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Solvent-free Biginelli Reaction Catalyzed to Synthesis of Biologically Active 3,4-dihydropyrimidin-2-(1*H*) –ones/ thiones Derivatives

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Abstract

A convenient and highly efficient procedure for the synthesis of 3,4-dihydropyrimidin-2-(1*H*)-one/thione derivatives *via* one-pot three-component Biginelli condensation of aryl aldehydes,urea/thiourea and ethyl/methyl acetoacetate in the presence of $ZnSO_4.7H_2O$ as an efficient, readily and inexpensive catalyst under solvent-free conditions have been studied. This protocol has advantages such as readily available and non-toxic catalyst, short reaction times, good to high yields, solvent-free conditions, facile reaction profiles, high atom-economy and simple work-up.

Keywords: *$ZnSO_4.7H_2O$, 3,4-dihydropyrimidin-2-(1H)-one/thione derivatives, Low-cost and non-toxic catalyst, Solvent-free conditions.*

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Introduction

Synthesis of heterocyclic compounds has attracted great interests due to their wide applicability in life and nature. The compounds with Pyrimidinone derivatives ring systems have been used as calcium channel blockers, α-1a-antagonists [1], a mitotic kinesin Eg5 inhibition [2], anti cancer (Mal3-101) [3], anti HIV agent [4], antibacterial and antifungal [5], antiviral [6], antioxidative [7]. The representatives such as batzelladines, ptilomycalines and crambescidines exhibit many biological activities such as anticancer, antifungal, anti HIV etc [8].

In the last decades, a number of methodologies for the preparation of these compounds have been reported,including various catalysts such as calcium fluoride [9], copper(II)sulfamate [10], bakers' yeast [11], hydrotalcite [12], hexaaquaaluminium (III) tetrafluoroborate [13], TBAB [14] and copper (II) tetrafluoroborate [15], Copper (II) acetate [16], [Btto][*p*-TSA] [17], triethylammonium acetate [18], *p*-dodecylbenzenesulfonic acid [19], TMSPTPOSA [20]. Some of the limitations of these methodologies are low yields, toxic organic solvents and catalyst, harsh reaction conditions and expensive materials. In recent years, the design and development of bioactive heterocyclic compounds synthesis performed through multi-component reactions (MCRs) [21-25], involving three or more reactants in one-pot, have attracted considerable interest since such processes; improve atom economy, efficiency and convergence. Based on the above-considerations and in continuation of our efforts to develop eco-safe methodologies [26-28], we reported herein $ZnSO_4.7H_2O$ as an efficient, readily, inexpensive and eco-safe catalytic system for the synthesis of 3, 4-dihydropyrimidin-2-(1*H*)-one/thione derivatives *via* three-component Biginelli [29] reaction between *β*-keto esters, aldehyde derivatives and urea/thiourea under solvent-free conditions with good to high yields and short reaction times under solvent-free conditions.

Experimental

General

Melting points all compounds were determined using an Electro thermal 9100 apparatus. ^{1}H NMR spectra were recorded on a Bruker DRX-400 Avance instrument with DMSO-d_6 as solvents. All reagents and solvents were purchased from Merck, Fluka and Acros chemical companies were used without further purification.

General procedure for preparation of 3, 4-dihydropyrimidin-2-(1H)-one/thione derivatives **(4a- p)**

A mixture of aldehyde derivatives (**1**, 1.0 mmol) and urea/thiourea (**2**, 1.5 mmol), ethyl/methyl acetoacetate (**3**, 1.0 mmol) were heated under solvent-free conditions at 80 °C for appropriate time

in the presence of $ZnSO_4.7H_2O$ (25 mol %). After completion of the reaction (by thin layer chromatography TLC) the mixture was cooled to room temperature before cold water was added. The precipitation was separated by filtration and the solid washed with ethanol (3×2 mL). The catalyst soluble in ethanol and was separated. Then, the precipitated recrystallized from ethanol to afford the pure products (**4a- p**). Spectra data products are represented below:

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

Yield: 88%; m.p. 198-200 °C; ^{1}H NMR (400 MHz, DMSO-d_6): 1.10 (3H, t, *J*= 7.2 Hz, $\underline{CH_3}CH_2$), 2.26 (3H, s, CH_3), 3.99 (2H, q, *J*=7.2 Hz, CH_2O), 5.15 (1H, s, $H_{benzylic}$), 7.26 (3H, d, *J*= 7.2 Hz, H_{Ar}), 7.33 (2H, t, *J*=7.2 Hz, H_{Ar}), 7.76 and 9.21 (2H, 2s, 2NH).

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

Yield: 90%; m.p. 176-178 °C; ^{1}H NMR (300 MHz, DMSO-d_6): 1.11 (3H , t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.25 (3H, s, CH_3), 3.99 (2H, q, *J*=9.6 Hz, CH_2O), 5.14 (1H, s, CHN), 7.13-7.20 (2H, m, ArH), 7.24-7.29 (2H, m, ArH), 7.78 and 9.25 (2H, 2s, 2NH).

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

Yield: 89%; m.p. 253-255 °C; ^{1}H NMR (300 MHz, DMSO-d_6): 1.12 (3H, t, *J*= 9.2 Hz, $\underline{CH_3}CH_2$), 2.26 (3H, s, CH_3), 2.85 (6H, s, $2CH_3$), 3.99 (2H, q, *J*=9.2 Hz, CH_2O), 5.04 (1H, s, CHN), 6.66 (2H, d, *J*=11.6 Hz, ArH), 7.42 (2H, d, *J*=11.6Hz, ArH), 7.61 and 9.11 (2H, 2s, 2NH).

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

Yield: 76%; m.p. 235-237 °C; ^{1}H NMR (400 MHz, DMSO-d_6): 1.11 (3H , t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.50 (3H, s, CH_3), 3.98 (2H, q, *J*=9.6 Hz, CH_2O), 5.04 (1H, s, $H_{benzylic}$), 6.68-7.04 (4H, m, H_{Ar}), 7.64 and 9.13 (2H, 2s, 2NH), 9.35 (1H, s, OH).

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

Yield: 91%; m.p. 209-211 °C; ^{1}H NMR (400 MHz, DMSO-d_6): 1.10 (3H, t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.28 (3H, s, CH_3), 3.99 (2H, q, *J*=9.6 Hz, CH_2O), 5.27 (1H, s, $H_{benzylic}$), 7.50-7.53 (2H, m, H_{Ar}), 7.23 (2H, d, *J*= 9.2Hz, H_{Ar}), 7.92 and 9.38 (2H, 2s, 2NH).

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

Yield: 78%; m.p. 212-214 °C; ^{1}H NMR (300 MHz, DMSO-d_6): 1.10 (3H , t, *J*= 9.2 Hz, $\underline{CH_3}CH_2$), 2.26 (3H, s, CH_3), 4.00 (2H, q, *J*=9.2 Hz, CH_2O), 5.14 (1H, s, CHN), 7.25 (2H, d, *J*= 11.2 Hz, ArH), 7.41 (2H, d, *J*= 11. 2 Hz, ArH), 7.79 and 9.27 (2H, 2s, 2NH).

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

Yield: 80%; m.p. 189-191°C; ^{1}H NMR (300 MHz, DMSO-d_6): 1.11 (3H , t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.26 (3H, s, CH_3), 4.01 (2H, q, *J*=9.6 Hz, CH_2O), 5.15 (1H, s, CHN), 7.19-7.26 (2H, m, ArH), 7.31-7.41 (2H, m, ArH), 7.83 and 9.30 (2H, 2s, 2NH).

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

Yield: 84%; m.p. 221-223 °C; ^{1}H NMR (400 MHz, DMSO-d_6): 1.00 (3H , t, *J*= 9.2 Hz, $\underline{CH_3}CH_2$), 2.31 (3H, s, CH_3), 4.02 (2H, q, *J*=9.2 Hz, CH_2O), 5.63 (1H, s, $H_{benzylic}$), 7.25-7.34 (3H, m, H_{Ar}), 7.41 (1H, d, *J*=8.8 Hz, H_{Ar}), 7.73 and 9.29 (2H, 2s, 2NH).

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

Yield: 88%; m.p. 204-206 °C; ^{1}H NMR (400 MHz, DMSO-d_6): 1.11 (3H, t, *J*= 7.2 Hz, $\underline{CH_3}CH_2$), 2.25 (3H, s, CH_3), 2.27 (3H, s, CH_3), 3.99 (2H, q, *J*=7.2 Hz, CH_2O), 5.11 (1H, s, CHN), 7.13 (4H, s, ArH), 7.70 and 9.17 (2H, 2s, 2NH).

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

Yield: 85%; m.p. 202-203°C; ^{1}H NMR (400 MHz, DMSO-d_6): 1.11 (3H , t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.24 (3H, s, CH_3), 3.73 (3H, s, OCH_3), 3.99 (2H, q, *J*=9.6 Hz, CH_2O), 5.09 (1H, s, $H_{benzylic}$), 6.89 (2H, d, *J*= 8.4Hz, H_{Ar}), 7.15 (2H, d, *J*= 8.8Hz, H_{Ar}), 7.70 and 9.18 (2H, 2s, 2NH).

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

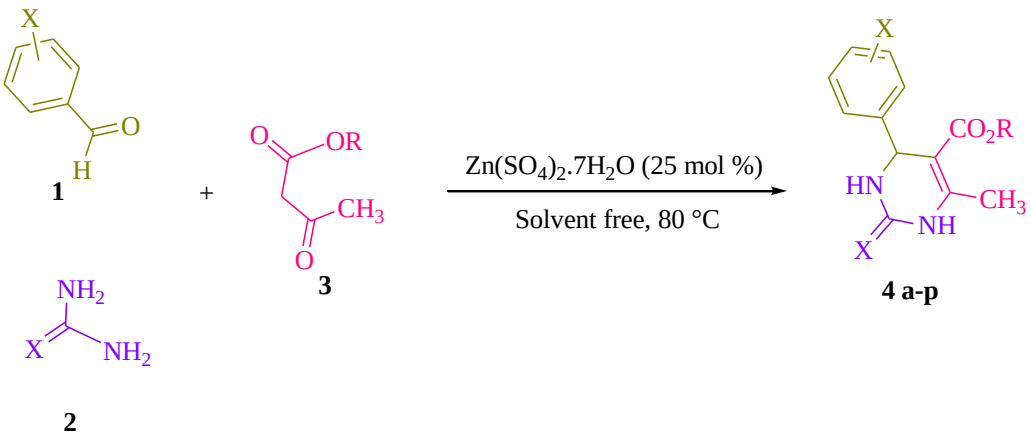
Yield: 89%; m.p. 204-206°C; ^{1}H NMR (400 MHz, DMSO-d_6): 1.13 (3H , t, *J*= 9.6 Hz, $\underline{CH_3}CH_2$), 2.26 (3H, s, CH_3), 3.74 (3H, s, OCH_3), 4.01 (2H, q, *J*=9.6 Hz, CH_2O), 5.13 (1H, s, $H_{benzylic}$), 6.78-6.86 (3H, m, H_{Ar}), 7.26 (1H, t, *J*= 10.4Hz, H_{Ar}), 7.76 and 9.20 (2H, 2s, 2NH).

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

Yield: 83%; m.p. 250-252 °C; ^{1}H NMR (400 MHz, DMSO-d_6): 2.31 (3H, s, CH_3), 3.46 (3H, s, OCH_3), 5.62 (1H, s, $H_{benzylic}$), 7.28-7.34 (3H, m, H_{Ar}), 7.42 (1H, d, *J*=7.2 Hz, H_{Ar}), 7.72 and 9.36 (2H, 2s, 2NH).

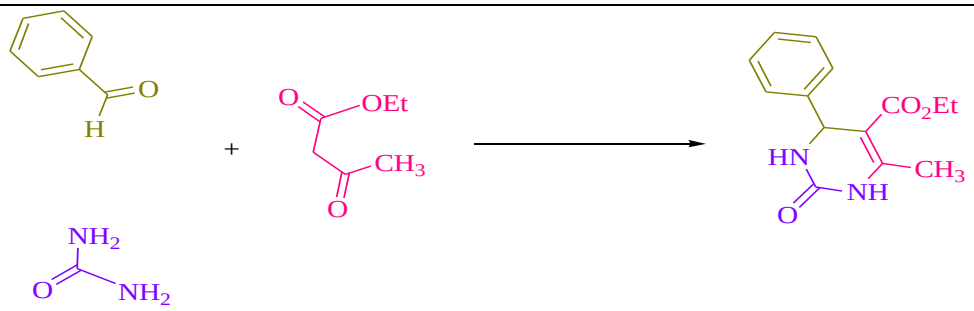
Results and discussion

In order to carry out preparation of 3,4-dihydropyrimidin-2-(1*H*)-one/thione derivativesin a more efficient way, the reaction of benzaldehyde (1.0 mmol), urea (1.5 mmol) and ethyl acetoacetate (1.0 mmol)were selected as a model system under thermal solvent-free conditions to find optimization of reaction conditions. The preparation of 3,4-dihydropyrimidin-2-(1*H*)-one/thione derivatives was studied at different amounts of $ZnSO_4.7H_2O$ as a low-cost, readily and efficient catalyst (5, 10, 15, 20, 25 and 30 mol%) and different reaction temperatures (rt, 40, 60, 70, 80 and 90°C) (Table 1). The reaction did not occur in the absence of catalyst (Table 1, entry 1). The best result was obtained using 25 mol% of $ZnSO_4.7H_2O$ at 80 °C (Table 1, entry 6). Using the optimized reaction conditions, the scope and efficiency of these procedures were explored for the synthesis of a wide variety of 3,4-dihydropyrimidin-2-(1*H*)-one/thione derivatives (Scheme 1). The results are summarized in Table 2. As shown in Table 2, the direct one-pot three-component reactions worked well with a variety of arylaldehydes including those bearing electron-withdrawing and electron-donating groups such as Cl, Me, NO_2, OMe, …, and the desired compounds were obtained in good to high yields. This methodology offers significant improvements such as simplicity in operation with no necessity of chromatographic purification steps, low-cost and eco-friendly catalyst.



Scheme1. Synthesis of 3, 4-dihydropyrimidin-2-(1*H*)-one/thione derivatives.

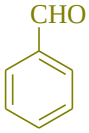
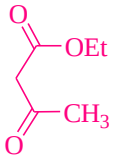
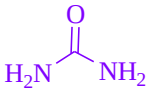
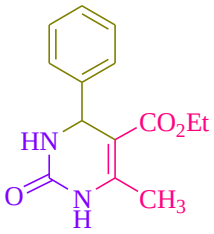
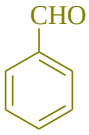
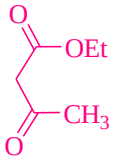
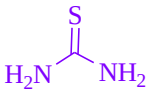
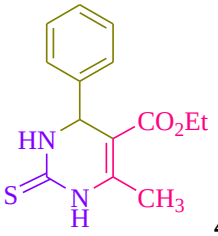
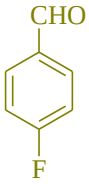
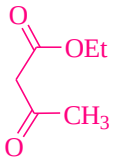
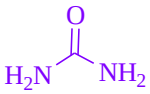
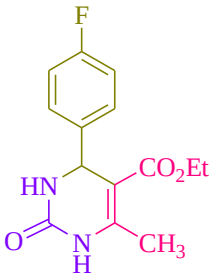
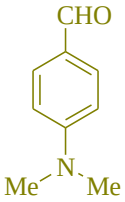
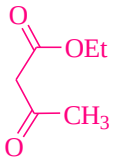
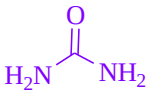
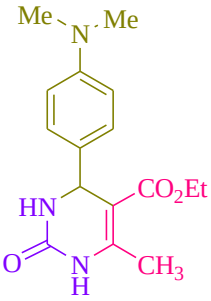
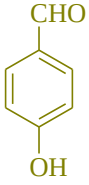
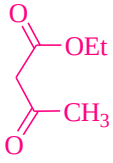
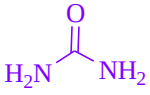
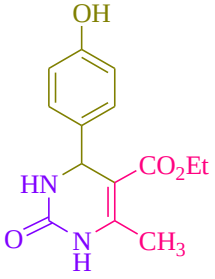
Table 1. Optimization of the reaction condition on the synthesis of **4a**[a]

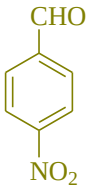
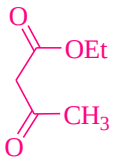
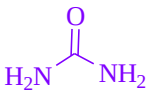
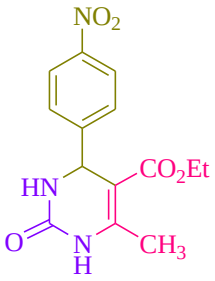
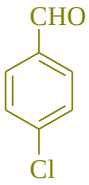
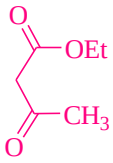
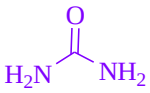
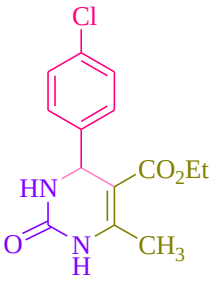
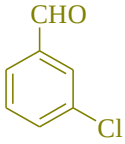
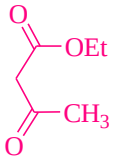
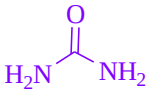
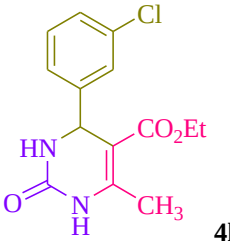
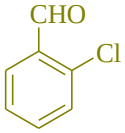
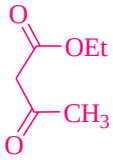
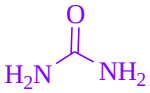
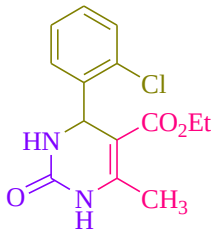
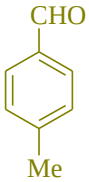
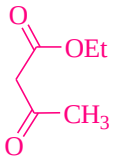
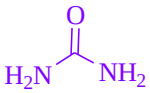
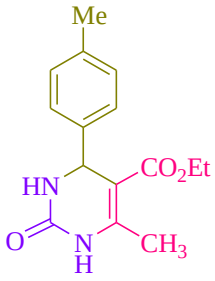
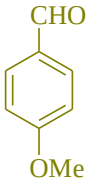
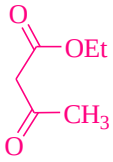
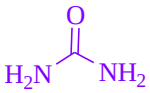
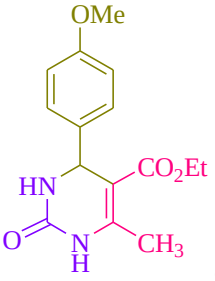


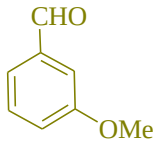
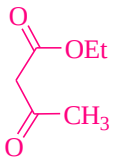
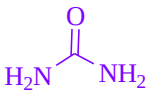
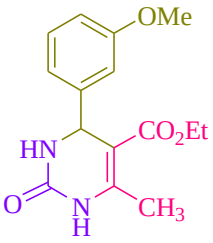
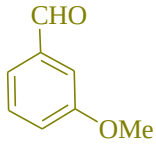
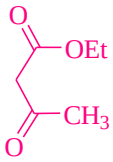
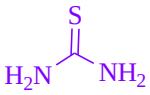
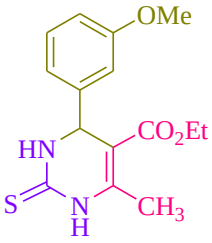
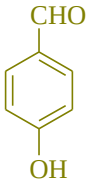
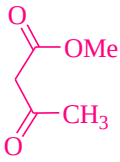
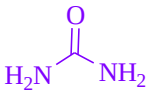
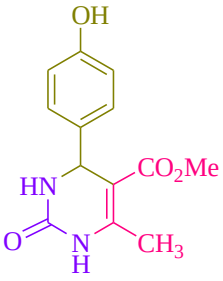
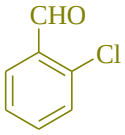
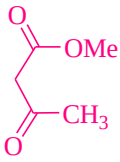
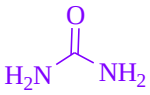
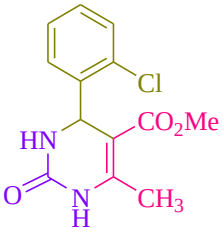
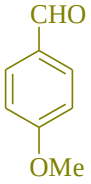
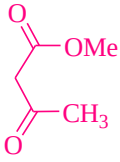
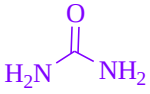
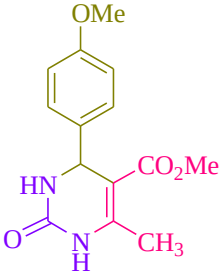
Entry	$ZnSO_4.7H_2O$ (mol %)	Temperature (°C)	Time (min)	Isolated Yields (%)
1	Catalyst free	80	360	No product
2	5	80	90	27
3	10	80	75	41
4	15	80	50	60
5	20	80	30	75
6	**25**	**80**	**20**	**88**
7	25	rt	360	No product
8	25	40	65	42
9	25	60	40	53
10	25	70	30	68
11	25	90	20	89
12	30	80	20	90

[a] Reaction conditions: benzaldehyde (1.0 mmol), ethyl acetoacetate (1.0 mmol), urea (1.5mmol) and $ZnSO_4.7H_2O$were heated under various temperatures for the appropriate time.

Table 2. Synthesis of 3,4-dihydropyrimidin-2-(1*H*)-one/thione derivatives.

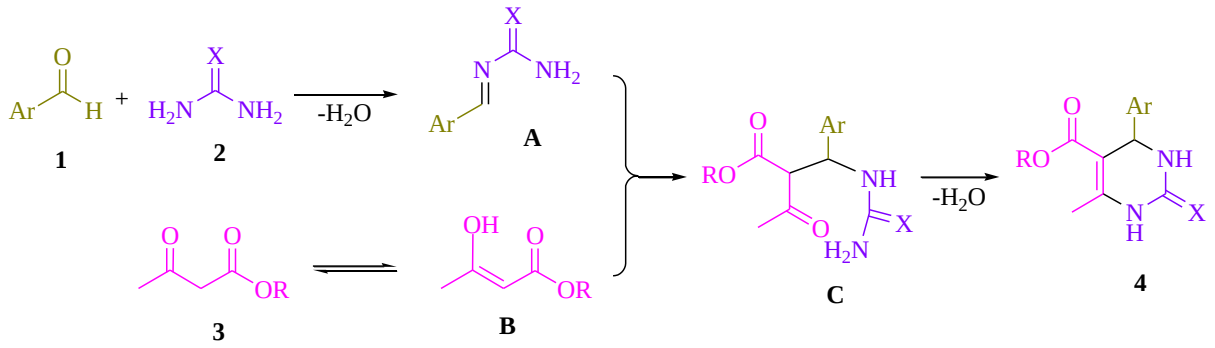
Entry	Substrate	Substrate	Substrate	Product [a]	Time (min)	Yield %[b]	m.p.˚C	Lit. m.p.˚C
1				 **4a**	20	88	198-200	200-202 [10]
2				 **4b**	20	85	207-209	208-210 [10]
3				 **4c**	20	90	176-178	174-176 [14]
4				 **4d**	25	89	253-255	254-256 [16]
5				 **4e**	30	76	235-237	234-236 [16]

6				 **4f**	15	91	209-211	207-209 [10]
7				 **4g**	30	78	212-214	214-215 [12]
8				 **4h**	30	80	189-191	191-193 [10]
9				 **4i**	25	84	221-223	220-223 [10]
10				 **4j**	15	88	204-206	204-205[11]
11				 **4k**	20	85	202-203	203-205 [17]

12				 **4l**	20	89	204-206	205-206[11]
13				 **4m**	20	82	151-153	150-151 [11]
14				 **4n**	30	79	245-247	245-246 [11]
15				 **4o**	20	83	250-252	248-252 [10]
16				 **4p**	20	87	190-192	190-194 [15]

[a] Isolated yield.[b]Reaction conditions: Aryl aldehyde derivatives (1.0 mmol), ethyl/methyl acetoacetate (1.0 mmol), urea/thiourea (1.5 mmol) and $ZnSO_4.7H_2O$ (25 mol %) were heated at 80 °C.

And proposed mechanistic route of 3,4-dihydropyrimidin-2-(1*H*)-one/thione synthesis in the presence of $ZnSO_4.7H_2O$ are shown in scheme 2. In this probable mechanism, the $ZnSO_4.7H_2O$ catalyzed Biginelli condensation via acylimin intermediate (**A**) is presented in Scheme 2. The reaction of aldehydes (**1**) and urea (**2**) generates an acylimin intermediate (**A**), which further reacts with the activated 1,3-dicarbonyl compound (**B**) producing an open-chain ureide (**C**) undergoing subsequent cyclization and dehydration to give the major product (**4**).



Scheme2. Proposed mechanistic route for the synthesis of 3,4-dihydropyrimidin-2-(1*H*)-ones/thiones.

Comparison of catalytic ability some of catalysts reported in the literature for synthesis of 3, 4-dihydropyrimidin-2-(1*H*)-one/thione derivatives are shown in Table 3. This study reveals that $ZnSO_4.7H_2O$ has shown its extraordinary potential to be an alternative eco-safe, readily and highly efficient catalyst for the Biginelli reaction. In Addition, the use of solvent-free conditions with good to high yields and short reaction times in the reaction with both urea and thiourea are the notable advantages this eco-safe and simple procedure.

Table 3. Comparison of catalytic ability some of catalysts reported in the literature for synthesis of 3, 4-dihydropyrimidin-2-(1*H*)-one/thione derivatives[a]

Entry	Catalyst	Conditions	Time/Yield (%)	References
1	Baker's yeast	Room temperature	24h/84	[11]
2	Hydrotalcite	Solvent-free, 80 °C	35 min/84	[12]
3	$[Al(H_2O)_6](BF_4)_3$	MeCN, Reflux	20 h/81	[13]
4	$Cu(BF_4)_2.xH_2O$	Room temperature	30 min/90	[15]

5	[Btto][*p*-TSA]	Solvent-free, 90 °C	30 min/96	[17]
6	triethylammonium acetate	Solvent-free, 70 °C	45min/90	[18]
7	*p*-dodecylbenzenesulfonic acid	Solvent-free, 80 °C	3 h/94	[19]
8	TMSPTPOSA	EtOH/Reflux	3 h/95	[20]
9	**$ZnSO_4.7H_2O$**	**Solvent-free, 80 °C**	**20 min/88**	**This work**

[a]Based on the three-component reaction of benzaldehyde, ethyl acetoacetate and urea.

Conclusion

In summary, a cost-effective, facile and one-pot synthetic route for the Biginelli synthesis of 3,4-dihydropyrimidin-2-(1*H*)-one/thione derivatives catalyzed by $ZnSO_4.7H_2O$ under solvent-free conditions was developed. Highly efficient, easy to handle and low-cost catalyst,high atom-economy, one-pot procedure, short reaction times, good to high yields, simple work-up and facile reaction profile are key features of this methodology.

Acknowledgments

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