$LiClO_4$-Catalyzed One-Pot Synthesis of Dihydropyrimidinones: An Improved Protocol for Biginelli Reaction[1]

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Abstract: Lithium perchlorate efficiently catalyzes the three-component condensation reaction of aldehyde, β-keto ester and urea in refluxing acetonitrile to afford the corresponding dihydropyrimidinones in high yields under neutral conditions. LiOTf is also found to be an efficient catalyst for the synthesis of dihydropyrimidinones from aldehyde, β-keto ester and urea.

Keywords: Biginelli reaction, lithium perchlorate, lithium triflate, dihydropyrimidinones

Dihydropyrimidinones are an important class of compounds which are becoming increasingly interesting due to their therapeutic and pharmacological activities.[2] Several functionalized dihydropyrimidines have been found to exhibit a wide spectrum of biological effects[3] including antiviral, antitumor, antibacterial and anti-inflammatory activities. In addition, 4-aryldihydropyrimidines have emerged[4] as potent calcium channel blockers, antihypertensive, α_{1a}-adrenergic antagonists and neuropeptide antagonists. Further, dihydropyrimidinone-5-carboxylate core unit is found in many marine natural products[5] including batzelladine alkaloids, which are found to be potent-HIV gp-120-CD_4 inhibitors. The simple and direct method originally reported by Biginelli[6] for the synthesis of dihydropyrimidinones involves three component condensation reactions (i.e., aldehyde, β-keto ester and urea) often suffer from low yields practically in case of substituted aromatic and aliphatic aldehydes.[7] Even though, high yields could be achieved by following complex multi-step procedures,[8] these methods lack the simplicity of original one-pot Biginelli protocol. Therefore, Biginelli reaction continues to attract the attention of researchers for the discovery of a milder and efficient procedure for the synthesis of dihydropyrimidinones. Recently, $BF_3{\cdot}OEt_2$, PPE, KSF clay, $InCl_3$, $FeCl_3$ and lanthanide triflates are found to be effective[9] for this transformation. In addition, microwave irradiations are also found to accelerate[10] the reaction. However, many of these one-pot procedures generally require strong protic or Lewis acids, prolonged reaction times and high temperatures. Thus, the use of neutral alternative would extend the scope of useful one-pot Biginelli reaction for the synthesis of dihydropyrimidinones.

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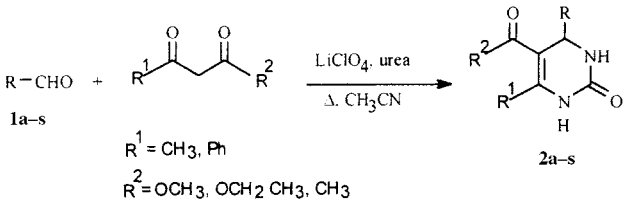
In recent years lithium perchlorate has received considerable attention as a powerful reaction medium for effecting various transformations[11] such as cycloaddition reactions, sigmatropic rearrangements, ring opening reactions of epoxides, glycosidation reactions, selective carbonyl protection as dithioacetals, Michael reactions and aldol reactions. Lithium perchlorate provides a convenient procedure to carry out the reactions under essentially neutral and workup conditions. In addition, lithium perchlorate is found to retain its activity even in the presence of nitrogen containing compounds. These special properties inherent to lithium perchlorate prompted us to explore this catalyst for the synthesis of dihydropyrimidinones.

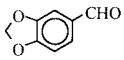
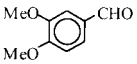
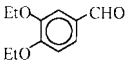
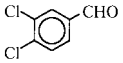
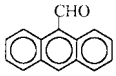
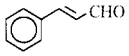
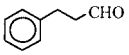
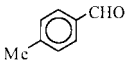
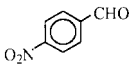
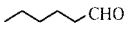
In this report, we describe a simple and practical method for the Biginelli reaction using catalytic amount of lithium perchlorate in acetonitrile. The reaction of benzaldehyde, ethyl acetoacetate and urea in the presence of 20 mol% lithium perchlorate in refluxing acetonitrile resulted in the formation of dihydropyrimidinone in 90% yield. Similarly, several aromatic, aliphatic and heterocyclic aldehydes reacted well under the present reaction conditions to give the corresponding dihydropyrimidinones in excellent yields (Scheme 1).



Scheme 1

The three component condensation reaction proceeded smoothly in refluxing acetonitrile and was also completed with in 5–8 h of reaction time. Many of the pharmacological relevant substitution patterns on the aromatic ring could be introduced with high efficiency. Aromatic aldehydes carrying either electron-donating or -withdrawing substituents afforded high yields of products in high purity. Acid sensitive aldehyde such as furfural worked well without the formation of any side products, which are normally observed either in the presence of protic or Lewis acids. In addition to its simplicity and milder reaction conditions, this method is effective even with aliphatic and α,β-unsaturated aldehydes which normally give poor yields in the presence of either protic or Lewis acids due to thier decomposition or polymerization under acidic

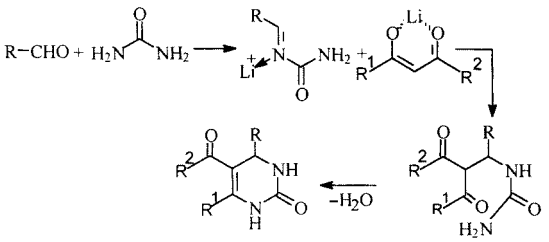
Table $LiClO_4$-Catalyzed Formation of Dihydropyrimidinones[a]

Substrate	R	R^1	R^2	Reaction Time (h)	Product	Yield[b] (%)	Mp (° C)[c] Found	Mp (° C)[c] Reported
1a		Me	OEt	6	**2a**	89	201–203	202[10]
1b		Me	OEt	7	**2b**	90	188–190	187[10]
1c		Me	OEt	8	**2c**	87	174–176	177[10]
1d		Me	OEt	8	**2d**	90	164–165	–
1e		Me	OEt	10	**2e**	85	222–223	223[9]
1f		Me	OEt	8	**2f**	87	247–248	246[10]
1g		Me	OEt	12	**2g**	81	251 (dec.)	–
1h		Me	OEt	6	**2h**	90	193–194	–
1i		Me	OEt	7	**2i**	83	229–231	232[9]
1j		Me	OEt	5	**2j**	90	207–208	208[10]
1k		Me	OEt	5	**2k**	85	206–208	205[9]
1l		Me	OEt	7	**2l**	81	182–184	186[7]
1m		Me	OEt	8	**2m**	87	237–238	237[7]
1n		Me	OEt	7	**2n**	89	170–171	172[9]
1o		Me	OEt	5	**2o**	90	206–207	208[9]
1p		Me	OEt	7	**2p**	82	161–162	163[10]
1q		Ph	OEt	10	**2q**	75	158	159[9]
1r		Me	OMe	6	**2r**	90	208–210	209[9]
1s		Me	Me	7	**2s**	88	241–243	–

[a] All products were characterized by ^1H NMR, IR and mass spectra and also by comparison of physical characteristics with authentic samples.
[b] Isolated and unoptimized yields.
[c] Melting points were uncorrected.

conditions. Another important feature of this procedure is the survival of a variety of functional groups such as olefins, ethers, esters, nitro and halides under the reaction conditions. The advantage of the use of $LiClO_4$ for this reaction lies in its simplicity. No additive or protic or Lewis acid is required in this procedure. This procedure not only preserves the simplicity of Biginelli reaction but also produces excellent yields of the dihydropyrimidinones. Thus

this procedure offers an easy access to substituted dihydropyrimidinones with a variety of substitution patterns. Among various solvents like acetonitrile, methanol, diethyl ether, THF and nitromethane used for this transformation, methanol and acetonitrile were the solvents of choice as best results were obtained with them. Several examples illustrating this novel and general method for the synthesis of dihydropyrimidinones are summarized in the Table. The reaction may proceed through the acylimine formation from aldehydes and urea, which is stabilized by lithium ion. Subsequent addition of the β-keto ester enolate to the acylimine followed by cyclodehydration would afford dihydropyrimidine (Scheme 2).



Scheme 2

In summary, the present method for the synthesis of DHPMs by lithium perchlorate catalyzed three-component condensation of β-keto ester, aldehyde and urea provides a novel and improved protocol for Biginelli reaction. In addition to its simplicity and neutral reaction conditions, this method is effective for aromatic, aliphatic, α,β-unsaturated and heterocyclic aldehydes.

3,4-Dihydro-2(1*H*)-pyrimidinones 2; General Procedure

A mixture of aldehyde (5 mmol), β-keto ester (5 mmol), urea (10 mmol) and LiClO4 or LiOTf (20% w/w of aldehyde) was stirred in refluxing MeCN for an appropriate time (Table). After completion of the reaction as indicated by TLC, the reaction mixture was poured into crushed ice and the resulting solid was filtered under suction and recrystallized from hot methanol to afford the pure product.

2a

Solid; mp 201–203 °C (Lit.[7] mp 202 °C).

^{1}H NMR (DMSO-d_6): δ = 1.17 (t, 3 H, *J* = 6.8 Hz), 2.30 (s, 3 H), 4.05 (q, 2 H, *J* = 6.8 Hz), 5.25 (s, 1 H), 7.15–7.35 (m, 6 H), 8.95 (br s, NH).

EIMS: *m*/*z* = 260 (M^+), 232, 184, 156, 138, 43.

IR (KBr): ν = 3245, 3120, 2950, 2925, 1720, 1702, 1640, 1590 cm^{-1}.

2b

Solid; mp 188–190 °C (Lit.[10] mp 187 °C).

^{1}H NMR (DMSO-d_6): δ = 1.15 (t, 3 H, *J* = 7.0 Hz), 2.35 (s, 3 H), 4.10 (q, 2 H, *J* = 7.0 Hz), 5.25 (s, 1 H), 5.50 (br s, 1 H, NH), 5.95 (s, 2 H), 6.75 (m, 3 H), 7.95 (br s, 1 H, NH).

EIMS: *m*/*z* (%) = 304 (M^+), 275, 258, 231, 183, 155, 137, 110, 69.

IR (KBr): ν = 3356, 3222, 2975, 1700, 1642, 1490, 1225, 1093, 795 cm^{-1}.

2c

Solid; mp mp 174–176 °C (Lit.[10] mp 177 °C).

^{1}H NMR (DMSO-d_6): δ = 1.15 (t, 3 H, *J* = 7.0 Hz), 2.30 (s, 3 H), 3.80 (s, 6 H), 4.05 (q, 2 H, *J* = 7.0 Hz), 5.25 (s, 1 H), 6.80 (m, 2 H), 6.85 (m, 1 H), 7.25 (br s, 1 H, NH), 8.95 (br s, 1 H, NH).

EIMS: *m*/*z* (%) = 320 (M^+), 292, 276, 247, 232, 183, 155, 137, 97, 69.

IR (KBr): ν = 3253, 3118, 2956, 1723, 1682, 1654, 1519, 1461, 1237, 1139, 1095, 790 cm^{-1}.

2d

Solid; mp 164–165 °C.

^{1}H NMR (DMSO-d_6): δ = 1.20 (t, 3 H, *J* = 6.9 Hz), 1.45 (t, 6 H, *J* = 6.9 Hz), 2.30 (s, 3 H), 4.05 (m, 6 H), 5.20 (s, 1 H), 6.80 (m, 2 H), 6.85 (m, 1 H), 7.15 (br s, 1 H, NH), 8.85 (br s, 1 H, NH). EIMS: m/z 348 (M^+), 319, 303, 275, 252, 231, 183, 155, 137.

IR (KBr): ν = 3256, 3118, 2978, 2933, 1717, 1705, 1653, 1519, 1226, 1091, 775 cm^{-1}.

2e

Solid; mp 222–223 °C (Lit.[9] mp 223 °C).

^{1}H NMR (DMSO-d_6): δ = 1.10 (t, 3 H, *J* = 6.9 Hz), 2.25 (s, 3 H), 4.05 (q, 2 H, *J* = 6.9 Hz), 5.20 (s, 1 H), 7.15 (m, 1 H), 7.35 (m, 2 H), 7.40 (br s, 1 H, NH), 9.0 (br s, 1 H, NH).

EIMS: *m*/*z* (%) = 328 (M^+), 300, 254, 183, 155, 137, 69.

IR (KBr): ν = 3352, 3225, 3109, 2975, 1697, 1645, 1469, 1400, 1321, 1228, 1099, 1032, 790 cm^{-1}.

2f

Solid; mp 247–248 °C (Lit.[10] mp 246 °C).

^{1}H NMR (DMSO-d_6): δ = 0.90 (t, 3 H, *J* = 7.0 Hz), 2.40 (s, 3 H), 3.90 (q, 2 H, *J* = 7.0 Hz), 6.10 (s, 1 H), 7.40–7.55 (m, 5 H), 7.80 (t, 1 H, *J* = 8.2 Hz), 7.85 (d, 1 H, *J* = 8.2 Hz), 8.30 (d, 1 H, *J* = 8.2 Hz), 9.10 (br s, NH).

EIMS: *m*/*z* (%) = 310 (M^+), 217, 176, 133, 119, 91, 69.

IR (KBr) : ν = 3245, 3118, 2977, 1698, 1647, 1431, 1231, 1088, 790 cm^{-1}.

2g

Solid; mp 251 °C (dec.).

^{1}H NMR (DMSO-d_6): δ = 1.19 (t, 3 H, *J* = 7.0 Hz), 2.28 (s, 3 H), 4.18 (q, 2 H, *J* = 7.0 Hz), 5.30 (br s, 1 H), 7.15–7.75 (m, 9 H).

EIMS: *m*/*z* = 360 (M^+), 332, 316, 288, 240, 212, 177.

IR (KBr): ν = 3247, 2978, 1716, 1696, 1497, 1261 cm^{-1}.

2h

Solid; mp 193–194 °C.

^{1}H NMR (DMSO-d_6): δ = 1.15 (t, 3 H, *J* = 6.8 Hz), 2.35 (s, 3 H), 4.10 (q, 2 H, *J* = 6.8 Hz), 5.35 (s, 1 H), 5.80 (br s, 1 H, NH), 6.85 (m, 1 H), 7.05 (m, 5 H), 7.45 (m, 3 H), 8.40 (br s, 1 H, NH).

EIMS: *m*/*z* = 352 (M^+), 323, 279, 183, 155, 137, 91, 69.

IR (KBr): ν = 3243, 3113, 2981, 1712, 1654, 1582, 1487, 1245, 1097, 785 cm^{-1}.

2i

Solid; mp 229–231 °C (Lit.[9] mp 232–235 °C).

^{1}H NMR (DMSO-d_6): δ = 1.05 (t, 3 H, *J* = 7.0 Hz), 2.50 (s, 3 H), 3.95 (q, 2 H, *J* = 7.0 Hz), 4.25 (d, 1 H, *J* = 6.0 Hz), 6.05 (dd, 1 H, *J* = 16.4 Hz), 6.2 (d, 1 H, *J* = 16.4 Hz), 7.25 (m, 5 H), 7.45 (d, 1 H, NH, *J* = 1.7 Hz), 8.95 (br s, NH).

EIMS: *m*/*z* (%) = 286 (M^+), 252, 224, 196, 149, 84.

IR (KBr): ν = 3335, 3242, 3098, 2978, 1689, 1642, 1492, 1373, 1218, 1121, 785 cm^{-1}.

2j
Solid; mp 207–208 °C (Lit.[10] mp 208 °C).

^1H NMR (DMSO-d_6): δ = 1.15 (t, 3 H, *J* = 6.9 Hz), 2.30 (s, 3 H), 4.20 (q, 2 H, *J* = 6.9 Hz), 5.30 (d, 1 H, *J* = 1.8 Hz), 6.85 (m, 2 H), 7.15 (m, 1 H), 7.30 (br s, NH), 8.90 (br s, NH).

EIMS: *m*/*z* = 266 (M^+), 238, 221, 193, 145, 117, 83.

IR (KBr): ν = 3245, 3120, 1720, 1690, 1535, 1460 cm^{-1}.

2k
Solid; mp 206–208 °C (Lit.[9] mp 205 °C).

^1H NMR (DMSO-d_6): δ = 1.17 (t, 3 H, *J* = 7.0 Hz), 2.35 (s, 3 H), 4.18 (q, 2 H, *J* = 7.0 Hz), 5.30 (d, 1 H, *J* = 2.0 Hz), 6.10 (d, 1H, *J* = 2.1 Hz), 6.33 (s, 1 H), 7.30 (m, 1 H), 7.82 (br s, NH), 9.21 (br s, NH).

EIMS: *m*/*z* = 250 (M^+), 221, 205, 177, 134, 80, 67.

IR (KBr): ν = 3268, 3118, 1716, 1681, 1591, 1485 cm^{-1}.

2l
Solid; mp 182–184 °C (Lit.[7] mp 186–187 °C).

^1H NMR (DMSO-d_6): δ = 1.25 (t, 3 H, *J* = 7.2 Hz), 1.80 (m, 2 H), 2.25 (s, 3 H), 2.60–2.70 (m, 2 H), 4.15 (m, 2 H), 4.25 (m, 1 H), 7.10–7.25 (m, 6 H), 8.80 (br s, 1 H, NH).

EIMS: *m*/*z* (%) = 288 (M^+), 183, 155, 137, 91.

IR (KBr): ν = 3248, 3117, 2975, 1724, 1652, 1495, 1287, 1226, 1092, 778 cm^{-1}.

2m
Solid; mp 237–238 °C (Lit.[7] mp 235–237 °C).

^1H NMR (DMSO-d_6): δ = 1.05 (m, 4 H), 1.25 (t, 3 H, *J* = 6.8 Hz), 1.45 (m, 3 H), 1.75 (m, 4 H), 2.3 (s, 3 H), 4.15 (m, 3 H), 6.15 (br s, 1 H, NH), 8.45 (br s, 1 H, NH).

EIMS: *m*/*z* (%) = 266 (M^+), 183, 137, 155, 40.

IR (KBr): ν = 3236, 3118, 2920, 2850, 1726, 1702, 1647, 1450, 1230, 1095, 789 cm^{-1}.

2n
Solid; mp 170–171 °C (Lit.[9] 172 °C).

^1H NMR (DMSO-d_6): δ = 1.25 (t, 3 H, *J* = 6.9 Hz), 2.32 (s, 3 H), 2.43 (s, 3 H), 4.10 (q, 2 H, *J* = 6.9 Hz), 5.40 (d, 1H, *J* = 2.2 Hz), 6.95–7.20 (m, 4 H), 7.61 (br s, NH), 9.10 (br s, NH).

EIMS: *m*/*z* = 274 (M^+), 245, 229, 201, 155, 138, 91.

IR (KBr): ν = 3240, 3110, 1695, 1665, 1560 cm^{-1}.

2o
Solid; mp 206–207 °C (Lit.[9] 208 °C).

^1H NMR (DMSO-d_6): δ = 1.20 (t, 3 H, *J* = 7.0 Hz), 2.28 (s, 3 H), 4.05 (q, 2 H, *J* = 7.0 Hz), 5.39 (d, 1H, *J* = 2.4 Hz), 7.52 (d, 2H, *J* = 8.7 Hz), 7.35 (br s, NH), 8.23 (d, 2 H, *J* = 8.7 Hz), 9.35 (br s, NH).

EIMS: *m*/*z* = 306 (M^+), 276, 232, 183, 155, 137.

IR (KBr): ν = 3235, 3109, 2975, 1701, 1645, 1558 cm^{-1}.

2p
Solid; mp 161–162 °C (Lit.[10] mp 163 °C).

^1H NMR (DMSO-d_6): δ = 0.95 (t, 3 H, *J* = 6.8 Hz), 1.25–1.40 (m, 9 H), 1.55 (m, 2 H), 2.30 (s, 3 H), 4.15 (m, 2 H), 4.25 (m, 1 H), 6.05 (br s, 1 H, NH), 8.55 (br s, 1 H, NH).

EIMS: *m*/*z* = 253 (M^+), 209, 183, 155, 137, 91, 40.

IR (KBr): ν = 3249, 2933, 1730, 1646, 1433, 1331, 1288, 1086, 779 cm^{-1}.

2q
Solid; mp 158 °C (Lit.[9] mp 159 °C).

^1H NMR (DMSO-d_6): δ = 0.90 (t, 3 H, *J* = 6.8 Hz), 3.85 (q, 2 H, *J* = 6.8 Hz), 5.45 (d, 1 H, *J* = 1.8 Hz), 6.55 (br s, 1 H, NH), 7.30–7.40 (m, 10 H), 7.80 (br s, 1 H, NH).

EIMS: *m*/*z* (%) = 322 (M^+), 294, 278, 249, 185, 157, 138, 91, 77, 69.

IR (KBr): ν = 3215, 3085, 2978, 1697, 1650 cm^{-1}.

2r
Solid; mp 210–211 °C (Lit.[9] mp 209 °C).

^1H NMR (DMSO-d_6): δ = 2.25 (s, 3 H), 3.60 (s, 3 H), 5.25 (d, 1 H, *J* = 1.7 Hz), 7.20–7.35 (m, 6 H), 9.05 (br s, 1 H, NH).

EIMS: *m*/*z* (%) = 246 (M^+), 219, 161, 134, 91, 69, 42.

IR (KBr): ν = 3255, 3120, 2958, 1725, 1680, 1653, 1518, 1462, 1238, 1140, 1095, 790 cm^{-1}.

2s
Solid; mp 241–243 °C.

^1H NMR (DMSO-d_6): δ = 2.10 (s, 3 H), 2.25 (s, 3 H), 5.30 (d, 1 H, *J* = 1.8 Hz), 7.20–7.30 (m, 5 H), 7.60 (br s, 1 H, NH), 9.05 (br s, 1 H, NH).

EIMS: *m*/*z* (%) = 230 (M^+), 188, 154, 144, 115, 77, 43.

IR (KBr): ν = 3286, 3251, 2920, 1703, 1675, 1598, 1414, 1327, 1235, 1105, 997, 965, 768 cm^{-1}.

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