

Four New Phenolic Compounds from *Curculigo crassifolia* (Hypoxidaceae)

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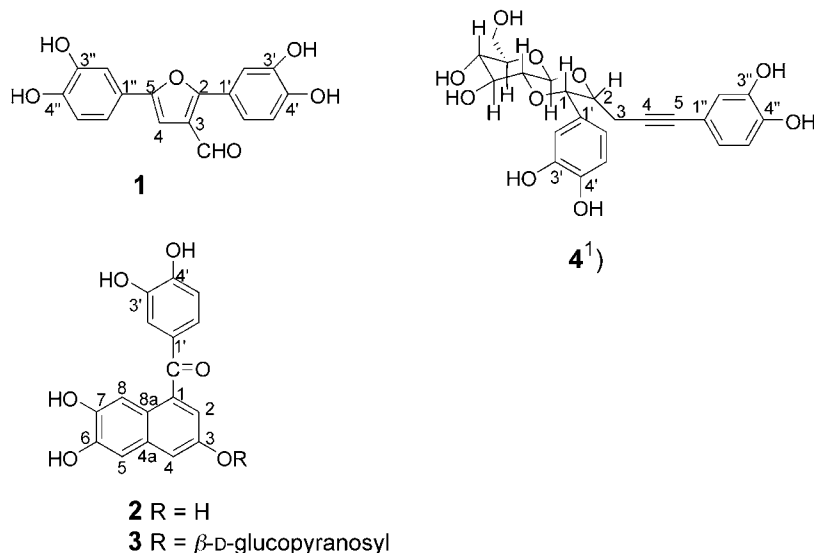
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Four new phenolic compounds, named crassifogenin A (**1**), crassifogenin B (**2**), crassifoside A (**3**), and crassifoside B (**4**), were isolated from the EtOH extract of the rhizomes of *Curculigo crassifolia*, and based on the chemical transformation and modern spectroscopic experiments, including 2D-NMR techniques (HMQC, HMBC, COSY, HMQC-TOCSY, and NOE), their structures were elucidated as 2,5-bis(3,4-dihydroxyphenyl)furan-3-carbaldehyde (**1**), (3,4-dihydroxyphenyl)(3,6,7-trihydroxynaphthalen-1-yl)methanone (**2**), (3,4-dihydroxyphenyl)[3-(β -D-glucopyranosyloxy)-6,7-dihydroxynaphthalen-1-yl]methanone (**3**), and 1,2-*O*-[2-(3,4-dihydroxyphenyl)-1-[3-(3,4-dihydroxyphenyl)prop-2-ynyl]ethane-1,2-diyl]- β -D-glucopyranose (**4**).

1. Introduction. – Several species of the genus *Curculigo* are well known for their use in medicine. Previous phytochemical and pharmacological studies on the species of the genus had been reported [1–4]. *Curculigo crassifolia* (BAK.) HOOK. f. (Hypoxidaceae) is distributed in the western and southern regions of China [5]. The rhizomes of this plant are used as a folk medicine for treating child pneumonitis [5]. However, so far, no extensive studies of this plant with respect to its chemical characteristics have been reported. The interesting immense medicinal importance of this genus encouraged us to undertake the phytochemical investigation on *Curculigo crassifolia*. This paper mainly describes the isolation and structural identification of the four new phenolic compounds **1–4** from the EtOH extract of the rhizomes of *C. crassifolia*.

2. Results and Discussion. – Crassifogenin A (**1**) was obtained as yellow powder. The positive-ion HR-FAB-MS showed a quasi-molecular-ion peak at m/z 313.0705 ($[M + H]^+$), in accordance with the molecular formula $C_{17}H_{13}O_6^+$ (calc. 313.0712). The IR spectrum indicated the presence of OH groups (3431 cm^{-1}) and a carbonyl group (1652 cm^{-1}). The $^1\text{H-NMR}$ spectrum showed the presence of two 3,4-disubstituted aromatic rings. 1D-NMR and HMBC experiments suggested the presence of a furan-ring moiety. Thus, the structure of **1** was elucidated as 2,5-bis(3,4-dihydroxyphenyl)-furan-3-carbaldehyde.

In the HMBC spectrum of **1**, the signal of H–C(4) at δ 7.39 was correlated with that of C(5) at δ 151.5, C(2) at δ 156.2, and CHO at δ 178.9 (Table 1). The signal of CHO at δ 9.37 was correlated with that of C(3) at δ 122.6, C(2) at δ 156.2, C(4) at δ 128.2, and C(5) at δ 151.5, which suggested the presence of a furan-ring moiety with a carbaldehyde located at C(3). The HMBC experiment also showed the long-range couplings of H–C(2') (δ 7.10) and H–C(6') (δ 7.02) with C(2) at δ 156.2, and of H–C(2'') (δ 6.81) and H–C(6'') (δ 6.71) with C(5) at δ



151.5, which suggested that two 3,4-disubstituted aromatic rings were connected with C(2) and C(5). On acetylation of **1** with Ac_2O in pyridine, the positive-ion FAB-MS of the acetate of **1** showed an ion at m/z 481 ($[\text{M}(\mathbf{1}) + 1 + 4 \text{ Ac}]^+$), which suggested that **1** had 4 free OH groups located at C(3'), C(4'), C(3''), and C(4'').

Table 1. ^1H - and ^{13}C -NMR and HMBC Data (CD_3OD) of Compound **1**. δ in ppm, J in Hz.

	$\delta(\text{C})^{\text{a}}$	$\delta(\text{H})^{\text{b}}$	HMBC ^b (H \rightarrow C)
C(2)	156.2	–	
C(3)	122.6	–	
H–C(4)	128.2	7.39 (<i>s</i>)	C(2), C(5), CHO, C(1'')
C(5)	151.5	–	
CHO	178.9	9.37 (<i>s</i>)	C(2), C(3), C(4), C(5)
C(1')	125.5	–	
H–C(2')	115.2	7.10 (<i>d</i> , $J = 2.1$)	C(2), C(1'), C(3'), C(6')
C(3')	146.4 ^c	–	
C(4')	148.3	–	
H–C(5')	115.9	6.72 (<i>d</i> , $J = 8.4$)	C(1'), C(3'), C(4')
H–C(6')	120.8	7.02 (<i>dd</i> , $J = 8.4, 2.1$)	C(2), C(2'), C(4')
C(1'')	125.3	–	
H–C(2'')	116.9	6.81 (<i>d</i> , $J = 2.0$)	C(5), C(3''), C(4''), C(6'')
C(3'')	146.4 ^c	–	
C(4'')	146.6 ^c	–	
H–C(5'')	116.8	6.79 (<i>d</i> , $J = 8.1$)	C(1''), C(3''), C(4'')
H–C(6'')	121.5	6.71 (<i>dd</i> , $J = 8.1, 2.0$)	C(5), C(2''), C(4'')

^a) Recorded at 125 MHz. ^b) Recorded at 500 MHz. ^c) Values may be interchanged.

¹⁾ Arbitrary numbering; for systematic names, see *Exper. Part*.

Crassifogenin B (**2**) was obtained as pale yellow powder. The positive-ion HR-FAB-MS showed a quasi-molecular-ion peak at m/z 313.0701 ($[M + H]^+$), in accordance with the molecular formula $C_{17}H_{13}O_6^+$ (calc. 313.0712). The IR spectrum indicated the presence of OH groups (3433 cm^{-1}) and a carbonyl group (1652 cm^{-1}). The ^1H - and ^{13}C -NMR data (Table 2) indicated the presence of a naphthalene-ring moiety in **2**. This was further confirmed by the HMBC spectrum. From these results and the spectral data, compound **2** was determined as (3,4-dihydroxyphenyl)(3,6,7-trihydroxynaphthalen-1-yl)methanone.

Table 2. ^1H - and ^{13}C -NMR and HMBC Data (CD_3OD) of Compound **2**. δ in ppm, J in Hz.

	$\delta(\text{C})^{\text{a}}$	$\delta(\text{H})^{\text{b}}$	HMBC ^b (H \rightarrow C)
C(1)	137.9	–	
H–C(2)	116.9	6.85 (d , $J = 2.4$)	C(4), C(8a), C=O
C(3)	153.2	–	
H–C(4)	111.6	7.03 (d , $J = 2.4$)	C(2), C(3), C(5), C(8a)
H–C(5)	109.6	6.99 (s)	C(4), C(6), C(8a)
C(6)	146.4	–	
C(7)	148.6	–	
H–C(8)	108.7	7.05 (s)	C(1), C(6), C(7), C(4a)
C(8a)	122.6	–	
C(4a)	132.9	–	
C=O	199.5	–	
C(1')	131.2	–	
H–C(2')	118.0	7.33 (d , $J = 2.0$)	C(3'), C(4'), C(6'), C=O
C(3')	146.2	–	
C(4')	152.8	–	
H–C(5')	115.9	6.80 (d , $J = 8.3$)	C(1'), C(3'), C(4')
H–C(6')	125.8	7.21 (dd , $J = 8.3, 2.0$)	C(2'), C(4'), C=O

^a) Recorded at 100 MHz. ^b) Recorded at 400 MHz.

In the HMQC spectrum of **2**, two d at δ 6.85 ($J = 2.4$ Hz, H–C(2)) and 7.03 ($J = 2.4$ Hz, H–C(4)) had connectivities with the C-atoms at δ 116.9 (C(2)) and 111.6 (C(4)), and two s at δ 6.99 (H–C(5)) and 7.05 (H–C(8)) had connectivities with C-atoms at δ 109.6 (C(5)) and 108.7 (C(8)). The NMR data revealed the presence of a 3,4-disubstituted aromatic ring linked to a C=O group by the presence of two d (each 1 H) at δ 7.33 ($J = 2.0$ Hz, H–C(2')) and 6.80 ($J = 8.3$ Hz, H–C(5')) along with a dd (1 H) at δ 7.21 ($J = 8.3, 2.0$ Hz, H–C(6')). The C-atoms connected to these protons were observed at δ 118.0 (C(2')), 115.9 (C(5')), and 125.8 (C(6')) in the HMQC spectrum. The HMBC experiment displayed correlations between H–C(2) and the C=O group and between H–C(2'), H–C(6'), and the C=O group, which suggested that the C=O group was linked to C(1) and C(1'), respectively. On acetylation of **2** with Ac_2O in pyridine, the positive-ion FAB-MS of the acetate of **2** showed an ion peak at m/z 523 ($[M(\mathbf{2}) + 1] + 5\text{ Ac}]^+$), which suggested that **2** had five free OH groups located at C(3), C(6), C(7), C(3'), and C(4').

Crassifoside A (**3**) was obtained as white powder. The negative-ion HR-FAB-MS showed a quasi-molecular-ion peak at m/z 473.1092 ($[M - H]^-$), in accordance with the molecular formula $C_{23}H_{21}O_{11}$ (calc. 473.1083). Its IR (see *Exper. Part*) and the ^1H - and ^{13}C -NMR data (Table 3) of the aglycone of **3** were closely similar to those of compound **2**, indicating that they have the same skeleton. On acidic hydrolysis of **3**, crassifogenin B (**2**) and glucose were detected by TLC and comparison on paper chromatography with an authentic sample of glucose. Thus the structure of **3** was determined to be (3,4-dihydroxyphenyl)[3-(β -D-glucopyranosyloxy)-6,7-dihydroxynaphthalen-1-yl]methanone.

Table 3. ^1H - and ^{13}C -NMR and HMBC Data (CD_3OD) of Compound **3**. δ in ppm, J in Hz.

	$\delta(\text{C})^{\text{a}}$	$\delta(\text{H})^{\text{b}}$	HMBC ^b (H \rightarrow C)
C(1)	137.8	–	
H–C(2)	117.9	7.10 (<i>d</i> , $J=2.3$)	C(1), C(3), C(4), C(8a), C=O
C(3)	153.9	–	
H–C(4)	114.0	7.41 (<i>d</i> , $J=2.3$)	C(2), C(3), C(5), C(8a)
H–C(5)	110.6	7.12 (<i>s</i>)	C(4), C(6), C(8a)
C(6)	147.3	–	
C(7)	148.7	–	
H–C(8)	108.7	7.11 (<i>s</i>)	C(1), C(6), C(7), C(8a), C(4a)
C(8a)	124.2	–	
C(4a)	132.4	–	
C=O	199.1	–	
C(1')	131.1	–	
H–C(2')	118.0	7.33 (<i>d</i> , $J=2.0$)	C(3'), C(4'), C(6'), C=O
C(3')	146.5	–	
C(4')	152.8	–	
H–C(5')	115.9	6.80 (<i>d</i> , $J=8.3$)	C(1'), C(3'), C(4')
H–C(6')	125.8	7.21 (<i>dd</i> , $J=8.3, 2.0$)	C(2'), C(4'), C=O
Glc: H–C(1'')	102.9	4.99 (<i>d</i> , $J=7.3$)	C(3)
H–C(2'')	71.4	3.47 (<i>m</i>)	
H–C(3'')	78.2	3.45 (<i>m</i>)	
H–C(4'')	75.0	3.46 (<i>m</i>)	
H–C(5'')	78.0	3.49 (<i>m</i>)	
$\text{CH}_2(6)$	62.5	3.90 (<i>dd</i> , $J=12.1, 2.0$), 3.71 (<i>dd</i> , $J=12.1, 5.3$)	

^a) Recorded at 100 MHz. ^b) Recorded at 400 MHz.

The ^1H - and ^{13}C -NMR spectrum of **3** indicated the presence of a hexose unit. A *d* (1 H) at δ 4.99 ($J=7.3$ Hz, H–C(1'') (Glc)) had connectivities with C-atoms at δ 102.9 (C(1'') (Glc)) in the HMQC spectrum. From the coupling constant of the anomeric H–C(1'') and ^{13}C -NMR chemical shifts due to the sugar moiety, the glucose unit should be in the β -D form. The other positions of the glucose unit were confirmed by ^1H , ^1H -COSY, HMBC, HMQC-TOCSY, and NOE experiments. In the HMBC spectrum of **3**, the signal of the anomeric H–C(1'') at δ 4.99 was correlated with that of C(3) at δ 153.9, which suggested that the glucose was connected at C(3).

Crassifoside B (**4**) was obtained as white powder. The negative-ion HR-FAB-MS showed a quasi-molecular-ion peak at m/z 459.2289 ($[M - \text{H}]^-$), in accordance with the molecular formula $\text{C}_{23}\text{H}_{23}\text{O}_{10}$ (calc. 459.1369). The ^1H -NMR spectrum showed the presence of two 3,4-disubstituted aromatic rings. By a selective ^1H -decoupling experiment and the HMBC spectrum, the norlignan sequence $\text{PhCH}(\text{O})\text{CH}(\text{O})\text{CH}_2\text{C}\equiv\text{CPh}$ was established. The remaining C-atoms presumably belong to a hexose unit. On acidic hydrolysis of **4**, glucose was detected by comparison on paper chromatography with an authentic sample. A detailed NMR-data analysis (Table 4) and 2D-NMR experiments (including ^1H , ^1H -COSY, HMBC, HMQC-TOCSY, and NOESY) suggested that **4** was a glucopyranose-fused norlignan with the structure of 1,2-*O*-{2-(3,4-dihydroxyphenyl)-1-[3-(3,4-dihydroxyphenyl)prop-2-ynyl]ethane-1,2-diy]- β -D-glucopyranose.

In the HMQC spectrum of **4**, a *d* (1 H) at δ 4.83 ($J=7.7$ Hz, H–C(1) (Glc)) had connectivities with C-atoms at δ 97.1 (C(1) (Glc)). From the coupling constant of the anomeric H–C(1) and ^{13}C -NMR chemical shifts,

Table 4. ^1H - and ^{13}C -NMR and HMBC Data (CD_3OD) of Compound **4**. δ in ppm, J in Hz. Arbitrary numbering¹.

	$\delta(\text{C})^{\text{a}}$	$\delta(\text{H})^{\text{b}}$	HMBC ^b (H \rightarrow C)
H–C(1)	77.6	4.81 (<i>d</i> , $J = 4.7$)	C(2'), C(6'), C(2), C(2) (Glc)
H–C(2)	75.4	4.56 (<i>m</i>)	C(1), C(3), C(4), C(1) (Glc)
CH ₂ (3)	23.5	2.85 (<i>dd</i> , $J = 17.1, 6.0$)	C(1), C(2), C(5)
		2.65 (<i>dd</i> , $J = 17.1, 6.0$)	
C(4)	83.3	–	
C(5)	84.3	–	
C(1')	131.5	–	
H–C(2')	116.2	6.98 (<i>d</i> , $J = 2.2$)	C(1), C(3'), C(6')
C(3')	146.0	–	
C(4')	146.3	–	
H–C(5')	116.2	6.76 (<i>d</i> , $J = 8.1$)	C(1'), C(3'), C(4')
H–C(6')	120.7	6.87 (<i>dd</i> , $J = 8.1, 2.2$)	C(1), C(2), C(4')
C(1'')	115.9	–	
H–C(2'')	119.5	6.79 (<i>d</i> , $J = 1.7$)	C(5), C(3''), C(4''), C(6'')
C(3'')	146.5	–	
C(4'')	147.0	–	
H–C(5'')	116.2	6.66 (<i>d</i> , $J = 8.1$)	C(1''), C(3''), C(4'')
H–C(6'')	125.0	6.73 (<i>dd</i> , $J = 8.1, 1.7$)	C(5), C(1''), C(2''), C(4'')
Glc: H–C(1)	97.1	4.83 (<i>d</i> , $J = 7.7$)	C(2)
H–C(2)	73.9	3.51 (<i>m</i>)	C(1)
H–C(3)	75.8	3.54 (<i>m</i>)	
H–C(4)	71.9	3.33 (<i>m</i>)	
H–C(5)	79.5	3.42 (<i>m</i>)	
CH ₂ (6)	62.5	3.85 (<i>dd</i> , $J = 12.0, 2.2$) 3.68 (<i>dd</i> , $J = 12.0, 5.6$)	

^a) Recorded at 125 MHz. ^b) Recorded at 500 MHz.

the glucose unit should be in the β -D form. The other linkages of the glucose unit were confirmed by ^1H , ^1H -COSY, HMBC, HMQC-TOCSY, and NOE experiments. In the HMBC spectrum of **4**, the signal of the anomeric H–C(1) at δ 4.83 was correlated with that of C(2) at δ 75.4, and the signal of H–C(2) (Glc) at δ 3.51 was correlated with that of C(1) at δ 77.6, which suggested that the glucose was connected at C(2) and C(1). The configuration of **4** was revealed by a NOESY experiment. The correlations H–C(1)/H–C(2) and H–C(1)/H–C(2) (Glc) were clearly observed, but no NOE was detected for H–C(2)/H–C(1) (Glc), establishing the axial orientation of H–C(1) and the equatorial orientation of H–C(2). On acetylation of **4** with Ac_2O in pyridine, the positive-ion FAB-MS analysis of the acetate of **4** showed an ion peak at m/z 755 ($[\text{M}(\mathbf{4}) + 1 + 7 \text{Ac}]^+$), which suggested that the glucose unit had only three free OH groups and that the other four AcO groups were replacing the phenolic OH groups.

The authors wish to thank the members of the analytical group of the State Key Laboratory of Phytochemistry and Plant Resources in West China for spectral measurements.

Experimental Part

General. Column chromatography (CC): *Qingdao* silica gel (200–300 mesh), eluent $\text{MeOH}/\text{CHCl}_3$; *Sephadex LH-20* gel, eluent EtOH. Optical rotations: *Jasco DIP-370* digital polarimeter; in MeOH. UV Spectra: *UV-210A* spectrometer; in MeOH; λ_{max} ($\log \epsilon$) in nm. IR Spectra: *Bio-Rad FTS-135* spectrometer; KBr pellets; $\tilde{\nu}$ in cm^{-1} . 1D- and 2D-NMR Spectra: *Bruker AM-400* or *Bruker DRX-500* spectrometer in CD_3OD with SiMe_4 as internal standard; δ in ppm, J in Hz. MS: *Autospec 3000* spectrometer, negative-ion or positive-ion mode; in m/z .

Plant Material. The plant material was collected in Eshan Prefecture, Yunnan Province, China, in October 2002 and identified as *Curculigo crassifolia* by Prof. Ping-Hua Yu, Kunming Institute of Botany, Chinese Academy of Science, where a voucher specimen is deposited.

Extraction and Isolation. The air-dried and powdered rhizomes of *C. crassifolia* (10 kg) were extracted with 95% EtOH (3×50 l) at r.t., and the combined extracts were evaporated to afford a residue (562 g). The residue was suspended in H₂O and then passed through a *D101*-resin column eluting with H₂O and 95% EtOH. The EtOH eluent was evaporated to give a residue (500 g), which was fractionated by CC (silica gel (3000 g, 200–300 mesh), CHCl₃/MeOH, 9:1): *Fractions 1–5*. *Fr. 2* (13 g) was purified by repeated CC (silica gel, CHCl₃/MeOH 9.5:0.5 and 8.5:1.5; then *Sephadex LH-20*, EtOH): pure **1** (150 mg). *Fr. 3* (40 g) was purified by CC (silica gel, CHCl₃/MeOH 9:1; then repeated *Sephadex LH-20*, EtOH): pure **2** (48 mg). *Fr. 5* (210 g) was purified by repeated CC (*Sephadex LH-20*, EtOH): pure **3** (23 mg) and **4** (25 mg).

Crassifogenin A (=2,5-Bis(3,4-dihydroxyphenyl)furan-3-carbaldehyde; **1**). Yellow powder. IR (KBr): 3431, 2928, 1652, 1490, 1280, 1112, 1053, 868, 794, 582. UV (MeOH): 203 (4.50), 261 (4.13), 292 (4.05), 363 (4.11). ¹H-NMR (CD₃OD, 500 MHz): see *Table 1*. ¹³C-NMR (CD₃OD, 125 MHz): see *Table 1*. FAB-MS (pos.): 313 ([*M* + H]⁺). HR-FAB-MS (pos.): 313.0705 ([*M* + H]⁺, C₁₇H₁₃O₆⁺; calc. 313.0712).

Tetraacetate of crassifogenin A: FAB-MS (pos.): 481 ([*M* + H]⁺).

Crassifogenin B (= (3,4-Dihydroxyphenyl)(3,6,7-trihydroxynaphthalen-1-yl)methanone; **2**). Pale yellow powder. IR (KBr): 3433, 2925, 2075, 1652, 1616, 1374, 1290, 1190, 1048, 875, 573. UV (MeOH): 233 (4.67), 285 (3.94), 324 (3.93). ¹H-NMR (CD₃OD, 400 MHz): see *Table 2*. ¹³C-NMR (CD₃OD, 100 MHz): see *Table 2*. FAB-MS (pos.): 313([*M* + H]⁺). HR-FAB-MS (pos.): 313.0701 ([*M* + H]⁺, C₁₇H₁₃O₆⁺; calc. 313.0712).

Pentaacetate of crassifogenin B: FAB-MS (pos.): 523 ([*M* + H]⁺).

Crassifoside A (= (3,4-Dihydroxyphenyl)[3-(β-D-glucopyranosyloxy)-6,7-dihydroxynaphthalen-1-yl]methanone; **3**). White powder. [α]_D²⁰ = –24.8 (*c* = 0.10, MeOH). IR (KBr): 3439, 2092, 1699, 1652, 1558, 1290, 1194, 1075, 1047, 880. UV (MeOH): 234 (4.61), 285 (3.93), 324 (3.88). ¹H-NMR (CD₃OD, 400 MHz): see *Table 3*. ¹³C-NMR (CD₃OD, 100 MHz): see *Table 3*. FAB-MS (neg.): 473 ([*M* – H][–]). HR-FAB-MS (neg.): 473.1092 ([*M* – H][–], C₂₃H₂₁O₁₁[–]; calc. 473.1083).

Crassifoside B (= 1,2-O-[2-(3,4-Dihydroxyphenyl)-1-[3-(3,4-dihydroxyphenyl)prop-2-ynyl]ethane-1,2-diol]-β-D-glucopyranose; **4**). White powder. [α]_D²⁰ = +74.7 (*c* = 0.15, MeOH). IR (KBr): 3422, 2926, 2052, 1610, 1521, 1445, 1370, 1289, 1115, 1044, 816, 618. UV (MeOH): 204 (4.73), 257 (4.18), 290 (3.82). ¹H-NMR (CD₃OD, 500 MHz): see *Table 4*. ¹³C-NMR (CD₃OD, 125 MHz): see *Table 4*. FAB-MS (neg.): 459 ([*M* – H][–]). HR-FAB-MS (neg.): 459.2289 ([*M* – H][–], C₂₃H₂₃O₁₀[–]; calc. 459.1369).

Heptaacetate of crassifoside B: FAB-MS (pos.): 755 ([*M* + H]⁺).

Acetylation of 1, 2, and 4. A soln. of each sample (1 mg) in pyridine (1 ml) was treated with Ac₂O (1 ml) and kept at 60–70° for 3–6 h. After evaporation, the residue was purified by prep. TLC (CHCl₃/MeOH, 9:1): acetate. Each acetate was subjected to FAB-MS analysis.

Acidic Hydrolysis. Compound **3** or **4** (3 mg) was dissolved in MeOH (2.0 ml) and 2M H₂SO₄ (2.0 ml) and refluxed on a boiling water bath for 2 h. The hydrolyzate was allowed to cool, diluted twofold with distilled H₂O, and partitioned between AcOEt and H₂O. The aq. layer was neutralized with aq. Ba(OH)₂ soln. and evaporated: residue. Crassifogenin B (**2**) was detected in the AcOEt extract of the acidic hydrolyzate of **3** by TLC comparison (CHCl₃/MeOH 5:1). Glucose was identified in the residue by comparison on paper chromatography (BuOH/AcOH/H₂O 5:1:5, upper layer) with an authentic sample.

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Received October 8, 2003