in the COSY-45 spectrum. The nature of the side chains was revealed by two sets of overlapped aromatic AMX and aliphatic ABX_2 spin systems, compatible with the presence of two 3,4-dihydroxyphenethoxyl moieties. This is also supported by the fragment ion **11** at *m/z* 154 in the eims of **1**. Cross comparison of the ^{13}C-nmr spectrum



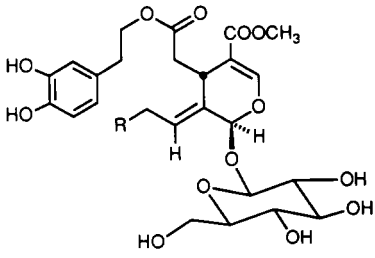
1 R=H, R_1=H
3 R=OH, R_1=H
5 R=H, R_1=Ac
6 R=OAc, R_1=Ac



7 R=H
8 R=Ac



9 R=H
10 R=Ac



2 R=OH
4 R=H

of **1** (Table 1) with that of **3** revealed close correspondence in every aspect, except that C-10 is replaced by a methyl group (δ 13.47) in **1**, and, as a result, the adjacent methine carbon C-8 (δ 124.85) undergoes an upfield shift. Strong resemblance was also observed in comparison with the spectrum of oleuropein [**4**] (4) among those carbon resonances of the secoiridoid glucoside residue, indicating identity at this part of the structure.

Acetylation of **1** provided an octaacetate **5**, $[\alpha]_D$ −58.5° ($CHCl_3$), $C_{48}H_{54}O_{23}$, by fabms based on the quasimolecular ions at *m/z* 999 $[M+H]^+$ and *m/z* 1021 $[M+Na]^+$. Besides those signals from the secoiridoid glucoside residue, the ^1H nmr of **5** displayed four aliphatic acetyl and four aromatic acetyl singlets, consistent with the presence of a β-D-glucopyranosyl and two 3,4-dihydroxyphenethoxyl moieties. The ^{13}C-nmr spectrum of **5** resembled closely that of multifloroside nonaacetate [**6**] (2), except for the resonance of the methyl carbon C-10 (δ 13.07) which appears in the aliphatic region. Thus, all these spectral evidences point to structure **1** for jasmultiside.

The stereochemistry was elaborated further by the following series of reactions. Mild alkaline hydrolysis of **1** provided the 3,4-dihydroxyphenethyl alcohol **7** and the secoiridoid glucoside moiety. The former compound was identified as its dimethyl ether **8** (5), and the latter residue was methylated with CH_2N_2 to yield the dimethyl ester **9**. The ^1H and ^{13}C nmr of **9** showed resonances of two carbomethoxy groups in addition to the signals from the secoiridoid glucoside residue. Further acetylation of **9** provided the dimethyl ester tetraacetate **10**. Its eims exhibited a fragment ion **12** at *m/z* 239, representing the secoiridoid nucleus, and ion **13** at *m/z* 331 indicative of the glucose tetraacetate unit. In addition to four acetyl signals, the proton spectrum of **9** revealed a characteristic A_3MX spin system originating from H-10, H-8, and H-1. Thus, the signal of methyl proton H-10 appears as a double doublet at δ 1.71 (J = 7.0, 1.4 Hz), showing couplings with H-1 at δ 5.67 (br s) and the vinylic proton H-8,

TABLE 1. Nmr Spectral Data for Compounds **1**, **3**, and **4**.

Carbon	Compound			
	1		**3**	**4**
	δ ^{1}H[a]	δ ^{13}C[b]	δ ^{13}C[c]	δ ^{13}C[d]
1	5.80 (br s)	94.35 (d)	94.57 (d)	95.2 (d)
3	7.42 (br s)	155.03 (d)	155.02 (d)	155.1 (d)
4		109.52 (s)	109.26 (s)	109.4 (s)
5	3.88 (dd, 9.5, 4.1)	31.64 (d)	32.17 (d)	31.7 (d)
6a	2.31 (dd, 13.9, 9.5)	41.10 (t)	41.07 (t)	41.2 (t)
6b	2.62 (dd, 13.9, 4.1)			
7		173.17 (s)	173.07 (s)	173.1 (s)
8	6.02 (q, 6.9)	124.85 (d)	129.33 (d)	124.8 (d)
9		130.40 (s)	130.64 (s)	130.5 (s)
10	1.58 (d, 6.9)	13.47 (q)	59.19 (t)	13.6 (q)
11		168.17 (s)	167.96 (s)	168.6 (s)
1′	4.21 (AB)[e]	66.76 (t)	66.88 (t)	66.8 (t)
2′	2.76 (X_2, 6.9)[f]	35.27 (t)[e]	35.29 (t)[e]	35.3 (t)
3′		131.02 (s)[f]	130.90 (s)[f]	130.7 (s)
4′	6.67 (d, 1.7)	116.42 (d)[g]	116.46 (d)[g]	116.5 (d)
5′		146.06 (s)	146.10 (s)[h]	146.1 (s)
6′		144.72 (s)	144.78 (s)	144.8 (s)
7′	6.69 (d, 8.0)	117.03 (d)	117.04 (d)[i]	117.0 (d)
8′	6.52 (dd, 8.0, 1.7)	121.33 (d)	121.32 (d)	121.3 (d)
1″	4.10 (AB)[e]	66.24 (t)	66.39 (t)	
2″	2.72 (X_2, 6.9)[f]	35.40 (t)[e]	35.39 (t)[e]	
3″		130.81 (s)[f]	130.92 (s)[f]	
4″	6.67 (d, 1.7)	116.41 (d)[g]	116.14 (d)[g]	
5″		146.06 (s)	146.78 (s)[h]	
6″		144.72 (s)	144.78 (s)	
7″	6.69 (d, 8.0)	117.03 (d)	116.97 (d)[i]	
8″	6.52 (dd, 8.0, 1.7)	121.33 (d)	121.32 (d)	
1‴	4.79 (d, 7.8)	100.92 (d)	100.82 (d)	100.9 (d)
2‴	3.2–3.5	74.69 (d)	74.78 (d)	74.7 (d)
3‴	3.2–3.5	78.18 (d)	78.30 (d)	78.3 (d)
4‴	3.2–3.5	71.43 (d)	71.37 (d)	71.4 (d)
5‴	3.2–3.5	77.86 (d)	78.14 (d)	77.9 (d)
6‴a	3.67 (dd, 10.3, 1.0)	62.69 (t)	62.40 (t)	62.7 (t)
6‴b	3.86 (d, 10.3)			
OMe				51.9 (q)

[a]Taken at 300 MHz in CD_3OD; *J* values in Hz.

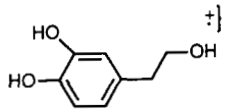
[b]Taken at 75.47 MHz in CD_3OD. The number of protons linked to each barbon in the ^{13}C spectra was determined by DEPT sequence and the number of carbons in each signal verified by inverse gated decoupling.

[c]Taken at 75.47 MHz in CD_3OD. Data in this column are from Shen *et al.* (2).

[d]Taken at 50.10 MHz in CD_3OD. Data in this column are from Inoue *et al.* (4).

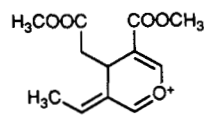
[e–i]Values with identical superscripts within a column may be interchanged.

which appears as a double quartet at δ 5.98 (J = 7.0, 1.0 Hz) due to allylic coupling with H-1. The carbon spectrum is also in complete agreement with structure **9**. Spectral comparisons showed **9** to be identical with oleoside 7,11-dimethyl ester tetraacetate, a known iridoid derivative prepared from oleuropein [**4**] and its analogues (5,6). Thus, this conversion unequivocally established the absolute structure of jasmultiside as **1**.



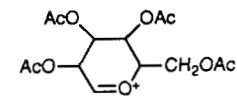
m/z 154

11



m/z 347

12



m/z 331

13

EXPERIMENTAL

General instrumental and experimental procedures were described in a previous paper (1). In addition to Si gel, RP C-18 (Merck) and Sephadex LH-20 were used in cc.

PLANT MATERIAL.—*J. multiflorum* was collected in a suburb of Taipei in July 1986. A voucher specimen is preserved in the Herbarium of the School of Pharmacy, National Taiwan University.

EXTRACTION AND ISOLATION.—The fresh plant aerial parts were extracted with 95% EtOH, and the extract was fractionated as described previously (2). Part of the *n*-BuOH-soluble fraction (95 g) was chromatographed on a Si gel (900 g) column and eluted with the lower layer of a $CHCl_3$-MeOH-H_2O (45:15:4) mixture, to yield fractions 1–8. An aliquot of fraction 3 (10.5 g) was separated on a Sephadex LH-20 (480 g) column eluted with MeOH to give 10-hydroxyoleuropein [**2**] (3.14 g) and a residue (4.65 g), shown to be a mixture by tlc. The latter residue was fractionated again on a Sephadex LH-20 (300 g) column and eluted with MeOH to afford an additional amount (0.9 g) of **2** and jasmultiside [**1**] (2.1 g). Compound **2** was identified by spectral comparisons (ir, uv, ^{1}H and ^{13}C nmr) with an authentic sample (2).

JASMULTISIDE [**1**].—Amorphous brown powder: $[\alpha]^{25}D$ −42.6° (*c* = 1.0, MeOH); uv λ max (MeOH) (log ε) 223.6 (4.42), 281.6 (3.85) nm; ir ν max (KBr) 3392, 2919, 1703, 1626, 1529, 1447, 1391, 1353, 1287, 1201, 1158, 1098, 1076, 1041, 926, 863 cm^{-1}; fabms (thioglycerol) *m/z* (rel. int.) $[M-H]^-$ 661 (87.5), $[M-H-C_8H_8O_2]^-$ 525 (14.4), $[M-H-C_6H_{10}O_5]^-$ 499 (4.9); eims *m/z* (rel. int.) $[M-Glc]^+$ 482 (3.1), 347 (4.9), 302 (10.2), $[M-Glc-154-CO]^+$ 300 (8.8), 289 (8.5), 288 (5.2), 287 (8.9), 286 (5.3), $[C_8H_{10}O_3]^+$ 154 (53.8), $[154-OH]^+$ 137 (43.3), 136 (82.9), 124 (10.0), $[154-MeO]^+$ 123 (100.0), 77 (6.7), 73 (29.5), 61 (9.3), 60 (14.0), 57 (10.0); ^{1}H and ^{13}C nmr see Table 1; selected COSY-45 data, ^{1}H-^{1}H [CD_3OD) regular H-5↔H-6a, H-6b, H-10↔H-8, H-1′↔H-2′, H-1″↔H-2″, H-1‴↔H-2‴, long range, H-10↔H-1, H-8↔H-1, H-3↔H-5, H-1↔H-3.

JASMULTISIDE OCTAACETATE [**5**].—Compound **1** (110.7 mg) was acetylated with Ac_2O/pyridine at room temperature. Usual workup provided a residue which was chromatographed on a Si gel (10 g) column and eluted with $CHCl_3$ and increasing amounts (1–5%) of MeOH to yield jasmultiside octaacetate [**5**] (130 mg) as white crystals from $CHCl_3$: mp 58–60°; $[\alpha]^{27}D$ −58.5° (*c* = 1.23, $CHCl_3$); uv λ max (MeOH) (log ε) 240 (4.15), 270 (3.43), 285 (3.00) nm; ir ν max (KBr) 1759, 1704, 1633, 1507, 1373, 1260, 1216 cm^{-1}; ^{1}H nmr δ (300 MHz, $CDCl_3$) 7.38 (1H, s, H-3), 7.08 (4H, m, H-4′, -8′, -4″, -8″), 7.02 (2H, d, *J* = 8.2, H-7′, -7″), 5.95 (1H, q, *J* = 6.9, H-8), 5.64 (1H, bs, H-1), 5.21 (1H, dd, *J* = 9.2, 8.4, H-3‴), 5.09 (1H, dd, *J* = 7.7, 8.4, H-2‴), 5.09 (1H, dd, *J* = 9.2, 9.5, H-4‴), 4.98 (1H, d, *J* = 7.7, H-1‴), 4.29 (4H, m, H-1′, -1″), 4.18 (1H, dd, *J* = 12.6, 7.6, H-6‴a), 4.06 (1H, bd, *J* ≈ 12.6, H-6‴b), 3.92 (1H, dd, *J* = 4.5, 9.0, H-5), 3.74 (1H, m, H-5‴), 2.89 (4H, m, H-2′, -2″), 2.65 (1H, dd, *J* = 14.5, 4.5, H-6b), 2.28 (1H, dd, *J* = 14.5, 9.0, H-6a) (coupling pattern of H-6a was determined from the COSY-45 spectrum), 2.24 (12H, s, OAc), 1.99 (12H, s, OAc), 1.64 (3H, d, *J* = 6.9, H-10); ^{13}C nmr δ (75.47 MHz, $CDCl_3$), 93.77 (d, C-1), 152.96 (d, C-3), 108.55 (s, C-4), 30.09 (d, C-5), 39.59 (t, C-6), 170.09 (s, C-7), 124.41 (d, C-8), 128.11 (s, C-9), 13.07 (q, C-10), 165.77 (s, C-11), 64.21 (t, C-1′), 34.24 (t, C-2′), 136.59 (s, C-3′), 123.09 (d, C-4′), 141.88 (s, C-5′), 140.61 (s, C-6′), 123.50 (d, C-7′), 126.56 (d, C-8′), 63.93 (t, C-1″), 34.08 (t, C-2″), 136.34 (s, C-3″), 123.09 (d, C-4″), 141.88 (s, C-5″), 140.61 (s, C-6″), 123.50 (d, C-7″), 126.56 (d, C-8″), 97.02 (d, C-1‴), 70.71 (d, C-2‴), 72.44 (d, C-3‴), 68.26 (d, C-4‴), 72.04 (d, C-5‴), 61.58 (t, C-6‴), 20.19 (×8) (q, $OCOCH_3$), 167.68 (×2), 167.76 (×2), 168.90, 169.00, 169.67, 170.62 (s, OCOMe); fabms (nitrobenzyl alcohol) *m/z* (rel. int.) $[M+Na]^+$ 1021 (1.8), $[M+H]^+$ 998 (4.5), $[M-C_{14}H_{19}O_{10}]^+$ 651 (30.9), $[M-C_{12}H_{13}O_5]^+$ 761 (0.9), $[M-H-C_6H_{10}O_5]^+$ 499 (4.9), $[M-C_{12}H_{13}O_5-C_6H_{10}O_5-H]^+$ 413 (13.7), $[C_{14}H_{19}O_9]^+$ 331 (91.5).

ALKALINE HYDROLYSIS OF JASMULTISIDE [**1**].—A solution of **1** (254 mg) in MeOH (2 ml) was mixed with 0.5 M NaOH (25 ml) and stirred at room temperature for 2 h. The mixture was acidified with Amberlite IR-120 (H^+ form) and extracted with EtOAc (50 ml × 4). The combined EtOAc extracts were dried and evaporated to give a brown syrup (43 mg), which showed identical behavior (^{1}H nmr and ltc)

with 2-(3,4-dihydroxyphenyl)ethyl alcohol [**7**] (7). Methylation with CH_2N_2/Et_2O at 0°for 24 h gave, on crystallization from Et_2O, needles of **8** (20 mg), mp 40–41°, identical with an authentic sample of 2-(3,4-dimethoxyphenyl)ethyl alcohol (mmp, ^{1}H nmr, ir, and tlc). The aqueous layer was concentrated to dryness to give a residue (192 mg), which was treated with CH_2N_2/Et_2O at 0° for 2 days. The reaction mixture was chromatographed on a Si gel (5 g) column and eluted with $CHCl_3$ and increasing amounts (1–15%) of MeOH to yield **9** (61 mg) as an oil: $[\alpha]^{22}D$ −156.8° (*c* = 1.63, $CHCl_3$); uv λ max (MeOH) (log ε) 236.4 (3.60) nm; ir ν max (neat) 3634, 1725, 1634, 1500, 1413, 1267, 1192, 1080, 933 cm^{-1}; ^{1}H nmr δ (300 MHz, $CDCl_3$) 5.76 (1H, br s, H-1), 7.45 (1H, s, H-3), 3.91 (1H, dd, *J* = 2.6, 3.3, H-5), 2.32 (1H, dd, *J* = 13.1, 2.6, H-6a), 2.68 (1H, dd, *J* = 13.1, 3.3, H-6b), 6.02 (1H, q, *J* = 7.0, H-8), 1.62 (3H, d, *J* = 7.0, H-10), 4.75 (1H, d, *J* = 7.0, H-1′), 3.67 (3H, s, OMe), 3.58 (3H, s, OMe); ^{13}C nmr δ (75.47 MHz, $CDCl_3$) 94.99 (d, C-1), 153.63 (d, C-3), 108.64 (s, C-4), 30.78 (d, C-5), 40.12 (t, C-6), 171.92 (s, C-7), 124.42 (d, C-8), 129.01 (s, C-9), 13.25 (q, C-10), 166.84 (s, C-11), 100.51 (d, C-1′), 73.41 (d, C-2′), 76.64 (d, C-3′), 70.22 (d, C-4′), 76.51 (d, C-5′), 62.11 (t, C-6′), 51.37 (q, OAc), 51.63 (q, OAc). Acetylation of compound **9** (55 mg) with Ac_2O/pyridine and usual workup provided a residue (65 mg), which was chromatographed on a Si gel (5 g) column and eluted with $CHCl_3$/MeOH mixtures (1–15%). Final purification with preparative tlc [1 mm plate, $CHCl_3$-MeOH (50:1)] yielded **10** (19 mg), $[\alpha]^{27}D$ −120.3 (*c* = 1.2, $CHCl_3$). The spectral data ([α]D, ir, uv, ^{1}H nmr) of this compound are identical with those of oleoside 7,11-dimethyl ester tetraacetate (5,6). Additional data: high resolution ^{1}H-nmr δ (300 MHz, $CDCl_3$) 5.67 (1H, br s, H-1), 7.43 (1H, s, H-3), 3.95 (1H, dd, *J* = 4.4, 8.9, H-5), 2.36 (1H, dd, *J* = 14.2, 8.9, H-6a), 2.73 (1H, dd, *J* = 14.2, 4.4, H-6b), 5.98 (1H, dq, *J* = 7.0, 1.0, H-8), 1.71 (3H, dd, *J* = 7.0, 1.4, H-10), 4.30 (1H, dd, *J* = 12.4, 4.6, H-6′a), 4.18 (1H, dd, *J* = 12.4, 2.6, H-6′b), 3.60 (3H, s, OMe), 3.70 (3H, s, OMe), 2.02, 2.04, 1.98 (×2) (12H, s, OAc); selected COSY 45 data ^{1}H-^{1}H [CD_3OD] regular H-5↔H-6a, H-6b, H-10↔H-8, H-5′↔H-6′a, H-6′b, long range, H-10↔H-1, H-8↔H-1, H-3↔H-5, H-1↔H-3; ^{13}C nmr δ (75.47 MHz, $CDCl_3$) 93.92 (d, C-1), 153.00 (d, C-3), 108.88 (s, C-4), 30.33 (d, C-5), 39.84 (t, C-6), 171.34 (s, C-7), 124.69 (d, C-8), 128.45 (s, C-9), 13.31 (q, C-10), 167.01 (s, C-11), 97.22 (d, C-1′), 70.97 (d, C-2′), 72.64 (d, C-3′), 68.55 (d, C-4′), 73.32 (d, C-5′), 61.90 (t, C-6′), 51.37, 51.25 (q, C-7, -11 OMe), 20.47 (×4) (q, $OCOCH_3$), 168.9, 169.23, 169.93, 170.35 (s, OCOMe); fabms (nitrobenzyl alcohol) *m/z* (rel. int.) $[M + Na]^+$ 609 (12.1), $[M + H]^+$ 587 (8.5), 331 $[C_6H_7O(OAc)_4]^+$ (100), 239 $[M - C_6H_7O_2(OAc)_4]^+$ (35.5), 169 (78.0); eims *m/z* (rel. int.) $[M - Glc]^+$ 332 (6.0), 331 $[C_6H_7O(OAc)_4]^+$ (42.9), 271 (13.7), 239 $[M - C_6H_7O_2(OAc)_4]^+$ (5.6), 211 (6.2), 170 (8.2), 169 (100), 145 (4.3), 139 (3.2), 109 (25.6).

ACKNOWLEDGMENTS

We are grateful for financial support (NSC-79-0412-B002-13 and NSC-79-0420-B002-16) from the National Science Council, R.O.C. We also thank Dr. Shoei-sheng Lee for his help and consultation in running some reactions.

LITERATURE CITED

1. Y.C. Shen and C.H. Chen, *J. Nat. Prod.*, **52**, 1060 (1989).
2. Y.C. Shen, C.Y. Lin, and C.H. Chen, *Phytochemistry*, **29**, 2905 (1990).
3. L.J. Ed-Naggar and J.L. Beal, *J. Nat. Prod.*, **43**, 649 (1980).
4. K. Inoue, T. Nishioka, T. Tanahashi, and H. Inouye, *Phytochemistry*, **21**, 2305 (1982).
5. H. Inouye, K. Inoue, T. Nishioka, and M. Kaniwa, *Phytochemistry*, **14**, 2029 (1975).
6. H. Inouye and T. Nishioka, *Tetrahedron*, **28**, 4231 (1972).
7. T. Kubota, N. Ichikawa, and T. Kamikawa, *J. Chem. Soc. Jpn.*, **89**, 72 (1968).

Received 11 February 1991