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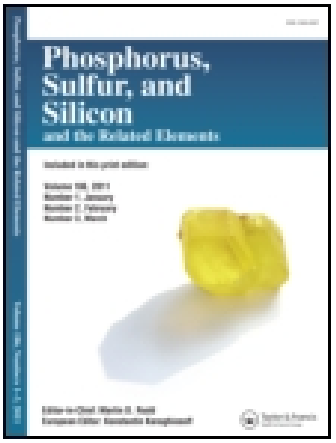

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One-Pot Synthesis of Dihydropyrimidine-Thione Derivatives Using Tungstate Sulfuric Acid (TSA) as a Recyclable Catalyst

Bahador Karami [a], Zahra Haghighijou [a], Mahnaz Farahi [a] & Saeed Khodabakhshi [a]
[a] Department of Chemistry, Yasouj University, Yasouj, Iran
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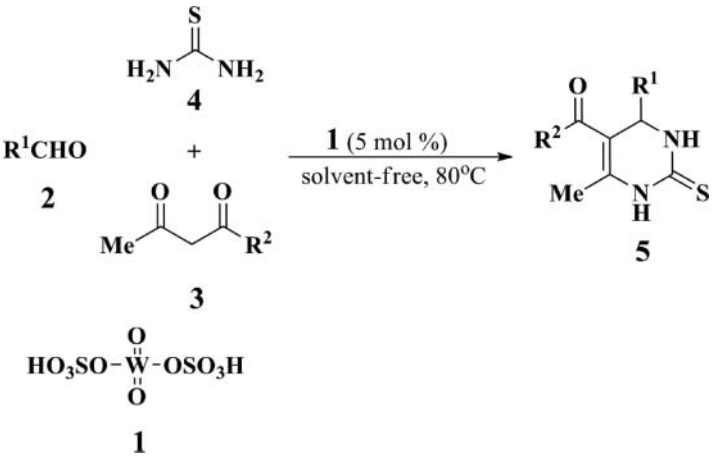
ONE-POT SYNTHESIS OF DIHYDROPYRIMIDINE-THIONE DERIVATIVES USING TUNGSTATE SULFURIC ACID (TSA) AS A RECYCLABLE CATALYST

Bahador Karami, Zahra Haghighijou, Mahnaz Farahi, and Saeed Khodabakhshi
Department of Chemistry, Yasouj University, Yasouj, Iran

GRAPHICAL ABSTRACT



Abstract *An efficient, simple, and environmentally benign procedure for the one-pot synthesis of 3,4-dihydropyrimidine-2-(1H) thione derivatives that entails the three-component condensation of appropriate aryl aldehydes, β-dicarbonyls, and thiourea in the presence of catalytic amount of tungstate sulfuric acid under solvent-free conditions was reported. This method has many advantages including excellent yields, short reaction time, and simple work-up procedure.*

Supplemental materials are available for this article. Go to the publisher's online edition of Phosphorus, Sulfur, and Silicon and the Related Elements *for the following free supplemental resource: 1H, 13C NMR and Mass Spectra for 5b, 5g, 5j, 5i, 5l (S2-S16). Additional Characterization Data (S17-18).*

Keywords Tungstate sulfuric acid; 3,4-dihydropyrimidine-2-(1H) thione; β-dicarbonyls; thiourea

INTRODUCTION

The chemistry of molecules containing sulfur atom attracts continuous attention as a consequence of the potential biological activity of this class of compounds.[1] For instance,

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Address correspondence to Bahador Karami, Department of Chemistry, Yasouj University, P.O. Box 353, Yasouj 75918-74831, Iran. E-mail: karami@mail.yu.ac.ir

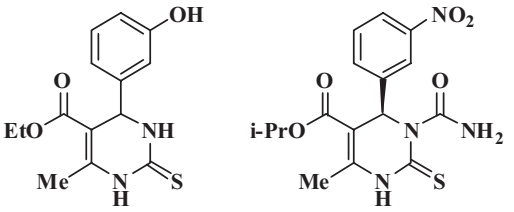

Figure 1 Two types of biologically active dihydropyrimidine compounds.

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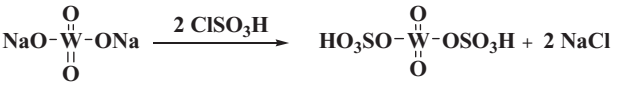
Monastrol, as one of the most important dihydropyrimidine-thione derivatives, is a new class of anticancer agents acting as cell division (mitosis) blockers and the (R)-SQ 32,926 has been identified as a potent orally active antihypertensive agent (Figure 1).[2] Considering the use of functionalized dihydropyrimidine-2(1H)-one/thione as potent calcium channel blockers,[3] antihypertensive agents,[4] and neuropeptide Y antagonist[5] and also, because of their diversified activities such as antibacterial,[6] antiviral,[7] and antitumor properties,[8] it can be concluded that these heterocyclic compounds play a significant role in synthetic, therapeutic, and bioorganic chemistry.

For preparing 3,4-dihydropyrimidine-2-(1H) one/thione derivatives, numerous methods are available in the literature. Generally, these compounds could be prepared via the Biginelli condensation of aldehydes with β-diketones and urea or thiourea derivatives in refluxing ethanolic HCl,[9] but the major disadvantage in this protocol, particularly for substituted aromatic and aliphatic aldehydes, is low yield of the products.[10,11] Due to the importance of Biginelli reaction products, several kinds of improved routes including the use of new catalysts, reagents, and techniques have been developed.[12] However, some of the reported methods have drawbacks such as unsatisfactory yield, long reaction time, and the use of complex or expensive catalysts, organic solvents, and many heavy metallic salts that can pollute the environment[12c–g].

RESULTS AND DISCUSSION

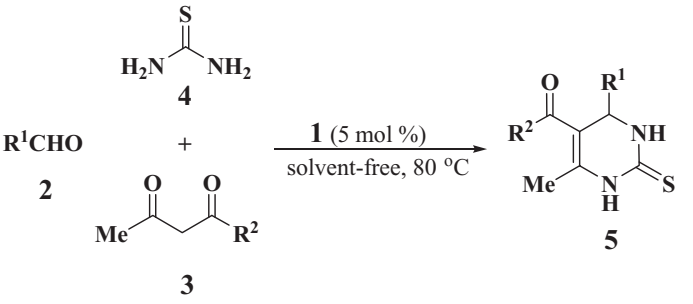
Recently, silica sulfuric acid and Nafion-H[13] have been used for a wide variety of reactions.[14] Accordingly, we found that anhydrous sodium tungstate reacts with chlorosulfonic acid (1:2 mole ratio) to give tungstate sulfuric acid (TSA **1**). The reaction is easy, clean, and performed without any workup (Scheme 1). It is to be noted that there is no gas production during the reaction. More details (particularly the synthetic method and characterization) of this process have been fully described in our previous article.[15a]



1

Scheme 1

In connection with our previous programs on developing the TSA (**1**) in organic transformations,[15] in this work, we wish to report a simple and convenient route for the condensation of aryl aldehydes (**2**) with β-dicarbonyls (**3**) and thiourea (**4**) using tungstate



Scheme 2

sulfuric acid as a catalyst under solvent-free conditions to afford the 3,4-dihydropyrimidine-2-(1H) thione derivatives (**5**) (Scheme 2).

To exploit simple and suitable conditions for the preparation of (**5**) using (**1**) as a solid acid catalyst, the treatment of benzaldehyde, ethyl acetoacetate, and thiourea was chosen as a model reaction. At first, we found that in the absence of (**1**), the reaction did not proceed even at a high temperature. After examining the various amounts of (**1**) according to Table 1 and a wide range of temperatures (Table 2), it was found that the condensation reaction can be efficiently carried out by adding 5 mol% of the catalyst at 80 °C under solvent-free conditions in a short time span of 60 min. The use of excessive amounts of the catalyst does not increase the yield or reaction rate.

In comparison with previous methods, two parallel reactions series, one according to classical Biginelli reaction (Method B) and another in our conditions (Method A), were designed. As shown in Table 3, the product yields were moderate (40%–75%) after 370–660 min, when the reactions were carried out with HCl (aq.) alone (classical Biginelli conditions), whereas the same reactions in the presence of tungstate sulfuric acid gave good to excellent yields (73%–90%) after 45–75 min under solvent-free conditions.

In order to prove the versatility of this method (summarized in Table 3), we extended these reaction conditions to a series of aryl aldehydes and two types of β-dicarbonyl compounds such as ethyl acetoacetate and acetylacetone. Both electron-donating and electron-withdrawing groups on the aryl aldehyde (R^1) reacted well in this process to afford the corresponding products (**5**) in good to excellent yields (Entries 1–13, Table 3).

The main advantages of the presented protocol over existing methods can be seen by comparing our results with those of some recently reported procedures, as shown in Table 4. In many cases, in order to achieve fast synthesis in organic process, a catalyst must be used. The need to implement green chemistry principles (e.g., safer solvents, less hazardous

Table 1 Optimization of the amounts of TSA (**1**) under solvent-free conditions at 80 °C for the synthesis of (**5k**)

Entry	Catalyst amount (mol%)	Time (min)	Yield (%)[a]
1	—	300	10[b]
2	1	300	35[b]
3	2	300	40[b]
4	5	60	80
5	10	70	75
6	20	100	75

[a]Isolated yields.
[b]Not completed.

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Table 2 Optimization of the temperature under solvent-free using TSA **(1)** (5 mol%) for the synthesis of (**5k**)

Entry	Temperature (°C)	Time (min)	Yield (%)[a]
1	25	360	10
2	60	360	25
3	80	60	80
4	120	55	75

[a]Isolated yields.

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Table 3 Synthesis of (**5**) through our and classical Biginelli method

				Yield (%)[a]		Time (min)		
Entry	Product	R^1	R^2	A[b]	B[c]	A[b]	B[c]	mp (°C) [L]
1	**5a**	C_6H_5	Me	80	55	60	660	223–225[16a,b]
2	**5b**	2-OH-C_6H_4	Me	80	40	60	660	224–226[d]
3	**5c**	2-Cl-C_6H_4	Me	73	75	45	600	174–175[16c]
4	**5d**	4-Cl-C_6H_4	Me	76	75	45	480	189–191[16d]
5	**5e**	2,6-Cl_2-C_6H_3	Me	88	65	75	600	133–135[16d]
6	**5f**	4-NO_2-C_6H_4	Me	87	65	75	370	156–158[16e]
7	**5g**	2-OMe-C_6H_4	Me	82	70	45	460	155–157[d]
8	**5h**	4-OMe-C_6H_4	Me	76	68	45	390	187–189[16d]
9	**5i**	3,4,5-OMe_3-C_6H_2	Me	87	64	60	600	152–153[d]
10	**5j**	4-Me-C_6H_4	Me	90	70	60	420	153–155[d]
11	**5k**	C_6H_5	OEt	86	50	60	600	206–207[16d]
12	**5l**	2-OH-C_6H_4	OEt	80	45	60	600	221–223[16f]
13	**5m**	3-NO_2-C_6H_4	OEt	85	65	75	490	206–208[16g]

[a]Isolated yields.
[b]Method A: new reaction conditions under solvent-free (cat. tungstate sulfuric acid, 45–75 min) conditions.
[c]Method B: classical Biginelli conditions (cat. HCl in EtOH, reflux).
[d]Novel compounds.

Table 4 Comparison of our method with other methods for the synthesis of (**5k**) based on the condensation of aldehydes (1 mmol), ethylacetoacetate (1 mmol), and thiourea (1.5 mmol)

Entry	Catalyst	Catalyst amount	Condition	Time (h)/Yield (%)/[Ref.]
1	HCl in EtOH	One drop	Reflux	10/50/[This work]
2	Na_2SeO_4	0.025 g	Solvent-free, 80 °C	1.5/77/[17a]
3	$ClCH_2CO_2H$	0.1 mmol	Solvent-free, 90 °C	5/79–87/[17b]
4	$Zn(BF_4)_2$	Aqueous solution (40% w/v) (0.06 mmol)	RT	6/65/[17c]
5	$Ca(HSO_4)_2$	0.5 mmol	Solvent-free, 90 °C	2.5/74/[17d]
6	P_2O_5/SiO_2	1 g	Solvent-free, 80 °C	2.5/83/[17e]
7	$NaBF_4$	0.2 mmol	Ethanol/Reflux	4.5/89/[17f]
9	TSA	0.05 mmol	Solvent-free, 80 °C	1/86/[This work]

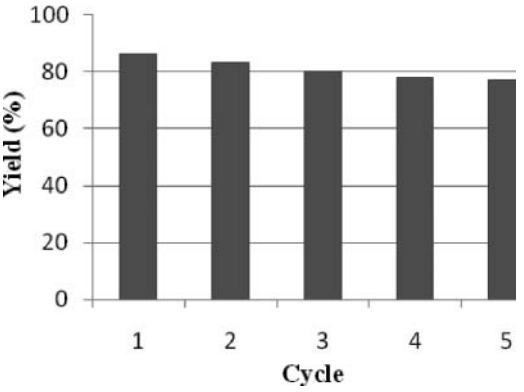
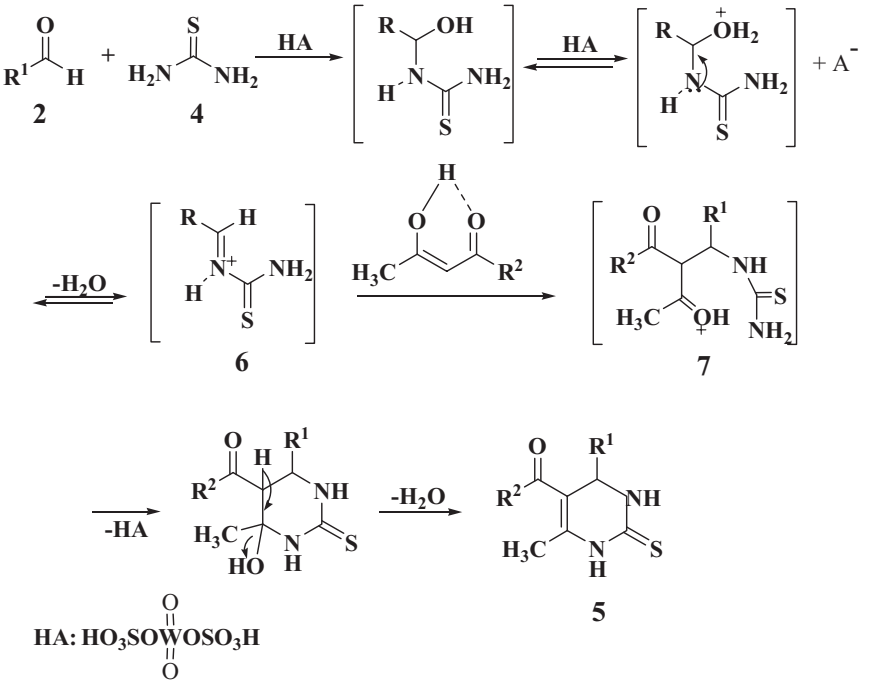


Figure 2 Recyclability of TSA as a catalyst for the synthesis of (5k) on the condensation of aldehydes (1 mmol) and ethylacetoacetate (1 mmol) under solvent-free conditions at 80 °C. Reaction time = 60–70 min.

chemical synthesis, atom economy, and catalysis) is a driving force toward the development of recoverable and reusable catalysts and avoidance of the use of toxic organic solvents.

Not only the ecological profile (through helping to decrease hazardous industrial waste) but also the economic profile (through the elimination of expensive organic solvent) is further improved if the catalyst is recyclable and reaction conditions are solvent-free. In this process, as indicated in Figure 2, the recycled catalyst was used for five cycles during which a little appreciable loss was observed in the catalytic activities.



Scheme 3

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Considering the general mechanistic pathway, which was proposed in a previous report,[18] the suggested mechanism for the formation of (**5**) using (**1**) is shown in Scheme 3. Protonation of the carbonyl group by Brønsted acid (TSA) generates an electrophilic center on the carbonyl carbon atom, which is easily attacked by the thiourea to form minimum intermediate (**6**), which is the key rate-limiting step. Interception of this minimum intermediate (**6**) by β-dicarbonyl produces an open chain thioureide (**7**), which subsequently cyclizes through dehydration process yielding compounds (**5a–n**).

CONCLUSION

In this study, we presented a powerful and modified method for the one-pot reaction of aldehydes, β-dicarbonyls, and thiourea by employing the tungstate sulfuric acid as commercially available, inexpensive, and reusable solid acid catalyst under solvent-free conditions. Higher yields and shorter reaction times than the classical Biginelli method, and simple work-up procedure are the appealing attributes of this project. Moreover, the use of the nonhygroscopic and ecofriendly catalyst such as TSA (**1**) is another advantage that makes this method a valid contribution to the existing methodologies.

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EXPERIMENTAL

General

All chemicals were purchased from Merck, Fluka, and Aldrich. The reactions were monitored by thin layer chromatography (TLC; silica-gel 60 F_{254}, *n*-hexane: ethyl acetate). IR spectra were recorded on a FT-IR JASCO-680 and the ^{1}H NMR spectra were obtained on a Bruker-Instrument DPX-400 MHz Avance 2 model. The varioEl CHNS Isfahan Industrial University was used for elemental analysis. Mass spectra were recorded on a Shimadzu Gc-MS QP 100 Ex spectrometer.

General procedure for the preparation of 3,4-dihdropyrimidine-2(1H)-thiones (5)

A mixture of aryl aldehyde **2** (1 mmol), β-dicarbonyl compound **3** (1 mmol), thiourea **4** (1.5 mmol), and TSA **1** (5 mol%) was stirred and heated at 80 °C in a preheated oil bath for the appropriate time (Table 3). After completing the reaction as indicated by TLC, the reaction mixture was cooled to room temperature and filtered, then washed with cold water, and recrystallized from ethanol to afford the pure product **(5)**. Many of the products were identified by comparing their spectral and physical data with those for authentic samples.[18] ^{1}H, ^{13}C NMR, and mass spectra for **5b**, **5g**, **5i**, **5j**, and **5l** can be found in the Supplemental Materials (Figures S 1–S 15).

Spectral and physical data for novel compounds

5-Acetyl-6-methyl-4-(2-hydroxyphenyl)-3,4-dihydropyrimidine-2(1H)-thione (5b): FT-IR (KBr) cm^{-1}: 3229, 3146, 1609, 1588, 1195, 760, 719; ^{1}H NMR (400 MHz, DMSO-d_6): δ_H (ppm) 9.06 (s, 1H), 9.01 (s, 1H), 7.23–7.18 (m, 2H), 6.94 (t, $J = 7.2$ Hz, 1H), 6.82 (d, $J = 8$ Hz, 1H), 4.74 (d, $J = 2.4$ Hz, 1H), 3.42 (s, 1H), 2.27 (s, 1H), 1.67 (s, 3H); ^{13}C NMR (100 MHz): δ_c (ppm) 176.93, 151.06, 130.09, 129.40, 124.72, 121.13,

116.86, 48.47, 47.89, 29.62, 23.24; Anal. Calcd. for $C_{13}H_{14}N_2O_2S$: C, 59.52; H, 5.38; N, 10.88; S, 12.22. Found: C, 60.83; H, 5.53; N, 10.72; S, 12.34; MS (*m/z*: 262).

5-Acetyl-6-methyl-4-(2-methoxylphenyl)-3,4-dihydropyrimidine-2(1H)-thione (5g): FT-IR (KBr) cm^{-1}: 3325, 3184, 1671, 1592, 1387, 754; ^1H NMR (400 MHz, DMSO-d_6): δ_H (ppm) 10.21 (s, 1H), 9.35 (s, 1H), 7.26 (d, $J = 8$ Hz, 1H), 7.03–6.92 (m, 2H), 6.89 (d, $J = 8$ Hz, 1H), 5.58 (d, $J = 3.6$ Hz, 1H), 3.76 (s, 3H), 2.27 (s, 3H), 2.07 (s, 3H); ^{13}C NMR (100 MHz): δ_c (ppm) 195.57, 174.61, 156.65, 144.35, 130.51, 129.74, 127.76, 120.91, 111.83, 109.61, 55.91, 49.54, 30.10, 18.30; Anal. Calcd. for $C_{14}H_{16}N_2O_2S$: C, 60.85; H, 5.84; N, 10.14; S, 11.60. Found: C, 59.98; H, 5.58; N, 9.37; S, 12.10; MS (*m/z*: 276).

5-Acetyl-6-methyl-4-(3,4,5-trimethoxyphenyl)-3,4-dihydropyrimidine-2(1H)-thione (5i): FT-IR (KBr) cm^{-1}: 3297, 3002, 1613, 1593, 1567, 1187, 1124, 1018, 685; ^1H NMR (400 MHz, DMSO-d_6): δ_H (ppm) 10.28 (s, 1H), 9.71 (s, 1H), 6.51 (s, 2H), 5.25 (s, 1H), 3.71 (s, 6H), 3.62 (s, 3H), 2.33 (s, 3H), 2.16 (s, 3H); ^{13}C NMR (100 MHz): δ_c (ppm) 195.47, 174.56, 153.38, 145.06, 138.86, 137.48, 110.51, 104.23, 60.43, 56.28, 54.23, 30.84, 18.65; Anal. Calcd. for $C_{16}H_{20}N_2O_4S$: C, 57.12; H, 5.99; N, 8.33; S, 9.53. Found: C, 57.45; H, 5.98; N, 8.33; S, 10.44; MS (*m/z*: 336).

5-Acetyl-6-methyl-4-(4-metheylphenyl)-3,4-dihydropyrimidine-2(1H)-thione (5j): FT-IR (KBr) cm^{-1}: 3329, 3177, 1671, 1571, 1454, 1341, 749; ^{1}HNMR (400 MHz, DMSO-d_6): δ_H (ppm) 10.24 (s, 1H), 9.71 (s, 1H), 7.14–7.08 (2d $Jc = 7{,}6$ and 7,6 Hz, 4H), 5.23 (s, 1H), 2.31 (s, 3H), 2.25 (s, 3H), 2.12 (s, 3H); ^{13}C NMR (100 MHz): δ_c (ppm) 195, 28, 174.42, 144.84, 140.45, 137.43, 129.62, 126.96, 110.86, 54.02, 30.78, 21.13, 18.67; Anal. Calcd. for $C_{14}H_{16}N_2OS$: C, 64.58; H, 6.19; N, 10.76; S, 12.32. Found: C, 65.97; H, 6.33; N, 10.84; S, 13.02; MS (*m/z*: 260).

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