Biginelli-like Three Component Reaction: Synthesis of some New Ethyl 6-ethoxycarbonylmethyl-4-aryl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate Derivatives

Javad Azizian,* Ali A. Mohammadi, M. Kohshari, Ali R. Karimi, Mohammad R. Mohammadizadeh

Department of Chemistry, Faculty of Science, Shahid Beheshti University, P. O. Box 19395-4716, Tehran, Iran, *E-mail:* E-mail: **j-azizian@cc.sbu.ac.ir**

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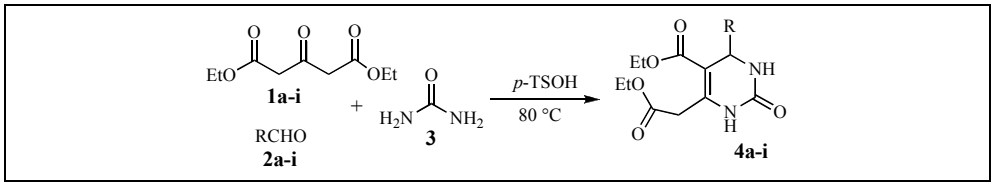

In the context of our high-throughput organic synthesis program, we have studied the reactivity of special β-keto esters toward the Biginelli reaction. We have found that a diethyl-3-oxoglutarate reacts with one molecule of urea and one molecule of aldehyde under solvent-free conditions to give a new family of 3,4-dihydropyrimidin-2(1*H*)-ones in good yields.

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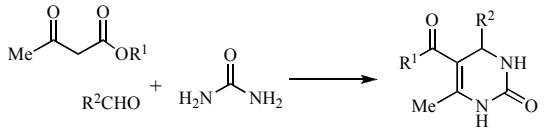
3,4-Dihydropyrimidin-2(1*H*)-ones (DHPMs) are an important class of heterocyclic compounds having important biological activities. During the last decade such compounds have shown interesting pharmacological properties like antiviral [1,2], antitumour [3], antibacterial, anti-inflammatory [4], and antihypertensive [5] activities. One of the most important methods for synthesis of this class of compounds is the one pot condensation of an aldehyde, β-ketoester and urea under acidic conditions, first reported by Biginelli (Scheme 1).

Scheme 1



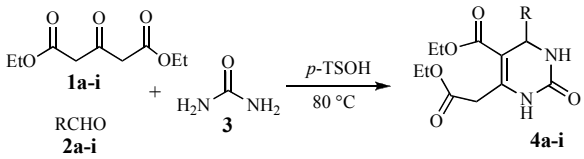
Because of intense interest in the biological activity of these compounds, in recent years, several synthetic procedures for preparing of DHPMs have been reported including classical conditions with microwave irradiation [6,9c] and by using Lewis acids as well as protic acids as promoters such as concentrated HCl [7,20a,b], $BF_3.OEt_2$ [8], montmorillonite [9], $InCl_3$ [10], $LaCl_3$ [11], $La(OTf)_3$ [12], $Yb(OTf)_3$ [12], $InBr_3$ [13], $BiCl_3$ [14], $Bi(OTf)_3$ [15], $In(OTf)_3$ [16], *p*-TSOH [17], $Cu(OTf)_2$ [18], $CeCL_3$ [19], $FeCl_3$ [20a,c], $ZnCl_2$ [20,21], $MgBr_2$ [22], $MgCl_2$ [23], $CdCl_2$ [24], $Zn(OTf)_2$ [25], $KHSO_4$ [26], have been used for this reaction.

In view of this and also in continuation to our interest on Multi-Component reactions (MCRs) [27], we report herein, a simple, facile, rapid and efficient MCRs for the preparation of some new 6-substituted DHPMs derivatives with *p*-TSOH as a nontoxic, inexpensive, very soluble in water, and easily available reagent.

Herein we describe a higher yielding protocol for the Biginelli-like three-component reactions of aldehydes, diethyl-3-oxoglutarate and urea for the synthesis of the new 6-substituted DHPMs **4a-i** using *p*-TSOH under mild conditions.

From a mixture of diethyl-3-oxoglutarate **1**(1 mmol), benzaldehyde **2a** (1 mmol), urea **3** (1.2 mmol), and *p*-TSOH (2 mol %), DHPMs **4a** was isolated in 98% yield (Scheme 2). The reaction was carried out at 80 ºC for 3 h (until the diethyl-3-oxoglutarate disappeared, as shown by TLC analysis). Then the Biginelli-like reactions of other various aldehydes **2b-i** under a established protocol

Scheme 2



Table

The Reaction of Aldehydes, Diethyl-3-oxoglutarate and Urea

Mp (°C)	Yield (%)	Time (h)	R	Products[a] **4**
178-180	98	3	Ph	**a**
156-158	95	3	2-BrC_6H_4	**b**
138-140	95	2.5	3-ClC_6H_4	**c**
136-138	97	2.5	4-ClC_6H_4	**d**
215-217	90	3	2-HOC_6H_4	**e**
229-231	91	3	4-HOC_6H_4	**f**
136-138	92	3.5	4-$CH_3C_6H_4$	**g**
148-150	94	2.5	3-$NO_2C_6H_4$	**h**
184-186	97	3.5	4-$NO_2C_6H_4$	**i**

[a]All products were characterized by ^1H, ^{13}C NMR, IR, MS and elemental analyses.

wherein we used a 0.02:1:1:1.2 ratio of *p*-TSOH, diethyl-3-oxoglutarate, aldehyde, and urea, respectively, furnished the respective DHPMs **4b-i** in good yields. The optimized results are summarized in Table 1.

In summary, we have described a mild, convenient method for the preparation of some new 6-substituted DHPMs by the Biginelli-like three-component cyclocondensation reaction of diethyl-3-oxoglutarate, aldehydes, and urea using cheap, non-toxic, very soluble in water, and easily available *p*-TsOH catalyst. Additionally, this new reaction might be a useful tool for high-throughput organic synthesis.

EXPERIMENTAL

Melting points were measured on the Electro thermal 9100 apparatus and are uncorrected. IR spectra were measured on a Shimadzu IR-470 Spectrophotometer. ^{1}H NMR and ^{13}C NMR spectra were determined on Bruker 500 DRX AVANCE instrument at 500 and 125 MHz, respectively. Elemental analyses were performed using a Heracus CHN-O-Rapid analyzer.

General Procedure for the Preparation of Ethyl 6-ethoxycarbonylmethyl-4-aryl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate 4a-i. A mixture of the appropriate aldehyde (1 mmol), diethyl-3-oxoglutarate (1 mmol), urea (1.2 mmol), and *p*-TsOH (0.02 mmol) was heated with stirring at 80 °C for the time period as indicated in Table 1. After completion of the reaction (TLC analysis), water was added to the mixture, and the crude products were directly filtrated through a sintered funnel. The crude products were further purified by recrystalization (EtOH) to afford products 4a-i (Table 1). All products were characterized by ^{1}H, ^{13}C NMR, IR, MS spectral data and by elemental analyses.

Ethyl 6-ethoxycarbonylmethyl-4-phenyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4a). White powder, yield 98%, mp 178-180 °C, IR(KBr), (ν_{max}/cm^{-1}): 3315, 3100, 2975, 1739, 1701, 1678, 1651.^{1}H NMR ($CDCl_3$) δ_H: 1.14 (t, 3H, *J*=7.2 Hz, CH_3), 1.26 (t, 3H, *J*=7.0 Hz, CH_3), 3.67 (d, 1H, *J*=16.8 Hz, CH_2), 3.94 (d, 1H, *J*=16.8 Hz, CH_2), 4.06 (q, 2H, *J*=7.2 Hz, CH_2), 4.18 (q, 2H, *J*=7.0 Hz, CH_2), 5.43 (s, 1H, CH), 6.02 (s, 1H, NH), 7.28-7.41 (m, 5H, arom), 8.88 (s, 1H, NH). ^{13}C NMR ($CDCl_3$) δ_C: 14.01 (CH_3), 14.0 (CH_3), 37.34 (CH_2), 55.74 (CH), 60.32 (CH_2), 61.37 (CH_2), 103.06 (C=C), 126.81, 128.07, 128.77 (arom), 142.56 (C=C), 143.28 (arom), 153.32, 165.07, 168.79 (CO). MS (m/z, %): 332 (M$^+$, 20), 286 (35), 275 (20), 255 (100), 245 (30), 229 (25), 213 (65), 209 (75), 186 (45), 181 (85), 153 (25), 132 (25), 106 (15), 77 (15). *Anal.* Calcd. for $C_{17}H_{20}N_2O_5$: C, 61.44; H, 6.02; N, 8.43. Found: C, 61.42; H, 6.00; N, 8.40.

Ethyl 6-ethoxycarbonylmethyl-4-(2-bromophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4b). White powder, yield 95%, mp 156-158 °C, IR (KBr), (ν_{max}/cm^{-1}): 3325, 3225, 3100, 2970, 1727, 1693, 1646. ^{1}H NMR ($CDCl_3$) δ_H: 1.06 (t, 3H, *J*=7.1 Hz, CH_3), 1.31 (t, 3H, *J*=7.1 Hz, CH_3), 3.56 (d, 1H, *J*=16.8 Hz, CH_2), 4.21 (d, 1H, *J*=16.8 Hz, CH_2), 4.01 (q, 2H, *J*=7.1 Hz, CH_2), 4.23 (q, 2H, *J*=7.1 Hz, CH_2), 5.77 (s, 1H, CH), 5.90 (s, 1H, NH), 7.14-7.58 (m, 4H, arom), 8.82 (s, 1H, NH). ^{13}C NMR ($CDCl_3$) δ_C: 13.93 (CH_3), 14.19 (CH_3), 37.39 (CH_2), 54.45 (CH), 60.34 (CH_2), 61.48 (CH_2), 101.27 (C=C), 122.77, 128.55, 129.71, 132.94 (arom), 140.80 (C=C), 144.44 (arom), 152.84, 164.77, 168.79 (CO). MS (m/z, %): 411 (M$^+$, 10), 367 (20), 365 (20), 339 (45), 337 (50), 293 (25), 291 (25), 286 (40), 255 (100), 239 (15), 209 (70), 181 (80), 155 (25), 137 (25), 102 (25), 75 (15). *Anal.* Calcd. for $C_{17}H_{19}N_2BrO_5$: C, 49.64; H, 4.62; N, 6.81. Found: C, 49.61; H, 4.59; N, 6.79.

Ethyl 6-ethoxycarbonylmethyl-4-(3-chlorophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4c). White powder, yield 95%, mp 138-140 °C, IR (KBr), (ν_{max}/cm^{-1}): 3303, 3115, 2980, 1738, 1701, 1674, 1651. ^{1}H NMR ($CDCl_3$) δ_H: 1.04 (t, 3H, *J*=7.1 Hz, CH_3), 1.18 (t, 3H, *J*=7.1 Hz, CH_3), 3.58 (d, 1H, *J*=16.9 Hz, CH_2), 3.86 (d, 1H, *J*=16.9 Hz, CH_2), 3.95 (q, 2H, *J*=7.0 Hz, CH_2), 4.10 (q, *J*=7.0 Hz, CH_2), 5.17 (d, 1H, *J*=3.2 Hz, CH), 7.28-7.40 (m, 4H), 7.87 (s, 1H, NH), 9.36 (s, 1H, NH). ^{13}C NMR ($CDCl_3$) δ_C: 14.32 (CH_3), 14.53 (CH_3), 37.54 (CH_2), 54.10 (CH), 60.10 (CH_2), 61.01 (CH_2), 100.64 (C=C), 125.71, 126.92, 127.90, 130.92, 133.46 (arom), 145.67 (C=C), 147.12 (arom), 152.90, 165.20, 169.19 (CO). MS (m/z, %): 366 (M$^+$, 20), 321 (30), 320 (25), 295 (30), 294 (25), 293 (100), 279 (20), 265 (30), 255 (100), 249 (25), 247 (50), 220 (30), 209 (65), 183 (45), 181 (75), 140 (20), 111 (15). *Anal.* Calcd. for $C_{17}H_{19}N_2ClO_5$: C, 55.66; H, 5.18; N, 7.64. Found: C, 55.66; H, 5.18; N, 7.64.

Ethyl 6-ethoxycarbonylmethyl-4-(4-chlorophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4d). White powder, yield 97%, mp 136-138 °C, IR (KBr), (ν_{max}/cm^{-1}): 3315, 3145, 2980, 1739, 1704, 1673, 1650. ^{1}H NMR ($CDCl_3$) δ_H: 1.05 (t, 3H, *J*=7.0 Hz, CH_3), 1.18 (t, 3H, *J*=7.0 Hz, CH_3), 3.62 (d, 1H, *J*=16.9 Hz, CH2), 3.81 (d, 1H, *J*=16.9 Hz, CH_2), 3.94 (q, 2H, J=7.0 Hz, CH_2), 4.10 (q, 2H, *J*=7.0 Hz, CH_2), 5.17 (s, 1H, CH), 7.34 (d, 2H, *J*=8.4 Hz, CH), 7.41 (d, 2H, J=8.4 Hz, CH) 7.84 (s, 1H, NH), 9.33 (s, 1H, NH). ^{13}C NMR ($CDCl_3$) δ_C: 14.33 (CH_3), 14.51 (CH_3), 37.49 (CH_2), 53.92 (CH), 60.03 (CH_2), 60.98 (CH_2), 100.93 (C=C), 128.84, 128.89, 132.46 (arom), 143.76 (C=C), 145.44 (arom), 152.16, 165.24, 169.07(CO). MS (m/z, %): 367 (M$^+$, 25), 321 (30), 320 (25), 293 (100), 279 (25), 255 (90), 247 (55), 220 (40), 209 (75), 181 (70), 166 (50), 140 (45), 110 (20). *Anal.* Calcd. for $C_{17}H_{19}N_2ClO_5$: C, 55.66; H, 5.18; N, 7.64. Found: C, 55.63; H, 5.14; N, 7.60.

Ethyl 6-ethoxycarbonylmethyl-4-(2-hydroxyphenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4e): White powder, yield 90%, mp 215-217 °C, IR (KBr), (ν_{max}/cm^{-1}): 3370, 3225, 3090, 2940, 1728, 1698, 1654, 1608, 1590. ^{1}H NMR (DMSO-d_6) δ_H: 1.01 (t, 3H, *J*=7.0 Hz, CH_3), 1.18 (t, 3H, *J*=7.0 Hz, CH_3), 3.62 (d, 1H, *J*=16.9 Hz, CH_2), 3.84 (d, 1H, *J*=16.9 Hz, CH_2), 3.90 (q, 2H, *J*=7.0 Hz, CH_2), 4.11 (q, 2H, *J*=7.0 Hz, CH_2), 5.52 (s, 1H, CH), 6.71 (t, 1H, *J*=7.3 Hz, CH), 6.80 (d, 1H, *J*=7.9 Hz, CH), 7.06 (t, 1H, *J*=6.9 Hz, CH), 7.17 (s, 1H, OH), 7.22 (d,1H, J=7.41 Hz, CH) 9.18 (s, 1H, NH), 9.68 (s, 1H, NH). ^{13}C NMR (DMSO-d_6) δC: 13.81 (CH_3), 14.02 (CH_3), 37.02 (CH_2), 48.29 (CH), 59.33 (CH_2), 60.47 (CH_2), 99.87 (C=C), 115.16, 118.73, 127.19, 128.38, 129.40 (arom), 144.87 (C=C), 152.15 (arom), 154.42, 164.96, 168.74 (CO). MS (m/z, %): 348 (M$^+$, 30), 302 (45), 301 (50), 275(50), 274 (45), 261 (50), 255 (100), 229 (80), 228 (50), 215 (50), 209 (55), 201 (50), 181 (70), 148 (25), 122 (25), 91 (20). *Anal.* Calcd. for $C_{17}H_{20}N_2O_6$: C, 58.62; H, 5.75; N, 8.05. Found: C, 58.58; H, 5.71; N, 8.01.

Ethyl 6-ethoxycarbonylmethyl-4-(4-hydroxyphenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4f). White powder, yield 91%, mp 229-231°C, IR (KBr), (ν_{max}/cm^{-1}): 3300, 3100, 2965, 1728, 1692, 1659, 1608, 1508. ^{1}H NMR (DMSO-d_6) δ_H: 0.90 (t, 3H, *J*=6.9 Hz, CH_3), 1.03 (t, 3H, *J*=7.0 Hz, CH_3),

3.45 (d, 1H, *J*=16.9 Hz, CH_2), 3.64 (d, 1H, *J*=16.9 Hz, CH_2), 3.78 (q, 2H, *J*=7.0 Hz, CH_2), 3.94 (q, 2H, *J*=6.9 Hz, CH_2), 4.92 (s, 1H, CH), 6.54 (d, 2H, *J*=8.1 Hz, CH), 6.98 (d, 2H, *J*=8.1 Hz, CH), 7.52 (s, 1H, OH), 9.04 (s, 1H, NH), 9.24 (s, 1H, NH). ^{13}C NMR (DMSO-d_6) δ_C: 13.86 (CH_3), 14.00 (CH_3), 36.98 (CH_2), 53.45 (CH), 59.41 (CH_2), 60.44 (CH_2), 101.35 (C=C), 114.92, 127.68, 134.92 (arom), 143.99 (C=C), 151.90 (arom), 156.60, 164.97, 168.67 (CO). MS (m/z, %): 348(M^+, 20), 319 (15), 303 (50), 302(50), 275 (100), 274 (50), 273 (50), 261 (75), 255 (75), 229 (100), 209 (70), 202 (70), 201 (50), 181 (75), 148 (50), 122 (45). *Anal.* Calcd. for $C_{17}H_{20}N_2O_6$: C, 58.62; H, 5.75; N, 8.05. Found: C, 58.60; H, 5.72; N, 8.02.

Ethyl 6-ethoxycarbonylmethyl-4-(4-methylphenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4g). White powder, yield 92%, mp 136-138 °C, IR (KBr), (ν_{max}/cm^{-1}): 3310, 3105, 2980, 1737, 1701, 1675, 1650. ^{1}H NMR ($CDCl_3$) δ_H: 1.15 (t, 3H, *J*=7.1 Hz, CH_3), 1.27 (t, 3H, *J*=7.1 Hz, CH_3), 2.34 (d, 3H, *J*=6.9, CH_3), 3.68 (d, 1H, *J*=16.8 Hz, CH_2), 3.94 (d, 1H, *J*=16.8 Hz, CH_2), 4.06 (q, 2H, *J*=7.1 Hz, CH_2), 4.19 (q, 2H, *J*=7.1 Hz, CH_2), 5.39 (s, 1H, CH), 5.69 (s, 1H, NH), 7.14 (d, 2H, *J*=7.6 Hz), 7.27 (d, 2H, *J*=7.9 Hz), 8.49 (s, 1H, NH).^{13}C NMR (CDCl3) δ_C: 14.05 (CH3), 14.12(CH_3), 21.14(CH_3), 37.36(CH_2), 55.46(CH), 60.29(CH_2), 61.35(CH_2), 103.14 (C=C), 126.71, 129.41, 137.74 (arom), 140.51 (C=C), 142.43 (arom), 153.35, 165.15, 168.81(CO). MS (m/z, %): 346 (M^+, 5), 301 (10), 300 (10), 273 (45), 255 (45), 227 (25), 209 (30), 181 (45), 146 (25), 110 (15), 91 (25), 81 (20), 65 (50), 56 (25), 45 (50), 43 (50), 32 (100), 31 (25), 30 (20). *Anal.* Calcd. for $C_{18}H_{22}N_2O_5$: C, 62.46; H, 6.36; N, 8.09. Found: C, 62.42; H, 6.31; N, 8.05.

Ethyl 6-ethoxycarbonylmethyl-4-(3-nitrophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4h). White powder, yield 94%, mp 148-150 °C, IR (KBr), (ν_{max}/cm^{-1}): 3235, 3105, 2975, 1736, 1714, 1692, 1645, 1517. ^{1}H NMR (DMSO-d_6) δ_H: 1.04 (t, 3H, *J*=6.2 Hz, CH_3), 1.16 (t, 3H, *J*=6.26 Hz, CH_3), 3.61 (d, 1H, *J*=16.9 Hz, CH_2), 3.85 (d, 1H, *J*=16.9 Hz, CH_2), 3.94 (q, 2H, *J*=7.0 Hz, CH_2), 4.10 (q, 2H, *J*=6.4 Hz, CH_2), 5.33 (s, 1H, CH), 7.66-7.69 (m, 1H, arom), 7.77-7.80 (m, 1H, arom), 7.97 (S, 1H, NH), 8.13-8.18 (m, 2H, arom), 9.45 (S, 1H, NH), ^{13}C NMR (DMSO-d_6) δ_C: 13.76, 13.97, 37.04, 53.53, 59.66, 60.53, 99.90, 121.31, 122.48, 130.14, 133.14, 145.57, 146.45, 147.77, 151.49, 164.60, 168.49, MS (m/z, %): 377 (M^+, 20), 360 (45), 332 (30), 304 (70), 274 (25), 258 (45), 255 (100), 231 (20), 209 (75), 181 (100), 163 (20), 137 (20), 110 (15). *Anal.* Calcd. for $C_{17}H_{19}N_3O_7$: C, 54.11; H, 5.04; N, 11.14. Found: C, 54.09; H, 5.01; N, 11.10.

Ethyl 6-ethoxycarbonylmethyl-4-(4-nitrophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4i). White powder, yield 97%, mp 184-186 °C, IR (KBr), (ν_{max}/cm^{-1}): 3305, 3095, 2970, 1727, 1702, 1692, 1654, 1527. ^{1}H NMR (DMSO-d_6) δ_H: 1.04 (t, 3H, *J*=6.9 Hz, CH_3), 1.17 (t, 3H, *J*=7.0 Hz, CH_3), 3.64 (d, 1H, *J*=16.9 Hz, CH_2), 3.82 (d, 1H, *J*=16.9 Hz, CH_2), 3.94 (q, 2H, *J*=6.9 Hz, CH_2), 4.10 (q, 2H, *J*=7.0 Hz, CH_2), 5.30 (d, 1H, CH), 7.59 (d, 2H, CH), 7.98 (s, 1H, NH), 8.21 (d, 2H, *J*=8.2 Hz, CH), 9.44 (s, 1H, NH). ^{13}C NMR (DMSO-d_6) δ_C: 13.80(CH_3), 14.02(CH_3), 37.00(CH_2), 53.60(CH), 59.69(CH_2), 60.55(CH_2), 99.82 (C=C), 123.78, 127.82 (arom), 145.53 (C=C), 146.79, 151.41(arom), 151.48, 164.60, 168.53(CO). MS (m/z, %): 377 (M^+, 25), 332 (45), 331 (30), 305 (20), 304 (98), 303 (45), 290 (25), 276 (25), 274 (25), 258 (55), 255 (95), 231 (30), 209 (90), 181 (100), 163 (30), 137 (25), 110 (15). *Anal.* Calcd. for $C_{17}H_{19}N_3O_7$: C, 54.11; H, 5.04; N, 11.14. Found: C, 54.08; H, 5.00; N, 11.11.

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