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MICROWAVE MEDIATED SYNTHESIS OF 3,4,4a,11a-TETRAHYDRO-5-PHENYL/5-(4’-SUBSTITUTEDPHENYL)-2*H*-INDENO[1,2-*b*]PYRANO-[3,2-*e*]PYRAN-6(5*H*)-ONES

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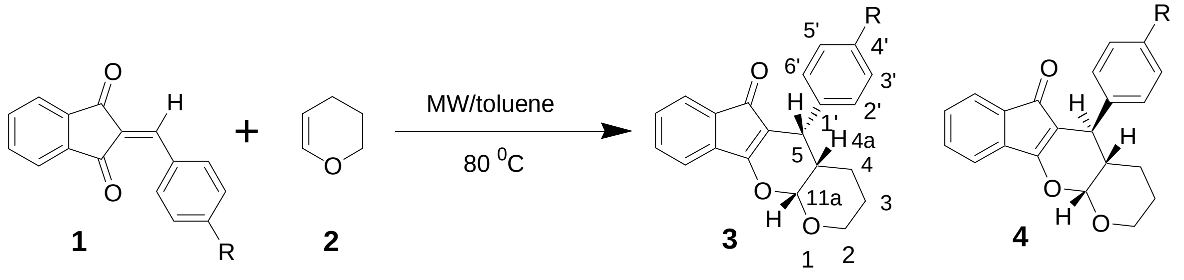
Abstract — Synthesis of 3,4,4a,11a-tetrahydro-5-phenyl/5-(4’-substituted phenyl)-2*H*-indeno[1,2-*b*]pyrano[3,2-*e*]pyran-6(5*H*)-ones have been reported under microwave conditions. The substrates for this being 2–benzylideneindane-1,3-dione (**1**) and 3,4-dihydro-2*H*-pyran (**2**); the product formation is governed by inverse Diels-Alder reaction conditions.

INTRODUCTION

Both inter- and intramolecular[1-7] Diels-Alder reactions have been extensively used in the synthesis of multicyclic ring systems primarily due to their ability to generate simultaneously upto four chiral centres in a highly stereoselective and largely predictable manner. In these reactions, α,β-unsaturated carbonyl compounds have been used both as dienes and as dienophiles depending upon the magnitude of electron density in the other component. In conventional Diels-Alder reactions, α,β-unsaturated compounds act as the dienophile while in inverse electron demand Diels-Alder reactions[8-9] these act as dienes. In view of our continuous interest in the synthesis of novel multicyclic heterocyclic ring systems, we describe in this paper, the synthesis and spectral studies of hitherto unknown 2*H*-indeno[1,2-*b*]pyrano[3,2-*e*]pyran-6(5*H*)-one ring system using inverse electron demand Diels-Alder reactions between 2-benzylideneindane-1,3-dione (**1**) and 3,4-dihydro-2*H*-pyran (**2**) under microwave conditions.

RESULTS AND DISCUSSION

The reaction was carried out between 2-benzylideneindane-1,3-dione (**1**) and 3,4-dihydro-2*H*-pyran (**2**) in toluene for 10-20 min at 80 °C under microwave conditions. The chromatographic separation of the resulting reaction mixture afforded two isomeric cycloadducts (**3**) and (**4**) in good yields (Scheme 1). The reaction was also carried out in the conventional way in acetonitrile, but the yield of the desired products was poor (Table 1). The structures of these cycloadducts have been ascertained through their IR, ^{1}H NMR and mass spectra.



a R=H
b R=CH_3
c R=OCH_3
d R=F
e R=CO_2CH_3
f R=NO_2

Scheme 1

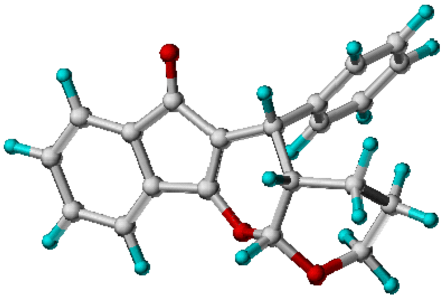
Table 1. Comparison of Inverse Diels-Alder reaction results under thermal and microwave condition.

R	**Time (min)/MW**	**Yield (%)**		**Time (h)/Δ**	**Yield *(%)**	
	Solvent (toluene/ MeCN)	**3**	**4**	Solvent (toluene/ MeCN)	**3**	**4**
a = H	20/180)	45	47	48/5	17.6	14.6
b = CH_3	45 / 180	0	0	72/10	0	0
c = OCH_3	60 /180	0	0	72/10	0	0
d = F	15 /120	45	48	24/3	15.2	5.6
e = CO_2CH_3	10 /150	48	43	24/3.5	14.9	6.2
f = NO_2	10 /180	50	41	36/4.0	16.7	8.2

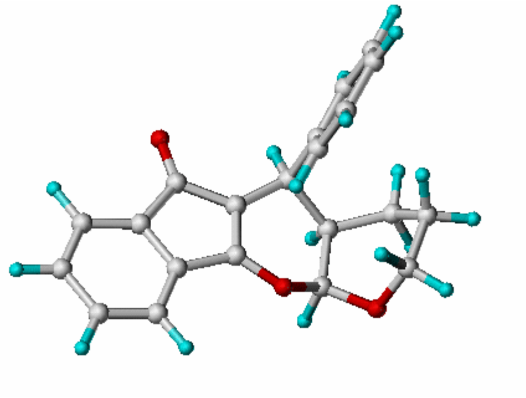
The regiochemistry and stereochemistry (*cis*) of cycloadducts could be explained on the basis of Woodward-Hoffmann rules,[10] further supported by ^{1}H-NMR chemical shifts of the protons and their coupling constants. In cycloadduct (**3f**), the protons C_{4a}-H, C_{11a}-H and C_5-H appeared in the regions δ 2.22-2.30, δ 5.75-5.77 (J=2.4Hz) and δ 4.28-4.30 (J=6.0Hz) respectively. In cycloadduct (**4f**), C_{4a}-H and C_{11a}-H appeared approximately in the same region except C_5-H that appeared at δ 3.75 as a singlet (little broadened at the base presumably due to its low coupling constant). The dihedral angle between the planes defined by the atoms 4a-H, C_{4a}-H and C_5; and C_{4a}, C_5 and 5-H in cycloadduct **3f** is 42.8° as calculated using Tripos Alchemy III computer programme; and thus $J_{4a,5}$ is expected to have a value between 4-6 Hz while the dihedral angle between the same planes in cycloadduct **4f** is 84.6° and thus $J_{4a,5}$ is expected to have a value between 0-1Hz.

These assignments for chemical shifts of C_5-H, C_{4a}-H and C_{11a}-H and coupling constants ($J_{4a,5}$) are in excellent agreement with those reported for cycloadducts[11,12] and further corroborated by HSQC (Figure 2) and the decoupling experiments of cycloadduct (**4f**). Upon double irradiation of C_{4a}-H in cycloadduct (**3f**, R=NO_2), the doublet in the region δ 4.28 – 4.30 due to C_5-H (J=6.0Hz) and doublet at δ 5.75-5.77 due to C_{11a}-H (J=2.4 Hz) collapsed to sharp singlets. Similarly, when C_{4a}-H in cycloadduct (**4f**, R=NO_2) was double irradiated, not only was the broadened singlet at δ 3.75 due to C_5-H was reduced to a sharp singlet, but even the doublet in the region δ 5.45-5.46 (J=2.6 Hz) due to C_{11a}-H, also changed to a sharp singlet. These two isomeric cycloadducts differ in stereochemistry at C_5-phenyl / substituted phenyl group. The stereochemical relationship between these three protons, viz., C_{4a}-H, C_5-H and C_{11a}-H in cycloadducts **3a** (*cis*) and **4a** (*trans*) have been confirmed through their molecular models (Figure 1) generated in their respective low energy orientation using Tripos Alchemy III computer programme.

Figure 1. Molecular models of cycloadducts of **3a** and **4a**

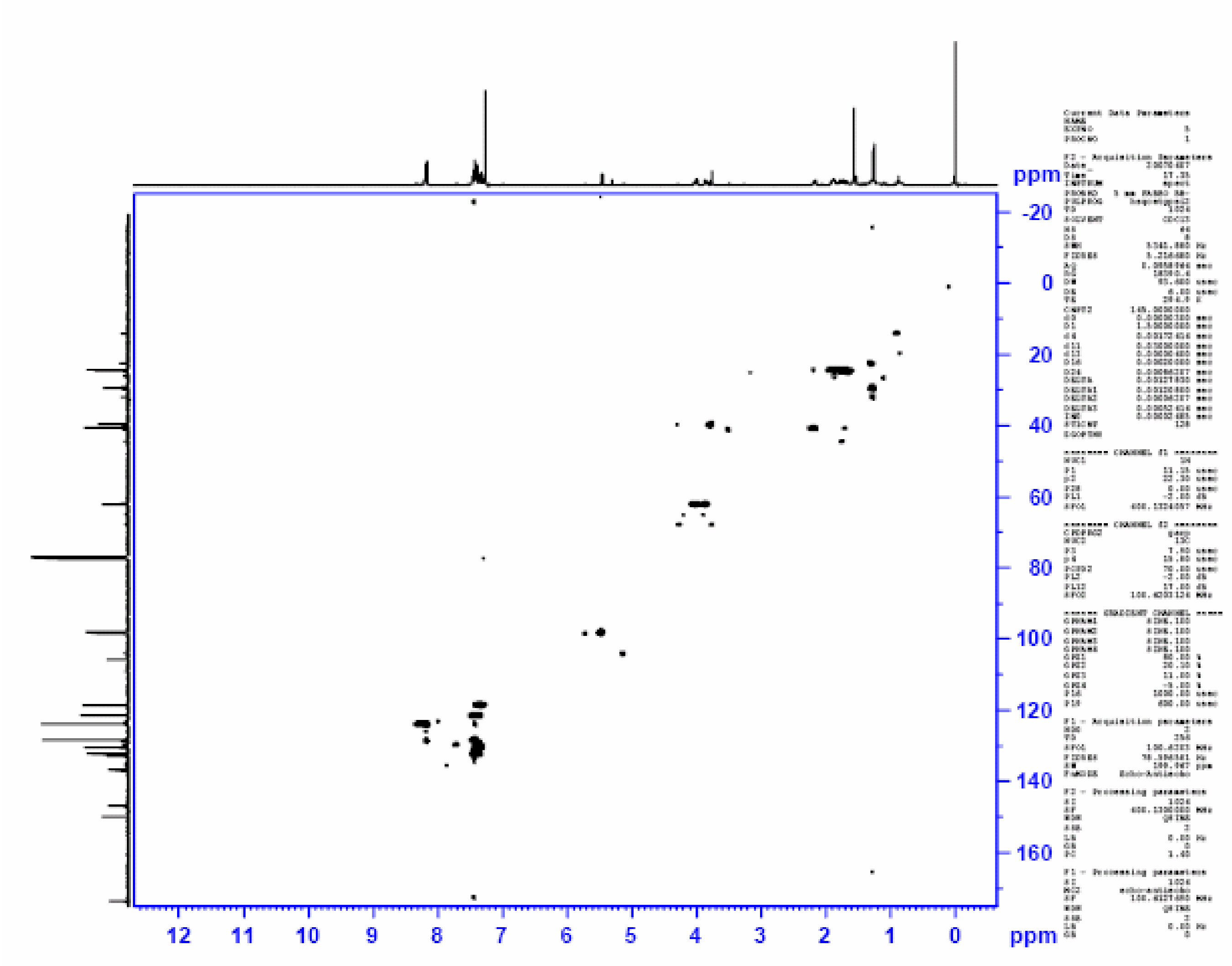


3a



4a

Figure 2. HSQC spectrum of cycloadduct **4f**



The formation of cycloadduct (**3f**) could be explained on the basis of *endo* approach[10] of 3,4-dihydro-2*H*-pyran in case of (**3f**) and *exo* approach in case of (**4f**). The steric reasons during the reaction might be responsible for the low yields of (**4f**).

Further, the effects of electron donating and withdrawing substituents have been explored on product formation. In case of electron donating groups, no formation of cycloadduct has been observed as it increases the energy gap between LUMO (diene) and HOMO (dienophile). The reaction was also tried in MeCN to obtain the comparative results with the thermal condition.

From the above discussion, we conclude that incorporation of electron-withdrawing groups at *p*-position of benzylidene moiety of 2-benzylideneindane-1,3-diones favours the cycloaddition between 1,3-diones and 3,4-dihydro-2*H*-pyran while incorporation of electron donating groups such as methyl and methoxy inhibit the cycloaddition reaction. Further, the formation of cycloadducts is controlled by the approach of 3,4-dihydro-2*H*-pyran from the less hindered site.

EXPERIMENTAL

Melting points were determined in open capillaries using a sulfuric acid bath and are uncorrected. IR spectra were recorded in Nujol mulls on a Perkin-Elmer 842 spectrophotometer and ^{1}H-NMR spectra were recorded on 90 MHz Perkin-Elmer R-32 instrument and ^{13}C-NMR were recorded on Bruker AV-400 (Top spin) in $CDCl_3$ using Me_4Si as internal standard. Low resolution mass spectra were recorded at 70eV using a VG-70S instrument. Elemental analysis was performed on Heraeus CHN-Rapid analyzer. Microwave reactions were carried out in Biotage initiator with power range of 0-400 W from magnetron at 2.45GHz.

General procedure for cycloaddition between 2-benzylideneindane-1,3-diones 1 and 3,4-dihydro-2*H*-pyran 2.

Microwave Method: A solution of 2-benzylideneindane-1,3-dione (**1**, 1 g, 4.3 mmol) and 3,4-dihydro-2*H*-pyran (**2**, 0.36 g, 4.4 mmol) in toluene (2 mL) was subjected to microwave irradiations for 10-20min in 2-5 mL vial in Biotage initiator with 'high' absorption level, fixed hold time 'on' and pre-stirring for 30 sec adjusted in the instrument. The reaction mass was concentrated under vacuum and the crude product purified by column chromatography (silica gel, 100-200 mesh; eluent hexane: benzene).

Conventional Method: A solution of 2-benzylideneindane-1,3-dione (**1**, 1 g, 4.3 mmol) and 3, 4-dihydro-2*H*-pyran (**2**, 0.34 g, 4 mmol) in anhydrous acetonitrile (80 mL) was refluxed for 4.5 h. The progress of the reaction monitored by TLC. The reaction mass was concentrated under vacuum and the crude product purified by column chromatography (silica gel, 100-200 mesh; eluent hexane: benzene) to obtain (**3a**) as yellow solid, which upon crystallization from benzene-pet. Ether mixture afforded bright yellow crystals. Subsequent elution's afforded another adduct (**4a**); crystallized from benzene-pet. Ether mixture to yield yellow crystals.

3a: (4aR*,5R*,11aS*) 3,4,4a,11a-Tetrahydro-5-phenyl-2*H*-indeno[1,2-*b*]pyrano[3,2-*e*]pyran-6(5*H*)-one. mp 165-166 °C; IR (KBr) 1695, 1630 cm^{-1}; ^{1}H-NMR (90MHz, $CDCl_3$): δ 1.33-1.48 (m, 2H, C_4-H), 1.54-1.75 (m, 2H, C_3-H), 2.17-2.25 (m, 1H, C_{4a}-H), 3.82-3.86 (m, 1H, C_2-H), 3.99-4.08 (m, 1H, C_2-H), 4.17-4.19 (d, *J* = 6.0 Hz, 1H, C_5-H), 5.72-5.75 (d, J=2.6Hz, 1H, C_{11a}-H), 7.08-7.40 (m, 9H, $C_{2'}$-H, $C_{3'}$-H, $C_{4'}$-H, $C_{5'}$-H, $C_{6'}$-H, C_7-H, C_8-H, C_9-H, and C_{10}-H); C^{13}-NMR (100MHz, $CDCl_3$): δ 189.8 (C-6), 171.7 (C-10b), 136.9 (C-1'), 134.9 (C-6a), 131.1 (C-5a), 132.4 (C-7), 130.6 (C-9), 128.5 (C-2' & C-6'), 125.8 (C-3' & 5'), 123.0 (C-4'), 120.4 (C-8), 116.4 (C-10), 100.2 (C-5), 98.2 (C-11a), 62.1 (C-2), 40.8 (C-5a), 39.7 (C-4a), 29.6 (C-4), 24.5 (C-3); MS (EI) m/z (int %) 318 ($M^{+\cdot}$, 38.7), 235 (63.4), 234 (21), 233 (47.0) and 84 (100). *Anal.* Calcd for $C_{21}H_{18}O_3$: C, 79.22; H, 5.70. Found: C, 79.26; H, 5.73.

4a: (4aR*,5S*,11aS*) 3,4,4a,11a-Tetrahydro-5-phenyl-2*H*-indeno[1,2-*b*]pyrano[3,2-*e*]pyran-6(5*H*)-

one. mp 168-169 °C; IR (KBr) 1695, 1630 cm^{-1}; ^{1}H-NMR (90MHz, $CDCl_3$): δ 1.61-1.73 (m, 2H, C_4-H), 1.78-1.89 (m, 2H, C_3-H), 2.14-2.20 (m, 1H, C_{4a}-H), 3.66 (br s, 1H, C_5-H), 3.78-3.84 (m, 1H, C_2-H), 3.93-4.02 (m, 1H, C_2-H), 5.47-5.50 (d, J=2.6Hz, 1H, C_{11a}-H), 7.18-7.51 (m, 9H, $C_{2'}$-H, $C_{3'}$-H, $C_{4'}$-H, $C_{5'}$-H, $C_{6'}$-H, C_7-H, C_8-H, C_9-H, and C_{10}-H); C^{13}-NMR (100MHz, $CDCl_3$): δ 189.8 (C-6), 171.7 (C-10b), 136.9 (C-1'), 134.9 (C-6a), 131.1 (C-5a), 132.4 (C-7), 130.6 (C-9), 128.5 (C-2' & C-6'), 125.8 (C-3' & 5'), 123.0 (C-4'), 120.4 (C-8), 116.4 (C-10), 100.2 (C-5), 98.2 (C-11a), 62.1 (C-2), 40.8 (C-5a), 39.7 (C-4a), 29.6 (C-4), 24.5 (C-3); MS (EI) m/z (int %) 318 ($M^{+\cdot}$, 37.2), 235 (64.1), 234 (18.9), 233 (45.1) and 84 (100). *Anal.* Calcd for $C_{21}H_{18}O_3$: C, 79.22; H, 5.70. Found: C, 79.25; H, 5.73.

3d: (4aR*,5R*,11aS*) 3,4,4a,11a-Tetrahydro-5-(4'-fluorophenyl)-2*H*-indeno[1,2-*b*]pyrano[3,2-*e*]-pyran-6(5*H*)-one. mp 182-183 °C; IR (KBr) 1695, 1620 cm^{-1}; ^{1}H-NMR (90MHz, $CDCl_3$): δ 1.58-1.70 (m, 2H, C_4-H), 1.89-2.18 (m, 2H, C_3-H), 2.19-2.25 (m, 1H, C_{4a}-H), 3.71-3.86 (m, 1H, C_2-H), 3.98-4.07 (m, 1H, C_2-H), 4.15-4.17 (d, J=6.0Hz, 1H, C_5-H), 5.71-5.74 (d, J=2.1Hz, 1H, C_{11a}-H), 6.95-7.38 (m, 8H, $C_{2'}$-H, $C_{3'}$-H, $C_{5'}$-H, $C_{6'}$-H, C_7-H, C_8-H, C_9-H and C_{10}-H); C^{13}-NMR (100MHz, $CDCl_3$): δ 188.8 (C-6), 172.7 (C-10b), 135.9 (C-1'), 132.9 (C-6a), 131.0 (C-10a), 131.4 (C-7), 130.6 (C-9), 128.5 (C-2' & C-6'), 126.8 (C-3' & 5'), 124.0 (C-4'), 120.4 (C-8), 116.4 (C-10), 102.2 (C-5) 99.2 (C-11a), 60.1 (C-2), 40.8 (C-5a), 39.7 (C-4a), 29.6 (C-4), 24.6 (C-3); MS (EI) m/z (int %) 336 ($M^{+\cdot}$, 22.8), 253 (34.7), 252 (16.8), 251 (30.9) and 84 (100). *Anal.* Calcd for $C_{21}H_{17}O_3F$: C, 74.99; H, 5.09; F, 5.65. Found: C, 74.99; H, 5.11; F, 5.68.

4d: (4aR*,5S*,11aS*) 3,4,4a,11a-Tetrahydro-5-(4'-fluorophenyl)-2*H*-indeno[1,2-*b*]pyrano[3,2-*e*]-pyran-6(5*H*)-one. mp 188-189 °C; IR (KBr) 1695, 1620 cm^{-1}; ^{1}H-NMR (90MHz, $CDCl_3$): δ 1.61-1.76 (m, 2H, C_4-H), 1.81-1.90 (m, 2H, C_3-H), 2.09-2.16 (m, 1H, C_{4a}-H), 3.64 (br s, 1H, C_5-H), 3.79-3.84 (m, 1H, C_2-H), 3.94-4.02 (m, 1H, C_2-H), 5.46-5.47 (d, J=2.4Hz,1H, C_{11a}-H), 6.98-7.43 (m, 8H, $C_{2'}$-H, $C_{3'}$-H, $C_{5'}$-H, $C_{6'}$-H, C_7-H, C_8-H, C_9-H and C_{10}-H); C^{13}-NMR (100MHz, $CDCl_3$): δ 189.4 (C-6), 172.7 (C-10b), 156.1 (C-4'), 141.9 (C-1'), 137.9 (C-6a), 133.1 (C-10a), 132.4 (C-7), 130.6 (C-9), 127.5 (C-2' & C-6'), 122.8 (C-3' & 5'), 121.5 (C-8), 118.5 (C-10), 105.7 (C-5), 98.2 (C-11a), 62.1 (C-2), 40.8 (C-5a), 39.7 (C-4a), 29.6 (C-4), 24.5 (C-3); MS (EI) m/z (int %) 336 ($M^{+\cdot}$, 20.2), 253 (32.0), 252 (9.1), 251 (9.1) and 84 (100). *Anal.* Calcd for $C_{21}H_{17}O_3F$: C, 74.99; H, 5.09; F, 5.65. Found: C, 74.98; H, 5.11; F, 5.68.

3e: (4aR*,5R*,11aS*) 3,4,4a,11a-Tetrahydro-5-(4'-carbomethoxy)phenyl-2*H*-indeno[1,2-*b*]pyrano-[3,2-*e*]pyran-6(5*H*)-one. mp 170-171 °C; IR (KBr) 1720, 1690 cm^{-1}; ^{1}H-NMR (90MHz, $CDCl_3$): δ 1.51-1.63 (m, 2H, C_4–H), 1.67-1.86 (m, 2H, C_3-H), 2.18-2.25 (m, 1H, C_{4a}–H), 3.65-3.73 (m, 1H, C_2-H), 3.89 (s, 3H, -CO_2CH_3), 3.91-3.96 (m, 1H, C_2-H), 4.21-4.23 (d, J=6.0Hz, 1H, C_5-H), 5.71-5.73 (d, J=1.4Hz, 1H, C_{11a}-H), 7.21-7.39 (m, 6H, $C_{2'}$-H, $C_{6'}$-H, C_7-H, C_8-H, C_9-H and C_{10}-H), 7.94-7.96 (d, J =

8.4Hz, 2H, $C_{3'}$-H, $C_{5'}$-H); C^{13}-NMR (100MHz, $CDCl_3$): δ 191.2 (C-6), 172.5 (C-10b), 167.1 (CO), 148.0 (C-4'), 146.8 (C-1'), 136.8 (C-6a), 134.0 (C-10a), 132.4 (C-7), 130.6 (C-9), 126.5 (C-2' & C-6'), 121.8 (C-3' & 5'), 121.5 (C-8), 118.5 (C-10), 104.8 (C-5), 98.2 (C-11a), 62.1 (C-2), 51.1 (CH_3), 40.8 (C-5a), 39.7 (C-4a), 29.6 (C-4), 24.5 (C-3) MS (EI) m/z (int %) 376 ($M^{+\cdot}$, 20.1), 293 (26.4), 292 (3.5), 233 (21.0), 84 (100). *Anal.* Calcd for $C_{23}H_{20}O_5$: C, 73.39; H, 5.36. Found: C, 73.41; H, 5.38.

4e: (4aR*,5S*,11aS*) 3,4,4a,11a-Tetrahydro-5-(4'-carbomethoxyphenyl)-2*H*-indeno[1,2-*b*]pyrano-[3,2-*e*]pyran-6(5*H*)-one. mp 178-179 °C; IR (KBr) 1720, 1690 cm^{-1}; ^{1}H-NMR (90MHz, $CDCl_3$): δ 1.63-1.73 (m, 2H, C_4-H), 1.79-1.91 (m, 2H, C_3-H), 2.13-2.19 (m, 1H, C_{4a}-H), 3.70 (br s, 1H, C_5-H), 3.79-3.85 (m, 1H, C_2-H), 3.89 (s, 3H, -CO_2CH_3), 3.94-4.02 (m, 1H, C_2-H), 5.46-5.47 (d, J=2.4Hz, 1H, C_{11a}-H), 7.26-7.44 (m, 6H, $C_{2'}$-H, $C_{6'}$-H, C_7-H, C_8-H, C_9-H and C_{10}-H), 7.95-7.98 (d, J=8.4Hz, 2H, $C_{3'}$-H and $C_{5'}$-H); C^{13}-NMR (100MHz, $CDCl_3$): δ 191.4 (C-6), 172.7 (C-10b), 167.0 (CO), 148.0 (C-4'), 146.9 (C-1'), 136.8 (C-6a), 134.0 (C-10a), 132.4 (C-7), 130.6 (C-9), 126.5 (C-2' & C-6'), 121.8 (C-3' & 5'), 121.5 (C-8), 118.5 (C-10), 105.8 (C-5), 98.2 (C-11a), 62.1 (C-2), 51.1 (CH_3), 40.8 (C-5a), 39.7 (C-4a), 29.6 (C-4), 24.5 (C-3); MS (EI) m/z (int %) 376 ($M^{+\cdot}$, 24.3), 293 (27.0), 292 (3.8), 233 (23.3) and 84 (100). *Anal.* Calcd for $C_{23}H_{20}O_5$: C, 73.39; H, 5.36. Found: C, 73.39; H, 5.39.

3f: (4aR*,5R*,11aS*) 3,4,4a,11a-Tetrahydro-5-(4'-nitrophenyl)-2*H*-indeno[1,2-*b*]pyrano[3,2-*e*]-pyran-6(5*H*)-one. mp 190-191°C; IR (KBr) 1700, 1620,1510, 1340 cm^{-1}; ^{1}H-NMR (90MHz, $CDCl_3$): δ 1.60-1.64 (m, 2H, C_4-H), 1.68-1.78 (m, 2H, C_3-H), 2.22-2.30 (m, 1H, C_{4a}-H), 3.85-3.89 (m, 1H, C_2-H), 4.00-4.06 (m, 1H, C_2-H), 4.28-4.30 (d, J=6.0Hz,1H, C_5-H), 5.75-5.77 (d, J=1.4Hz, 1H, C_{11a}-H), 7.29-7.42 (m, 6H, $C_{2'}$-H, $C_{6'}$-H, C_7-H, C_8-H, C_9-H and C_{10}-H), 8.16-8.20 (d, 2H, J=8.7Hz, $C_{3'}$-H, $C_{5'}$-H); C^{13}-NMR (100MHz, $CDCl_3$): δ 192.4 (C-6), 173.7 (C-10b), 150.0 (C-4'), 146.9 (C-1'), 136.9 (C-6a), 133.1 (C-5a), 132.4 (C-7), 130.6 (C-9), 128.5 (C-2' & C-6'), 123.8 (C-3' & 5'), 121.4 (C-8), 118.4 (C-10), 104.1 (C-5), 98.2 (C-11a), 62.1 (C-2), 40.8 (C-5a), 39.7 (C-4a), 29.6 (C-4), 24.5 (C-3); MS (EI) m/z (int %) 363 ($M^{+\cdot}$, 31.6), 333 (18.9), 249 (72.3), 248 (37.4) and 84 (100). *Anal.* Calcd for $C_{21}H_{17}NO_5$: C, 69.41; H, 4.72; N, 3.85. Found: C, 69.42; H, 4.74; N, 3.87.

4f: (4aR*,5S*,11aS*) 3,4,4a,11a-Tetrahydro-5-(4'-nitrophenyl-2*H*-indeno[1,2-*b*]pyrano[3,2-*e*]-pyran-6(5*H*)-one. mp 198-199 °C; IR (KBr) 1700,1620, 1510, 1340 cm^{-1}; ^{1}H-NMR (90MHz, $CDCl_3$): δ 1.68-1.85 (m, 2H, C_4-H), 1.86-1.90 (m, 2H, C_3-H), 2.14-2.19 (m, 1H, C_{4a}-H), 3.75 (br s, 1H, C_5-H), 3.82-3.87 (m, 1H, C_2-H), 3.96-4.04 (m, 1H, C_2-H), 5.45-5.46 (d, J=2.5Hz, 1H, C_{11a}-H), 7.35-7.45 (m, 6H, $C_{2'}$-H, $C_{6'}$-H, C_7-H, C_8-H, C_9-H, and C_{10}-H), 8.16-8.20 (d, J=8.7Hz, 2H, $C_{3'}$-H, $C_{5'}$-H); C^{13}-NMR (100MHz, $CDCl_3$): δ 192.4 (C-6), 173.7 (C-10b), 150.0 (C-4'), 146.9 (C-1'), 136.9 (C-6a), 133.0 (C-10a), 132.4 (C-7), 130.6 (C-9), 128.5 (C-2' & C-6'), 123.8 (C-3' & 5'), 121.5 (C-8), 118.5 (C-10), 105.7 (C-5), 98.2 (C-11a), 62.1 (C-2), 40.8 (C-5a), 39.7 (C-4a), 29.6 (C-4), 24.5 (C-3); MS (EI) m/z (int %) 363 ($M^{+\cdot}$, 28.8), 333 (13.1), 249 (71.9), 248 (41.7) and 84 (100). *Anal.* Calcd for $C_{21}H_{17}NO_5$: C, 69.41; H, 4.72; N, 3.85. Found: C, 69.43; H, 4.75; N, 3.86.

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