General procedure for the preparation of 3,4-dihydropyrimidin-2-(1H)-ones/thiones

A mixture of aldehyde (1 mmol), ethyl acetoacetate (1 mmol), urea or thiourea (1.0 mmol) and [HMIm][OAc] (5 mol%) was heated with stirring at 100 °C. The progress of the reaction was monitored by TLC. After completion of reaction, twice cool water was added to the reaction mixture and decanted to remove the ionic liquid. Then, the crude product was dissolved in hot ethanol and the product recrystallized from ethanol. Various aromatic aldehyde derivatives were put under the same condition and melting point of the obtained products were determined by the open capillary tube method in electrothermal melting point apparatus (Table 4). The obtained Biginelli products (DHPMs) were also confirmed by spectroscopy analysis.

5-Ethoxycarbonyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1*H*)-one (4a)

mp 198–199 °C (Lit [1], 198-200 °C); IR (KBr, cm^{-1}) 3244 (NH), 3116 (CH arom), 2931 (CH aliph), 1701 (C=O), 1651 (C=O); ^{1}H NMR (250 MHz, DMSO-d_6/TMS) δ_{ppm}: 1.10 (t, J = 7.2 Hz, 3H, $OCH_2\underline{CH_3}$), 2.27 (s, 3H, CH_3), 3.70 (q, J = 7.2 Hz, 2H, $O\underline{CH_2}CH_3$), 5.24 (d, J = 3.6 Hz, 1H, CH), 7.85 (s, 1H, NH), 7.16-7.39 (m, 5H, Ph), 9.29 (s, 1H, NH); ^{13}C NMR (62.9 MHz, DMSO-d_6/TMS) δ_{ppm}: 13.90; 15.24, 54.56, 59.04, 100.68, 125.40, 126.96, 128.28, 129.08, 135.13, 136.53, 141.52, 148.67, 152.33, 165.14.

5-Ethoxycarbonyl-6-methyl-4-(4-methylphenyl)-3,4-dihydropyrimidin-2(1*H*)-one (4b)

mp 216-217 °C (Lit [1], 212-215 °C); IR (KBr, cm^{-1}) 3244 (NH), 3116 (CH arom), 2927 (CH aliph), 1705 (C=O), 1651 (C=O); ^{1}H NMR (250 MHz, DMSO-d_6/TMS) δ_{ppm}: 1.10 (t, J =7.1 Hz, 3H, $OCH_2\underline{CH_3}$); 2.24 (s, 3H, CH_3), 2.26 (s, 3H, CH_3), 3.98 (q, J = 7.0, 2H, $O\underline{CH_2}CH_3$), 5.10 (s, 1H, CH), 7.02-7.15 (m, 4H), 7.72 (s, 1H, NH), 9.19 (s, 1H, NH), ^{13}C NMR (62.9 MHz, DMSO-d_6) δ_{ppm}: 14.21, 17.79, 20.86, 53.78, 59.13, 99.65, 126.31, 129.08, 136.43, 142.19, 148.20, 152.43, 165.53.

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

mp 206-208 °C (Lit [1], 205-207 °C); IR (KBr, cm^{-1}) 3226 (NH), 3100 (CH arom), 2929 (CH aliph), 1710 (C=O), 1651 (C=O); ^{1}H NMR (250 MHz, DMSO-d_6) δ_{ppm}: 1.09 (t, J = 6.8 Hz, 3H, $OCH_2\underline{CH_3}$), 2.23 (s, 3H, CH_3), 3.7 (s, 3H, $O\underline{CH_3}$), 3.96 (q, J = 6.8 Hz, 2H, $O\underline{CH_2}CH_3$), 5.07 (s, 1H, CH), 6.84-7.15 (m, 4H), 7.66 (s, 1H, NH), 9.14 (s, 1H, NH); ^{13}C NMR (62.9 MHz, DMSO-d_6) δppm: 14.00, 17.56, 53.32, 54.80, 58.69, 99.95, 113.46, 127.13, 136.90, 147.54, 152.00, 158.00, 165.23.

5-Ethoxycarbonyl-6-methyl-4-(4-chlorophenyl)-3,4-dihydropyrimidin-2(1*H*)-one (4d)

mp 205-206 °C (Lit [2], 204-206 °C); IR (KBr, cm^{-1}) 3243 (NH), 3116 (CH arom), 2980 (CH aliph), 1705 (C=O), 1648 (C=O); ^{1}H NMR (500 MHz, DMSO-d_6) δ_{ppm}: 0.92 (t, J = 7.0 Hz, 3H, $OCH_2\underline{CH_3}$), 2.1 (s, 3H, CH_3), 3.82 (q, J = 7.0 Hz, 2H, $O\underline{CH_2}CH_3$), 5.0 (s, 1H, CH), 7.09 (d, J = 8.5 Hz, 2H, AA'), 7.22 (d, J = 8.5 Hz, 2H, BB'), 7.63 (s, 1H, NH), 9.11 (s, 1H, NH); ^{13}C NMR (125 MHz, DMSO-d_6) δ_{ppm}: 14.49, 18.26, 53.92, 59.70, 99.33, 128.65, 128.83, 132.28, 144.24, 149.15, 152.47, 165.65.

5-Ethoxycarbonyl-6-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1*H*)-one (4e)

mp 211-212 °C (Lit [1], 204-206 °C); IR (KBr, cm^{-1}) 3232 (NH), 3117 (CH arom), 2916 (CH aliph), 1705 (C=O), 1647 (C=O); ^{1}H NMR (500 MHz, DMSO-d_6) δ_{ppm}: 1.08 (t, J = 7.0 Hz, 3H, $OCH_2\underline{CH_3}$), 2.25 (s, 3H, CH_3), 3.97 (q, J = 7.0 Hz, 2H, $O\underline{CH_2}CH_3$), 5.26 (s, 1H, CH), 7.50 (d, J = 7.35 Hz, AA', 2H), 7.89 (s, 1H, NH), 8.23 (d, J = 7.35 Hz, BB', 2H), 9.35 (s, 1H, NH); ^{13}C NMR (62.9 MHz, DMSO-d_6) δ_{ppm}: 14.45, 18.43, 54.12, 59.78, 98.87, 124.16, 128.18, 147.20, 149.80, 152.19, 152.49, 165.54.

5-Ethoxycarbonyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1*H*)-thione (4f)

mp 205-207 °C (Lit [1], 210-212 °C); IR (KBr, cm^{-1}) 3328 (NH), 3174 (CH arom), 1670 (C=O), 1573 (C=O); ^{1}H NMR (500 MHz, DMSO-d_6) δ_{ppm}: 1.09 (t, J = 7.0 Hz, 3H, $OCH_2\underline{CH_3}$), 2.28 (s, 3H, CH_3), 4.0 (q, J = 7.0 Hz, 2H, $O\underline{CH_2}CH_3$), 5.16 (s, 1H, CH), 7.19-7.35 (m, 5H, C_6H_5), 9.64 (s, 1H, NH), 10.33 (s, 1H, NH); ^{13}C NMR (125 MHz, DMSO-d_6) δ_{ppm}: 13.96, 17.07, 55.01, 59.47, 100.88, 113.79, 127.55, 135.64, 144.68, 158.66, 165.09, 173.94.

5-Ethoxycarbonyl-6-methyl-4-(4-methylphenyl)-3,4-dihydropyrimidin-2(1*H*)-thione (4g)

mp 194-195 °C (Lit [1], 191-193 °C); IR (KBr, cm^{-1}) 3255 (NH), 1659 (C=O), 1652 (C=O); ^{1}H NMR (250

MHz, DMSO-d_6) δ_{ppm}: 1.11 (t, J = 7.2 Hz, 3H, $OCH_2\underline{CH_3}$), 2.27 (s, 3H, CH_3), 2.28 (s, 3H, CH_3), 4.0 (q, J = 7.2 Hz, 2H, $O\underline{CH_2}CH_3$), 5.13 (s, 1H, CH), 7.08-7.16 (m, 4H), 9.61 (s, 1H, NH), 10.29 (s, 1H, NH); ^{13}C NMR (62.9 MHz, DMSO-d_6) δ_{ppm}: 12.92, 16.16, 53.43, 59.65, 78.47, 100.18, 125.02, 126.68, 127.82, 129.33, 136.38, 140.54, 144.36, 164.65, 173.66.

5-Ethoxycarbonyl-6-methyl-4-(4-methoxyphenyl)-3,4-dihydropyrimidin-2(1*H*)-thione (4h)

mp 156-157 °C (Lit [1], 151-153 °C); IR (KBr, cm^{-1}) 3250 (NH), 1651 (C=O), 1598 (C=S); ^{1}H NMR (250 MHz, DMSO-d_6) δ_{ppm}: 1.09 (t, J = 7.0 Hz, 3H, $OCH_2\underline{CH_3}$), 2.27 (s, 3H, CH_3), 3.71 (s, 3H, OCH_3), 3.99 (q, J = 7.0 Hz, 2H, $O\underline{CH_2}CH_3$), 5.10 (s, 1H, $\underline{CH}$), 6.87-7.14 (m, 4H, C_6H_4), 9.59 (s, 1H, NH), 10.29 (s, 1H, NH); ^{13}C NMR (62.9 MHz, DMSO-d_6) δ_{ppm}: 14.1, 17.2, 53.2, 55.1, 59.1, 101.1, 127.7, 135.8, 144.7, 158.8, 165.2, 174.

5-Ethoxycarbonyl-6-methyl-4-(4-chlorophenyl)-3,4-dihydropyrimidin-2(1*H*)-thione (4i)

mp 182-183 °C (Lit [3], 179-180 °C); IR (KBr, cm^{-1}) 3255 (NH), 1657 (C=O), 1560 (C=S); ^{1}H NMR (250 MHz, DMSO-d_6) δ_{ppm}: 1.08 (t, J = 7.0 Hz, 3H, $OCH_2\underline{CH_3}$), 2.28 (s, 3H, $\underline{CH_3}$), 4.0 (q, J = 7.0 Hz, 2H, $O\underline{CH_2}CH_3$), 5.16 (s, 1H, $\underline{CH}$), 7.19-7.43 (m, 4H, C_6H_4), 9.65 (s, 1H, NH), 10.36 (s, 1H, CH); ^{13}C NMR (125MHz, DMSO-d_6) δ_{ppm}: 13.93, 17.12, 53.38, 59.59, 100.21, 128.25, 128.53, 132.20, 142.31, 145.32, 164.92, 174.17.

5-Ethoxycarbonyl-6-methyl-4-(4-nitrophenyl)-3,4-dihydropyrimidin-2(1*H*)-thione (4j)

mp 207-208 °C (Lit [1], 206-208 °C); IR (KBr, cm^{-1}) 3253 (NH), 1655 (C=O), 1548 (C=S); ^{1}H NMR (250 MHz, DMSO-d_6) δ_{ppm}: 1.12 (t, J = 7.0 Hz, 3H, $OCH_2\underline{CH_3}$), 2.30 (s, 3H, $\underline{CH_3}$), 4.0 (q, J = 7.0 Hz, 2H, $O\underline{CH_2}CH_3$), 5.26 (s, 1H, $\underline{CH}$), 7.45-8.25 (m, 4H, C_6H_4), 9.62 (s, 1H, NH), 10.40 (s, 1H, CH).