## SiO$_2$-I catalyzed ultrasound assisted one-pot three-component synthesis of 3,4-dihydropyrimidine-2-(1H)-ones/-thiones

Abstract

An energy efficient and one-pot three-component reaction for a competent preparation of 3,4-dihydropyrimidine-2-(1H)-ones/-thiones using SiO$_2$-I (Silica Iodide) as a reliable and reusable heterogeneous catalyst has been developed. The reaction proceeds *via* condensation of araldehydes, urea/thiourea and ethyl acetoacetate in an ethanol as a medium under ultrasonic condition to afford the target molecules in the best yields. The reaction proceeds in 30 min; and the heterogeneous catalyst: SiO$_2$-I, has showed high proficiency in performing this one-pot multicomponent Biginelli reaction through recoverability, recyclability and minimization of the waste.
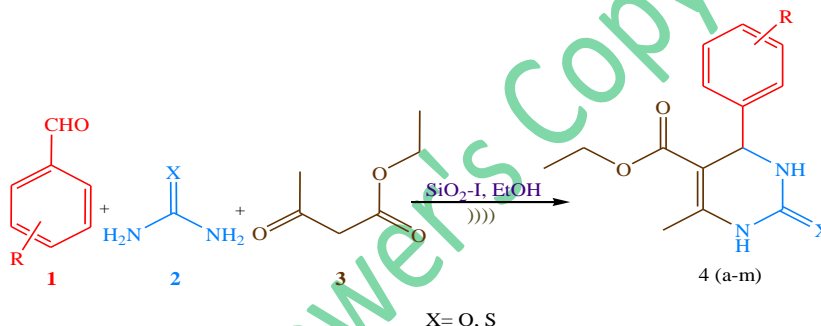
Keywords: 3,4-Dihydropyrimidine-2-(1H)-ones/-thiones; SiO$_2$-I; araldehydes; urea/thiourea; ethyl acetoacetate; ultrasonication.

1. Introduction

Ultrasonication has been recognized as a substantial mode for green and sustainable synthetic organic processes, [1–3] and provides several advantageous like tumbling time, minimization of waste, very high yields of the product by enhancing the rate and lowering activation energy in micro surroundings. [4,5] The viability of one-pot multicomponent reactions (MCRs) under ultrasonication usingthe heterogeneous SiO$_2$-Ias a catalyst has shown considerable progress in their efficiency from implementation and environmental points of view. [6,7]One of the significant, vital and biologically essential heterocyclic scaffolds is pyrimidine, and numerous natural products are found to possess this most familiar key motif. Molecules which are having pyrimidine skeleton are found to exhibit unique and valuable therapeutic properties, and play an essential role in biochemical processes [8]. Pyrimidine and its derivatives have occupied a characteristic place in the field of organic and medicinal chemistry in the design of biologically active compounds since decades [9]. They possess wide-range of

pharmacological activities such as: calcium channel blocking property, as antifungals, antimalarials, antibacterials, antihypertensive, anti-inflammatory agents, and are the inhibitors of fatty acid transporters, α1a-antagonists, neuropeptide Y antagonists and work as mitotic kinesin inhibitors [10–14]. A few marine polycyclic alkaloids such as: batzelladine A and B, ptilocaulin and saxitoxin, due to the presence of dihydropyrimidine (DHPM) moiety in themare known to inhibit the binding of HIV gp-120 to CD4 cells in AIDS therapy [15]. Functionalized dihydropyrimidine analogues of novel 4-aryl-5-isopropoxycarbonyl-6-methyl-3,4-dihydropyrimidinones have emerged as anti-microbical agents [16].

In 1893, Biginelli synthesized 3,4-dihydropyrimidine-2(1*H*)-ones for the first time *via* an acid catalysed one-pot multicomponent reaction of an aldehyde, α,β-ketoester and urea [17]. The reported protocol has drawbacks such as: prolonged reaction times, low yield of the products and tolerance of sensitive functional groups throughout the reaction. This difficulty has led to the growth of one-pot multi-step synthetic approaches towards the synthesis of DHPMs and a number of modifications have been developed to carry out the Biginelli condensation reaction with various types of catalysts such as: Lewis acids [18], Brønsted acids [19], polymer supported catalysts [20], ion-exchange resins [21], phase transfer catalysis [19], ionic liquids [20], Brønsted bases [22], solid phase reagents [23] and heterogeneous catalysts [24], and various conditions such as microwave irradiation [25], ultrasonication [26], using other green approach synthesis [27], under solvent-free condition [28], grindstone technique [29], ZnO nanoparticles embedded in SBA-15 [30] and dendrimer-attached phosphotungstic acid nanoparticles immobilized on nano silica [31]. Many of these protocols involve harsh reaction conditions, tedious work-up procedures, long reaction durations, involve expensive reagents, non-recoverability of the catalysts, strongly acidic and basic conditions, environmental contamination, undesirable yields and non-tolerance of certain functional groups. Thus, the progress of devising alternate, mild and eco-friendly methods, which can overcome those drawbacks are of great significance towards the synthesis of DHPs. The ultrasonication approach offers copious advantages such as: better yields of the target molecules, superior reaction rates, works under mild and energy efficient reaction conditions, and minimization of waste takes place when compared with conventional methods.

2. Results and discussion

To determine the viability and generality of the present SiO_2-I [32] catalyzed ultrasonic one-pot MCR, the influence of reaction parameters such as reaction medium, temperature, catalyst, feed ratio of the catalyst and the energy efficiency were studied to examine their roles in increasing the rate of the reactions and yield of the products by taking 3-methoxybenzaldehyde (1 mmol), urea (1 mmol) and ethyl acetoacetate (1 mmol) as model substrates.

2.1 Effect of catalyst

Various catalysts were screened under different reaction conditions (28 °C, reflux temperature of the solvent and ultrasonication) to authenticate the right selection and the results of this study are shown in **Table 1**. To study the activity of the catalyst, the present one-pot three-component reaction was first carried out without catalyst wherein a maximum yield of only 35% could be obtained (entry 1). It was further observed that, the yield of the product hardly enhanced in the presence of catalysts like NaI, SiO_2, TiO_2, $CeCl_3$, $ZnCl_2$, K_2CO_3 and ZnO (entries 2–8), whereas the use of SiO_2–I as a catalyst accelerated the reaction and gave the product in excellent yield (96%, entry 9). Hence, SiO_2–I under ultrasonic condition was selected for our further studies.

Table 1: Effect of various catalysts on the synthesis of 4-(3′-methoxyphenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (**4a**) under ultrasonic condition.

Entry	Catalyst	Time (min)	Yield (%)
1	No catalyst	360	20
2	NaI[a]	90	27
3	SiO_2[a]	90	24
4	TiO_2[a]	90	48
5	$CeCl_3$[a]	90	59
6	$ZnCl_2$[a]	90	62
7	K_2CO_3[a]	90	76
8	Nano ZnO[a]	90	87
9	**SiO_2-I**[b]	**30**	**96**

[a]10 mol% catalyst in EtOH (5 mL); [b]0.1 g in EtOH (5 mL);

2.2 Solvent effect

We then started the evaluation of the effect of various solvents such as: polar aprotic and polar protic solvents and nonpolar solvents; and the result are presented in the **Table 2**. To demonstrate the effect of the solvent, the model reaction of 3-methoxybenzaldehyde (1 mmol), urea (1 mmol) and ethyl acetoacetate (1 mmol) was first studied under solvent-free condition to get a maximum yield of 35% under ultrasonic condition (entry 1), In nonpolar solvents the reaction rate was found to be very slow and we recorded low yields (entries 2,3), and in the case of polar solvents such as: 1,4-dioxane, DMSO, DMF and THF, moderate yields were obtained (entries 4−7); to our enchantment, the most promising enhancement was observed when we used protic solvents such as: MeOH, H_2O and ethanol, and the yields were excellent (entries 8−10); and among these three solvents, ethanol was found to be the best solvent in terms of acceleration

of the rate of the reaction (30 min) and yield of the product (96%, entry 10). Hence, for our further studies we used ethanol as solvent under ultrasonic condition.

Table 2: Solvent effect on the SiO_2-I catalyzed synthesis of 4-(3′-methoxyphenyl)-6-methyl-2-oxo-3,4-dihydro-(1*H*)-pyrimidine-5-ethyl carboxylate (**4a**)

		Reaction		Condition[c]			
		RT (28 °C)		Reflux		Ultrasound	
Entry	Solvent[a]	Time	Yield[b]	Time	Yield[b]	Time	Yield[b]
		(min)	%	(min)	%	(min)	%
1	No solvent	300	15	300	24	**30**	30
2	*n*-Hexane	300	18	300	42	**30**	46
3	CH_3CN	300	22	300	38	**30**	45
4	1,4-dioxane	300	30	300	47	**30**	50
5	DMSO	300	25	300	30	**30**	35
6	DMF	300	20	300	26	**30**	30
7	THF	300	20	300	28	**30**	30
8	H_2O	300	35	300	60	**30**	70
9	MeOH	300	40	300	60	**30**	70
10	Ethanol	300	50	300	80	**30**	**95**

[a]5 mL; [b]Isolated yield; [c]3-methoxybenzaldehyde (1 mmol), urea (1 mmol), ethyl acetoacetate (1 mmol) and SiO_2-I (0.1 g).

2.3 Catalyst feed ratio

A study on the effect of catalyst-load on the progress of this successful reaction under ultrasonic condition was then taken up and the results are encapsulated in the **Table 3**.When the reaction was carried out by using 0.05, 0.06, 0.07, 0.08, 0.09 and 0.10 g of SiO_2–I, with the increase in the amount of the catalyst from 0.05 g to 0.1 g the yield of the product got enhanced gradually from 57% with 0.05 g to 96% when 0.1 g of the catalyst (entry 6). Increase in the amount of SiO_2–I did not show much variation in the yield of the product **4a**.

Table 3: Effect of the catalyst-load on the synthesis of 4-(3′-methoxyphenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-carboxylate (**4a**) in ethanol

Entry	Catalyst loading (g)	Yield[a] (%)
1	0.05	57
2	0.06	62
3	0.07	67
4	0.08	75
5	0.09	84
6	**0.10**	**96**

[a] Isolated yield.

0.1 g of SiO_2–I as catalyst in ethanol as a medium under ultrasonic condition was thus, used to prepare a variety of 3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylates from different substituted araldehydes, ethyl acetoacetate and urea/thiourea as shown in the **Table 4**. To our fortune, SiO_2–I worked as a best catalyst irrespective of the presence of electron donating or electron withdrawing groups in the nucleus of the araldehydes, and the reactions went to completion within 30 min. and afforded differentially substituted 3,4-dihydropyrimidin-2-(1H)-ones/-thiones in excellent yields.

Table 4: SiO_2–I catalysed synthesis of 3,4-dihydropyrimidin-2 (1H)-ones/-thiones (**4a–4m**)

X= O, S
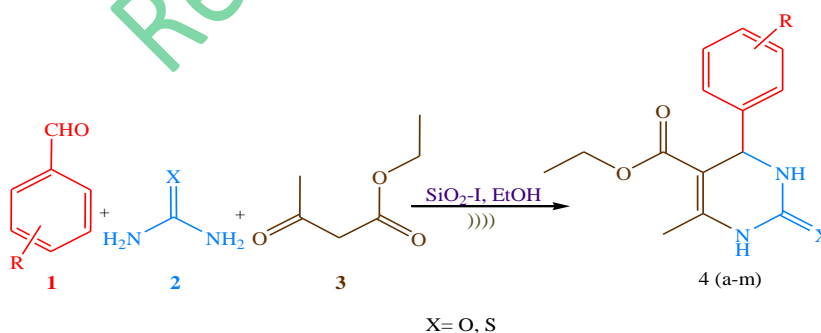
Entry	Aldehyde	X	Product[a]	Time (min)	Yield[b] (%)	M.p (°C)
1	3-$CH_3OC_6H_4CHO$	O	**4a**	30	96	205−207
2	3,4-$CH_3OC_6H_3CHO$	O	**4b**	30	94	178−180
3	3-$NO_2C_6H_4CHO$	O	**4c**	30	92	225−227
4	4-ClC_6H_4CHO	O	**4d**	30	96	212−214
5	4-$CH_3C_6H_4CHO$	O	**4e**	30	95	210−212

6	4-NO$_2$C$_6$H$_3$CHO	O	**4f**	30	94	208−210
7	2-NO$_2$C$_6$H$_3$CHO	O	**4g**	30	93	218−220
8	2-ClC$_6$H$_4$CHO	O	**4h**	30	94	215−217
9	2,3-ClC$_6$H$_3$CHO	O	**4i**	30	93	244−246
10	2-CF$_3$C$_6$H$_4$CHO	O	**4j**	30	94	202−204
11	C$_6$H$_5$CHO	S	**4k**	30	92	207−208
12	4-ClC$_6$H$_4$CHO	S	**4l**	30	89	192−194
13	3-NO$_2$C$_6$H$_4$CHO	S	**4m**	30	86	206−207

[a]Compared on TLC with the standard samples; and characterized by IR,^{1}H NMR/^{13}C NMR/LC-Mass spectral analysis; [b]Isolated yield.

From the above results, it is evident that, SiO$_2$-I may activate the carbonyl group of the araldehyde and eases the attack of urea/thiourea (**2**) to form an acyl imine. The active methylene present in ethyl acetoacetate (**3**) may then attack the intermediate imine to produce ureide. This on subsequent cyclization may lead to the corresponding 2,3-dihydropyrimidinones/-thiones.

3. Experimental

3.1. Materials and apparatus

All commercially available reagents were used without any purification, except liquid aldehydes which were purified by distillation before use. Melting points were found out using a Raaga, Chennai made melting point apparatus. The progress of the reactions was monitored by thin layer chromatography [silica gel plates (Merck 60 F250), observed under the UV lamp]. Infrared spectra were recorded on an Agilent make Cary 630 FT-IR spectrophotometer. ^{1}H NMR spectra were recorded on a Varian Mercury instrument working at 400 MHz in CDCl$_3$ as a solvent and ^{13}C NMR spectra were recorded on a Bruker AMX instrument (100 MHz) in DMSO-d_6 as a solvent and TMS as an internal standard. Liquid chromatography-Mass spectra were recorded on an Agilent Technologies (1200 series) instrument. Ultrasonic reactions were performed using a SIDILU, Indian make sonic bath working at a constant frequency of 35 kHz and maintained at 25 °C by continuously circulating water.

3.2. General experimental procedure for the synthesis of 3,4-dihydropyrimidin-2-(1H)-ones/ -thiones

To a mixture of araldehyde (1 mmol), urea/thiourea (1 mmol), ethyl acetoacetate(1 mmol) in ethanol (5 mL), taken in a 50-mL conical flask was added SiO$_2$-I (0.1 g) and placed in an ultrasonic bath working at a constant frequency of 35 kHz for 30 min. The completion of the reaction was followed by thin layer chromatography using 3:7; Ethyl acetate/n-hexane as an eluent. The reaction mixture, after the completion of the reaction, was quenched with crushed ice and filtered. The residue was washed with ethanol (5 mL × 2), and the solid SiO$_2$–I was collected

and dried at 100 °C for 2 h and kept aside for reuse. The product present in the filtrate was recovered by removing the solvent by distillation and recrystallized from hot aq. ethanol. The structures of all the products were established either by Infrared, [1]H Nuclear magnetic resonance, [13]C Nuclear magnetic resonance and Mass spectral analysis, from their melting points or by the comparison on TLC with the standard samples.

3.3. Reusability of SiO$_2$–I:

The reusability results of the catalyst are given in the form of a graph as shown in the **Figure 1**. It is clear from this graph that, SiO$_2$–I can be reused successfully for at least five runs, and the yield of **4a** was found to be 96 %, 95 %, 90 %, 85 % and 80 %, respectively for the first to fifth cycle. The decrease in the yield is due to the loss of the catalyst at the time of recovery by filtration during workup of the reactions.
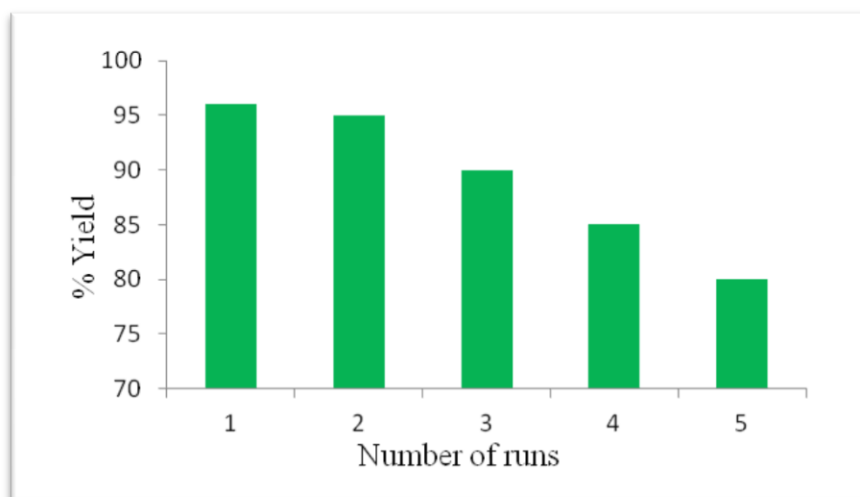
Figure 1: Catalyst reusability graph

3.4. Spectral data

4-(3′-Methoxyphenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (**4a**)**:**
IR (ATR, υ cm^{-1}): 3237, 3100, 2982, 1700, 1647, 1038;
^{1}H NMR: δ (ppm) = 1.07–1.11 (t, J = 7.2 Hz, 3H, CH$_3$), 2.23 (s, 3H, CH$_3$), 3.70 (s, 3H, CH$_3$), 3.95–4.00 (q, J = 7.2 Hz, 2H, CH$_2$), 5.10 (d, J = 2.4 Hz, 1H, CH), 6.76–7.24 (m, 4H, Ar-H), 7.67 (s, 1H, NH), 9.13 (s, 1H, NH);
^{13}C NMR: δ (ppm) = 14.5, 18.2, 53.7, 55.5, 59.6, 106.0, 111.1, 112.1, 119.5, 127.8, 148.4, 152.6, 158.9, 160.8, 167.1;
Mass (m/z): 291.1[M+H]$^+$.

4-(3′,4′-Dimethoxyphenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (**4b**)**:**

IR (ATR, υ cm⁻¹): 3247, 3107, 2955, 1706, 1680, 1024;

^{1}H NMR: δ (ppm) = 1.07–1.16 (t, J = 6.8 Hz, 3H, CH$_3$), 2.23 (s, 3H, CH$_3$), 3.69 (s, 6H, 2 × OCH$_3$), 3.95–4.00 (q, J = 6.8 Hz, 2H, CH$_2$), 5.07 (d, J = 2.8 Hz, 1H,CH), 6.75–6.77 (d, J = 8.4 Hz, 1H, Ar-H), 6.823 (s, 1H, Ar-H), 6.85–6.88 (d, J = 8.4 Hz, 1H,Ar-H), 7.62 (s, 1H, NH), 9.09 (s, 1H, NH);

^{13}C NMR: δ (ppm) = 14.5, 18.0, 55.9, 59.0, 60.1, 106.0, 111.6, 118.2, 118.3, 136.6, 148.2, 148.3, 149.2, 152.6, 168.5;

Mass (m/z): 321.1 [M+H]⁺.

4-(3′-Nitrophenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4c):

IR (ATR, υ cm⁻¹): 3226, 3105, 2964, 1685, 1636, 1523;

^{1}HNMR: δ (ppm) = 1.06–1.09 (t, J = 6.8 Hz, 3H, CH$_3$), 2.25 (s, 3H, CH$_3$), 3.96–4.01 (q, J = 6.8 Hz, 2H, CH$_2$), 5.28 (d, J = 2.4 Hz, 1H, CH), 7.61–7.68 (m, 2H, Ar-H), 7.85 (s, 1H, NH), 8.06 (s, 1H, Ar-H), 8.10–8.12 (d, J = 7.6 Hz, 1H, Ar-H), 9.32 (s, 1H, NH);

^{13}C NMR: δ (ppm) = 14.2, 17.0, 54.1, 58.0, 59.3, 108.0, 122.6, 123.5, 128.0, 130.5, 145.3, 148.8, 148.9, 150.0, 164.2;

Mass (m/z): 306.1 [M+H]⁺.

4-(4'-Chlorophenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4d):

IR (KBr, υ cm⁻¹): 3329, 1670, 1580, 1540, 1498, 1432, 1335, 1303, 1234, 1199, 1138, 1084, 1025, 928, 877, 752, 690;

^{1}H NMR: δ (ppm) = 1.05 (t, J = 6.9 Hz, 3H), 2.22 (s, 3H), 3.93 (q, J = 7.3 Hz, 2H), 5.22 (s, 1H), 7.45 (d, J = 8.7 Hz, 2H), 7.86 (s, 1H, NH), 8.19 (d, J = 8.7 Hz, 2H), 9.32 (s, 1H, NH);

^{13}CNMR: δ (ppm) = 14.6, 18.4, 54.2, 59.2, 59.9, 98.2, 123.8, 124.4, 127.8, 128.2, 147.2, 152.3, 152.5, 158.2, 165.5.

4-(4'-Methylphenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4e):

[33]

IR (KBr, υ cm⁻¹): 3220, 3100, 1720 (sh), 1700;

^{1}H NMR: δ (ppm) = 1.12 (t, J = 7.5 Hz, 3H), 2.28, 2.30 (2 s, 6H, 2 × CH$_3$), 4.00 (q, J = 7.5 Hz, 2H), 5.11 (d, J = 3.0 Hz, 1H), 7.12 (s, 4H), 7.70 (s, 1H, NH), 9.19 (s, 1H, NH).

4-(4'-Nitrophenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4f): [33]
IR (KBr, υ cm⁻¹): 3230, 3120, 1730, 1710, 1650;

^{1}H NMR: δ (ppm) = 1.11 (t, J = 7.5 Hz, 3H), 2.29 (s, 3H), 4.00 (q, J = 7.5 Hz, 2H), 5.29 (d, J = 3.0 Hz, 1H), 7.51 (d, J = 10 Hz, 2 H), 7.91 (br s, 1H), 8.23 (d, J = 10.0 Hz, 2H), 9.37 (br s, 1H).

4-(2'-Nitrophenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4g): [33]
IR (KBr, υ cm⁻¹): 3240, 3100, 1710, 1650;

^{1}H NMR: δ (ppm) = 0.94 (t, J = 7.5 Hz, 3H), 2.30 (s, 1H), 3.88 (q, J = 7.5 Hz, 2H), 5.81 (d, J = 3.0 Hz, 1H), 7.49–7.98 (m, 5H), 9.39 (br s, 1H).

4-(2'-Chlorophenyl)-6-methyl-2-oxo-3,4-dihydro(1H)pyrimidine-5-ethyl carboxylate (4h): [33]

IR (KBr, υ cm^{-1}): 3360, 3220, 3100, 1690, 1640;

^{1}H NMR: δ (ppm) = 1.08 (t, J = 7.5 Hz, 3H), 2.32 (s, 3H), 3.91 (q, J = 7.5Hz, 2H), 5.67 (d, J = 2.5 Hz, 1H), 7.22–7.46 (m, 4H), 7.72 (br s, 1H, NH), 9.30 (br s, 1H, NH).

4-(2',3'-Dichlorophenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4i): [33]

IR (KBr, υ cm^{-1}): 3360, 3100, 1700, 1690, 1640;

^{1}H NMR: δ (ppm) = 0.97 (t, J = 7.5 Hz, 3H), 2.31 (s, 3H), 3.89 (q, J = 7.5 Hz, 2H), 5.69 (br s, 1H), 7.25–7.43 (m, 2H), 7.50–7.61 (m, 1H), 7.80 (br s, 1H, NH), 9.32 (br s, 1H, NH).

4-(2'-Trifluoromethylphenyl)-6-methyl-2-oxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4j): [33]

IR (KBr, υ cm^{-1}): 3230, 3100, 1700, 1640;

^{1}H NMR: δ (ppm) = 0.97 (t, J = 7.5 Hz, 3H), 2.45 (s, 3H), 3.97 (q, J = 7.5 Hz, 2H), 5.37 (s, 1H, CH), 5.82 (br s, 1H, NH), 7.32–7.70 (m, 4H), 8.46 (br s, 1H, NH).

4-Phenyl-6-methyl-2-thioxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4k): [34]

IR (KBr, υ cm^{-1}): 3243, 1711, 1627;

^{1}HNMR: δ (ppm) = 1.10 (t, J = 7.06 Hz, 3H, CH$_3$), 2.29 (s, 3H, CH$_3$), 4.00 (q, J = 7.0 Hz, 2H, CH$_2$), 5.18 (s, 1H, CH), 7.28 (m, 5H, Ar-H), 9.63 (s, 1H, NH), 10.30 (s, 1H, NH);

^{13}CNMR: δ (ppm) = 12.2, 15.5, 52.2, 57.8, 99.2, 124.7, 125.8, 126.6, 143.1, 163.4, 172.6;

Mass (m/z): 277.1 [M+H]$^+$.

4-(4'-Chlorophenyl)-6-methyl-2-thioxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4l): [34]

IR (KBr, υ cm^{-1}): 3242, 1705, 1638;

^{1}HNMR: δ (ppm) = 1.12 (t, J = 7.1 Hz, 3H, CH$_3$), 2.27 (s, 3H, CH$_3$), 4.02 (q, J = 7.1 Hz, 2H, CH$_2$), 5.16 (s, 1H, CH), 7.28 (d, J = 8.6 Hz, 2H, Ar-H), 7.45 (d, J = 0.8 Hz, 2H, Ar-H), 9.75 (s, 1H, NH), 10.58 (s, 1H, NH);

Mass (m/z): 311.06 [M+H]$^+$.

4-(3'-Nitrophenyl)-6-methyl-2-thioxo-3,4-dihydro-(1H)-pyrimidine-5-ethyl carboxylate (4m): [34]

IR (KBr, υ cm^{-1}): 3170, 1715, 1661, 1593, 1540;

^{1}HNMR: δ (ppm) = 1.11 (t, J = 7.5 Hz, 3H, CH$_3$), 2.34 (s, 3H, CH$_3$), 4.04 (q, J = 7.6 Hz, 2H, CH$_2$), 5.36 (s, 1H, CH), 7.65–7.73 (m, 2H, Ar-H), 8.08 (s, 1H, Ar-H), 9.80 (s, 1H, NH), 10.56 (s, 1H, NH);

Mass (m/z): 322.08 [M+H]$^+$.

4. Conclusions

In conclusion, we have developed a versatile, SiO$_2$-I catalysed, energy efficient, one-pot three-component, green protocol for the synthesis of 3,4-dihydropyrimidin-2-(1H)-ones/-thiones in ethanol as a solvent under ultrasonic condition. The synthesis of the target heterocyclic compounds has several advantages such as: mild reaction condition, short reaction duration, ease of isolation and best yields of the products. The heterogeneous catalyst SiO$_2$-I can be recycled for a minimum of five times without loss of activity.

5. Acknowledgement

6. References

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