

# Notes

## New Cembranoid Lactones from the Indian Ocean Soft Coral *Sinularia flexibilis*

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A detailed chemical examination of the Indian Ocean soft coral *Sinularia flexibilis* yielded two new cembranolid  $\epsilon$ -lactones, sandensolide monoacetate (**1**) and flexibolide (**4**), in addition to the previously reported cembranolid  $\delta$ -lactones, flexibilolide and dihydroflexibilolide, and the known cembranoid derivatives, sandensolide, cembrene A, and flexibile. Their structures have been established by a study of their spectral data (UV, IR,  $^1\text{H}$  and  $^{13}\text{C}$  NMR, 2D NMR ( $^1\text{H}$ - $^1\text{H}$ , COSY and NOESY), and mass spectrometry.

Soft corals (Coelenterata, Octocorallia, Alcyonaceae) are a rich source of terpenoids, polyhydroxysteroids, and other compounds. Soft corals of the genus *Sinularia* are prolific, with 90 known species of which about 35 have been chemically examined.<sup>1,2</sup> *Sinularia flexibilis* (Alcyoniidae) occurs in different seas, and several collections have been chemically examined by workers who have reported the isolation of a range of cembranoid diterpenes.<sup>3–9</sup>

In our continuing search for bioactive secondary metabolites from soft corals of the Indian Ocean,<sup>10–18</sup> we examined *Sinularia flexibilis* collected from Diglipur Island of the Andaman and Nicobar group of islands of the Indian Ocean and reported the isolation of the two new cembranolid  $\delta$ -lactones flexibilolide and dihydroflexibilolide from its EtOAc extract.<sup>18</sup> A detailed further examination of the same extract furnished the five additional compounds A–G, two of which are new.

Compounds A and B were identified as cembrene A and flexibile,<sup>3,7</sup> respectively, by a comparison of their physical and spectral data with those reported.

Compound C analyzed for  $\text{C}_{22}\text{H}_{32}\text{O}_5$  and had a peak at  $m/z$  316 ( $\text{M}^+ - \text{AcOH}$ ) in its mass spectrum. It was recognised as the new cembranolid lactone (**1**) by a close examination of its physical and spectral data (Table 1), which were very similar to those of sandensolide (**2**).<sup>12</sup> It was a monoacetate with one hydroxyl group ( $\nu_{\text{max}}$  3600  $\text{cm}^{-1}$ ) and an acetate ( $\nu_{\text{max}}$  1720, 1250  $\text{cm}^{-1}$ ), while sandensolide is a dihydroxy derivative ( $\nu_{\text{max}}$  3680, 3610  $\text{cm}^{-1}$ ). This difference was also evident from their  $^1\text{H}$  NMR spectra, where sandensolide showed two carbinol methine protons at  $\delta$  3.80 (d,  $J = 15$  Hz) and 4.20 (dd,  $J = 13, 6$  Hz), while compound C showed a hydroxymethine proton at  $\delta$  3.80 and an acetoxy methine proton at  $\delta$  5.56 (dd,  $J = 13, 6$  Hz). The relationship between the two compounds was confirmed by acetylation of compound C to give sandensolide diacetate (**3**), identical with material obtained by acetylation of san-

densolide (**2**). Of the two carbinol methine protons in sandensolide, it was the downfield allylic proton at  $\delta$  4.20 that moved to 5.52 in compound C, indicating that C is the 13-acetate of sandensolide (**1**). This finding was also supported by the resonance of C-13 in sandensolide at 76.8 (d) and in sandensolide acetate at 78.6 (d).

The structure of C was further supported by its  $^1\text{H}$ - $^1\text{H}$  COSY and NOESY connectivities. The 5-H carbinol methine proton at  $\delta$  3.80 showed COSY connectivity with only one of the two neighboring methylene protons appearing at  $\delta$  1.38 (with no coupling to the other methylene proton, dihedral angle  $90^\circ$ ) as in sandensolide. The 13-H acetoxy methine proton at 5.56 showed allylic coupling with 20-H<sub>3</sub> on the trisubstituted double bond. The same 13-H showed  $^1\text{H}$ - $^1\text{H}$  COSY and NOESY connectivity with 14-H<sub>2</sub>. Similarly 20-H<sub>3</sub> showed COSY and NOESY connectivity with 11-H, which in turn showed connectivity to 10 $\beta$ -H at 3.14, and 10 $\beta$ -H showed connectivity to 10 $\alpha$ -H and vice versa. Because the structure of sandensolide was established by extensive spectral data, including X-ray analysis, the structure of compound C was established as (1*S*,4*S*,5*R*,13*R*,8*E*,11*E*)-5-hydroxy-13-acetoxy-cembra-8,11,15-trien-16,4-olide (**1**).

Compound D was identified as sandensolide (**2**), the cembranolid  $\delta$ -lactone recently reported from our laboratory from another soft coral species, *Simularia sandensis* Versveldt.<sup>12</sup>

Compound E,  $\text{C}_{20}\text{H}_{30}\text{O}_5$   $m/z$  332 ( $\text{M}^+ - \text{H}_2\text{O}$ ) was considered from its physical and spectral characteristics as a new seven-membered cembranolid  $\epsilon$ -lactone and was designated flexibolide (**4**). It exhibited IR absorptions for two hydroxyls (3400 and 3360  $\text{cm}^{-1}$ ), olefinic unsaturation at 1640 and 900  $\text{cm}^{-1}$ , and an  $\alpha,\beta$ -unsaturated  $\epsilon$ -lactone (1720  $\text{cm}^{-1}$ ); the presence of the latter group was supported by its UV absorption at 217 nm. Its  $^1\text{H}$ -NMR spectrum, taken in  $d_5$ -pyridine (Table 2), was reminiscent of a cembranoid lactone and showed two singlet methyls at  $\delta$  1.24 and 1.46 connected to oxygenated carbons. One of these, together with the exomethylene protons at  $\delta$  5.54 and 6.42 (each 1H, s) is

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**Table 1.** <sup>1</sup>H NMR (400 MHz) and <sup>13</sup>C NMR (100 MHz) Data for Compounds **1** and **2** (CDCl<sub>3</sub>, δ in ppm, *J* in Hz)

position	δ <sub>H</sub>		δ <sub>C</sub> <sup>a</sup>	
	<b>1</b>	<b>2</b>	<b>1</b>	<b>2</b>
1	2.15 (m)	2.15 (m)	35.5, D	35.6, D
2	2.61 (m)	2.68 (m)	32.0, T	31.9, T
3	2.18 (dd, <i>J</i> = 13, 10, αH)	2.19 (dd, <i>J</i> = 13, 10, αH)	33.4, T	33.6, T
	1.96 (dd, <i>J</i> = 13, 10, βH)	1.98 (dd, <i>J</i> = 13, 10, βH)		
4			86.9, S	86.7, S
5	3.80 (d, <i>J</i> = 15)	3.80 (d, <i>J</i> = 15)	67.3, D	67.2, D
6	1.88 (dd, <i>J</i> = 15, 8, αH)	1.88 (dd, <i>J</i> = 15, 8, αH)	26.9, T	26.4, T
	1.38 (dd, <i>J</i> = 15, 8, βH)	1.38 (dd, <i>J</i> = 15, 8, βH)		
7	2.31 (m)	2.32 (m)	34.9, T	34.8, T
8			134.5, S	134.0, S
9	5.38 (t, <i>J</i> = 9)	5.37 (t, <i>J</i> = 9)	124.2, D	124.2, D
10	2.45 (q, <i>J</i> = 14, αH)	2.46 (q, <i>J</i> = 14, αH)	29.1, T	29.0, T
	3.13 (q, <i>J</i> = 15, βH)	3.14 (q, <i>J</i> = 15, βH)		
11	5.32 (t, <i>J</i> = 9)	5.37 (t, <i>J</i> = 9)	128.8, D	128.0, D
12			132.0, S	132.8, S
13	5.56 (dd, <i>J</i> = 13, 6)	4.20 (dd, <i>J</i> = 13, 6)	78.6, D	76.8, D
14	2.22 (m)	2.23 (m)	26.8, T	26.9, T
15			144.6, S	144.8, S
16			169.5, S	169.5, S
17	5.48 (s, Hα)	5.53 (s, Hα)	124.5, T	124.8, T
	6.27 (s, Hβ)	6.27 (s, Hβ)		
18	1.30 (s)	1.30 (s)	22.7, Q	22.8, Q
19	1.54 (s)	1.54 (s)	14.9, Q	15.0, Q
20	1.65 (s)	1.66 (q)	10.2, Q	9.8, Q

<sup>a</sup> Assignment of carbon types (S = C, D = CH, T = CH<sub>2</sub>, Q = CH<sub>3</sub>) was made by DEPT.

**Table 2.** <sup>1</sup>H-NMR (400 MHz) and <sup>13</sup>C-NMR (100 MHz) Data for Compounds **4–6** (δ in ppm, *J* in Hz)

position	δ <sub>H</sub>			δ <sub>C</sub> <sup>a</sup>		
	<b>4</b> <sup>b</sup>	<b>5</b> <sup>c</sup>	<b>6</b> <sup>c</sup>	<b>4</b> <sup>b</sup>	<b>5</b> <sup>c</sup>	<b>6</b> <sup>c</sup>
1	2.12 (m)	2.1 (m)		39.2, d	34.7, d	35.7, d
2	1.98 (m) 1.97 (m)	2.2–2.5 (m)		33.3, t	33.2, t	33.6, t
3	2.54 (m) 2.5 (m)	2.2–2.5 (m)		33.7, t	33.6, t	34.0, t
4				87.4, s	88.2, s	88.2, s
5	4.48 (dd, <i>J</i> = 10, 6.5)	4.36 (dd, <i>J</i> = 10, 6.5)	4.37 (dd, <i>J</i> = 8.4, 3.6)	85.2, d	83.8, d	83.4, d
6	2.98 (m) 2.96 (m)			30.0, t	29.8, t	30.4, t
7	3.10 (dd, <i>J</i> = 13, 6.5)	3.08 (dd, <i>J</i> = 13, 6.5)		34.6, t	34.9, t	34.7, t
8				85.1, s	85.4, s	86.4, s
9	3.82 (d, <i>J</i> = 13)	5.23 (d, <i>J</i> = 13)	5.17 (d, <i>J</i> = 11)	77.8, d	77.7, d	77.9, d
10	2.56 (dd, <i>J</i> = 12, 6)	2.55 (dd, <i>J</i> = 12, 6)		38.0, t	36.6, t	27.3, t
	2.11 (dd, <i>J</i> = 12, 6)	2.10 (dd, <i>J</i> = 12, 6)				
11	5.77 (m)	5.63 (m)		128.0, d	126.2, d	34.3, t
12				134.8, s	132.8, s	59.9, s
13	4.20 (dd, <i>J</i> = 13, 6.5)	4.92 (dd, <i>J</i> = 12.5, 6)	3.40 (dd, <i>J</i> = 10, 6.4)	76.8, d	78.3, d	60.8, d
14	1.92 (m)	1.92 (m)		28.0, t	27.5, t	26.7, t
15				146.8, s	145.5, s	144.4, s
16				170.1, s	170.2, s	169.5, s
17	5.54 (s)	5.45 (s)	5.45 (s)			
	6.42 (s)	6.21 (s)	6.26 (s)	123.8, t	123.6, t	123.6, t
18	1.46 (s)	1.26 (s)	1.24 (s)	19.4, q	16.7, q	16.4, q
19	1.24 (s)	1.23 (s)	1.24 (s)	30.2, q	29.5, q	29.3, q
20	1.74 (s)	1.64 (s)	1.16 (s)	11.6, q	11.4, q	17.1, q
–OCOCH <sub>3</sub>		2.02 (s)	2.06 (s)		21.1, q	21.1, q
–OCOCH <sub>3</sub>					171.4, s	171.2, s
–OCOCH <sub>3</sub>		2.08 (s)			21.2, q	
–OCOCH <sub>3</sub>					170.2, s	

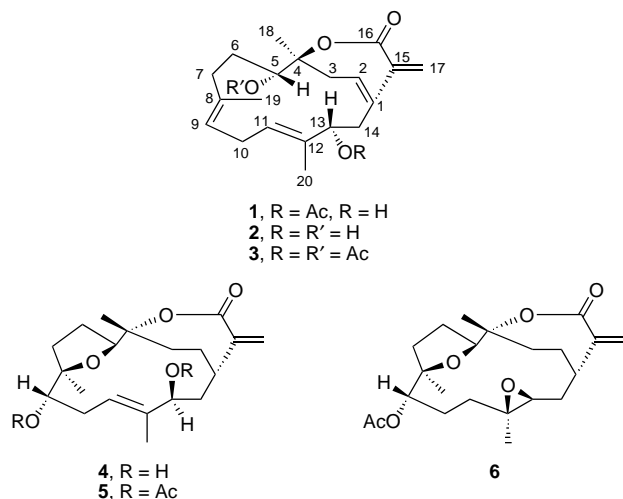
<sup>a</sup> Assignment of carbon types (S = C, D = CH, T = CH<sub>2</sub>, Q = CH<sub>3</sub>) was made by DEPT. <sup>b</sup> Spectra were recorded in *d*<sub>5</sub>-pyridine. <sup>c</sup> Spectra were recorded in CDCl<sub>3</sub>.

characteristic of a seven-membered α,β-unsaturated α-methylene lactone,<sup>3–7</sup> as found in compounds C and D. A methyl group on a trisubstituted double bond at δ 1.74 (s) was evident, with the corresponding olefinic proton appearing at δ 5.77 (m). The compound also exhibited three carbinol methine protons at δ 3.82 (d, *J* = 13 Hz), 4.20 (dd, *J* = 13, 6.5 Hz), and 4.48 (dd, *J* = 10, 6.5 Hz), and it formed a diacetate (**5**) in which two of the above carbinolic methine protons shifted downfield from 3.82 to 5.23 and from 4.20 to 5.52 ppm, supporting their secondary nature. The unchanged third proton connected to an oxygenated carbon suggests the presence of a hindered secondary hydroxyl group

or an ether bridge involving this oxygen. In the absence of an epoxide this could be an ether bridge as in 5,8-epoxycembranolide<sup>5</sup> isolated earlier from the same species. Consistently, the double-bond equivalence also requires the molecule to be tricyclic.

The <sup>13</sup>C NMR spectrum (Table 2) coupled with the DEPT spectrum supported the presence of all the functionalities, five oxygenated carbons [δ 87.4 (s), 85.2 (d), 85.1 (s), 77.8 (d), and 76.8 (d)], four olefinic carbons [128.0 (d), 134.8 (s), 146.8 (s), 123.8 (t)], and a lactone carbonyl (170.1, s). The spectral characteristics of flexibolide compared very well with those of 5,8-epoxycembranolide<sup>5</sup> (**6**) except for expected differences due to

their different functionalities. A comparison of the  $^1\text{H}$ - and  $^{13}\text{C}$ -NMR spectral data (Table 2) of flexibolide (**4**) and its diacetate (**5**) with those of the **6** from C1 to C9 and C15 to C18 and with those of sandensolide (**2**) from C10 to C14 confirmed the structural assignment as 5,8-epoxy-9,13-dihydroxycembra-11,15-dien-16,4-olide and suggested the stereochemical assignment as (1*R*, 4*R*, 5*S*, 8*R*, 9*S*, 13*S*, 11*E*). The stereochemistry of the hydroxyl group at C13 was tentatively assigned as (*S*) on assumed stereospecific ring opening of the 12,13 $\beta$ -epoxide ring.



Compounds F and G were identified as the two recently reported<sup>18</sup> trihydroxycembranolide  $\delta$ -lactones, flexibilolide and dihydroflexibilolide, respectively.

## Experimental Section

**General Experimental Procedures.** Elemental analyses were determined on a Carlo-Erba 1108 instrument. UV spectra were recorded on a Milton Roy Spectronic 1201 instrument. IR spectra were recorded on a Perkin-Elmer 841 spectrophotometer.  $^1\text{H}$ - and  $^{13}\text{C}$ -NMR spectra were measured on Bruker 400 MHz and JEOL JNM EX-90 spectrometers, using  $\text{CDCl}_3$  and  $d_5$ -pyridine as solvents and TMS as internal standard. MS were obtained on a JEOL JMS-300 spectrometer. Mps were determined on VEB-analytic Dreder HMK hot plate and are uncorrected. Optical rotations were determined on a Perkin-Elmer model 141 polarimeter.

**Animal Material.** The soft coral *Simularia flexibilis* was collected in April 1992, at Diglipur Island (91.31°E, 10.31°N) of the Andaman and Nicobar group of islands of the Indian Ocean. Voucher specimens are preserved at the Department of Organic Chemistry, Andhra University, and the Museum of (NIO), Goa.

**Extraction and Isolation.** Slices of the soft coral (ca. 10 kg wet wt) were soaked in EtOH (10 L) at room temperature, and the extract was decanted carefully. This process was repeated eight times. The combined MeOH extract was concentrated to a small volume and fractionated into EtOAc. The residue from the EtOAc extract (30 g) was chromatographed over a Si gel column (300 g, 100–200 mesh) using solvents of increasing polarity from hexane to EtOAc. The selected fractions were further purified by passing over Si gel columns or recrystallization from appropriate solvents to yield seven pure compounds (A–G).

**Compound A:** colorless oil; 30 mg (0.0015%) [ $\alpha$ ] $^{27}_D$   $-3.5^\circ$  (*c* 0.35,  $\text{CHCl}_3$ ) identified as (3*E*,7*E*,11*E*)-cembra-3,7,11,15-tetraene (cembrene A) by comparison of its  $^1\text{H}$  NMR data with literature data.<sup>7,9</sup>

**Compound B:** colorless oil; 20 mg (0.001%), identified as flexibilene by comparison of its  $^1\text{H}$  NMR spectral data with literature data.<sup>3,7</sup>

**Compound C:** (1*S*,4*S*,5*R*,13*R*,11*E*)-5-hydroxy-13-acetoxycembra-8,11,15-trien-16,4-olide (sandensolide-13-acetate, **1**); colorless needles ( $\text{Me}_2\text{CO}$ -hexane); 30 mg (0.0015%); mp 163–165 °C; [ $\alpha$ ] $^{27}_D$   $+36.5^\circ$  (*c* 0.85,  $\text{CHCl}_3$ ); Anal. Calcd for  $\text{C}_{22}\text{H}_{32}\text{O}_5$ : C, 70.21, H, 8.51. Found: C, 70.11; H, 8.16;  $\lambda_{\text{max}}$  ( $\text{CHCl}_3$ ) 217 nm;  $\nu_{\text{max}}$  ( $\text{CHCl}_3$ ) 3600 (OH), 2980, 1720, 1665, 1360, 1250, 1130, and 910  $\text{cm}^{-1}$ ;  $^1\text{H}$ - and  $^{13}\text{C}$ -NMR data (see Table 1); EIMS (70eV) *m/z* 316 ( $\text{M}^+ - \text{AcOH}$ , 30.1), 301 (316 –  $\text{CH}_3$ , 14.5), 298 (316 –  $\text{H}_2\text{O}$ , 32.4), 218 (11.1), 190 (10.1), 160 (20.9), 108 (60.5), and 93 (80).

**Acetylation of Compound C: Sandensolide Diacetate (3).** To compound C (10mg) in pyridine (0.5 mL) was added  $\text{Ac}_2\text{O}$  (0.5 mL), and the mixture was allowed to stand at room temperature for 12 h. The diacetate obtained after usual workup crystallized from  $\text{Me}_2\text{CO}$ -hexane as colorless needles, 8 mg;  $\text{M}^+$  418; mp 148–150 °C; [ $\alpha$ ] $^{27}_D$   $+45.5^\circ$  (*c* 0.95,  $\text{CHCl}_3$ ); Anal. Calcd for  $\text{C}_{24}\text{H}_{34}\text{O}_6$ : C, 68.9; H, 8.13. Found: C, 68.55; H, 7.95;  $\lambda_{\text{max}}$  ( $\text{CHCl}_3$ ) 217 nm;  $\nu_{\text{max}}$  ( $\text{CHCl}_3$ ) 2980, 1720, and 1250 (OAc), 1720, 1440, 1190, 1140, and 930  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{CDCl}_3$ , 90 MHz)  $\delta$  1.32, 1.54, 1.63, 2.02, 2.10 (each 3H, s), 2.50 (1H, m), 3.16 (1H, q, *J* = 15 Hz), 5.10 to 5.40 (3H, m), 5.52 (1H, m), 5.43, and 6.28 (each 1H, s).

**Compound D:** colorless needles, 850 mg (0.0425%); mp 188–190 °C; [ $\alpha$ ] $^{27}_D$   $+55.5^\circ$  (*c* 0.90,  $\text{CHCl}_3$ ) identified as (1*S*,4*S*,5*R*,13*R*,8*E*,11*E*)-5,13-dihydroxycembra-8,11,15-trien-16,4-olide (sandensolide, **2**) by comparison of its  $^1\text{H}$ - and  $^{13}\text{C}$ -NMR spectral data with literature data.<sup>12</sup>

**Acetylation of Compound D: Sandensolide Diacetate (3).** Compound D (10 mg) on acetylation with  $\text{Py}-\text{Ac}_2\text{O}$ , as described above, furnished sandensolide diacetate identical in every respect with the acetate obtained from compound C.

**Compound E. (1*R*,4*R*,5*S*,8*R*,9*S*,13*R*,11*E*)-5,8-Epoxy-9,13-dihydroxycembra-11,15-dien-16,4-olide (Flexibolide, **4**):** colourless needles; 25 mg (0.00125%); mp 198–200 °C; [ $\alpha$ ] $^{27}_D$   $+31.3^\circ$  (*c* 1.33, MeOH); Anal. Calcd for  $\text{C}_{20}\text{H}_{30}\text{O}_5$ : C, 68.58; H, 8.58. Found: C, 68.12; H, 8.50;  $\lambda_{\text{max}}$  (MeOH) 217 nm;  $\nu_{\text{max}}$  (KBr) 3400 and 3360 (OH), 1720, 1640, 1465, 1380, 1080, and 920  $\text{cm}^{-1}$ ;  $^1\text{H}$ - and  $^{13}\text{C}$ -NMR data (see Table 2); EIMS *m/z* 332 ( $\text{M}^+ - \text{H}_2\text{O}$  31.1), 314 (21), 288 (21.3), 236 (15.1), 218 (17), 166 (11.6), 134 (67.9), 122 (75.9), 108 (92.2).

**Acetylation of Compound E: Flexibolide Diacetate (5).** Compound E (10 mg) was acetylated in pyridine (0.5 mL) with  $\text{Ac}_2\text{O}$  (0.5 mL) as described above to give the acetate as colorless oil, 7 mg; *m/z*  $\text{M}^+$  434; [ $\alpha$ ] $^{27}_D$   $+20.4^\circ$ ;  $\lambda_{\text{max}}$  (MeOH) 217 nm; Anal. Calcd for  $\text{C}_{24}\text{H}_{34}\text{O}_7$ : C, 66.36; H, 7.83. Found: C, 66.10; H, 7.52;  $\nu_{\text{max}}$  ( $\text{CHCl}_3$ ) 2950, 1725, and 1250 (OAc), 1720, 1440, 1190, 1142, and 930  $\text{cm}^{-1}$ ;  $^1\text{H}$  NMR ( $\text{C}_5\text{D}_5\text{N}$ , 90 MHz) 1.24, 1.48, 1.76, 2.02, and 2.08 (each 3H, s); 4.46 (1H, m); 5.23, 5.52, and 5.65 (each 1H, m); 5.45 and 6.42 (each 1H, s).

**Compound F:** colorless needles; 80 mg (0.004%) mp

178–180 °C;  $[\alpha]^{27}_D -40.7^\circ$  (*c* 0.9, MeOH). The compound was found to be identical in every respect with (1*R*,3*S*,4*S*,8*R*,11*S*,12*E*)-4,8,11-trihydroxycembra-12,15-diene-16,3-olide (flexibililolide).<sup>18</sup>

**Compound G:** colorless needles; 80 mg (0.004%) mp 160–163 °C;  $[\alpha]^{27}_D -22.0^\circ$  (*c* 0.99, MeOH). The compound was found to be identical in every respect with (1*R*,3*S*,4*S*,8*R*,11*S*,15*S*,12*E*)-4,8,11-trihydroxycembra-12-ene-16,3-olide (dihydroflexibililolide).<sup>18</sup>

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### References and Notes

- (1) Faulkner, D. J. *Nat. Prod. Rep.* **1995**, *12*, 223–269, and the earlier references cited therein.
- (2) Anjaneyulu, A. S. R.; Rao, G. V. *J. Sci. Ind. Res.*, **1995**, *54*, 637–649.
- (3) Herin, M.; Tursch, B.; *Bull. Soc. Chim. Belg.* **1976**, *85*, 707–719.
- (4) Tursch, B.; Braekman, J. C.; Daloze, D.; Herin, M.; Karlsson, R.; Losman, D. *Tetrahedron*, **1975**, *31*, 129–133.
- (5) Mori, K.; Suzuki, S.; Iguchi, K.; Yamada, Y. *Chem. Lett.* **1983**, 1515–1516.
- (6) Weinheimer, A. J.; Matson, J. A. *Tetrahedron Lett.* **1977**, *34*, 2923–2926.
- (7) Kazlauskas, R.; Murphy, P. T.; Wells, R. J.; Schonholzer, P.; Coll, J. C. *Aust. J. Chem.* **1978**, *31*, 1817–1824.
- (8) Guerrero, P. P.; Read, R. W.; Batley, M.; Janairo, G. C. *J. Nat. Prod.* **1995**, *58*, 1185–1191.
- (9) Kazlauskas, R.; Marwood, J. F.; Wells, R. J. *Aust. J. Chem.* **1980**, *33*, 1799–1803.
- (10) Anjaneyulu, A. S. R.; Sagar, K. S.; Venugopal, M. J. R. V. *Tetrahedron* **1995**, *51*, 10 997–11 010.
- (11) Anjaneyulu, A. S. R.; Sagar, K. S. *J. Chem. Res. (S)* **1995**, 142–143.
- (12) Anjaneyulu, A. S. R.; Rao, G. V.; Sagar, K. S.; Ravikumar, K.; Chandramohan, K. *Nat. Prod. Lett.* **1995**, *7*, 183–190.
- (13) Anjaneyulu, A. S. R.; Raju, K. V. S.; Rao, G. V. *Indian J. Chem.* **1995**, *34B*, 463–467.
- (14) Anjaneyulu, A. S. R.; Rao, G. V.; Raju, K. V. S.; Murty, M. V. R. K. *Indian J. Chem.* **1995**, *34B*, 1074–1079.
- (15) Anjaneyulu, A. S. R.; Sagar, K. S.; Prakash, C. V. S. *Indian J. Chem.* **1996**, *35B*, 819–825.
- (16) Anjaneyulu, A. S. R.; Rao, G. V.; Rao, N. S. K. *Indian J. Chem.* **1996**, *35B*, 815–818.
- (17) Anjaneyulu, A. S. R.; Rao, G. V. *Indian J. Chem.* **1996**, *35B*, 810–814.
- (18) Anjaneyulu, A. S. R.; Sagar, K. S. *Nat. Prod. Lett.* **1996** (in press).

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