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Heteropoly acid supported on activated natural clay-catalyzed synthesis of 3,4-dihydropyrimidinones/thiones through Biginelli reaction under solvent-free conditions

Karuppaiah Selvakumar, Thangamariyappan Shanmugaprabha, Murugan Kumaresan, and Ponnusamy Sami

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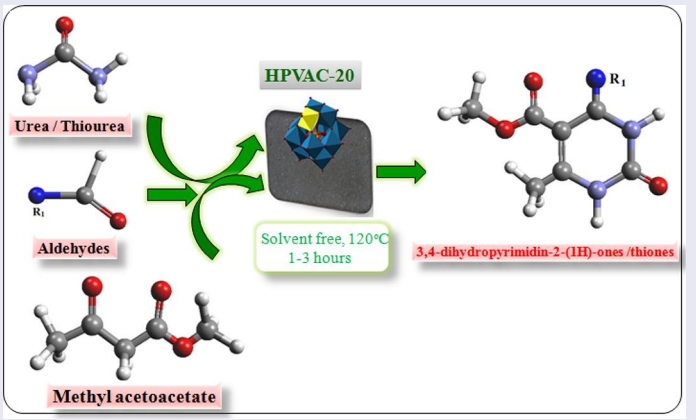
ABSTRACT

Dihydropyrimidinones/thiones (DHPM's) have been prepared by one-pot condensation of methyl acetoacetate, aldehydes, urea/thiourea in the presence of heteropoly-11-tungsto-1-vanadophosphoric acid, $H_4[PVW_{11}O_{40}] \cdot 32H_2O$, (HPV) supported on activated natural clay (HPVAC) under solvent-free reaction condition have been proposed. The DHPM derivatives were identified through elemental analysis and melting point measurements and characterized by FT-IR, ^{1}H-NMR, ^{13}C-NMR spectroscopic methods.

GRAPHICAL ABSTRACT



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KEYWORDS
3,4-Dihydropyrimidinones/thiones; heteropoly acid; multi-component synthesis; natural clay

Introduction

The Biginelli reaction is a one-pot three component reaction invented by Biginelli in 1893.[1] This reaction involves the cyclocondensation of an aldehyde, β-ketoester, and urea or thiourea using acidic catalysts to yield 3,4-dihydropyrimidin-2(1*H*)-ones/3,4-dihydropyrimidin-2(1*H*)-thiones (DHPMs). DHPMs are generally referred to as Biginelli compounds.[2] Aryl substituted DHPMs and their derivatives have been receiving much

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attention in recent years due to their applications in the field of drugs and pharmaceuticals. They exhibit a wide range of biological activities and are extensively used in pharmaceutical as calcium channel blockers, antihypertensive, antibacterial, antitumor, anti-inflammatory, and anti-cancer agents.[3,4]

In 1930, wool protection activity of Biginelli compounds was patented for the protection of wool against moths.[2] Moreover, the dihydropyrimidine scaffold is a key component in several alkaloids with marine sources, which also have interesting biological properties. Most notably among these are the batzelladine alkaloids, which are inhibitors for the binding of HIV gp-120 to CD4 cells for the treatment of AIDS.[5] These compounds have been becoming very interesting due to their wide spectrum of biological activities[6] and used as a starting point to prepare complex heterocyclic scaffolds with pharmacological properties.[7]

Many synthetic methods for the synthesis of this heterocyclic scaffold are now available. Recently, the synthesis of DHPM's is the direct condensation reaction of methyl acetoacetate, aldehydes, and urea/thiourea using catalysts such as PS-PEG-SO_3H, AlKIT-5, ZnO, Al-M41, $Sn(HPO_4)_2 \cdot H_2O$, Fe_3O_4@mesoporous SBA-15, [VSim][HSO_4], pyridiniumtriflate, CuO-CeO_2, iodine, cellulose sulfuric acid, PPF-SO_3H, PDAG-Co, citric acid, $[PyPS]_3PW_{12}O_{40}$, Fe_3O_4@SiO_2-imid-$H_3[PMo_{12}O_{40}]$, Nafion-Ga, phytic acid, Cu@PMO-IL, zeolite-ZSM-5, Fe@SBIL-BPMO, NFS-PRS, [Btto][*p*-TSA], aluminatesulfonic acid, SiO_2-$BaCl_2$, NH_4SCN, zirconia sulfuric acid, d-xylonic acid, and $Ga(OTf)_3$.[8–36]

Despite of the successful methods discussed above, unfortunately, many of these catalysts suffer from one or more limitations, such as long reaction times, low yields, occurrence of several side reactions, drastic reaction conditions and tedious workup procedure. Some of the catalysts are expensive, complex or unavailable and in some of the reaction organic solvents are also used. With the increase of environmental consciousness in chemical research and industry, the solvent-free Biginelli reaction has attracted much attention recently. These factors stimulate us to search for a better catalyst, which has to show a higher activity for the synthesis of DHPMs under mild reaction conditions. Recently the catalytic application of heteropoly-11-tungsto-1-vanadophosphoric acid, $H_4[PVW_{11}O_{40}].32H_2O$, (HPV) supported on activated natural clay (HPVAC) toward the synthesis of bis(indolyl)methanes, *N,N′*-alkylidene bisamides and imidazoles have been reported.[37,38] In continuation of this work a green route for the preparation of 3,4-dihydropyrimidin-2(1*H*)-ones and 3,4-dihydropyrimidin-2(1*H*)-thiones using HPVAC as a mild and reusable catalyst is reported in this paper.

Results and discussion

Effect of catalysts

The condensation of methyl acetoacetate, benzaldehyde with thiourea in the presence of catalysts was chosen as a model reaction to study the effectiveness of the catalysts chosen for the present study. The effectiveness of the catalyst was evaluated with the aid of percentage yield of the products. The results on the screening of catalysis are given in Table 1. Among the different catalysts used, HPVAC produced high yield of 86%. This optimization study tempted us to investigate on HPVAC with different loadings of HPV.

Table 1. Catalyst screening for the synthesis of 3,4-dihydropyrimidin-2(1*H*)-thione.[a]

Run	Catalyst	Catalyst amount (g)	Yield[b] (%)
1	Activated natural clay (AC)	0.05	48
2	$H_3[PW_{12}O_{40}]$	0.03	60
3	$H_4[PVW_{11}O_{40}]$	0.03	63
4	$H_5[PV_2W_{10}O_{40}]$	0.03	66
5	10% $H_3[PW_{12}O_{40}]$/AC	0.05	75
6	10% $H_4[PVW_{11}O_{40}]$/AC	0.05	86
7	10% $H_5[PV_2W_{10}O_{40}]$/AC	0.05	87

[a]Reaction conditions: methyl acetoacetate (1 mmol), benzaldehyde (1 mmol), thiourea (1.25 mmol), solvent-free condition, 120 °C, 1.5 h.
[b]Isolated yields.

Effect of HPV loadings

The dependency of amount of loading of HPV on the catalytic activity of HPVAC was also investigated. The efficiency of the HPVAC-10, 20, and 30 was evaluated for the condensation reaction of methyl acetoacetate, benzaldehyde with thiourea in terms of the percentage yield of product. The HPVAC-20 showed higher efficiency compared to that of HPVAC-10 and comparable efficiency with that of HPVAC-30. Hence HPVAC-20 has been optimized as the catalyst to perform the transformation (Table 2).

Effect of solvents

The title reaction was tried in several solvent media such as toluene, dichloromethane, 1,2-dichloroethane, ethyleneglycol, dimethylsulfoxide, and dimethylforamide and the results are given in Table 3. The results reveal that toluene is found to be the best solvent among all the solvents. The yield of the reaction under solvent free condition is found to be

Table 2. Effect of catalyst loading in the synthesis of 3,4-dihydropyrimidin-2(1*H*)-thione.[a]

Run	Catalyst	Catalyst amount (g)	Yield[b] (%)
1	HPVAC-10	0.05	86
2	HPVAC-20	0.05	91
3	HPVAC-30	0.05	92

[a]Reaction conditions: methyl acetoacetate (1 mmol), benzaldehyde (1 mmol), thiourea (1.25 mmol), solvent-free condition, 120 °C, 1.5 h.
[b]Isolated yields.

Table 3. Effect of solvents on the reaction of methyl acetoacetate, benzaldehyde, thiourea catalyzed by HPVAC-20.[a]

Run	Solvent	Time (h)	Yield[b] (%)
1	Dichloromethane	5	60
2	1,2-Dichloroethane	5	68
3	Toluene	5	75
4	Ethylene glycol	5	62
5	Dimethyl sulfoxide	5	61
6[c]	Solvent free condition[c]	1.5	91

[a]Reaction conditions: methyl acetoacetate (1 mmol), benzaldehyde (1 mmol), thiourea (1.25 mmol), HPVAC-20 (0.05 g), solvent (5 mL), reflux.
[b]Isolated yield.
[c]120 °C.

Table 4. Effect of reusability of HPVAC-20 catalyst on the 3,4-dihydropyrimidin-2(1*H*)-thione yield.[a]

Run	Cycle	Amount of catalyst (g)	Percentage of loss of catalyst (%)	Yield[b] (%)
1	0	0.050	–	91
2	1	0.048	4	89
3	2	0.046	8	88
4	3	0.043	14	87

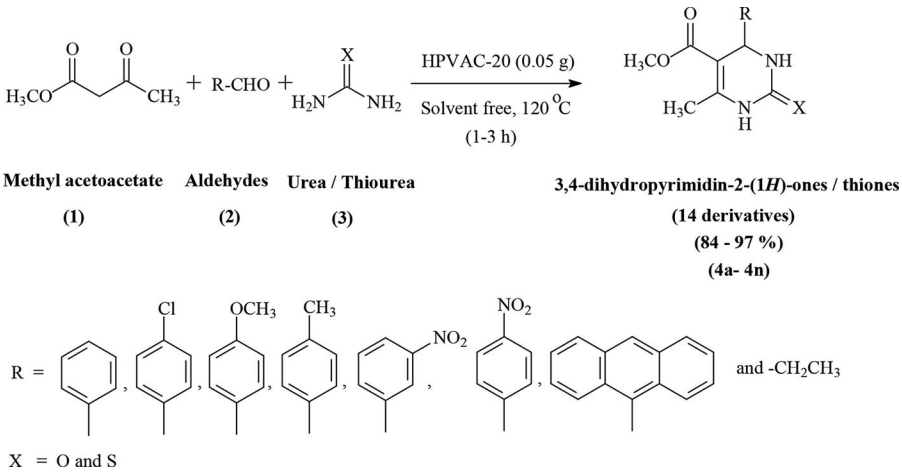
[a]Reaction conditions: methyl acetoacetate (1 mmol), benzaldehyde (1 mmol), thiourea (1.25 mmol), HPVAC-20 (0.05 g), solvent free, 120 °C, 1.5 h.
[b]Isolated yield.

higher than all the other media. It is also interesting to note that the reaction time is reduced from 5 to 1.5 h under solvent free condition.

Reusability of the catalyst

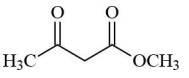
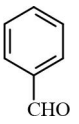
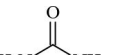
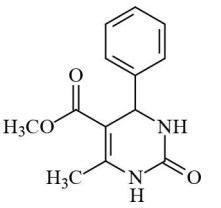
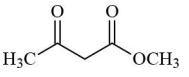
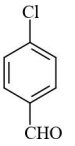
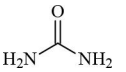
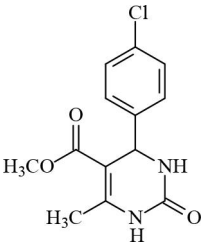
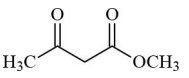
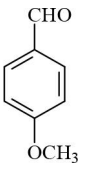
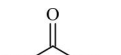
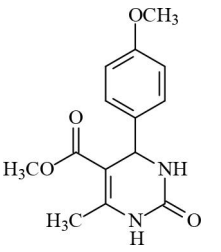
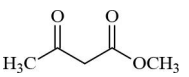
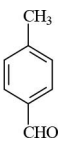
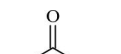
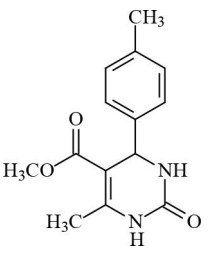
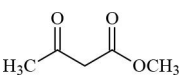
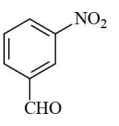
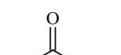
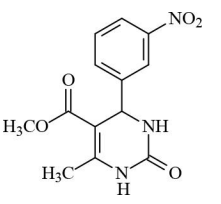
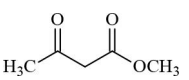
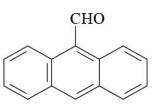
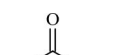
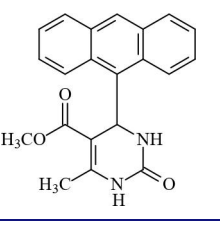
The reusability of the catalyst was evaluated for HPVAC-20 catalyst, by separating the solid from the reaction mixture by simple filtration, washing with hot ethanol and drying in air oven at 120 °C for 3 h prior to reuse in subsequent reactions. Each time a loss of 4% of the catalyst has been found and the final loss of catalyst after its fourth consecutive usage was found to be 14%. No significant loss in the product yield was observed for about four times of its reuse (Table 4).

With the aid of the above experiments the best reaction condition for the title reaction is achieved using HPVAC-20 as catalyst. Using this procedure, different kinds of aromatic aldehydes were condensed with methyl acetoacetate and urea/thiourea to produce the corresponding 3,4-dihydropyrimidin-2-(1*H*)-ones/thiones at 120 °C under solvent free conditions (Scheme 1). Aromatic aldehyde with different functional groups was subjected to the condensation reaction and the corresponding products were obtained in good quantitative yields. In this manner 14 derivatives were prepared and the results in terms of reaction time and percentage of yield are given in Table 5. The functional groups in the aromatic ring of the aldehyde influence the yield of the reaction as well as the reaction



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

Table 5. The condensation reaction of methyl acetoacetate, aldehydes and urea/thiourea over HPVAC-20 by conventional heating.[a]

	Substrates					
Entry	Benzil	Aldehyde	Urea/thiourea	Product	Time (h)	Yield[b] (%)
4a					1.5	93
4b					1	97
4c					3	88
4d					2	92
4e					2	90
4f					3	88

(*Continued*)

Table 5. Continued.

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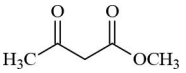
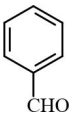
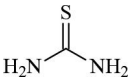
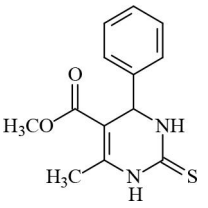
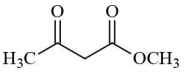
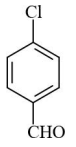
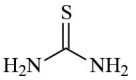
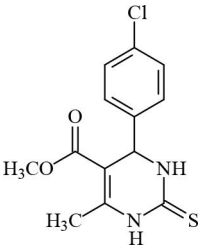
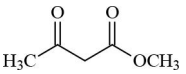
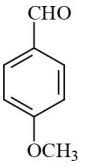
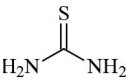
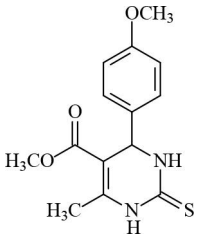
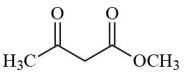
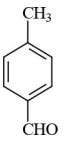
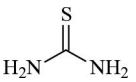
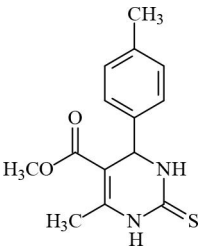
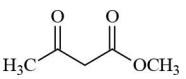
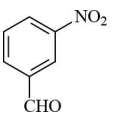
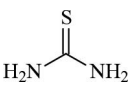
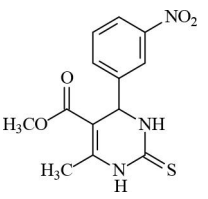
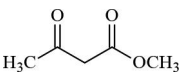
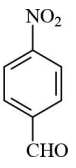
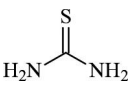
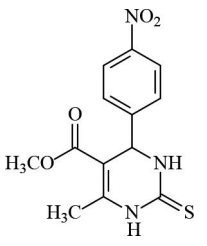
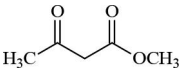
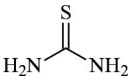
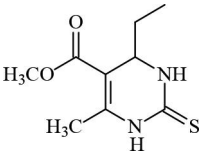
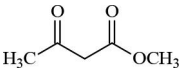
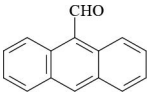
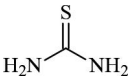
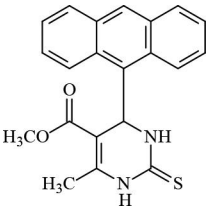
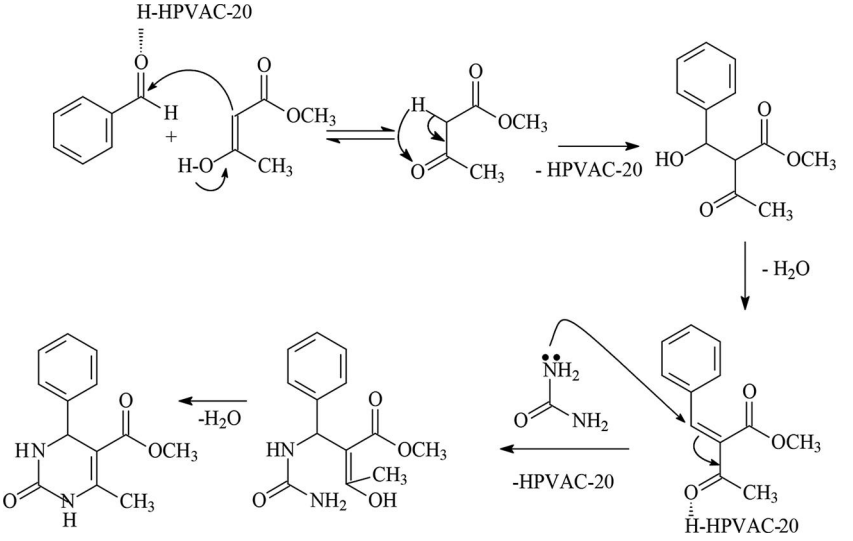
4g					1.5	91
4h					1	94
4i					3	87
4j					2	90
4k					2	89
4l					2	87

(*Continued*)

Table 5. Continued.

Entry	Substrates: Benzil	Substrates: Aldehyde	Substrates: Urea/thiourea	Product	Time (h)	Yield[b] (%)
4m		CH_3CH_2CHO			3	84
4n					3	86

[a]Reaction conditions: methyl acetoacetate (1 mmol), aldehyde (1 mmol), and urea/thiourea (1.25 mmol), HPVAC-20 (0.05 g), solvent free, 120 °C.
[b]Isolated yields.



Scheme 2. A plausible reaction mechanism for the synthesis of 3,4-dihydropyrimidin-2(1*H*)-one/thione using HPVAC catalyst.

Table 6. Comparative account of catalytic ability of HPVAC-20 with other catalysts toward the synthesis of 3,4-dihydropyrimidin-2(1*H*)-thione.

S. no.	Catalysts	Amount (g)	Condition	Reaction time	Yield (%)	Reference
1	AlKIT-5	0.15	CH_3CN solvent and reflux	4 h	82	[9]
2	PPF-SO_3H	0.25	Ethanol solvent and reflux	8 h	75	[20]
3	Iodine	0.1	Microwave (600 W, 60 °C) solvent free	15 min	95	[18]
4	$[PyPS]_3PW_{12}O_{40}$	0.20	Microwave (120 °C) solvent free	10 min	94	[23]
5	Phytic acid	0.01	Solvent free (100 °C)	30 min	85	[26]
6	HPVAC-20	0.05	Solvent free (120 °C)	1.5 h	91	This work

time. The electron withdrawing groups in the aldehyde increase the product yield and reduce the reaction time in comparison with electron donating functional groups.

The plausible mechanism for the title reaction has been proposed and is given in Scheme 2.

A comparative account of efficiency of HPVAC-20 with other reported catalysts for the synthesis of 3,4-dihydropyrimidin-2(1*H*)-thione namely methyl 4-(phenyl)-6-methyl-2-thioxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate was given in Table 6. The data clearly ascertain the versatility of the present protocol using HPVAC-20 as catalyst over the other catalysts.

Experimental

Materials and methods

All commercially available chemicals were obtained from Sigma Aldrich and used without further purification. A series of HPV supported activated natural clay catalysts (HPVAC) were prepared by varying the loading amount of HPV *viz.* 10, 20, and 30% (w/w) on to the activated natural clay and characterized by the literature procedure recently published from this laboratory.[37]

Melting points were measured on an electro-thermal melting point apparatus. FT-IR spectra were recorded using Shimadzu IR Affinity-1 FT-IR spectrophotometer as KBr discs. ^{1}H and ^{13}C-NMR spectra were recorded by Bruker 300 and 100 MHz NMR instrument with DMSO-d_6 as solvent and TMS as internal reference. Elemental analyses were performed on Elementar Vario EL III equipment.

General method for the synthesis of 3,4-dihydropyrimidin-2(1H)-ones/thiones

The catalytic activity of HPVAC toward the condensation reaction of methyl acetoacetate, aldehyde and urea / thiourea to form the 3,4-dihydropyrimidin-2(1*H*)-ones/thiones was established under solvent free condition. In a typical procedure methyl acetoacetate (1 mmol), aldehyde (1 mmol), urea/thiourea (1.25 mmol), and HPVAC-20 (0.05 g) were mixed and the mixture was heated in an oil bath at 120 °C for appropriate time. Progress of the reaction was monitored by TLC (ethylacetate/petroleum ether, 3:7). After completion of the reaction, hot ethanol (15 mL) was added and the catalyst was separated by filtration. The filtrate was poured into crushed ice with stirring. The crude product thus obtained was filtered and washed with cold water (20 mL). The product was recrystallized from 95% ethanol. Finally the pure products of 3,4-dihydropyrimidin-2(1*H*)-one/thione derivatives **(4a–n)** were also identified by melting point measurement and elemental analysis. Further the derivatives were characterized by FT-IR, ^{1}H-NMR, and ^{13}C-NMR spectroscopic methods. The general pattern of the reaction is depicted in Scheme 1.

Conclusion

In conclusion, the efficient synthesis of 3,4-dihydropyrimidin-2(1*H*)-one/thione derivatives was achieved by the condensation reaction of various aromatic aldehydes, methyl acetoacetate and urea/thiourea using HPVAC-20 as a heterogeneous catalyst under solvent free conditions at 120 °C for 1–3 h. This protocol can be considered as a green catalytic system for the synthesis of 3,4-dihydropyrimidin-2(1*H*)-one/thione derivatives. On the other hand, the catalyst can be reused for about four times without significant loss of its

catalytic activity. Organic derivatives were identified by melting point measurements and through elemental (CHNS) analysis and characterized by FT-IR, ^{1}H-NMR, and ^{13}C-NMR spectroscopic methods.

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