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## FOUR NEW PRENYLATED 2-ARYLBENZOFURANS FROM THE ROOT BARK OF *ARTOCARPUS PETELOTII*

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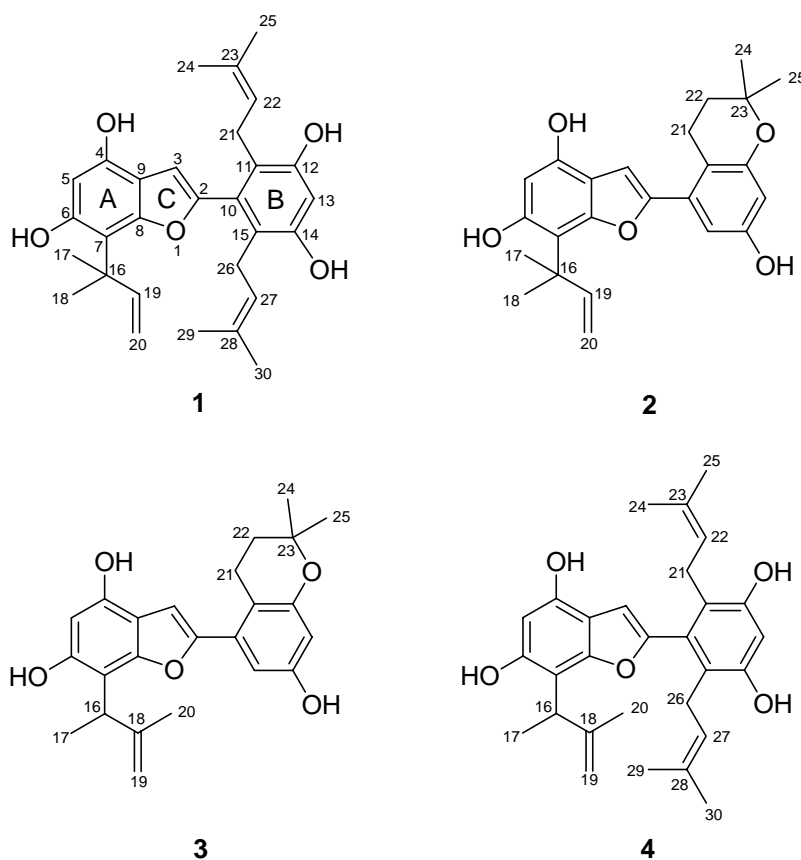
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**Abstract** – Four new prenylated 2-arylbenzofurans, artopetelins H–K (**1–4**), were isolated from the root bark of *Artocarpus petelotii* Gagnep. Their structures were elucidated by extensive 1D-, 2D-NMR, and MS spectral analysis.

### INTRODUCTION

*Artocarpus* species (Moraceae) are evergreen plants distributed over tropical regions of Asia, such as Indonesia, Thailand, and Sri Lanka. Primarily known for their large edible fruits, some members have important medicinal value. Especially in Indonesia, many *Artocarpus* plants are used as traditional folk medicine called “Jamu” against inflammation, malaria, fever, dysentery, and tuberculosis.<sup>1</sup> Previous research work on this genus provided a variety of prenylated flavonoids and some stilbenes and 2-arylbenzofurans with chemical and biological diversity.<sup>2–4</sup> In a program searching for bioactive prenylated phenols from Chinese *Artocarpus* plants,<sup>5–8</sup> we have investigated the chemical constituents of *Artocarpus chama* Buch.-Ham. and *Artocarpus petelotii* Gagnep, both plants being cultivated in Xishuangbanna, Yunnan province, China. It is interesting that their phenolic constituents found so far are different. The roots and stems of *A. chama* are rich in prenylated flavones with cytotoxicity, prenylated stilbenes and their biogenetic derivatives being minor constituents.<sup>5,6</sup> On the other hand, a series of prenylated 2-arylbenzofurans, artopetelins A–G, were isolated from the root bark of *A. petelotii*.<sup>7,8</sup> The limited distribution of 2-arylbenzofurans in *Artocarpus* genus and their interesting bioactivities, such as antibacterial,<sup>9</sup> antiviral,<sup>10</sup> cancer-chemopreventive, and cyclooxygenase-inhibitory effects,<sup>4</sup> have attracted our attention. Therefore, a re-examination of the EtOH extract from the root bark of *A. petelotii* yielded four new prenylated 2-arylbenzofurans, artopetelins H–K (**1–4**). This paper describes the isolation and

structure elucidation of these compounds.



**Figure 1.** Structures of Compounds (1–4)

## RESULTS AND DISCUSSION

Artopetelin H (**1**), a yellow amorphous powder, was assigned a molecular formula of  $C_{29}H_{34}O_5$  by HREIMS ( $m/z$  462.2414 [ $M$ ]<sup>+</sup>). The UV absorption maxima at 207 and 285 nm suggested the presence of a 2-arylbenzofuran skeleton.<sup>7,8,11</sup> The IR spectrum showed absorptions typical of hydroxyl groups ( $3446\text{ cm}^{-1}$ ) and aromatic rings ( $1614$  and  $1450\text{ cm}^{-1}$ ). The  $^1\text{H-NMR}$  spectrum showed signals of four hydroxyl groups [ $\delta_{\text{H}}$  8.41, 7.53 (each 1H, s) and 7.97 (2H, s)], three downfield singlets [ $\delta_{\text{H}}$  6.59, 6.58, and 6.31 (each 1H, s)], two symmetrical 3,3-dimethylallyl (prenyl) side chains [ $\delta_{\text{H}}$  5.09 (2H, br t,  $J = 6.7$  Hz), 3.17 (4H, br d,  $J = 6.7$  Hz), and 1.53, 1.40 (each 6H, br s)], and a 1,1-dimethylallyl group [ $\delta_{\text{H}}$  6.43 (1H, dd,  $J = 10.6, 17.5$  Hz), 5.03 (1H, br d,  $J = 17.5$  Hz), 4.92 (1H, br d,  $J = 10.6$  Hz), and 1.65 (6H, s)]. The  $^{13}\text{C-NMR}$  and HMQC spectra of **1** revealed the presence of twenty-nine carbon signals, corresponding to a triprenylated and tetrahydroxylated 2-arylbenzofuran with a symmetrical moiety on ring B. On the basis of HMQC and HMBC spectral analysis, we were able to fully assign all  $^1\text{H}$ - and  $^{13}\text{C}$ -NMR signals (Tables 1 and 2) and determine the position of the substituents. The HMBC correlations from 21, 26-H<sub>2</sub>

( $\delta_{\text{H}}$  3.17) to C-10 ( $\delta_{\text{C}}$  134.0), C-11, 15 ( $\delta_{\text{C}}$  121.6), and C-12, 14 ( $\delta_{\text{C}}$  154.7), as well as those from 12, 14-OH ( $\delta_{\text{H}}$  7.97) to C-11, 15, C-12, 14, and C-13 ( $\delta_{\text{C}}$  105.1), indicated that the two prenyl and two hydroxyl groups were located at C-11, 15 and C-12, 14, respectively (Figure 2). The HMBC correlations from 17, 18- $\text{H}_3$  ( $\delta_{\text{H}}$  1.65) to C-7 ( $\delta_{\text{C}}$  110.3), from 4-OH ( $\delta_{\text{H}}$  8.41) to C-4 ( $\delta_{\text{C}}$  149.9), C-5 ( $\delta_{\text{C}}$  100.4), and C-9 ( $\delta_{\text{C}}$  113.6), and from 6-OH ( $\delta_{\text{H}}$  7.53) to C-5, C-6 ( $\delta_{\text{C}}$  154.6), and C-7, supported the substitution of ring A. Thus, artopetelin H was elucidated as 7-(1,1-dimethylprop-2-enyl)-2-[3,5-dihydroxy-2,6-bis(3-methylbut-2-enyl)phenyl]-1-benzofuran-4,6-diol (**1**).

**Table 1.**  $^1\text{H-NMR}$  Data for Compounds (**1–4**) in acetone- $d_6$  ( $\delta$  in ppm,  $J$  in Hz)

Proton	<b>1</b> <sup>a)</sup>	<b>2</b> <sup>a)</sup>	<b>3</b> <sup>b)</sup>	<b>4</b> <sup>a)</sup>
3	6.59 (s)	6.98 (s)	6.98 (s)	6.56 (s)
5	6.31 (s)	6.35 (s)	6.38 (s)	6.34 (s)
13	6.58 (s)	6.28 (d, $J = 2.5$ )	6.28 (d, $J = 2.5$ )	6.57 (s)
15		6.97 (d, $J = 2.5$ )	6.95 (d, $J = 2.5$ )	
16			4.07 (q, $J = 7.2$ )	4.00 (q, $J = 7.4$ )
17	1.65 (s)	1.72 (s)	1.58 (d, $J = 7.2$ )	1.50 (d, $J = 7.4$ )
18	1.65 (s)	1.72 (s)		
19a			4.99 (br s)	4.82 (br s)
19b			4.84 (br s)	4.73 (br s)
19	6.43 (dd, $J = 10.6, 17.5$ )	6.48 (dd, $J = 10.6, 17.5$ )		
20a	5.03 (br d, $J = 17.5$ )	5.06 (dd, $J = 1.4, 17.5$ )		
20b	4.92 (br d, $J = 10.6$ )	4.96 (dd, $J = 1.4, 10.6$ )		
20			1.69 (br s)	1.65 (br s)
21	3.17 (br d, $J = 6.7$ )	2.89 (t, $J = 6.8$ )	2.88–2.92 (m)	3.13 (br d, $J = 6.5$ )
22	5.09 (br t, $J = 6.7$ )	1.85 (t, $J = 6.8$ )	1.84 (t, $J = 6.7$ )	5.05 (br t, $J = 6.5$ )
24	1.40 (br s)	1.34 (s)	1.33 (s)	1.36 (br s)
25	1.53 (br s)	1.34 (s)	1.33 (s)	1.51 (br s)
26	3.17 (br d, $J = 6.7$ )			3.13 (br d, $J = 6.5$ )
27	5.09 (br t, $J = 6.7$ )			5.05 (br t, $J = 6.5$ )
29	1.40 (br s)			1.36 (br s)
30	1.53 (br s)			1.51 (br s)
4-OH	8.41 (s)			8.93 (s)
6-OH	7.53 (s)			8.35 (s)
12-OH	7.97 (s)			8.49 (s)
14-OH	7.97 (s)			8.49 (s)

<sup>a)</sup> At 400 MHz. <sup>b)</sup> At 500 MHz.

Artopetelin I (**2**), a yellow amorphous powder, was assigned a molecular formula of  $\text{C}_{24}\text{H}_{26}\text{O}_5$  by HREIMS ( $m/z$  394.1796 [ $M$ ]<sup>+</sup>). The UV and IR data resembled those of 2-arylbenzofuran derivatives. The  $^1\text{H-NMR}$  spectrum showed signals of a 1,1-dimethylallyl group [ $\delta_{\text{H}}$  6.48 (1H, dd,  $J = 10.6, 17.5$  Hz), 5.06 (1H, dd,  $J = 1.4, 17.5$  Hz), 4.96 (1H, dd,  $J = 1.4, 10.6$  Hz), and 1.72 (6H, s)], two *meta*-coupled aromatic protons [ $\delta_{\text{H}}$  6.97 and 6.28 (each 1H, d,  $J = 2.5$  Hz)], two downfield singlets [ $\delta_{\text{H}}$  6.98 and 6.35

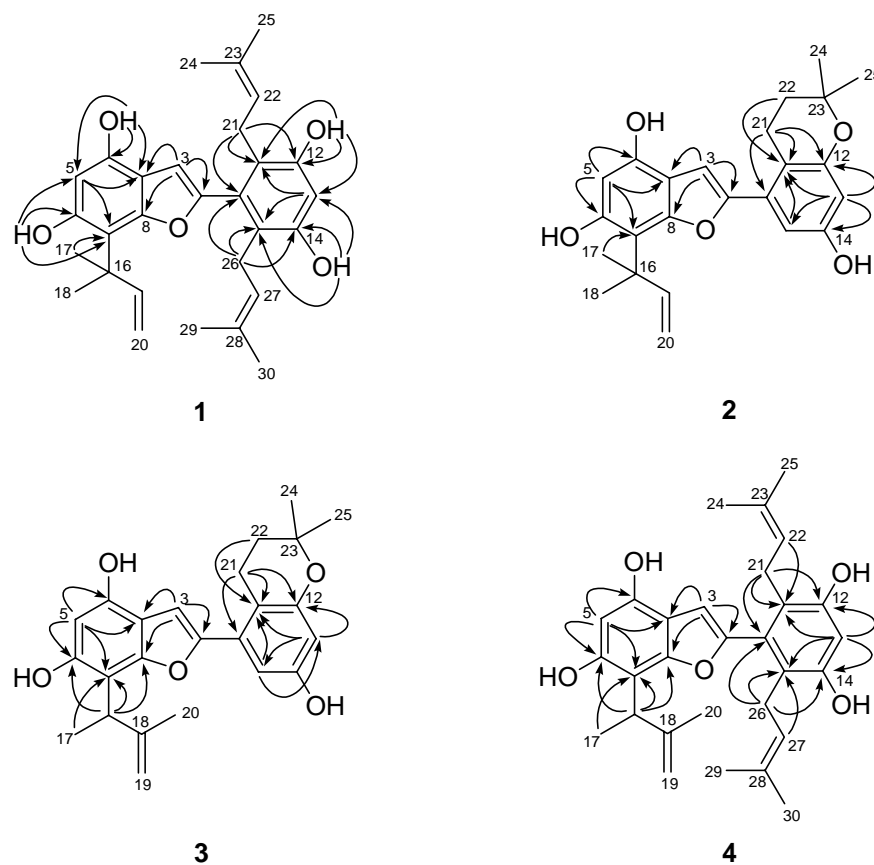
(each 1H, s)], and a 3,4-dihydro-2,2-dimethylpyran ring [ $\delta_{\text{H}}$  2.89, 1.85 (each 2H, t,  $J = 6.8$  Hz) and 1.34 (6H, s)]. Comparison of the NMR spectroscopic data of **2** and **1** (Tables 1 and 2) indicated that they should have the same moiety of rings A and C. However, **2** contained a 3,4-dihydro-2,2-dimethylpyran ring attached at ring B rather than two symmetrical prenyl groups in **1**. This pyranoid moiety was fused at C-11 and C-12, as established by the HMBC correlations from 21-H<sub>2</sub> ( $\delta_{\text{H}}$  2.89) to C-10 ( $\delta_{\text{C}}$  132.5), C-11 ( $\delta_{\text{C}}$  110.6), and C-12 ( $\delta_{\text{C}}$  156.5) and from 22-H<sub>2</sub> ( $\delta_{\text{H}}$  1.85) to C-11. The two *meta*-coupled aromatic protons ( $\delta_{\text{H}}$  6.97 and 6.28) were assigned to C-15 and C-13, respectively, supported by the HMBC correlations shown in Figure 2. Thus, the structure of artopetelin I was elucidated as 7-(1,1-dimethylprop-2-enyl)-2-(3,4-dihydro-7-hydroxy-2,2-dimethyl-2H-1-benzopyran-5-yl)-1-benzofuran-4,6-diol (**2**).

**Table 2.** <sup>13</sup>C-NMR Data for Compounds (**1–4**) in acetone-*d*<sub>6</sub> ( $\delta$  in ppm)

Carbon	<b>1</b> <sup>a)</sup>	<b>2</b> <sup>a)</sup>	<b>3</b> <sup>b)</sup>	<b>4</b> <sup>a)</sup>
2	152.2	152.6	152.9	152.2
3	103.9	103.9	103.6	104.0
4	149.9	150.2	149.9	149.3
5	100.4	100.4	98.7	98.8
6	154.6	155.61 <sup>c)</sup>	154.3	153.5
7	110.3	110.0	107.1	107.3
8	156.5	155.64 <sup>c)</sup>	155.8	156.5
9	113.6	113.7	112.7	112.4
10	134.0	132.5	132.3	133.5
11	121.6	110.6	110.4	121.2
12	154.7	156.5	156.2	154.4
13	105.1	104.8	104.5	104.6
14	154.7	157.6	157.2	154.4
15	121.6	108.0	107.6	121.2
16	41.6	41.5	36.4	36.7
17	29.6	29.4	18.0	17.9
18	29.6	29.4	149.4	149.5
19	150.5	150.5	109.2	109.4
20	110.1	109.7	22.48 <sup>d)</sup>	22.4
21	27.4	22.9	22.54 <sup>d)</sup>	27.0
22	125.8	34.0	33.6	125.4
23	130.2	74.4	74.1	129.9
24	18.2	27.2	26.8 <sup>e)</sup>	17.8
25	26.2	27.2	26.9 <sup>e)</sup>	25.8
26	27.4			27.0
27	125.8			125.4
28	130.2			129.9
29	18.2			17.8
30	26.2			25.8

<sup>a)</sup> At 100 MHz. <sup>b)</sup> At 125 MHz. <sup>c)</sup>, <sup>d)</sup>, <sup>e)</sup> Signals may be exchangeable.

Artopetelin J (**3**), a yellow amorphous powder, was assigned a molecular formula of  $C_{24}H_{26}O_5$  by HREIMS ( $m/z$  394.1770  $[M]^+$ ). The NMR data of **3** resembled those of **2** (Tables 1 and 2), except for signals due to a 1,2-dimethylallyl group:  $\delta_H$  4.99 and 4.84 (each 1H, br s, 19- $H_2$ ), 4.07 (1H, q,  $J = 7.2$  Hz, 16-H), 1.69 (3H, br s, 20- $H_3$ ), and 1.58 (3H, d,  $J = 7.2$  Hz, 17- $H_3$ );  $\delta_C$  36.4 (C-16), 18.0 (C-17), 149.4 (C-18), 109.2 (C-19), and 22.48 (C-20). The HMBC correlations from 16-H to C-6 ( $\delta_C$  154.3), C-7 ( $\delta_C$  107.1), and C-8 ( $\delta_C$  155.8) and from 17- $H_3$  to C-7 showed that the 1,2-dimethylallyl group was located at C-7 (Figure 2). Thus, the planar structure of artopetelin J was elucidated as 7-(1,2-dimethylprop-2-enyl)-2-(3,4-dihydro-7-hydroxy-2,2-dimethyl-2H-1-benzopyran-5-yl)-1-benzofuran-4,6-diol (**3**). Similarly to artopetelins D and E,<sup>8</sup> the configuration at C-16 remains to be determined.



**Figure. 2** Key HMBC (H  $\rightarrow$  C) correlations of Compounds (**1–4**)

Artopetelin K (**4**), a yellow amorphous powder, was assigned a molecular formula of  $C_{29}H_{34}O_5$  by HREIMS ( $m/z$  462.2416  $[M]^+$ ). The NMR characteristics of **4** (Tables 1 and 2) showed that it was a 2-arylbenzofuran with the same symmetrical substituents on ring B as **1** and the same moiety of ring A as **3**. This result was corroborated by the HMBC correlations shown in Figure 2. Thus, the planar structure of artopetelin K was elucidated as 7-(1,2-dimethylprop-2-enyl)-2-[3,5-dihydroxy-2,6-bis(3-methylbut-2-enyl)phenyl]-1-benzofuran-4,6-diol (**4**). The configuration at C-16 remains to be determined.

## EXPERIMENTAL

**General Experimental Procedures.** Melting points were measured with an XT-4 micro-melting point apparatus and are uncorrected. Optical rotations were measured with a JASCO P1030 polarimeter. UV spectra were obtained with a Shimadzu UV-2401PC spectrophotometer. IR spectroscopic data were recorded with a Nicolet Avatar 360 spectrometer with KBr pellets. EIMS and HREIMS data were recorded with a Finnigan MAT-95 mass spectrometer. NMR spectra were obtained with a Bruker DRX-400 and -500 instruments. Chemical shifts are reported with respect to acetone- $d_6$  ( $\delta_H = 2.04$ ,  $\delta_C = 206.0$  ppm). Column chromatography was performed on silica gel H (10–40  $\mu\text{m}$  and 200–300 mesh; Yantai Institute of Chemical Technology, China), Chromatorex PR-18 gel (20–45  $\mu\text{m}$ ; Fuji Silysia Chemical, Ltd., Kasugai, Japan), MCI gel CHP-20P (75–150  $\mu\text{m}$ ; Mitsubishi Chemical Corporation, Japan), Toyopearl HW-40C (Tosoh Corporation, Japan), and Sephadex LH-20 (Amersham Biosciences, GE Health Care). Preparative and analytical TLC was run on precoated silica gel GF<sub>254</sub> plates (10–40  $\mu\text{m}$ ; Yantai Institute of Chemical Technology, China).

**Plant Material.** The root bark of *A. petelotii* Gagnep was collected in Xishuangbanna, Yunnan, P. R. China, in July 1998, and air-dried. The plant was identified by Prof. Han-Dong Sun, Kunming Institute of Botany, and a voucher specimen (TCM 98-07-02 Hou) was deposited in the Herbarium of the Department of Pharmacognosy, School of Pharmacy, Fudan University.

**Extraction and Isolation.** The air-dried and powdered root bark (6.4 kg) of *A. petelotii* was percolated with 95% EtOH (60 L) at rt. The filtrate was concentrated to give a residue (800 g), which was suspended in H<sub>2</sub>O (2 L) and partitioned successively with petroleum ether (4×800 mL) and EtOAc (4×800 mL). The EtOAc extract (110 g) was subjected to column chromatograph (CC) on silica gel [petroleum ether-Me<sub>2</sub>CO (8 : 2 → 2 : 8)] to yield fractions A–I. Fraction F (9.5 g) was chromatographed over silica gel [CHCl<sub>3</sub>-EtOAc (30 : 1 → 1 : 2)] to give fractions F1–F6. Fraction F3 was separated by CC on silica gel [CHCl<sub>3</sub>-Me<sub>2</sub>CO (40 : 1)] to provide fractions F3.1–F3.6. Fraction F3.5 was subjected to CC on RP-18 [MeOH-H<sub>2</sub>O (6.5 : 3.5)] and MCI gel CHP-20P [MeOH-H<sub>2</sub>O (6.5 : 3.5)], followed by Sephadex LH-20 [CHCl<sub>3</sub>-MeOH (1:1)] to yield **4** (15 mg). Fraction F3.6 was separated by CC on RP-18 [MeOH-H<sub>2</sub>O (5.5 : 4.5)] and MCI gel CHP-20P [MeOH-H<sub>2</sub>O (6.5 : 3.5)] to give fractions F3.6.1–F3.6.4. Fraction F3.6.2 was further purified by preparative TLC [CHCl<sub>3</sub>-2-propanol (30 : 1)] to give **1** (3 mg). Fraction F3.6.3 was separated by preparative TLC [CHCl<sub>3</sub>-2-propanol (20 : 1)] to afford **2** (5 mg). Fraction F4 was isolated by CC on RP-18 [MeOH-H<sub>2</sub>O (6.5 : 3.5)] and Toyopearl HW-40C [MeOH-H<sub>2</sub>O (7.5 : 2.5)], followed by Sephadex LH-20 [CHCl<sub>3</sub>-MeOH (1 : 1)] to yield **3** (30 mg).

**Artopetelin H (1):** yellow amorphous powder, mp 138–140 °C; UV (MeOH)  $\lambda_{\text{max}}$  (log  $\epsilon$ ): 285 (3.86), 207 (4.47) nm; IR (KBr)  $\nu_{\text{max}}$ : 3446, 2924, 2853, 1614, 1450, 1145, 1087  $\text{cm}^{-1}$ ; EIMS  $m/z$  (*rel. int.* %): 462 ( $M^+$ , 100), 445 (11), 419 (26), 407 (12), 391 (12), 363 (24), 351 (17), 253 (15), 213 (12); HREIMS:

$m/z$  462.2414  $[M]^+$  (calcd for  $C_{29}H_{34}O_5$ : 462.2406);  $^1H$ -NMR and  $^{13}C$ -NMR data: shown in Tables 1 and 2, respectively.

**Artopetelin I (2)**: yellow amorphous powder, mp 146–148 °C; UV (MeOH)  $\lambda_{max}$  (log  $\epsilon$ ): 316 (4.15), 218 (4.26) nm; IR (KBr)  $\nu_{max}$ : 3423, 2971, 2925, 1614, 1445, 1421, 1368, 1327, 1279, 1152, 1119, 1055, 1023, 966  $cm^{-1}$ ; EIMS  $m/z$  (*rel. int.* %): 394 ( $M^+$ , 100), 379 (38), 339 (13), 108 (15), 101 (15), 90 (33); HREIMS:  $m/z$  394.1796  $[M]^+$  (calcd for  $C_{24}H_{26}O_5$ : 394.1780);  $^1H$ -NMR and  $^{13}C$ -NMR data: shown in Tables 1 and 2, respectively.

**Artopetelin J (3)**: yellow amorphous powder, mp 158–160 °C;  $[\alpha]_D^{20} +1.4^\circ$  ( $c$  0.38, acetone); UV (MeOH)  $\lambda_{max}$  (log  $\epsilon$ ): 314 (4.17), 217 (4.24) nm; IR (KBr)  $\nu_{max}$ : 3386, 2972, 2925, 1616, 1444, 1372, 1284, 1153, 1120, 1023  $cm^{-1}$ ; EIMS  $m/z$  (*rel. int.* %): 394 ( $M^+$ , 100), 379 (9), 339 (17), 338 (9), 323 (6); HREIMS:  $m/z$  394.1770  $[M]^+$  (calcd for  $C_{24}H_{26}O_5$ : 394.1780);  $^1H$ -NMR and  $^{13}C$ -NMR data: shown in Tables 1 and 2, respectively.

**Artopetelin K (4)**: yellow amorphous powder, mp 119–120 °C;  $[\alpha]_D^{20} +1.2^\circ$  ( $c$  0.34, acetone); UV (MeOH)  $\lambda_{max}$  (log  $\epsilon$ ): 288 (3.96), 207 (4.55) nm; IR (KBr)  $\nu_{max}$ : 3441, 2965, 2924, 1699, 1616, 1435, 1375, 1262, 1160, 1132, 1086, 1032  $cm^{-1}$ ; EIMS  $m/z$  (*rel. int.* %): 462 ( $M^+$ , 100), 419 (26), 391 (11), 363 (13), 337 (12), 253 (20), 225 (10), 207 (21); HREIMS:  $m/z$  462.2416  $[M]^+$  (calcd for  $C_{29}H_{34}O_5$ : 462.2406);  $^1H$ -NMR and  $^{13}C$ -NMR data: shown in Tables 1 and 2, respectively.

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