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Pumice as a Novel Natural Heterogeneous catalyst for Synthesis of 3,4-dihydropyrimidine-2-(*1H*)-ones/thiones *via* Biginelli reaction under solvent-free conditions

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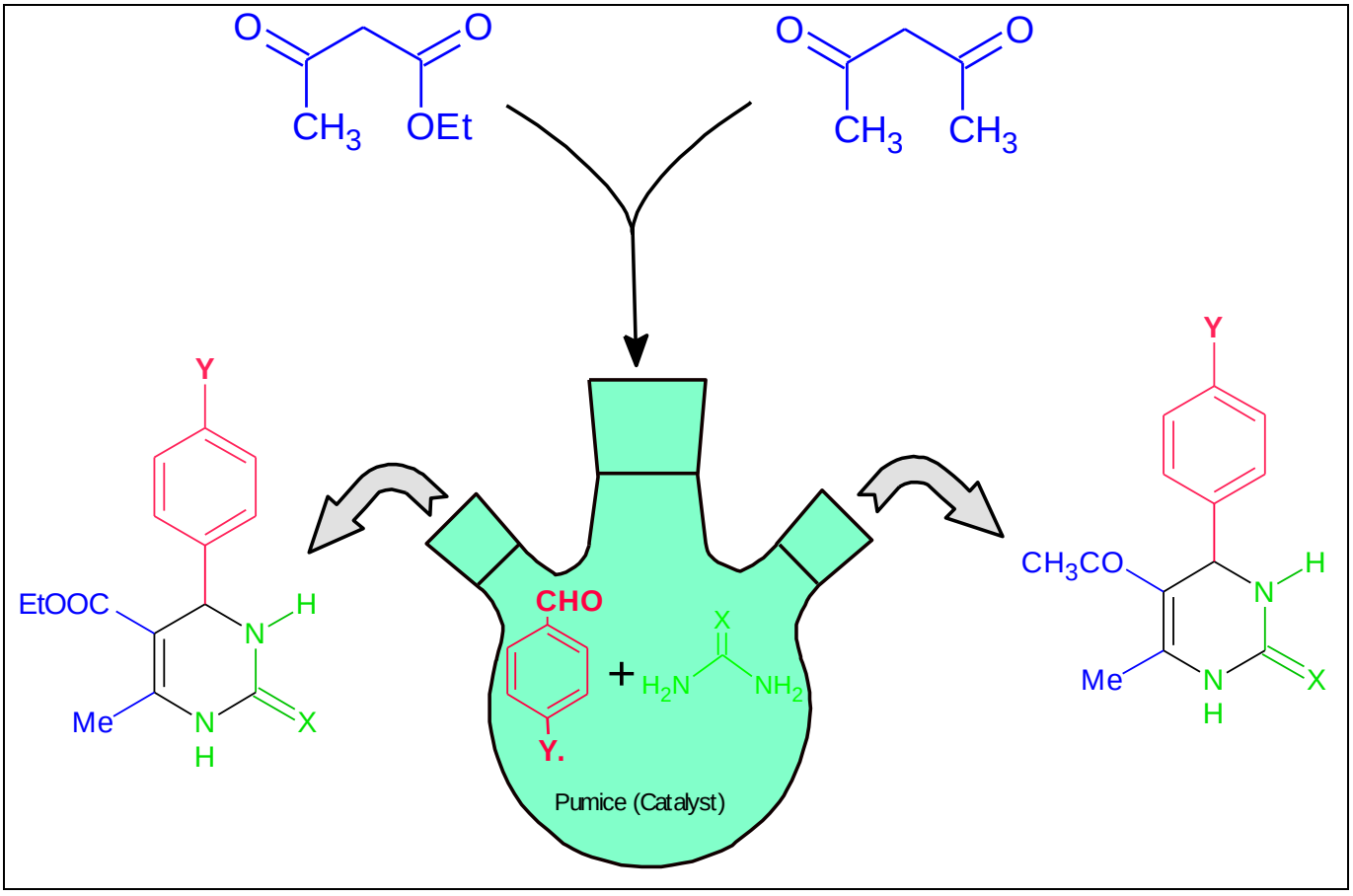
Graphic abstract:



Abstract:

An efficient and environmentally Pumice as a new catalyst for designing of 3,4-dihydropyrimidin-2(1H) ones / thiones *via* one-pot multi component condensation of aromatic aldehydes, urea/ thiourea and ethyl acetoacetate or acetyl acetone in excellent

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yields (96-99 %). The advantages using of this new catalyst is very cheap, available, non-toxic, stable under thermal conditions, easy work-up, improved yields, the product of reaction is very pure without using chromatographic methods and solvent free conditions.

KEYWORDS: One pot reaction, 3,4- Dihydropyrimidin-2(1H) ones / thiones and Pumice.

Introduction

Multi-component reactions is very important field in organic chemistry which is direct method from synthesis of the heterocyclic compounds like imidazoles, pyrazoles, pyridines and acridines [1-4]. Biginelli reaction is considered that the most famous multi-component reactions for the designing of dihydropyrimidinones which are known for exhibiting a extensive range of pharmaceutical and biological effectiveness like antitumor, antiviral, anti-inflammatory and antibacterial properties [5]. Also, these components have developed as potential calcium channel blockers [6], neuropeptide antagonists, α1a–adrenergic antagonists and antihypertensive activities. In addition that 2-oxodihydropyrimidine-5-carboxylate is also discovered in numerous marine natural products [7], which having batzelladine alkaloids, including that affect toward the potent HIV gp-120-CD_4 inhibitors [8,9]. In general, Biginelli reaction have need long reaction times (24 hours) and regularly undergoes from small yields of products in case of substituted aldehydes [10, 11]. hence the Biginelli reaction is continuing to attract the attention of scientist interested to find insignificant and more efficient procedures for the designing of dihydropyrimidinones. There are many catalysts have been tested to design of dihydropyrimidinones [12. 13]. A plethora of reagents/methods have been reported for this purpose such as Amberlyst-15, Nafion-H, KSF clay with dry acetic acid under microwave irradiation [10], ionic liquids [14, 15], cerric ammonium nitrate under ultrasonication [16], Lewis acids (such as BF_3OEt_2) in combination with transition metals and a suitable proton source [17], lanthanide triflates [18], lanthanide chloride [19], and indium chloride [20]. Although these methods each have their own merits, they also undergo from the drawbacks with respect to cost of reagent, reaction work-ups and reaction time, Consequently, the Biginelli reaction still requires an efficient protocol for the synthesis of pyrimidinone compounds.

Although a handful of the reported catalysts are environmentally friendly, many others are either toxic or corrosive and difficult to handle. Considering the current status of global issues, there is an ever increasing stress for replacing toxic solvents and catalysts with environmentally accepted competent. In this Letter, we report that a simple and effective

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procedure for the synthesis of 3,4-dihydropyrimidin-2(1H) ones / thiones *via* one-pot multi-component condensation of aromatic aldehydes, urea/thiourea and β-ketoesters by employing pumice as a green novel and natural catalyst.

Results and Discussion

In the last years, there are a developing demand for the improving of organic synthesis in ecofriendly media. Synthetic manipulations have to be made to reduce using of hazardous chemicals through substituting the toxic organic solvents in reactions and their subsequent workup with other friendly solvents. There needs to replace toxic solvents by green in industrial processes, huge amount of solvent gets wasted.

Some Special properties exhibited by heterogeneous catalyst pumice supported Red Sea Sand, are as eco-friendly Non-toxic nature, easily operational handling, thermal stability, mildness of the reaction conditions and economically least expensive attracted the attention of luminaries for organic synthetic.

We study the effect of catalyst amount for synthesis of 3,4- dihydropyrimidin-2(1H) ones / thiones, by heating aromatic aldehydes, namely; benzaldehyde, *p*-chlorobenzaldehyde, *p*-nitrobenzaldehyde, *p*-methoxybenzaldehyde, *N*,*N*-dimethylaminobenzaldehyde and 4-hydroxybenzaldehyde with urea/thiourea and ethyl acetoacetate or acetylacetone in presence of different amount of pumice (0.10 g - 0.50 g, within few minutes (1-3 minutes) (**Scheme 1**), we are obtained the expected products 3,4- dihydropyrimidin-2(1H) ones / thiones in excellent yield within short time (2-3 minutes) and problems associated with toxic solvents usage (safety, pollution and cost) were avoided in conventional protocol. The optimized results are summarized in **Table 2**, It has found that 0.40 g of pumice get the maximum yield of product 99% and when we increase the amount of pumice catalyst the yields are stable (**Table 2**).

Once the reaction completes the catalyst was recovered easily by heating the mixture in ethanol then filtration and reused successfully several times without loss of catalytic properties or its amounts.

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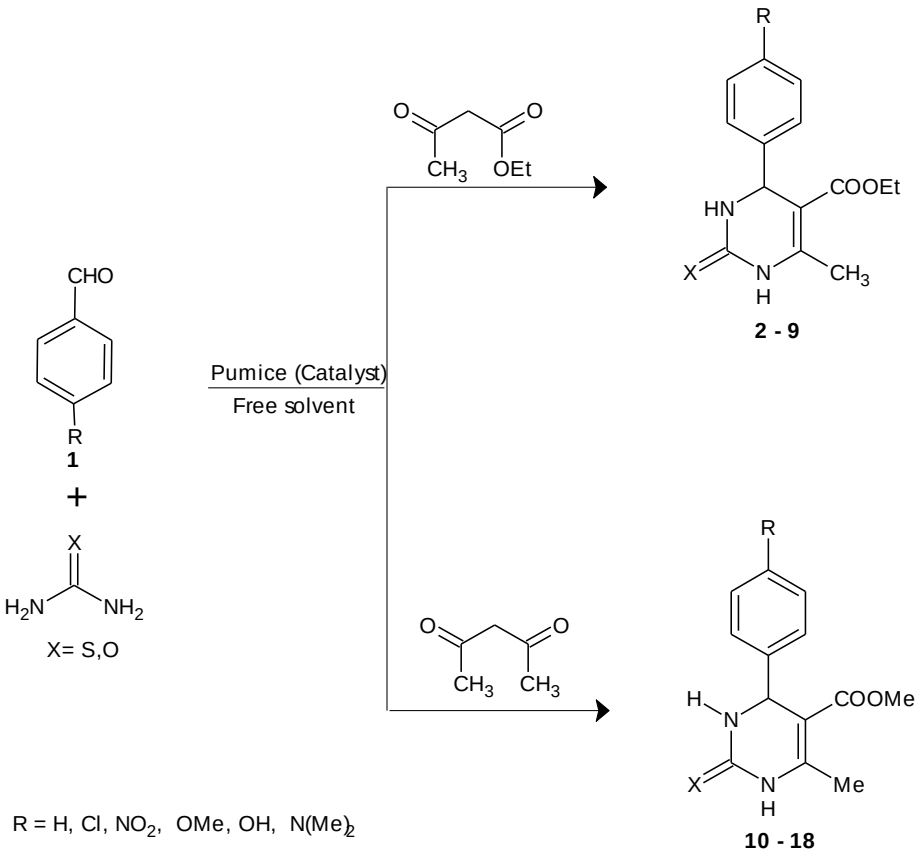

Scheme 1

Synthesis of 3,4-dihydropyrimidin-2(*H*) ones / thiones

Table 1: Synthesis of 3,4-dihydropyrimidin-2(1*H*) ones / thiones

Entry	R	X	Y	M.P.	Reported
2	**OEt**	**O**	H	202-203	201-202 [21]
3	**OEt**	**O**	Cl	214-215	212-214[21]
4	**OEt**	**O**	NO_2	209	209-210[21]
5	**OEt**	**O**	OMe	201	201-202[21]
6	**OEt**	**O**	$N(Me)_2$	2233	231–232[21]
7	**OEt**	**S**	H	208	207–209[23]
8	**OEt**	**S**	Cl	194	192–194[24]
9	**OEt**	**S**	OMe	153	150–151[22]
10	**Me**	**O**	H	228	232[22]
11	**Me**	**O**	Cl	224	224-226[23]
12	**Me**	**O**	NO_2	236	236-238[24]
13	**Me**	**O**	OMe	173	172[24]
14	**Me**	**O**	OH	256	256[26]

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15	**Me**	**S**	Cl	250	250-252 [29]
16	**Me**	**S**	NO_2	245	246-247 [29]
17	**Me**	**S**	OMe	182	182-183 [29]
18	**Me**	**S**	OH	253	251–252[31]

The reaction mechanism for formation of synthesis 3,4-dihydropyrimidin-2-(1H)-one/thione derivatives is beginning from the catalyst **1** which have acidic character (Pumice is a volcanic rock that consists of 70% SiO_2 and 13% Al_2O_3) followed by addition of aromatic aldehyde **2** at this time there are interaction between catalyst and aldehyde take place, subsequently, ethyl 3-oxobutanoate is appended, in this step, the aldehyde and catalyst were attaching and attacking with the enol form of the ethyl 3-oxobutanoate keto-enol **3** equilibrium. In this moment H_2O was eliminated through formation of adduct which combined with catalyst **4**, then urea or thiourea added to form C-N bond **5**, after that inter nucleophilic attack of NH_2 to C=O of $CH_3C=O$ **6,** followed by the catalyst was be sundered from the product **7**, dehydrated of H_2O molecule to from the target compound, now the catalyst is free to restart the process again. (**Scheme 2**)

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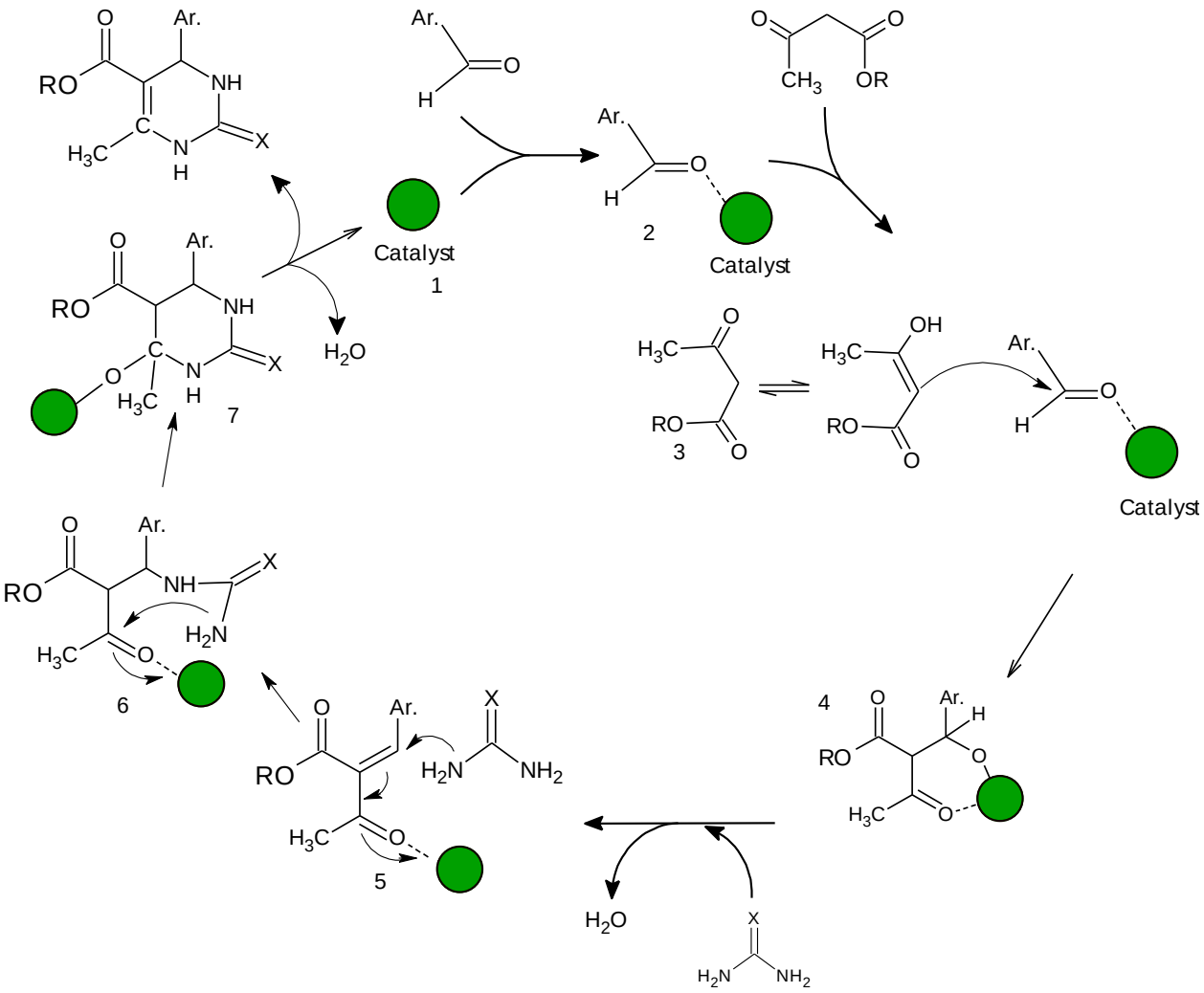

Scheme 2

The reaction mechanism for formation of 3,4-dihydropyrimidin-2-(1H)-one/thione derivatives

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TABLE 2. Comparison amount of pumice and yields for Biginelli reaction under free solvent conditions.

Compound	Amount of Pumic				
	0.1 g	0.2g	0.3g	0.4g	0.5 g
	yield				
2	58%	86%	93%	98%	98%
3	53%	82%	95%	99%	99%
4	61%	89%	92%	97%	97%
5	57%	81%	97%	97%	97%
6	64%	92%	99%	99%	99%
7	48%	85%	91%	98%	98%
8	70%	90%	96%	98%	98%
9	56%	91%	95%	96%	96%
10	63%	93%	94%	98%	98%
11	72%	90%	97%	97%	97%
12	61%	87%	96%	98%	98%
13	58%	89%	95%	98%	98%
14	72%	93%	97%	97%	97%
15	60%	92%	97%	98%	98%
16	59%	90%	96%	96%	96%
17	55%	91%	97%	98%	98%
18	71%	89%	97%	97%	97%

From table above, we sure as scientists and chemists can observe that the green chemistry method is considered as environ-friendly and economically procedure compared with the traditional procedure for synthesis 3,4-dihydropyrimidin-2-(1H)-one/thione derivatives.

Conclusion

In conclusion, we have successfully developed a convenient, efficient and quick method for the designing of 3,4-dihydropyrimidin-2-(1H)-one/thione derivatives *via* one-pot multi-component condensation of aromatic aldehydes, urea/thiourea and β-ketoesters by

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employing pumice as a novel heterogeneous green catalyst. The environmental advantages having that ignoring organic solvent, simplicity and generality of procedure, shorter reaction time, simple workup, catalyst-free and reusable catalysts conditions, and pure products in good to excellent yields

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Experimental

All melting points were recorded on Melt-Temp II melting point apparatus. IR spectra were measured as KBr pellets on a Shimadzu DR-8001 spectrometer. ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker at 400 MHz using TMS as an internal reference and DMSO-*d6* as a solvent.

General procedure for synthesis of 3,4-dihydropyrimidin-2(1*H*) ones / thiones 2-18.

To a mixture of aromatic aldehyde (10 mmol), ethyl acetoacetate or acetyl acetone (10 mmol), urea/ thiourea (15 mmol) and different amount of pumice catalyst (0.2g-0.6g) were heated under free solvent for 1-3 minutes. After completion of the reaction, the hot reaction mixture was poured in 10 ml ethanol and filtered to get catalyst recovered, and the filtered was kept at room temperature, and then filtered to obtain the products.

Ethyl 6-methyl-2-oxo-4-phenyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate (2).

Melting point = 202-203°C. IR (cm^{-1}): 3242, 3197, 1645, 1578; ^{1}H NMR: δ 9.18 (s, 1H, NH), 7.47 (s, 1H, NH), 7.36-7.20 (m, 5H, ArH), 5.09 (s, 1H, CH), 3.92 (q, 2H, OCH_2), 2.22 (s, 3H, CH_3), 1.17 (3H, $OCH_2\underline{CH_3}$); ^{13}C NMR: δ 167.12, 154.14, 149.07, 146.14, 129.18, 128.91, 127.12, 98.97, 60.16, 57.11, 18.93, 15.34.

Ethyl 4-(*p*-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (3).

Melting point = 214-215 °C. IR (cm^{-1}): 3285, 3142, 1649, 1576; ^{1}H NMR: δ 9.33 (s, 1H, NH), 7.43 (s, 1H, NH), 7.34-7.14 (m, 4H, ArH), 5.43 (s, 1H, CH), 3.87 (q, 2H, OCH_2), 2.34 (s, 3H, CH_3), 1.04 ($OCH_2\underline{CH_3}$); ^{13}C NMR: δ 163.23, 153.20, 144.29, 136.10, 134.85, 132.41, 131.21, 101,19, 59.16, 53.15, 19.11, 15.32.

Ethyl 6-methyl-4-(*p* -nitrophenyl)-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (4).

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Melting point = 209-210°C. IR (cm^{-1}): 3276, 3190, 1647, 1571; ^{1}H NMR: δ 9.43 (s, 1H, NH), 8.4598 (d, J = 9 Hz, 2H),7.90 (s, 1H, NH), 7.87-7.98 (m, 4H, ArH), 5.12 (s, 1H, CH), 3.97 (q, 2H, OCH_2), 2.34 (s, 3H, CH_3), 1.09 (t, 3H, OCH_2CH_3); ^{13}C NMR: δ 167.35, 160.71, 156.54, 150.12, 147.98, 140.54, 132.12, 128.21, 125.65, 99.89, 62.10, 59.19, 54.12, 18.23, 14.25.

Ethyl 4-(*p* -methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (5).

Melting point = 201-203°C. IR (cm^{-1}): 3276, 3153, 1642, 1568; ^{1}H NMR: δ 9.32 (s, 1H, NH), 7.90 (s, 1H, NH), 7.65-7.18 (m, 4H, ArH), 5.47 (s, 1H, CH), 3.99 (q, 2H, OCH_2), 3.89 (s, 3H, OCH_3), 2.65 (s, 3H, CH_3), 1.08 (t, 3H, $OCH_2\underline{CH_3}$); ^{13}C NMR: δ 167.29, 160.29, 156.87, 148.98, 140.45, 127.49, 114.62, 100.12, 59.41, 56.18, 53.90, 18.56, 14.35.

Ethyl 4-(*p* -(dimethylamino)phenyl)-6-methyl-2-oxo-1,2,3, 4-tetrahydropyrimidine-5-carboxylate (6).

Melting point = 233-234°C, IR (cm^{-1}): 3264, 3198, 1625, 1523; ^{1}H NMR:^{1}H NMR δ 9.14 (s, 1H, NH), 7.98 (s, 1H, NH), 7.61-6.32 (m, 4H, ArH), 5.01 (s, 1H, CH), 3.98 (q, 2H, OCH_2), 3.23 (s, 3H, CH_3), 3.51 (s, 3H, CH_3), 2.21 (s, 3H, CH3), 1.11 (t, 3H, OCH_2CH_3); ^{13}C NMR : δ 187.14, 168.49, 154.43, 152.22, 1494.9, 173.20, 127.24, 112.37, 111.65, 100.04, 59.86, 53.78, 18.29, 14.64.

Ethyl 6-methyl-2- thioxo -4-phenyl-1,2,3,4-tetrahydropyrimidine5-carboxylate (7).

Melting point = 208–209°C. IR (cm^{-1}): 3276, 3172, 1634, 1595; ^{1}H NMR: δ 9.78 (s, 1H, NH), 8.07 (s, 1H, NH), 7.56-7.34 (m, 5H, ArH), 5.11 (s, 1H, CH), 3.98 (q, J = 7.4 Hz, 2H, OCH_2), 2.21 (s, 3H, CH_3), 1.89 (t, 3H, $OCH_2\underline{CH_3}$); ^{13}C NMR: δ 174.73, 153.52, 150.13, 147.08, 132.08, 128.97, 127.18, 100.05, 59.87, 57.12, 19.87, 16.87.

Ethyl 4-(*p* -chlorophenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (8).

Melting point = 187-189°C. IR (cm^{-1}): 3246, 3107, 1673, 1563; ^{1}H NMR: δ 9.32 (s, 1H, NH), 7.87 (s, 1H, NH), 7.34-7.87 (m, 4H, ArH), 5.31 (s, 1H, CH), 3.98 (q, 2H, OCH_2), 2.34 (s, 3H, CH_3), 1.17 (t, 3H, $OCH_2\underline{CH_3}$); ^{13}C NMR : δ 166.76, 154.24, 152.23, 149.01, 138.87, 135.18, 122.76, 100.13, 59.17, 531.8, 18.12, 14.54.

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Ethyl 4-(*p* -methoxyphenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5 carboxylate (9).

Melting point = 153-154 °C. IR (cm^{-1}): 3278, 3195, 1650, 1560; ^{1}H NMR: δ 9.54 (s, 1H, NH), 7.98 (s, 1H, NH), 7.63-7.34 (m, 4H, ArH), 5.13 (s, 1H, CH), 3.98 (q, 2H, OCH_2), 3.86 (s, 3H, OCH_3), 2.35 (s, 3H, CH_3), 1.21 (t, , 3H, $OCH_2\underline{CH_3}$); ^{13}C NMR : δ 167.56, 159.97, 154.43, 149.75, 139.85, 137.34, 132.08, 114.32, 100.21, 59.56, 55.05, 53.68, 18.22, 14.75.

5-Acetyl-6-methyl-4-phenyl-3,4-dihydropyrimidin-2(1H)-one (10).

Melting point = 228-229 °C IR (cm^{-1}): 3246, 2197, 1642, ^{1}H NMR : 9.01 (s, 1H, NH), 8.23 (s, 1H, NH), 7.64 - 7.21 (m, 5H, ArH), 5.54 (d, CH), 2.32 (s, 3H, $COCH_3$), 2.23 (s, 3H, CH_3), ^{13}C NMR: 194.74, 154.60, 149.54, 144.90, 129.45, 128.81, 128.23, 109.04, 55.62, 39.53, 32.43, 19.32.

5- Acetyl -6-methyl-4-(*p* -chlorophenyl)-3,4-dihydropyrimidin-2(1H)-one (11).

Melting point = 224-225^{0}C. IR spectrum (cm-1): 3234, 3212, 3124, 2987, 1703, 1644,^{1}H NMR; 9.07 (s, 1H, NH), 7.89 (s, 1H, NH),7.98-7.46 (m, 4H, Ar), 5.02 (d, 1H, CH), 2.31 (s, 3H, $COCH_3$), 2.24 (s, 3H, CH_3); ^{13}C NMR: 187.26, 157.57, 150.25, 143.32, 138.10, 134.32, 130.02, 100.31, 53.34, 30.56, 19.54.

5- Acetyl –6-methyl-4-(*p* -nitrophenyl)-3,4-dihydropyrimidin-2(1H)-one (12).

Melting point = 236-238 °C. IR (cm-1): 3296, 3149, 2878, 1666, 1595; ^{1}H NMR; 9.11 (s, 1H, NH), 8.31 (s, 1H, NH), 7.87 - 7.34 (m, 4H, ArH), 5.50 (d, 1H J = 4Hz, CH), 2.28 (s, 3H, $COCH_3$), 2.16 (s, 3H, CH_3), ^{13}C NMR: 178.87, 154.32, 150.12, 144.31, 130.45, 129.98, 128.90, 108.89, 55.65, 39.92, 33.23, 19.52.

5- Acetyl -6-methyl-4-(*p* -methoxyphenyl)-3,4-dihydropyrimidin-2(1H)-one (13).

Melting point = 173-175 ^{0}C. IR (cm-¹): 3232, 3123, 2957, 1714, 1700 , 1631, 1234, 1029: 1H NMR δ 9.02 (s, 1H, NH), 7.83 (s, 1H, NH), 7.46-6.98 (m, 4H, ArH), 5.52 (s, 1H, Ar-CH), 3.87 (s, 3H, OCH),), 2.23 (s, 3H, $COCH_3$), 2.32 (s, 3H, CH_3); ^{13}C NMR: 184.44, 161.15, 153.20, 150.66, 138.52, 130.76, 119.56, 108.45, 56.93, 52.29, 31.31, 19.17.

5- Acetyl -6-methyl-4-(*p* -hydroxyphenyl)-3,4-dihydropyrimidin-2(1H)-one (14).

Melting point = 255-256 ^{0}C. IR (cm-1): 3245, 3107, 2941, 1700, 1614, ^{1}H NMR: 9.23 (s, 1H, NH), 9.02 (s, 1H, OH), 7.32 (s, 1H, NH), 7.21- 6.78 (m, 4H, ArH), 5.04 (s, 1H, Ar CH), 2.31 (s, 3H, $COCH_3$), 2.18 (s, 3H, CH_3), ^{13}C NMR: 190.14, 154.43, 150.23, 146.42, 135.06, 125.31, 118.43, 107.43, 50.34, 26.56, 17.98.

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5- Acetyl -6-methyl-4-(*p* -chlorophenyl)-3,4-dihydropyrimidine-2(1H)-thione (15).

Melting point = 251-252°C. IR (cm-1): 3252, 3128, 2987, 2962, 1631, 1538 ; ^{1}H NMR: 9.98 (s, 1H, NH), 9.06 (s, 1H, NH), 7.89-7.42 (m, 4H, ArCH), 5.86 (s, 1H, ArH), 2.42 (s, 3H, $COCH_3$), 2.20 (s, 3H, CH_3); ^{13}C NMR: 187.14, 174.51, 152.05, 148.23, 136.65, 128.84, 128.01, 110.81, 55.51, 30.62, 18.63.

5- Acetyl -6-methyl-4-(*p* -nitrophenyl)-3,4-dihydropyrimidine-2(1H)-thione (16).

Melting point = 245-246 ^{0}C. IR (cm-1): 3247, 3183, 2967, 2924, 1646; ^{1}H NMR: 10.08 (s, 1H, NH), 9.43 (s, 1H, NH), 8.03-7.64 (m, 4H, ArCH), 5.83 (d, 1H, J = 4Hz, CH), 2.28 (s, 3H, COCH3), 2.25 (s, 3H, CH_3), ^{13}C NMR: 184.07, 174.65, 154.86, 149.87, 146.01, 130.76, 127.03, 110.01, 54.06, 30.34, 19.74.

5- Acetyl -6-methyl-4-(*p* -methoxyphenyl)-3,4-dihydropyrimidine-2(1H)-thione (17).

Melting point = 182-183 °C. IR (cm^{-1}): 3236, 3158, 1645, 1578; ^{1}H NMR : 9.12 (s, 1H, NH), 8.06 (s, 1H, NH), 7.34 - 6.98(m, 4H, ArH), 5.67 (d, CH), 3.89 (s, 3H, OCH_3), 2.37 (s, 3H, $COCH_3$), 2.01 (s, 3H, CH_3), ^{13}C NMR: 185.28, 174.58 , 159.36, 144.55, 136.35, 132.27, 114.44, 110.20, 55.54,30.07, 19.21.

5- Acetyl -6-methyl-4-(*p* -hydroxyphenyl)-3,4-dihydropyrimidin-2(1H)-thione (18).

Melting point = 253–254. IR (cm-1): 3219, 3128, 2976, 1654, 1611; ^{1}H NMR: 9.54 (s, 1H, NH), 9.18 (s, 1H, OH), 7.65 (s, 1H, NH), 7.29- 6.85 (m, 4H, ArH), 5.08 (s, Ar CH), 2.33 (s, 3H, $COCH_3$), 2.19 (s, 3H, CH_3), ^{13}C NMR: 178.52, 153.65, 150.09, 147.87, 134.21, 125.35, 119.20, 108.00, 50.53, 28.13, 18.02.

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