

Supplementary Data:

5-Ethoxycarbonyl-4-(4-methoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (entry 1):

Mp 199-200 °C (Lit.^{8c} Mp 200-201 °C); IR (KBr): 3350, 1735, 1610 cm⁻¹; PMR (CDCl₃ + DMSO-d₆): δ_H = 1.17 (3H, t, *J* = 8 Hz, CO₂CH₂CH₃), 2.33 (3H, s, CH₃), 3.78 (3H, s, OCH₃), 4.02 (2H, q, *J* = 8 Hz, CO₂CH₂CH₃), 5.30(1H, bs, CH), 6.81 (2H, d, *J* = 8 Hz, ArHs), 7.24 (2H, d, *J* = 8 Hz, ArHs) 7.49 (1H, bs, NH), 8.78 (1H, bs, NH); CMR (CDCl₃ + DMSO-d₆): δ_C = 165.3, 158.0, 152.0, 147.5, 136.9, 127.3, 113.6, 99.9, 59.0, 53.2, 17.0 and 14.0.

4-(4-Isopropylphenyl)-5-methylcarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one.

(entry 6): Mp 165-167 °C; IR (KBr): 3285, 1678, 1602 cm⁻¹; PMR (CDCl₃ + DMSO-d₆): δ_H = 1.21[6H, d, *J* = 8Hz, CH(CH₃)₂], 2.11(3H, s, CH₃), 2.34(3H, s, COCH₃), 2.87 [1H, septet, CH(CH₃)₂], 5.31 (1H, bs, ArCH), 7.19 (4H, m, ArHs), 7.70 (1H, bs, NH), 8.96(1H, bs, NH); CMR (CDCl₃ + DMSO-d₆): δ_C = 196.08, 153.29, 149.0, 148.93, 142.25, 127.38, 110.78, 54.62, 33.99, 31.00, 24.68 and 19.74.

4-(4-Isopropylphenyl)-5-methylcarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-thione

(entry 7): Mp 140-142 °C; IR (KBr): 3422 (broad), 1660, 1187 cm⁻¹; PMR (CDCl₃ + DMSO-d₆): δ_H = 1.22 [6H, d, *J* = 8Hz, CH(CH₃)₂], 2.15 (3H, s, CH₃), 2.38 (3H, s, COCH₃), 2.87 [1H, septet, CH(CH₃)₂], 5.42 (1H, bs, ArCH), 7.26

(4H, m, ArHs), 7.58(1H, bs, NH), 8.18 (1H, bs, NH); CMR (CDCl₃ + DMSO-d₆): δ_C = 195.81,

173.7, 149.20, 126.8-142.57, 55.79, 33.70, 30.26, 23.82 and 19.02.

5-Ethoxycarbonyl-4-(3-nitrophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (entry 8):

Mp 225-227 °C (Lit.¹⁴ Mp 227-229 °C); IR (KBr): 3400, 1725, 1520, 1340 cm⁻¹; PMR (CDCl₃ + DMSO-d₆): δ_H 1.18 (3H, t, *J* = 7 Hz, CO₂CH₂CH₃), 2.37 (3H, s, CH₃), 3.78 (3H, s, OCH₃), 4.07 (2H, q, *J* = 7 Hz, CO₂CH₂CH₃), 5.46(1H, bs, CH), 7.69-8.19 (3H, m, ArHs), 8.97 (1H, bs, NH), 10.15 (1H, bs, NH); CMR (CDCl₃ + DMSO-d₆): δ_C = 165.74, 152.46, 150.13, 148.41, 147.67, 133.68, 130.93, 123.06, 121.69, 98.98, 60.08, 54.22, 18.54 and 14.69.

4-(2,5-Dimethylphenyl)-5-ethoxycarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one. (entry 10)

Mp 224-226 °C; IR (KBr): 3352, 1696, 1626 cm⁻¹; PMR (CDCl₃ + DMSO-d₆): δ_H = 1.07 (3H, t, *J* = 8Hz, CO₂CH₂CH₃), 2.25(3H, s, CH₃), 2.37 and 2.40 [3H each, s, ArCH₃], 3.96 (2H, q, *J* = 8Hz, CH₂CH₃), 5.53 (1H, s, ArCH), 6.93-7.03 (3H, m, ArHs), 7.67 (1H, s, NH) and 8.78 (1H, s, NH); CMR (CDCl₃ + DMSO-d₆): δ_C = 165.23, 151.62, 147.95, 127.02-142.86, 99.21, 58.93, 50.69, 20.75, 18.12, 17.60 and 13.79.

4-(2,5-Dimethylphenyl)-5-methylcarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one. (entry 11)

Mp 110-111 °C; IR (KBr): 3281, 1693, 1666, 1597 cm⁻¹; PMR (CDCl₃ + DMSO-d₆): δ_H 2.0 (3H, s, CH₃), 2.2 (3H, s, COCH₃), 2.32 (3H, s, ArCH₃),

2.42 (3H, s, ArCH₃), 5.62 (1H, s, ArCH), 6.9-7.15 (3H, m, ArHs), 7.6 (1H, s, NH) and 10.2 (1H, s, NH).

5- Ethoxycarbonyl - 6 - methyl - 4- styryl- 3, 4- dihydropyrimidin-2(1H)-one (entry 13):

Mp 230-232 °C (Lit.¹⁴ Mp 230-232 °C); IR (KBr): 3247, 3115, 2928, 1720, 1701, 1652 cm⁻¹; PMR (CDCl₃ + DMSO-d₆): δ_H = 1.21 (3H, t, *J* = 7 Hz, CO₂CH₂CH₃), 2.29 (3H, s, CH₃), 3.66 (2H, q, *J* = 7 Hz, CO₂CH₂CH₃), 5.37 (1H, bs, CH), 6.22(dd, 1H, *J* = 16 Hz and 4 Hz, ArCH=CHCH), 6.48 (1H, d, *J* = 16 Hz, ArCH=CHCH), 7.3-7.4(4H, m, ArHs), 7.51 (1H, bs, NH), 8.75 (1H, bs, NH); CMR (CDCl₃ + DMSO-d₆): δ_C = 165.1, 152.6, 148.5, 136.2, 130, 128.6, 128.1, 127.5, 126.2, 97.8, 59.2, 51.8, 17.7 and 14.2

5-Methylcarbonyl-6-methyl-4-styryl-3,4- dihydropyrimidin- 2(1H)-one. (entry 15)

Mp 223-226 °C (Lit^{17c} 224-226 °C); IR (KBr): 3277 (broad), 1675, 1603, 1229 cm⁻¹; ¹H NMR (CDCl₃ + DMSO-d₆): δ_H = 2.26 (3H, s, -CH₃), 2.28 (3H, s, -COCH₃), 4.85 (1H, brs, Ar-CH), 6.11-6.22 (1H, dd, *J* = 14 Hz and *J* = 4 Hz, HC=CH-Ar), 6.40 (1H, d, *J* = 14 Hz, HC=CH-Ar), 7.23-7.36 (5H, m, ArHs), 7.87 (1H, s, -NH), 9.06 (1H, s, -NH)

5- Methylcarbonyl- 6- methyl- 4- styryl- 3, 4-dihydropyrimidin- 2(1H)-thione. (entry 16)

Mp 229-231 °C; IR (KBr): 3276 (broad), 1675, 1603, 1229 cm⁻¹; ¹H NMR (CDCl₃ + DMSO-d₆): δ_H = 2.27 (3H, s, -CH₃), 2.31 (3H, s, -COCH₃), 4.91 (1H, brs, Ar-CH), 6.05 – 6.15 (1H, dd, *J* = 14 Hz and *J* = 4 Hz, HC=CH-Ar), 6.39 (1H, d, *J* = 14

Hz, HC=CH-Ar), 7.20-7.36 (5H, m, ArHs), 9.33 (1H, s, -NH) and 10.09 (1H, s, -NH).

5 - Ethoxycarbonyl- 4- (propyl)- 6- methyl- 3, 4- dihydropyrimidin-2(1H)-one (entry 19):

Mp 158-160 °C (Lit.^{16b} Mp 170-172 °C); IR (KBr): 3247, 3117, 1717, 1701, 1652 cm⁻¹; PMR (CDCl₃ + DMSO-d₆): δ_H = 0.75 (3H, t, *J* = 7 Hz, CH₂CH₃), 1.15 (3H, t, CO₂CH₂CH₃), 1.42 (2H, m, CH₂), 2.13 (3H, s, Ar-CH₃), 3.30 (1H, s, CH), 4.00(2H, m, -CH₂), 7.27 (1H, bs, NH) and 8.88 (1H, bs, NH).