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A rapid, efficient, and high-yielding synthesis of 4,4′-(arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ol) derivatives catalyzed by 12-tungstophosphoric acid ($H_3PW_{12}O_{40}$)

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Abstract A fast, green, and efficient method for high-yielding synthesis of 4,4′-(arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ols) by one-pot reaction of two equivalents of 3-methyl-1*H*-pyrazol-5(4*H*)-one with aryl aldehydes, using 12-tungstophosphoric acid ($H_3PW_{12}O_{40}$) as an effective and inorganic catalyst is described. The catalyst is inexpensive and readily available and can be recovered conveniently and reused efficiently such that a considerable catalytic activity still could be achieved after the tenth run. Compared to the other methods, the present methodology offers several advantages such as excellent yields, short reaction times, and mild reaction conditions with an easy work-up.

Keywords 4,4′-(Arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ols) · 3-Methyl-1*H*-pyrazol-5(4*H*)-one · Heteropoly acid · 12-Tungstophosphoric acid

Introduction

Pyrazoles, especially 1*H*-pyrazol-5(4*H*)-one derivatives including 4,4′-(arylmethylene)bis(1*H*-pyrazol-5-ols), are an important class of heterocyclic compounds with diverse and interesting biological activities. These compounds are reported to possess significant antipyretic [1], antidepressant [2], antiviral [3], antifungal [4], antitumor [5], and anti-inflammatory [6] activities. Moreover, a number of these compounds have been considered as fungicides [7], pesticides, and dyestuffs [8, 9], and as the chelating and extracting reagents for different metal ions [10]. Some of the pyrazolone derivatives are now included in many commercialized drugs for brain ischemia [11] and myocardial ischemia [12]. Furthermore, pyrazoles are not only of interest as intermediates in chemical synthesis but also as efficient analytical

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regents in the complexation of transition-metal ions [13]. On the other hand, 4,4′-(arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ols) are applied as important intermediates for the synthesis of fused heterocycles such as 1*H*-pyrano[2,3-c:6,5-c']dipyrazoles [14] and dipyrazolo[3,4-c:3′,4′-f][1,2]diazepines [15]. Additionally, some bispyrazoles are used as ligands for the synthesis of organometallic palladium(II) complexes [16].

In spite of extensive application of 4,4́-(arylmethylene)bis(1*H*-pyrazol-5-ols), a few methods have been reported for the preparation of these interesting compounds. The conventional chemical approach to the synthesis of these compounds involves the successive Knoevenagel synthesis of the corresponding arylidenepyrazolones in first step and its Michael reaction with 1-phenyl-3-methyl-1*H*-pyrazol-5(4*H*)-one in second step [17]. Nevertheless, one-pot synthetic methods for the preparation of 4,4́-(arylmethylene)bis(1*H*-pyrazol-5-ols) are known and employ the tandem Knoevenagel-Michael reaction of aryl aldehydes with two equivalents of 1,3-disubstituted-1*H*-pyrazol-5(4*H*)-ones performed using various catalysts such as 1,3,5-tris(hydrogensulfato)benzene [18], 3-aminopropylated silica gel [19], silica-bonded ionic liquid [20], silica-bonded *N*-propylpiperazine sulfamic acid [21], 2-hydroxy ethylammonium propionate [22], sodium dodecyl sulfate [23], sulfuric acid ([3-(3-silicapropyl)sulfanyl]propyl)ester [24], poly(ethylene glycol)-bound sulfonic acid [25], [HMIM]HSO_4 under ultrasonic irradiation [26], phosphomolybdic acid [27], LiOH.H_2O [28], cellulose sulfuric acid [29], silica-bonded *N*-propyltriethylenetetramine [30], silica-bonded *S*-sulfonic acid [31], and pyridine trifluoroacetate [32]. Catalysis by piperidine in ethanolic solution reported by Singh et al. [33] afforded the desired products in poor 15–35 % yields. The uncatalyzed reactions for the synthesis of these compounds have been also reported in poly(ethylene glycol)-400 (PEG-400) as reaction media at 110 °C [34] or in refluxing water [35]. Even though these uncatalyzed reactions afford the corresponding 4,4́-(arylmethylene)bis(1*H*-pyrazol-5-ols) in 72–94 % yields, the reactions require long reaction times (2–8 h) for completion. Furthermore, Elison et al. [36] applied the electrocatalytic procedure in the presence of sodium bromide as electrolyte to the synthesis of these compounds. However, many of these methodologies are not entirely satisfactory and suffer from limitations such as long reaction times, unsatisfactory yields, and using ultrasonic irradiation or electrocatalytic procedure for accelerated synthesis. Thus, the development of an alternate milder and clean procedure is highly demanding for the synthesis of 4,4′-(arylmethylene)bis(1*H*-pyrazol-5-ols), which surpasses those limitations.

The use of Keggin-type heteropolyacids (HPAs) as homogeneous or heterogeneous catalysts in organic synthesis has been developed as a result of their several advantages, such as environmental compatibility, reusability, non-corrosiveness, and relative lack of disposal problems, which make them economically and environmentally attractive [37, 38]. Being stronger acids, HPAs, especially 12-tungstophosphoric acid ($H_3PW_{12}O_{40}$), generally exhibit higher catalytic activities than conventional catalysts, such as mineral acids, ion-exchange resins, zeolites, etc. [39, 40]. On the other hand, the $H_3PW_{12}O_{40}$ is highly soluble in water and polar organic solvents, such as lower alcohols and carboxylic acids, and insoluble in hydrocarbons. To the best of our knowledge, there are no examples on

the use of $H_3PW_{12}O_{40}$ as homogeneous catalyst for the synthesis of 4,4′-(arylmethylene)bis(1*H*-pyrazol-5-ols). On the other hand, only two papers have been reported for the synthesis of 4,4′-(arylmethylene)bis(1-unsubstituted-1*H*-pyrazol-5-ols) [22, 23].

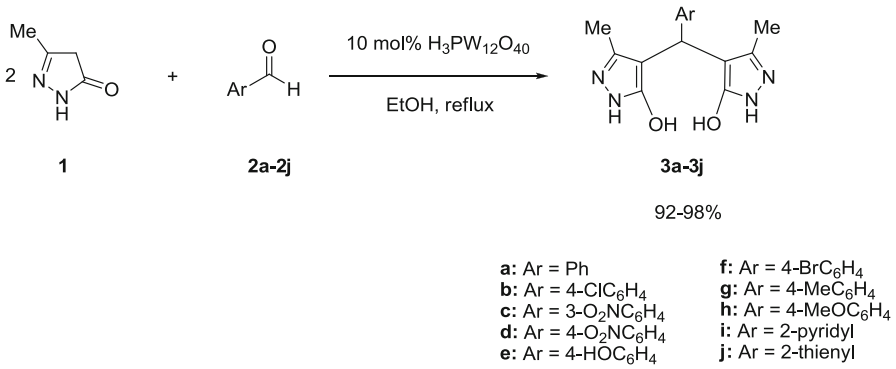
As part of our research program directed towards the development of highly expedient methods using reusable catalysts for the synthesis of organic compounds [41–44], we report here our results on the rapid, efficient, and high-yielding synthesis of 4,4′-(arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ols) by one-pot reaction of two equivalents of 3-methyl-1*H*-pyrazol-5(4*H*)-one [45], with aryl aldehydes in the presence of $H_3PW_{12}O_{40}$, the strongest HPA in the Keggin series [39], as a homogeneous catalyst (Scheme 1).

Experimental

Melting points were recorded using a Stuart SMP3 melting point apparatus. The FT-IR spectra of the products were obtained with KBr disks, using a Tensor 27 Bruker spectrophotometer. The ^{1}H NMR and ^{13}C-NMR spectra were recorded using a Bruker 400 spectrometer at 400- and 100-MHz frequencies, respectively.

General procedure for the synthesis of 4,4′-(arylmethylene)bis(3-methyl-1H-pyrazol-5-ols) **3a–3j**

A mixture of 3-methyl-1*H*-pyrazol-5(4*H*)-one **1** (2 mmol), an aryl aldehyde **2a-2j** (1 mmol), and $H_3PW_{12}O_{40}$ (10 mol % based on aryl aldehyde) in ethanol (2 ml) was heated under reflux for 10–60 min. During the procedure, the reaction was monitored by TLC. Upon completion of the transformation, the reaction mixture was cooled to room temperature. This resulted in the precipitation of the product, which was collected by filtration, washed repeatedly with cold water and recrystallized from ethanol to give products **3a–3j** in high yields.



Scheme 1 $H_3PW_{12}O_{40}$ catalyzed synthesis of 4,4′-(arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ols)

Selected ^{1}H and ^{13}C NMR and FT-IR data

4,4′-(Phenylmethylene)bis(3-methyl-1H-pyrazol-5-ol) ***3a*** ^{1}H NMR (400 MHz, d_6-DMSO): δ 2.08 (s, 6H), 4.83 (s, 1H), 7.08–7.17 (m, 3H), 7.19–7.25 (m, 2H), 11.24 (br., 4H); ^{13}C-NMR (100 MHz, d_6-DMSO): δ 10.8, 33.2, 104.7, 125.9, 127.9, 128.2, 140.2, 143.8, 161.5; IR (KBr disc): υ 3,296, 2971, 2,500–3,500, 1,614, 1,522, 1,492, 1,380, 1,275, 1,214, 1,050, 774, 718 cm^{-1}.

4,4′-((4-Chlorophenyl)methylene)bis(3-methyl-1H-pyrazol-5-ol) ***3b*** ^{1}H NMR (400 MHz, d_6-DMSO): δ 2.08 (s, 6H), 4.82 (s, 1H), 7.13 (d, $J = 8.4$ Hz, 2H), 7.27 (d, $J = 8.4$ Hz, 2H), 11.33 (br., 4H); ^{13}C-NMR (100 MHz, d_6-DMSO): δ 10.8, 32.6, 104.3, 128.1, 129.8, 130.4, 140.1, 142.8, 161.4; IR (KBr disc): υ 2,400–3,500, 1,601, 1,532, 1,491, 1,203, 1,172, 1,091, 1,015, 852, 799, 767 cm^{-1}.

4,4′-((4-Bromophenyl)methylene)bis(3-methyl-1H-pyrazol-5-ol) ***3f*** ^{1}H NMR (400 MHz, d_6-DMSO): δ 2.18 (s, 6H), 4.86 (s, 1H), 7.07 (d, $J = 8.4$ Hz, 2H), 7.42 (d, $J = 8.4$ Hz, 2H), 12.17 (br., 4H); ^{13}C-NMR (100 MHz, d_6-DMSO): δ 10.7, 32.2, 104.5, 119.4, 130.0, 131.2, 141.9, 142.4, 161.1; IR (KBr disc): υ 3,186, 2,400–3,500, 1,595, 1,485, 1,144, 1,079, 1,010, 979, 898, 817 cm^{-1}.

4,4′-((4-Methylphenyl)methylene)bis(3-methyl-1H-pyrazol-5-ol) ***3g*** ^{1}H NMR (400 MHz, d_6-DMSO): δ 2.13 (s, 6H), 2.23 (s, 3H), 4.79 (s, 1H), 6.95–7.05 (m, 4H), 11.83 (br., 4H); ^{13}C-NMR (100 MHz, d_6-DMSO): δ 10.8, 21.0, 32.5, 105.0, 127.7, 128.9, 134.9, 140.3, 141.1, 161.4; IR (KBr disc): υ 3,126, 2,922, 2,400–3,500, 1,599, 1,512, 1,449, 1,080, 961, 898, 820 cm^{-1}.

4,4′-((4-Methoxyphenyl)methylene)bis(3-methyl-1H-pyrazol-5-ol) ***3h*** ^{1}H NMR (400 MHz, d_6-DMSO): δ 2.16 (s, 6H), 3.69 (s, 3H), 4.80 (s, 1H), 6.79 (d, $J = 8.4$ Hz, 2H), 7.03 (d, $J = 8.4$ Hz, 2H), 12.32 (br., 4H); ^{13}C-NMR (100 MHz, d_6-DMSO): δ 10.7, 32.0, 55.4, 105.2, 113.7, 128.7, 135.0, 141.6, 157.8, 161.2; IR (KBr disc): υ 3,205, 2,930, 2,400–3,500, 1,603, 1,510, 1,463, 1,247, 1,178, 1,080, 1,033, 980, 898, 822 cm^{-1}.

4,4′-(Pyridin-2-ylmethylene)bis(3-methyl-1H-pyrazol-5-ol) ***3i*** ^{1}H NMR (400 MHz, d_6-DMSO): δ 2.12 (s, 6H), 5.04 (s, 1H), 7.22–7.30 (m, 1H), 7.37 (d, $J = 8.0$ Hz, 1H), 7.77 (td, $J = 7.6$, 1.6 Hz, 1H), 8.46 (d, $J = 4.5$ Hz, 1H), 11.81 (br., 4H); ^{13}C-NMR (100 MHz, d_6-DMSO): δ 10.8, 36.2, 103.6, 122.2, 122.9, 138.0, 141.0, 148.0, 160.7, 161.9; IR (KBr disc): υ 3,174, 2,927, 2,400–3,500, 1,608, 1,528, 1,471, 1,080, 980, 897, 820 cm^{-1}.

4,4′-(Thiophen-2-ylmethylene)bis(3-methyl-1H-pyrazol-5-ol) ***3j*** ^{1}H NMR (400 MHz, d_6-DMSO): δ 2.12 (s, 6H), 4.99 (s, 1H), 6.58–6.62 (m, 1H), 6.85 (dd, $J = 4.8$, 3.6 Hz, 1H), 7.24 (d, $J = 4.8$ Hz, 1H), 11.47 (br., 4H); ^{13}C-NMR (100 MHz, d_6-DMSO): δ 10.7, 29.3, 104.9, 124.2, 124.3, 126.9, 140.3, 149.1, 161.2; IR (KBr disc): υ 3,373, 3,104, 2,925, 2,300–3,500, 1,595, 1,524, 1,467, 1,080, 980, 897, 820 cm^{-1}.

Results and discussion

To search for the optimal conditions, the synthesis of compound **3b** was selected as a model reaction. The reaction was carried out by heating a mixture of 3-methyl-1*H*-

pyrazol-5(4*H*)-one **1** (2 mmol), and 4-cholorobenzaldehyde **2b** (1 mmol) in the absence or presence of $H_3PW_{12}O_{40}$ as catalyst in different solvents, including H_2O, EtOH, CH_3CN, $CHCl_3$, CH_2Cl_2, *n*-hexane and also under solvent-free conditions. The results are summarized in Table 1. Without the catalyst, no or a trace product was obtained at reflux temperature in solvent or 120 °C under solvent-free conditions even after reaction for 120 min (entries 1–7). Ethanol proved to be a much better solvent in terms of yield as well as reaction time than all the others. Although, there are no significant differences in yields between EtOH and H_2O, the reactions in EtOH have much shorter reaction times. The excellent yield of the product was obtained when the reaction was conducted in refluxing EtOH in the presence of 10 mol % of the $H_3PW_{12}O_{40}$ catalyst (entry 18). No significant

Table 1 Optimization of reaction conditions for the synthesis of compound **3b** catalyzed by $H_3PW_{12}O_{40}$

Entry	Catalyst	Mol %	Solvent	T (°C)	Time (min)	Isolated yield (%)
1	$H_3PW_{12}O_{40}$	–	H_2O	Reflux	120	–
2	$H_3PW_{12}O_{40}$	–	EtOH	Reflux	120	–
3	$H_3PW_{12}O_{40}$	–	CH_3CN	Reflux	120	–
4	$H_3PW_{12}O_{40}$	–	$CHCl_3$	Reflux	120	–
5	$H_3PW_{12}O_{40}$	–	CH_2Cl_2	Reflux	120	–
6	$H_3PW_{12}O_{40}$	–	*n*-hexane	Reflux	120	–
7	$H_3PW_{12}O_{40}$	–	–	120	120	–
8	$H_3PW_{12}O_{40}$	5	H_2O	Reflux	120	60
9	$H_3PW_{12}O_{40}$	5	EtOH	Reflux	30	65
10	$H_3PW_{12}O_{40}$	5	CH_3CN	Reflux	120	20
11	$H_3PW_{12}O_{40}$	5	$CHCl_3$	Reflux	120	Trace
12	$H_3PW_{12}O_{40}$	5	CH_2Cl_2	Reflux	120	Trace
13	$H_3PW_{12}O_{40}$	5	*n*-hexane	Reflux	120	Trace
14	$H_3PW_{12}O_{40}$	5	–	120	120	20
15	$H_3PW_{12}O_{40}$	7	H_2O	Reflux	90	81
16	$H_3PW_{12}O_{40}$	7	EtOH	Reflux	20	85
17	$H_3PW_{12}O_{40}$	10	H_2O	Reflux	90	94
18	$H_3PW_{12}O_{40}$	10	EtOH	Reflux	15	98
19	$H_3PW_{12}O_{40}$	15	EtOH	Reflux	15	97
20	$H_4SiW_{12}O_{40}$	5	H_2O	Reflux	120	43
21	$H_4SiW_{12}O_{40}$	5	EtOH	Reflux	30	44
22	$H_4SiW_{12}O_{40}$	5	CH_3CN	Reflux	120	17
23	$H_4SiW_{12}O_{40}$	7	H_2O	Reflux	90	58
24	$H_4SiW_{12}O_{40}$	7	EtOH	Reflux	20	60
25	$H_4SiW_{12}O_{40}$	10	H_2O	Reflux	90	72
26	$H_4SiW_{12}O_{40}$	10	EtOH	Reflux	15	74
27	$H_4SiW_{12}O_{40}$	15	EtOH	Reflux	15	74

Reaction conditions: 3-methyl-1*H*-pyrazol-5(4*H*)-one **1** (2 mmol), and 4-cholorobenzaldehyde **2b** (1 mmol)

improvement in the time of the reaction was observed using higher amount of the catalyst (entry 19). For comparison and to show the merit of the $H_3PW_{12}O_{40}$ catalyst, synthesis of the model compound, **3b**, was also investigated using another HPA, silicotungstic acid ($H_4SiW_{12}O_{40}$) as catalyst. The results are shown in Table 1 (entries 20–27). As depicted, $H_3PW_{12}O_{40}$ proved to be the better catalyst than $H_4SiW_{12}O_{40}$ in terms of reaction time and yield. Therefore, our optimized condition is 10 mol % of $H_3PW_{12}O_{40}$ in refluxing EtOH. All subsequent reactions were carried out in these optimized conditions.

Using these optimized reaction conditions, the scope and efficiency of this approach was explored for the synthesis of a wide variety of 4,4′-(arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ols) and the obtained results are summarized in Table 2. All the aforementioned reactions (Table 2) delivered excellent product yields and accommodated a wide range of aromatic aldehydes bearing both electron-donating and electron-withdrawing substituents. Also, heteroaromatic aldehydes performed well and produced a high yield of the corresponding product.

Although there are only two papers on the synthesis of 4,4′-(arylmethylene)bis(1-unsubstituted-1*H*-pyrazol-5-ols) [22, 23], to further evaluate the overall utility of the current methodology, we compared our results with those of the other methods reported for the synthesis of 4,4′-(arylmethylene)bis(1-substituted-1*H*-pyrazol-5-ols). This comparison is shown in Table 3. It is clear from the data that our method has short reaction times and provides higher yields of the products.

We also used our optimized reaction conditions to evaluate the reusability of the catalyst $H_3PW_{12}O_{40}$. After completion of the reaction, the reaction mixture was cooled to room temperature, the product was collected by filtration, and washed repeatedly with cold water. The combined filtrate was evaporated to dryness under reduced pressure. The solid catalyst was collected, dried at 70 °C under vacuum for 1 h, and reused for the same experiment. We found that the catalyst could be used at least ten times with only a slight reduction in activity (Fig. 1).

Furthermore, retention of the structure of the catalyst was confirmed by comparing the FT-IR spectra of the recovered catalysts (Fig. 2b–f) with that of the fresh catalyst (Fig. 2a) for the model reaction. As shown, these spectra are almost identical.

Considering the Brønsted acidic nature of $H_3PW_{12}O_{40} \equiv HA$, a possible mechanism is proposed as depicted in Scheme 2. The catalyst plays a significant role in increasing the electrophilic character of the electrophiles in the reaction. The reaction occurs via initial formation of the intermediate [**I**], prepared by Knoevenagel condensation of first equivalent of enolic form of 3-methyl-1*H*-pyrazol-5(4*H*)-one **1** and aryl aldehydes **2a–2j**, which reacts subsequently with second equivalent of enolic form of **1** to afford the desired products **3a–3j**. Under these conditions, attempts to isolate the intermediate [**I**] failed even after careful monitoring of the reactions.

In summary, we successfully developed a simple, efficient and ecofriendly method for the synthesis of 4,4′-(arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ols) by one-pot reaction of two equivalents of 3-methyl-1*H*-pyrazol-5(4*H*)-one with aryl aldehydes, using cheap and readily available $H_3PW_{12}O_{40}$ as homogeneous catalyst. Some attractive features of this protocol are excellent yields, simple procedure,

Table 2 $H_3PW_{12}O_{40}$ catalyzed synthesis of 4,4′-(arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ols) **3a–3j**

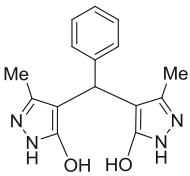
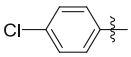
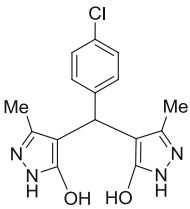
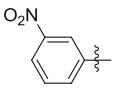
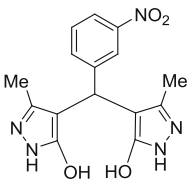
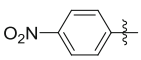
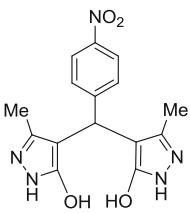
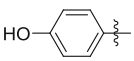
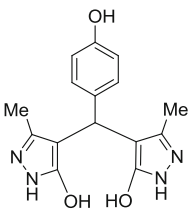
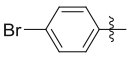
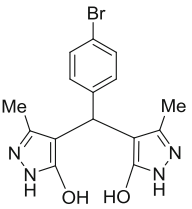
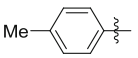
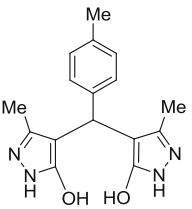
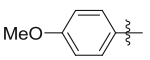
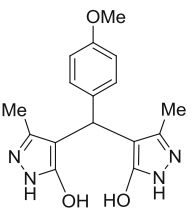
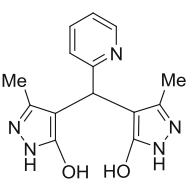
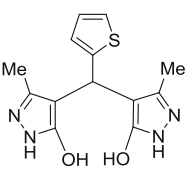
Entry	Ar	Product*	Time (min)	Isolated yield (%)	m.p. (°C)	
					Found	Reported
1		 **3a**	15	91	242–244	230–232 [23]
2		 **3b**	15	98	220–222	224–226 [23]
3		 **3c**	10	92	269–270	271–272 [23]
4		 **3d**	10	93	297–299	300–302 [23]
5		 **3e**	60	94	261–263	262–264 [23]

Table 2 continued

Entry	Ar	Product*	Time (min)	Isolated yield (%)	m.p. (°C)	
					Found	Reported
6		 **3f**	10	97	218–222	–
7		 **3g**	60	96	191–193	–
8		 **3h**	60	93	189–191	–
9		 **3i**	60	97	211–212	–
10		 **3j**	60	93	203–205	–

Reaction conditions: 3-Methyl-1*H*-pyrazol-5(4*H*)-one **1** (2 mmol), aryl aldehyde **2a–2j** (1 mmol), $H_3PW_{12}O_{40}$ (10 mol % based on aryl aldehyde), EtOH (2 ml), reflux

* Characterization of new products were performed according to their IR, ^{1}H NMR, and ^{13}C NMR data, and for known products by their IR spectral data and a comparison of their melting points with those of authentic samples

Table 3 Comparison of the efficiencies of different catalysts for the synthesis of 4,4′-(arylmethylene)bis(1-(un)substituted-1*H*-pyrazol-5-ols)

Catalyst	Conditions			Time (min)	Yield (%)	References
	Solvent	T/°C	Other			
Silica-bonded ionic liquid	EtOH	Reflux	–	60–270	77–90	[20]
Silica-bonded *N*-propylpiperazine sulfamic acid	–	80	–	35–60	80–93	[21]
2-Hydroxy ethylammonium propionate	–	90	–	10–80	77–96	[22]
Sodium dodecyl sulfate	H_2O	Reflux	–	60–240	78–92	[23]
Sulfuric acid ([3-(3-silicapropyl) sulfanyl]propyl)ester	EtOH	Reflux	–	130–260	74–90	[24]
Poly(ethylene glycol)-bound sulfonic acid	H_2O	Reflux	–	30–120	76–94	[25]
[HMIM]HSO_4	EtOH	r.t.	Ultrasonic irradiation	30–75	40–92	[26]
Phosphomolybdic acid	EtOH	Reflux	–	210–330	91–96	[27]
$LiOH.H_2O$	H_2O	90	–	60–75	79–86	[28]
Cellulose sulfuric acid	EtOH/ H_2O	Reflux	–	10–200	74–97	[29]
Silica-bonded *N*-propyltriethylenetetramine	EtOH	Reflux	–	15–120	72–93	[30]
Silica-bonded *S*-sulfonic acid	EtOH	Reflux	–	40–240	75–90	[31]
Pyridine trifluoroacetate	H_2O	70	–	300–900	75–95	[32]
$H_3PW_{12}O_{40}$	EtOH	Reflux	–	10–60	93–98	this work

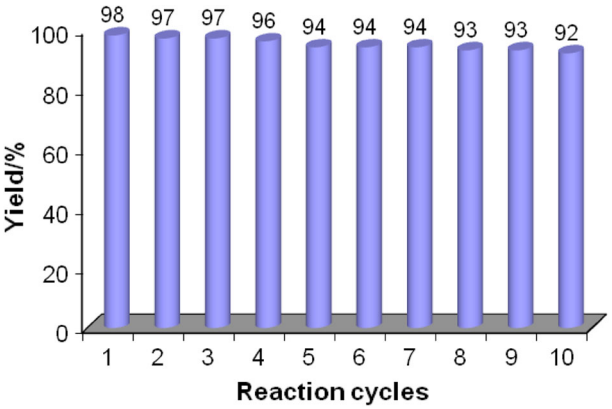


Fig. 1 Effect of recycling on catalytic performance of $H_3PW_{12}O_{40}$ in the synthesis of **3b** in model reaction

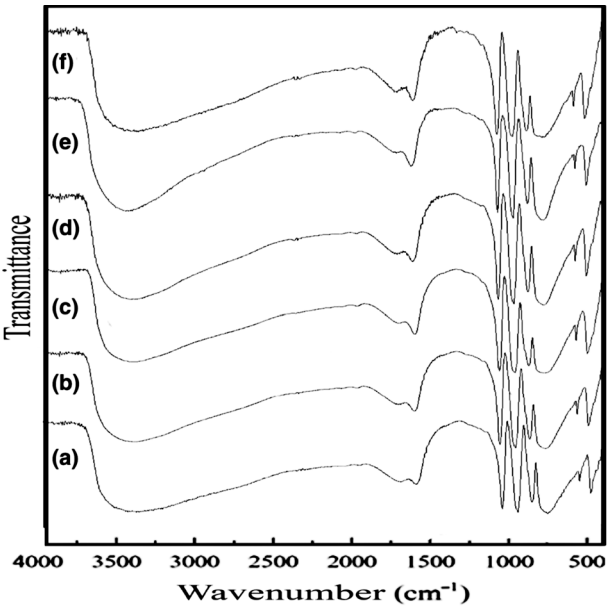
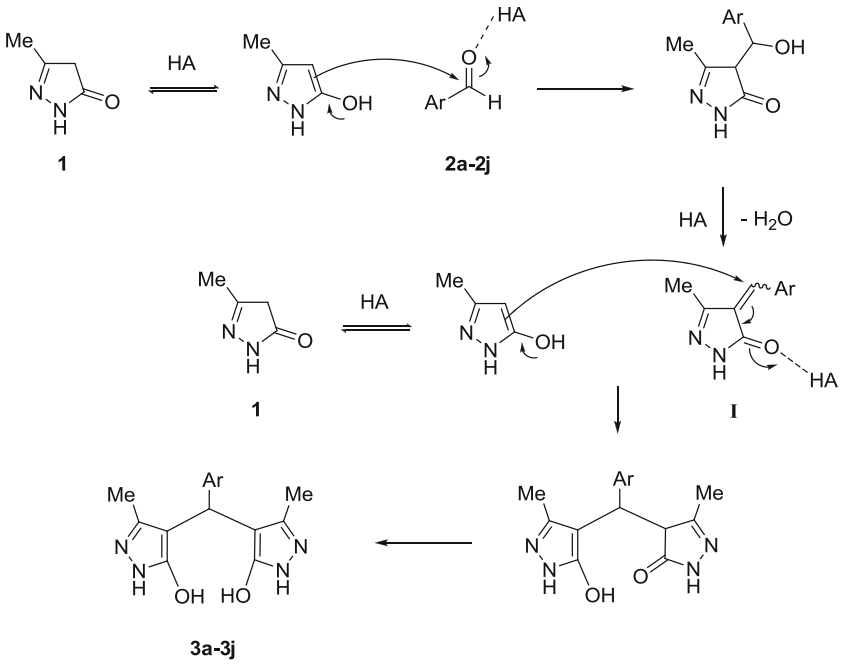


Fig. 2 FT-IR spectra of fresh catalyst $H_3PW_{12}O_{40}$ (run 1, *a*), and recovered catalysts (runs 2, 4, 6, 8 and 10, *b–f*, respectively) for synthesis of compound **3b** in model reaction



Scheme 2 Plausible mechanism for the formation of 4,4′-(arylmethylene)bis(3-methyl-1*H*-pyrazol-5-ols) in the presence of $H_3PW_{12}O_{40} \equiv$ HA as catalyst

short reaction times, easy work-up, high catalytic activity and recyclability, and reusability of the catalyst. The catalyst can be used at least ten times without substantial reduction in its catalytic activity.

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