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## Gypsum-Catalyzed One-Pot Synthesis of 3,4-Dihydropyrimidin-2(1*H*)-ones Under Solvent-Free Conditions

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**Abstract:** In view of the emerging importance of the green chemistry principles in chemical and pharmaceutical industries, we disclose, herein, a new economic approach producing the biologically active dihydropyrimidinones in good yields using the solventless one-pot Biginelli condensation in the presence of gypsum as an environmental friendly and recycled catalyst.

**Keywords:** 3,4-Dihydropyrimidin-2(1*H*)-ones, Solvent-free reactions, Gypsum, MCCs.

### Introduction

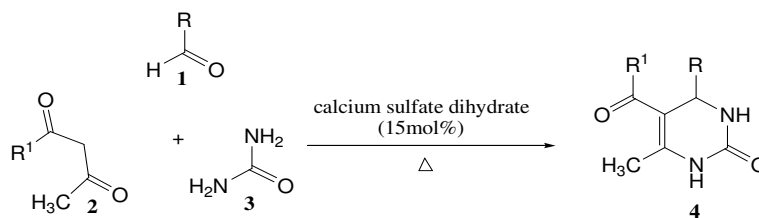
Due to environmental awareness, the development of simple, efficient and environmentally benign chemical processes or methodologies for the synthesis of complex structures are the major challenges for chemists' world over<sup>1</sup>. The one-pot multi-component condensations (MCCs) represent a possible instrument to perform a near ideal synthesis because they possess several qualities, namely the possibility of building-up complex molecules with maximum simplicity and brevity. Therefore, in recent years<sup>2</sup>, MCCs have attracted considerable interest and have been transformed into one of the most efficient, economic and environmentally friendly tools for combinatorial and parallel synthesis<sup>3</sup>.

Today, the one-pot Biginelli reaction, first described more than a century ago<sup>4</sup>, is considered as one of the most important multi-component reactions for generating complex compounds that possess diverse therapeutic and pharmacological properties<sup>4,6</sup>, therefore, their

synthesis has been the focus of much interest for organic and medicinal chemists. Thus, several improved synthetic methods have been reported using Lewis acids as well as protic acids as promoters under classical reflux<sup>7-13</sup> or solvent free conditions<sup>14-20</sup> and microwave<sup>21-24</sup> or ultrasonic irradiation<sup>25</sup>.

Due to the ever-mounting environmental concern in the field of chemistry, it is advisable to easily recover and recycle catalysts, especially toxic metallic ones. Additionally, the challenge for a sustainable environment calls for clean procedures to avoid harmful organic solvents.<sup>26</sup> However, a number of the reported protocols for Biginelli reaction use volatile toxic organic solvents, as reaction media and many heavy and unrecovered metallic salts as catalysts which are not acceptable in the context of green synthesis as they result in the pollution to environment to some extent. Therefore, there is still room for further searches for better catalysts that could be superior to the existing ones with regard to toxicity, handling and recyclability. In this respect, we are interested to introduce potential catalyst to overcome these limitations.

In pursuit of developing a methodology for the preparation of these biologically important compounds<sup>27-29</sup>, we herein describe a mild and efficient synthesis of dihydropyrimidinones, *via* the solventless one-pot Biginelli condensation protocol, using gypsum, a mineral composed of calcium sulfate with two molecules of water, as a recyclable, non toxic, easy available and potential Lewis acid catalyst (Scheme 1).



**Scheme 1**

## Experimental

Unless specified all solvents and reagents were of reagent grade and used without further purification. Melting points were determined in a capillary tube and are uncorrected. <sup>1</sup>H and <sup>13</sup>C NMR spectra were recorded as solutions in DMSO-*d*<sub>6</sub> and chemical shifts are reported in parts per million (ppm) on a BRUKER AVANCE DPX spectrometer at 250 and 62.9 MHz respectively using TMS as internal standard. Spectral patterns are designated as s, singlet; d, doublet; dd, double doublet; t, triplet; br, broad, m, multiplet. Coupling constants are reported in hertz (Hz). IR spectra were obtained as potassium bromide (KBr) pellets with a Shimadzu FT IR-8201 PC spectrometer.

### *General procedure for the synthesis of 3,4-dihydropyrimidinones (4)*

In a typical experiment, a mixture of aldehyde (5 mmol, 535 mg), urea (5 mmol, 300 mg), 1,3-dicarbonyl compound (5 mmol, 650 mg) and CaSO<sub>4</sub>·2H<sub>2</sub>O (15 mol%), was heated with stirring at 100 °C for the required time (Table 2) in a water bath. The reaction was monitored by TLC using ethyl acetate/hexane (4/6). After completion (100% conversion using aldehyde as the blank reactant), the resulting solidified mixture was diluted with hot EtOH (20 mL). The resulting suspension was filtered through a Büchner funnel to recover the catalyst and the filtrate was poured into ice cold water (30 mL). The resulting precipitate was collected and recrystallized in EtOH to give the pure product **4**. The recovered catalyst dried for investigation of the recyclability of the catalyst.

All the products **4a-4x** were characterized by IR,  $^1\text{H}$ - $^{13}\text{C}$  NMR, and also by comparing their physical characteristics with those in the literature.

#### Physical and spectral data of unknown products

##### *4-(3,5-Dimethoxyphenyl)-5-ethoxycarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one(4e)*

M.p. 205-208 °C,  $^1\text{H}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 9.26 (s, 1H, N1-H), 7.78 (d,  $J$  = 1.87, 1H, N3-H), 6.37-6.42 (m, 3H,  $\text{CH}_{\text{arom}}$ ), 5.10 (d,  $J$  = 3.38, 1H, CH), 3.71(s, 6H,  $\text{Ar-OCH}_3$ )<sub>2</sub>, 3.6 (s, 3H,  $\text{COOCH}_3$ ), 2.25 (s, 3H,  $\text{CH}_3$ );  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 166.3, 160.9, 152.7, 149.3, 147.2, 104.8, 99.0, 98.7, 55.5, 53.9, 51.3, 18.2.

##### *5-Ethoxycarbonyl-4-(3-fluorophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one(4k)*

M.p. 209-211 °C,  $^1\text{H}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 9.28 (s, 1H, N1-H), 7.88 (s, 1H, N3-H), 7.35-7.44 (m, 1H,  $\text{CH}_{\text{arom}}$ ), 6.98-7.13 (m, 3H,  $\text{CH}_{\text{arom}}$ ), 5.19 (d,  $J$  = 3.42 Hz, 1H, CH), 3.48 (s, 3H,  $\text{OCH}_3$ ), 2.27 (s, 3H,  $\text{CH}_3$ );  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 164.5, 166.2, 152.5, 149.6, 147.8, 131.0, 122.6, 114.4, 113.5, 98.9, 53.7, 51.3.

##### *4-(3,5-Dimethoxyphenyl)-5-methoxycarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one(4n)*

M.p. 191-193 °C,  $^1\text{H}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 9.26 (s, 1H, N1-H), 7.78 (d,  $J$  = 1.87, 1H, N3-H), 6.37-6.42 (m, 3H,  $\text{CH}_{\text{arom}}$ ), 5.10 (d,  $J$  = 3.38, 1H, CH), 3.71(s, 6H,  $\text{Ar-OCH}_3$ )<sub>2</sub>, 3.6 (s, 3H,  $\text{COOCH}_3$ ), 2.25 (s, 3H,  $\text{CH}_3$ );  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 166.3, 160.9, 152.7, 149.3, 147.2, 104.8, 99.0, 98.7, 55.5, 53.9, 51.3, 18.2.

##### *4-(3-Fluorophenyl)-5-methoxycarbonyl-6-methyl-3,4-dihydropyrimidin-2(1H)-one (4q)*

M.p. 226-229 °C,  $^1\text{H}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 9.28 (s, 1H, N1-H), 7.88 (s, 1H, N3-H), 7.35-7.44 (m, 1H,  $\text{CH}_{\text{arom}}$ ), 6.98-7.13 (m, 3H,  $\text{CH}_{\text{arom}}$ ), 5.19 (d,  $J$  = 3.42 Hz, 1H, CH), 3.48 (s, 3H,  $\text{OCH}_3$ ), 2.27 (s, 3H,  $\text{CH}_3$ );  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 164.5, 166.2, 152.5, 149.6, 147.8, 131.0, 122.6, 114.4, 113.5, 98.9, 53.7, 51.3.

##### *5-Acetyl-4-(3,5-dimethoxyphenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (4t)*

M.p. 196-198 °C,  $^1\text{H}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 9.22 (s, 1H, N1-H), 7.76 (s, 1H, N3-H), 6.49 (s, 3H,  $\text{CH}_{\text{arom}}$ ), 5.22 (d,  $^3J$  = 3.22 Hz, 1H, CH), 3.71 (s, 6H,  $\text{Ph-(OCH}_3$ )<sub>2</sub>), 2.24 (s, 3H,  $\text{COCH}_3$ ), 2.12 (s, 3H,  $\text{CH}_3$ );  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 194.8, 161.0, 152.6, 148.7, 146.7, 109.5, 105.1, 98.7, 55.5, 54.1, 30.7, 19.3.

##### *5-Acetyl-4-(2,4-dichlorophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one (4u)*

M.p. 225-227 °C,  $^1\text{H}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 9.35 (s, 1H, N1-H), 7.84 (s, 1H, N3-H), 7.61 (s, 1H,  $\text{CH}_{\text{arom}9}$ ), 7.28 (d,  $J$  = 8.4 Hz, 1H,  $\text{CH}_{\text{arom}11}$ ), 7.42 (d,  $J$  = 8.4 Hz, 1H,  $\text{CH}_{\text{arom}12}$ ), 5.63 (d,  $J$  = 3.27 Hz, 1H, CH), 2.34 (s, 3H,  $\text{COCH}_3$ ), 2.09 (s, 3H,  $\text{CH}_3$ );  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 194.3, 176.3, 149.5, 140.5, 133.3, 133.2, 130.3, 129.4, 128.4, 108.8, 51.6, 30.7, 19.4.

##### *5-Acetyl-4-(3-fluorophenyl)-6-methyl-3,4-dihydropyrimidin-2(1H)-one(4w)*

M.p. 254-256 °C,  $^1\text{H}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 9.27 (s, 1H, N1-H), 7.91 (s, 1H, N3-H), 7.35-7.44 (m, 3H,  $\text{CH}_{\text{arom}}$ ), 7.35-7.44 (m, 1H,  $\text{CH}_{\text{arom}}$ ), 5.28 (d,  $J$  = 3.50 Hz, 1H, CH), 2.30 (s, 3H,  $\text{COCH}_3$ ), 2.15 (s, 3H,  $\text{CH}_3$ );  $^{13}\text{C}$  NMR (DMSO- $d_6$ ):  $\delta$  (ppm) = 194.6, 164.5, 152.5, 149.1, 147.6, 147.5, 131.0, 130.9, 122.8, 109.8, 53.6, 30.9, 19.4.

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

M.p. 208-210 °C, <sup>1</sup>H NMR (DMSO-d<sub>6</sub>): δ (ppm) = 9.19 (s, 1H, N1-H), 7.72 (s, 1H, N3-H), 7.39-6.87 (m, 4H, CH<sub>arom</sub>), 5.48 (s, 1H, CH), 3.82 (s, 3H, COCH<sub>3</sub>), 2.11 (s, 3H, CH<sub>3</sub>); <sup>13</sup>C NMR (DMSO-d<sub>6</sub>): δ (ppm) = 195.0, 156.7, 152.6, 148.6, 131.5, 129.4, 127.2, 120.8, 111.7, 108.1, 55.8, 49.1, 30.1, 19.1.

**Results and Discussion**

To evaluate the effect of the catalyst under different reaction conditions, the reaction of benzaldehyde, ethyl acetoacetate and urea was selected as a model reaction and the results are presented in Table 1. The best result was achieved by carrying out the reaction with (1:1:1) mole ratio of benzaldehyde, ethyl acetoacetate and urea at 100 °C.

Although ethanol and acetonitrile (Table 1, entries 4 and 5) were found to be effective, considering stringent environmental regulations, solvent free reactions are desired. Therefore, we intended the Biginelli reaction under solvent free conditions to determine the optimum amount of catalyst (Table 1, entries 6-10). The activity of the recycled catalyst was also examined under the optimized conditions and the desired product were obtained in 85, 84, 85% yields after 1-3 runs, respectively (Table 1, entry 11).

**Table 1.** Effect of catalysts under different reaction conditions for condensation of benzaldehyde, ethyl acetoacetate and urea<sup>a</sup>.

Entry	Solvent	Amounts of CaSO <sub>4</sub> .2 H <sub>2</sub> O, mol %	Refluxing time, min	Yield, % <sup>b</sup>
1	THF	20	180	55
2	Toluene	20	180	70
3	Diethyl ether	20	180	65
4	Acetonitrile	20	180	88
5	Ethanol	20	180	75
6	Free solvent	20	120	85
7	Free solvent	05	120	45
8	Free solvent	10	120	70
9	Free solvent	15	120	85
10	Free solvent	25	120	85
11 <sup>c</sup>	Solvent-free	15	120	85,84, 85

<sup>a</sup>Reaction condition: benzaldehyde (5 mmol), ethylacetoacetate (5 mmol), urea (5 mmol) and catalyst in solvent, (5 mL, entries 1-5) at reflux. <sup>b</sup>Isolated yield. <sup>c</sup>Catalyst was recycled for three times.

Promoted by this success, we extended this reaction of urea with a range of other 1,3-dicarbonyl compounds and aldehydes, under similar conditions, furnishing the corresponding 3,4-dihydropyrimidin-2(1H)-ones (**4a-x**) in good yields. The generality and scope of the reaction are summarized in Table 2.

For comparison purposes, yields obtained for **4a**, **4c** and **4f** using Hekmatshoar<sup>37</sup> conditions (NiSO<sub>4</sub>.7H<sub>2</sub>O in CH<sub>3</sub>CN, reflux 1.5 h, method B), Shaabani<sup>38</sup> conditions (LiHSO<sub>4</sub>, solvent free, 4 h, method C) and Reddy<sup>39</sup> conditions (Montmonillonite KSF, 48 h, method D) are given in Table 2. It is remarkable to note that the present method (CaSO<sub>4</sub>.2H<sub>2</sub>O, solvent free at 100 °C, method A) gave good yields in shorter reaction times.

**Table 2.** CaSO<sub>4</sub>·2H<sub>2</sub>O-catalyzed synthesis of dihydropyrimidinones under solvent free conditions

Entry	R	R <sup>1</sup>	Time, min	DHPM <sup>a</sup>	Yield, %				Melting point, °C	
					A <sup>b</sup>	B <sup>c</sup>	C <sup>d</sup>	D <sup>e</sup>	Found <sup>e</sup>	Reported
1	C <sub>6</sub> H <sub>5</sub> -	OC <sub>2</sub> H <sub>5</sub>	90	<b>4a</b>	85	80	57	82	204-206	206-207 <sup>30,31</sup>
2	4-(CH <sub>3</sub> O)-C <sub>6</sub> H <sub>4</sub> -	OC <sub>2</sub> H <sub>5</sub>	95	<b>4b</b>	80				204-205	203-204 <sup>30</sup>
3	4-(CH <sub>3</sub> )-C <sub>6</sub> H <sub>4</sub> -	OC <sub>2</sub> H <sub>5</sub>	80	<b>4c</b>	85	80	66		218-220	215-216 <sup>30</sup>
4	2-(CH <sub>3</sub> )-C <sub>6</sub> H <sub>4</sub> -	OC <sub>2</sub> H <sub>5</sub>	120	<b>4d</b>	80				204-205	207-208 <sup>15</sup>
5	3,5-(CH <sub>3</sub> O) <sub>2</sub> C <sub>6</sub> H <sub>3</sub> -	OC <sub>2</sub> H <sub>5</sub>	90	<b>4e</b>	80				205-208	-
6	4-Cl-C <sub>6</sub> H <sub>4</sub> -	OC <sub>2</sub> H <sub>5</sub>	105	<b>4f</b>	82	80			211-213	212-214 <sup>15</sup>
7	4-N-(CH <sub>3</sub> ) <sub>2</sub> C <sub>6</sub> H <sub>4</sub> -	OC <sub>2</sub> H <sub>5</sub>	120	<b>4g</b>	75				257-259	256-258 <sup>32</sup>
8	4-(HO)-C <sub>6</sub> H <sub>4</sub> -	OC <sub>2</sub> H <sub>5</sub>	90	<b>4h</b>	85				230-233	230-232 <sup>33</sup>
9	3-(Cl)-C <sub>6</sub> H <sub>4</sub> -	OC <sub>2</sub> H <sub>5</sub>	60	<b>4i</b>	80				194-196	193-195 <sup>32</sup>
10	2,4-(Cl) <sub>2</sub> -C <sub>6</sub> H <sub>3</sub> -	OC <sub>2</sub> H <sub>5</sub>	90	<b>4j</b>	85				247-248	248-250 <sup>34</sup>
11	3-F-C <sub>6</sub> H <sub>4</sub> -	OC <sub>2</sub> H <sub>5</sub>	60	<b>4k</b>	85				209-211	-
12	C <sub>6</sub> H <sub>5</sub> -	OCH <sub>3</sub>	65	<b>4l</b>	80				211-214	211-213 <sup>35</sup>
13	4-(CH <sub>3</sub> O)-C <sub>6</sub> H <sub>4</sub> -	OCH <sub>3</sub>	120	<b>4m</b>	85				194-196	193-196 <sup>15</sup>
14	3,5(CH <sub>3</sub> O) <sub>2</sub> C <sub>6</sub> H <sub>3</sub> -	OCH <sub>3</sub>	55	<b>4n</b>	82				191-193	-
15	2-(CH <sub>3</sub> )-C <sub>6</sub> H <sub>4</sub> -	OCH <sub>3</sub>	60	<b>4o</b>	85				239-241	240-242 <sup>30,31</sup>
16	2,4-(Cl) <sub>2</sub> -C <sub>6</sub> H <sub>3</sub> -	OCH <sub>3</sub>	120	<b>4p</b>	80				257-259	255-257 <sup>35</sup>
17	3-F-C <sub>6</sub> H <sub>4</sub> -	OCH <sub>3</sub>	90	<b>4q</b>	80				226-229	-
18	4-(Cl)-C <sub>6</sub> H <sub>4</sub> -	OCH <sub>3</sub>	75	<b>4r</b>	88				203-206	204-205 <sup>35</sup>
19	C <sub>6</sub> H <sub>5</sub> -	CH <sub>3</sub>	65	<b>4s</b>	85				233-235	232-235 <sup>18</sup>
20	3,5-(CH <sub>3</sub> O) <sub>2</sub> -	CH <sub>3</sub>	60	<b>4t</b>	70				196-198	-
21	2,4-(Cl) <sub>2</sub> -C <sub>6</sub> H <sub>3</sub> -	CH <sub>3</sub>	120	<b>4u</b>	85				225-227	-
22	4-(Cl)-C <sub>6</sub> H <sub>4</sub> -	CH <sub>3</sub>	75	<b>4v</b>	85				227-229	230-232 <sup>36</sup>
23	3-F-C <sub>6</sub> H <sub>4</sub> -	CH <sub>3</sub>	70	<b>4w</b>	80				254-256	-
24	2-(CH <sub>3</sub> )-C <sub>6</sub> H <sub>4</sub> -	CH <sub>3</sub>	55	<b>4x</b>	85				208-210	-

<sup>a</sup>All products were characterized by <sup>1</sup>H, <sup>13</sup>C NMR and IR spectroscopy. <sup>b</sup>Method A: using our new conditions (cat. CaSO<sub>4</sub>·2H<sub>2</sub>O, solvent free at 100 °C). <sup>c</sup>Method B: using R. Hekmatshoar conditions (Cat. NiSO<sub>4</sub>·7H<sub>2</sub>O in CH<sub>3</sub>CN, reflux 1.5 h)<sup>37</sup>. <sup>d</sup>Method C: using A. Shaabani conditions (cat. LiHSO<sub>4</sub>, solvent free, 4 h)<sup>38</sup>. <sup>e</sup>Method D: using K. R. Reddy conditions (Montmonillonite KSF, 48 h)<sup>39</sup>. <sup>f</sup>Melting points are uncorrected.

Moreover, the condensation protocol is fairly general and several functionalities including chloro, hydroxyl, methoxy, fluoro and amino do survive during the course of the reaction to give the corresponding DHPMs in moderate to good yields with high purity.

## Conclusion

We have developed a potential catalyst for the synthesis of an important class of products known under the acronym of DHPMs. The easy handling and workup combined with the easy available, non expensive, non toxic and recycled catalyst; good yields, short reaction periods and the needless reaction solvents are salient features of the catalyst that render the presented procedure relatively environmentally acceptable. Moreover, the compatibility with various functional groups should make the present method useful and important in addition to the known methodologies for the Biginelli reaction.

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