Supplementary Material

An Efficient and Modified Biginelli type Synthesis of 3, 4-dihydro-1H-indeno [1, 2-d] pyrimidine-2, 5-dione Using Phosphorous Pentoxide.

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General Experimental Methods

The melting points were determined in open capillary were found to be uncorrected. The purity of sample checked by thin-layer chromatography using silica gel 60-F 254 plates. The reported compounds were confirmed by ¹H NMR and IR while unreported compounds were confirmed by, ¹H NMR, IR, ¹³C NMR and Mass analysis. The ¹H NMR and ¹³C NMR spectra were taken in DMSO-*d*₆ using Brucker 300 MHz and 75 MHz or Brucker 400 MHz and 100 MHz spectrophotometer with TMS as internal standard. IR spectra were recorded on Shimadzu IR-450 FT-IR spectrophotometer. All the data found were corrected with the proposed structure.

General experimental procedure and characterization data

The mixture of 1, 3 indanedione (1mmol), aromatic / aliphatic aldehydes (1mmol) and urea/thiourea (5mmol) and phosphorous pentoxide (0.2 g) in 25 ml round bottom flask were mixed thoroughly and refluxed on water bath using ethanol as a solvent. The reaction is completed within 4-5 hours monitored by TLC. After completion, the mixture was cooled, poured on the crushed ice. On stirring, desired product was separated. The separated product was then filtered, washed with water and pet ether, dried and recrystallised using ethanol to get pure product.

4-(2-hydroxyphenyl)-3,4-dihydro-1H-indeno[1,2-d]pyrimidine-2,5-dione (Entry 1)
orange solid, m.p.196-200 °C (Lit.200-204³³), yield (83 %), IR (KBr): 1637, 1701, 3251, 3407 cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 5.52 (s, 1H, -CH), 6.67-6.72 (m, 1H, H_{Ar}), 6.77-6.80 (d, 1H, H_{Ar}), 7.00- 7.04 (m, 2H, H_{Ar}), 7.20-7.33 (m, 3H, H_{Ar}), 7.54-7.56 (m, 1H, H_{Ar}), 7.96 (br.s, 2H, -NH), 10.60 (s, 1H, -OH)), Elemental Anal. Found: C (69.78%) H (4.08%) N (9.47%) Calcd for C₁₇H₁₂N₂O₃ = C (69.86%) H (4.14%) N (9.58%).

4-(4-methoxyphenyl)-3,4-dihydro-1H-indeno[1,2-d]pyrimidine-2,5-dione (Entry-3)
yellow solid,m.p.179-183⁰C (Lit.180-183³³), yield (87%), IR(KBr): 1681, 1722, 3396, 3441 cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 3.92 (s, 3H, -OMe), 6.93-6.95 (m, 1H, -CH), 7.76-7.90 (m, 2H, H_{Ar}), 7.91-7.96 (m, 6H, H_{Ar}), 8.69-8.70 (s, 1H, -NH), 10.57 (s, 1H, -NH) ¹³C NMR (300 MHz, DMSO-*d*₆): δ 55.59, 115.73, 116.88, 122.63, 122.75, 124.98, 125.05, 131.61, 135.35, 135.51, 139.19, 141.66, 146.80, 147.43, 153.29, 189.20, 189.96; M⁺= 321; Elemental Anal. Found: C (70.46%) H (4.57%) N (9.09%) Calcd for C₁₈H₁₄N₂O₃ = C (70.58%) H (4.61%) N (9.15%)

4-phenyl-3,4-dihydro-1H-indeno[1,2-d]pyrimidine-2,5-dione (Entry-4) yellow solid, m.p. 173-175⁰C (Lit.175-178³³), yield (83%), IR(KBr): 1653, 1683, 3220, 3383 cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 5.30-5.31 (br.s, 1H, -CH), 7.17-7.43 (m, 7H, H_{Ar}), 7.51-7.53 (d, 1H, H_{Ar}), 7.62 (s, 1H, H_{Ar}), 7.81 (s, 1H, -NH), 10.63 (s, 1H, -NH); Elemental Anal. Found: C (73.81%) H (4.27%) N (10.06%) Calcd for C₁₇H₁₂N₂O₂ = C (73.90%) H (4.38%) N (10.14%)

4-(3-nitrophenyl)-3,4-dihydro-1H-indeno[1,2-d]pyrimidine-2,5-dione (Entry-6) pale yellow solid,m.p.200-204⁰C, yield (93%), IR(KBr): 1684, 1731, 3095, 3414 cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 7.81-7.85 (m, 2H, H_{Ar}), 7.97-8.05 (m, 7H, -NH and H_{Ar}), 8.39- 8.42 (d, 1H, H_{Ar}), 8.66-8.68(d, 1H, H_{Ar}), 9.54(s, 1H, -NH), ¹³C NMR (300 MHz, DMSO-*d*₆): δ 123.24, 123.41, 126.65, 126.99, 130.13, 131.56, 133.99, 136.17, 136.26, 139.69, 139.72, 142.04, 142.19, 147.80, 188.47, 188.81; M⁺= 321; Elemental Anal. Found: C (63.43%) H (3.35%) N (13.02%) Calcd for C₁₇H₁₁N₃O₄ = C (63.55%) H (3.45%) N (13.08%)

4-(4-hydroxyphenyl)-3,4-dihydro-1H-indeno[1,2-d]pyrimidine-2,5-dione (Entry-7)

yellow solid, m.p.250-255⁰C, yield (88%), IR(KBr): 1669, 1721, 3095, 3246, 3464 cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 6.89-6.92 (d, 1H, -CH), 7.65-7.83 (d, 1H, H_{Ar}), 7.85-7.94 (m, 7H, H_{Ar}), 8.32(s, 1H, -OH), 9.52 (s, 1H, -NH), 10.44(s, 1H, -NH) ¹³C NMR (300 MHz, DMSO-*d*₆): δ 115.74, 120.61, 122.58, 122.60, 124.79, 125.06, 130.22, 135.24, 135.42, 139.16, 141.61, 145.25, 146.84, 52.62, 188.89, 189.98; M⁺= 292 (m/z = 266 100%); Elemental Anal. Found: C (69.79%) H (4.09%) N (9.49%) Calcd for C₁₇H₁₂N₂O₃ = C (69.86%) H (4.14%) N (9.58%)

4-phenyl-2-thioxo-1, 2, 3, 4-tetrahydro-5H-indeno [1, 2-d] pyrimidin-5-one (Entry-11)

orange solid, m.p.250-252⁰C (Lit 249-252³³), yield (82%), IR (KBr): 1635, 1676, 3185, 3266, cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 5.31 (s, 1H, -CH), 7.22-7.31 (m, 8H, H_{Ar}), 7.75-7.78 (d, 1H, H_{Ar}), 9.75 (s, 1H, -NH), 11.65 (s, 1H, -NH) ; Elemental Anal. Found: C (69.77%) H (4.03%) N (9.46%) Calcd for C₁₇H₁₂N₂OS = C (69.84%) H (4.14%) N (9.58%)

4-(4-hydroxyphenyl)-2-thioxo-1,2,3,4-tetrahydro-5H-indeno[1,2-d]pyrimidin-5-one

(Entry-13) orange solid, m.p.245-255⁰C, yield (81%), IR (KBr): 1669, 1723, 3341, cm⁻¹; ¹H NMR (300 MHz, DMSO-*d*₆): δ 6.92-6.94 (d, 2H, -CH & H_{Ar}), 7.75-7.89 (s, 1H, H_{Ar}), 7.90-7.95 (m, 6H, H_{Ar}), 8.51-8.54 (d, 2H,-NH), 10.84 (s, 1H, -OH), ¹³C NMR (300 MHz, DMSO-*d*₆): δ 115.97, 122.65, 122.70, 124.55, 125.14, 135.34, 135.51, 137.55, 139.19, 141.61, 146.24, 163.29, 188.97, 189.93; M⁺= 308 Elemental Anal. Found: C (66.13%) H (3.84%) N (9.03%) Calcd for C₁₇H₁₂N₂O₂S = C (66.22%) H (3.92%) N (9.08%)

¹H-NMR spectrum of entry 1

Current Data Parameters
NAME CSS
EXPNO 140
PROCNO 1

F2 - Acquisition Parameters
Date_ 20140221
Time 17.02
INSTRUM spect
PROBHD 5 mm BBO BB-1H
PULPROG zg30
TD 65536
SOLVENT DMSO
NS 16
DS 2
SWH 6188.119 Hz
FIDRES 0.094423 Hz
AQ 5.2953587 sec
RG 161
DW 80.800 usec
DE 6.00 usec
TE 300.0 K
D1 1.00000000 sec
TD0 1

===== CHANNEL f1 =====
NUC1 1H
P1 11.38 usec
PL1 0.00 dB
SFO1 300.1318534 MHz

F2 - Processing parameters
SI 32768
SF 300.1300000 MHz
WDW EM
SSB 0
LB 0.30 Hz
GB 0
PC 1.00

IR spectrum of entry 1

¹H-NMR spectrum of entry 3

¹³C-NMR spectrum of entry 3

Mass spectrum of entry 3

IR spectrum of entry 3

¹H-NMR spectrum of entry 4

IR spectrum of entry 4

¹H-NMR spectrum of entry 6

¹³C-NMR spectrum of entry 6

Mass spectrum of entry 6

IR spectrum of entry 6

¹H-NMR spectrum of entry 7

¹³C spectrum of entry 7

Mass spectrum of entry 7

IR spectrum of entry 7

¹H-NMR spectrum of entry 11

IR spectrum of entry 11

¹H-NMR spectrum of entry 13

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NAME      2_DEC_Warlekar
EXPNO     3
PROCNO    1
Date_     20131201
Time      19.33
INSTRUM   spect
PROBHD    5 mm FASBO BB-
PULPROG   zgpg30
TD         20768
SOLVENT   DMSO
NS         32
DS         0
SWH        6410.286 Hz
FIDRES     0.308660 Hz
AQ         1.6189540 sec
RG          203
DW         78.000 usec
DE         6.50 usec
TE         298.0 K
D1         2.00000000 sec
TD0        1

===== CHANNEL f1 =====
NUC1       1H
P1         14.10 usec
PL1        -3.00 dB
PL1W       13.42244830 W
SFO1       400.2320016 MHz
SI         65826
SF         400.2300099 MHz
WDW        EM
SSB        0
LB         0.00 Hz
GB         0
PC         1.00
    
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¹³C spectrum of entry 13

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NAME      2_DEC_Warlekar
EXPNO     11
PROCNO    1
Date_     20131202
Time      0.21
INSTRUM   spect
PROBHD    5 mm FASBO BB-
PULPROG   zgpg30
TD         32768
SOLVENT   DMSO
NS         1092
DS         4
SWH        24038.461 Hz
FIDRES     0.722596 Hz
AQ         0.6816244 sec
RG          203
DW         20.800 usec
DE         6.50 usec
TE         298.0 K
D1         3.00000000 sec
D11        0.02000000 sec
TD0        1

===== CHANNEL f1 =====
NUC1       13C
P1         9.62 usec
PL1        -2.00 dB
PL1W       58.25462341 W
SFO1       100.6494874 MHz

===== CHANNEL f2 =====
CPDPRG2   waltz16
NUC1       1H
PCPD2     90.00 usec
PL2        -3.00 dB
PL12       12.10 dB
PL13       16.00 dB
PL1W       13.42244830 W
PL12W      0.32948199 W
PL13W      0.16897857 W
SFO2       400.2316009 MHz
SI         121072
SF         100.6379678 MHz
WDW        EM
SSB        0
LB         1.00 Hz
GB         0
PC         1.40
    
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Mass spectrum of entry 13

IR spectrum of entry 13