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RESEARCH PAPER

Tungstophosphoric Acid Supported on Highly Organosoluble Polyamide (PW_{12}/PA): Highly Efficient Catalysts for the Synthesis of Novel 1,3,5-Triaryl-2-pyrazoline Derivatives

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Abstract: A novel compound constructed from polyoxometalate ($H_3PW_{12}O_{40}$, PW_{12}) and poly(amidoamine) (PA) was prepared at room temperature in an aqueous solution by an impregnation method. A series of novel 1,3,5-triaryl-2-pyrazoline derivatives was synthesized by the reaction between chalcone and phenylhydrazine in the presence of the title compound, PW_{12}/PA, in high yields. The structures of the compounds obtained were determined by IR and ^{1}H NMR spectra.

Key words: inorganic-organic hybrid; polyoxometalate; pyrazoline; phenylhydrazine; chalcone

Pyrazolines are well known and important nitrogen-containing five-membered heterocyclic compounds and various methods have been used for their synthesis [1–4]. Pyrazoline derivatives are attracting an increasing amount of interest by many researchers in medicinal chemistry because of their bioactivity such as antimicrobial [5,6], antiamoebic [7,8], antinociceptive [9], anticancer [10], antidepressant [11], and anti-inflammatory [12–16] activities, and also in conjugated fluorescent dyes that emit blue fluorescence with high fluorescence quantum yields [17,18] and electroluminescence yields [19–21]. After the pioneering work of Fischer and Knövenagel [22] in the late nineteenth century, the reactions of α,β-unsaturated aldehydes and ketones with hydrazines became one of the most popular methods for the preparation of 2-pyrazolines [23–30]. Among the various pyrazoline derivatives, 1,3,5-triaryl-2-pyrazolines seem to be the most frequently studied pyrazoline-type compounds. Several catalysts have been developed for the preparation of these heterocycles including a sodium acetate-acetic acid aqueous solution under ultrasound irradiation [31], a hot acetic acid solution [32], a K_2CO_3-mediated microwave irradiation [33], and $H_3PW_{12}O_{40}$ [34].

In continuation of our previous work on the catalytic properties of heteropoly acids (HPAs) [35–37], we now report a suitable synthesis of an inorganic-organic hybrid compound constructed from polyoxometalate ($H_3PW_{12}O_{40}$) and poly(amidoamine) (PW_{12}/PA) as a heterogeneous catalyst for the synthesis of 1,3,5-triaryl-2-pyrazolines derivatives.

1 Experimental

All materials were commercial reagent grade. Aldehydes, phenylhydrazine, and acetophenone were obtained from Merck or Aldrich. $H_3PW_{12}O_{40}$ was purchased from Merck. FT-IR

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spectra were obtained as potassium bromide pellets in the range 400–4000 cm^{-1} with a Nicolet Impact 400 D. ^1H NMR spectra were recorded with a Bruker Avance AQS 300 MHZ. The melting points were determined using an electrothermal digital melting point apparatus and are uncorrected. Reaction courses and product mixtures were monitored by thin layer chromatography (TLC).

1.1 Preparation of the catalyst

1.1.1 Synthesis of support monomer

5-[3-Phenyl-2-(9,10-dihydro-9,10-ethanoanthracene-11,12-dicarboximido)propanoyl-amino] isophthalic acid **1** was synthesized and purified as described elsewhere [38].

1.1.2 Synthesis of polymer

The PAs were prepared by the following general procedure: 0.10 g (1.70×10^{-4} mol) of diacid monomer **1** and 0.0338 g (1.70×10^{-4} mol) of diamine **2**(Benzidine) were dissolved in 0.35 g of [1,3-$(pr)_2$im]Br, and then 0.11 ml (4.26×10^{-4} mol) of triphenyl phosphate (TPP) was added. The mixture was heated at 110 °C for 2.5 h. As the reaction proceeded, the solution became viscous. The resulting product was poured into 30 ml of methanol, filtered, and dried under vacuum to give 0.120 g (90%) of PA, **3** (Scheme 1) [39].

1.1.3 Preparation of PW_{12}/PA

The PW_{12}/PA catalyst was prepared by the incipient wetness method. The adsorption was performed following the procedure described previously [40]. In a typical process, a 640 mg portion of PW_{12} was dissolved in deionized water and impregnated dropwise into 1600 mg of PA in 25 ml methanol with constant agitation. The resulting pastes were dried for 4 h at 110 °C.

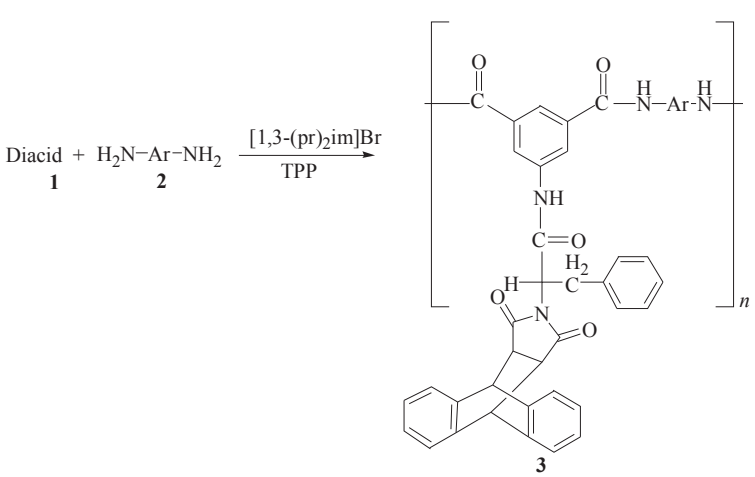
1.2 Typical procedure for the synthesis of 1,3,5-triaryl-2-pyrazoline

The starting chalcone **4** (Scheme 2) was easily prepared by Claisen-Schmidt condensation between acetophenone and aromatic aldehydes in the presence of an ethanolic solution of sodium hydroxide according to a literature procedure [26].

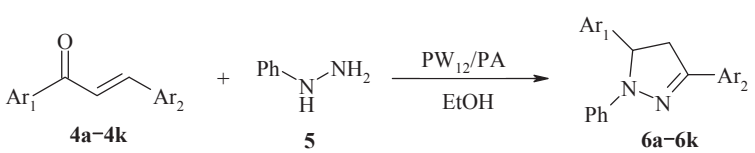
The typical procedure for catalytic synthesis of 1,3,5-triaryl-2-pyrazoline was illustrated in Scheme 2. To a reaction vessel was added chalcone (1 mmol), phenylhydrazine (1 mmol), and PW_{12}/PA (3 mol%). Reaction progress was monitored by TLC. At the end of the reaction, the reaction mixture was washed out of the vessel using CH_2Cl_2 (10 ml) and then filtered. The supported catalyst was easily recovered quantitatively by simple filtration. The filtrate was dried ($MgSO_4$) and evaporated. The crude product was purified on a silica gel plate or a silica gel column (20% ethyl acetate in hexane) to give 1,3,5-triaryl-2-pyrazoline. The products were identified by a comparison of their physical data with those prepared in accordance with the literature procedures.

1.3 Selected spectroscopic data

5-(4-Methylphenyl)-1,3-diphenyl-2-pyrazoline (**6c**). ^1H



Scheme 1. Synthesis of polymer **3**.



Scheme 2. Catalytic synthesis of 1,3,5-triaryl-2-pyrazoline derivatives (**6**) from chalcone and phenylhydrazine over PW_{12}/PA.

NMR ($CDCl_3$): δ 2.34 (s, 3H, CH_3), 3.14 (dd, J = 7.1, 17.0 Hz, 1H), 3.85 (dd, J = 12.1, 17.0 Hz, 1H), 5.27 (dd, J = 6.9, 12 Hz, 1H), 6.77–7.75 (m, 14H). Anal. calcd. for $C_{22}H_{20}N_2$: C 84.62, H 6.41, N 8.97; Found: C: 84.61, H 6.43, N 9.00. ^{13}C NMR ($CDCl_3$): δ 21.68, 44.09, 64.73, 113.69, 119.47, 126.23, 128.43, 128.88, 129.34, 129.57, 130.19, 133.07, 137.67, 141.53, 145.43, 147.17. IR (KBr, cm^{-1}): ν_{max} 1117, 1499, 1593.

5-(4-Chlorophenyl)-1,3-Diphenyl-2-Pyrazoline (**6d**). ^{1}H-NMR ($CDCl_3$): δ 3.04 (dd, J = 7.4, 17.6 Hz, 1H), 3.77 (dd, J = 11.6, 17.6 Hz, 1H), 5.67 (dd, J = 7.4, 11.6 Hz, 1H), 6.71–7.64 (m, 14H). Anal. Calcd. for $C_{21}H_{17}ClN_2$: C 75.78, H 5.15, N 8.41; Found: C 75.69, H 5.10, N 8.49%. ^{13}CNMR ($CDCl_3$): δ 43.4, 63.8, 113.4, 119.4, 125.7, 127.3, 128.5, 128.7, 128.9, 129.3, 129.9, 132.5, 133.3, 141.1, 144.6, 146.7. IR (KBr, cm^{-1}): ν_{max} 1123, 1495, 1598.

5-(3-Chlorophenyl)-1,3-diphenyl-2-pyrazoline (**6e**). ^{1}H NMR ($CDCl_3$): δ 3.08 (dd, J = 6.8, 17.0 Hz, 1H), 3.43 (dd, J = 12.2, 17.2 Hz, 1H), 5.63 (dd, J = 6.9, 12.4 Hz, 1H), 6.85–7.81 (m, 14H). Anal. calcd. for $C_{21}H_{17}N_2Cl$: C 85.85, H 5.72, N 9.43; Found: C 85.79, H 5.70, N 9.41. ^{13}C NMR ($CDCl_3$): δ 42.37, 61.54, 113.58, 119.60, 124.44, 127.79, 128.07, 128.92, 129.18, 129.48, 130.67, 132.53, 133.04, 135.04, 139.67, 144.84, 147.53. IR (KBr, cm^{-1}): ν_{max} 1127, 1501, 1593.

5-(2-Chlorophenyl)-1,3-diphenyl-2-pyrazoline (**6f**). ^{1}H NMR ($CDCl_3$): δ 3.06 (dd, J = 4.8, 17.6 Hz, 1H), 3.96 (dd, J = 11.2, 17.7 Hz, 1H), 5.64 (dd, J = 4.7, 11.0 Hz, 1H), 6.76–7.74 (m, 14H). Anal. Calcd. for $C_{21}H_{17}ClN_2$: C 75.78, H 5.15, N 8.41; Found: C 75.83, H 5.23, N 8.38%. ^{13}C NMR ($CDCl_3$): δ 41.9, 61.5, 113.3, 119.2, 125.9, 127.5, 127.9, 128.3, 128.5, 128.8, 129.3. 129.8, 131.4, 132.2, 139.5, 144.6, 147.4. IR (KBr, cm^{-1}): ν_{max} 1120, 1497, 1595.

5-(2,4-Dichlorophenyl)-1,3-diphenyl-2-pyrazoline (**6g**). ^{1}H NMR ($CDCl_3$): δ 3.02 (dd, J = 6.6, 17.6 Hz, 1H), 3.97 (dd, J = 12.5, 17.5 Hz, 1H), 5.59 (dd, J = 6.6, 12.2 Hz, 1H), 6.69–7.71 (m, 13H). Anal. Calcd. for $C_{21}H_{16}Cl_2N_2$: C 68.67, H 4.39, N 7.62; Found: C 68.73, H 4.39, N 7.71%. ^{13}CNMR ($CDCl_3$): δ 41.5, 60.9, 113.5, 119.5, 124.3, 125.6, 127.4, 127.7, 128.6, 128.5, 128.5, 129.2, 129.5, 132.1, 133.9, 137.7, 144.4, 147.5. IR (KBr, cm^{-1}): ν_{max} 1118, 1503, 1589.

5-(3-Bromophenyl)-1,3-diphenyl-2-pyrazoline (**6h**). ^{1}H NMR ($CDCl_3$): δ 3.08 (dd, J = 7.1, 17.0 Hz, 1H), 3.35 (dd, J = 12.1, 16.9 Hz, 1H), 5.68 (dd, J = 6.9, 12.7 Hz, 1H), 6.80–7.75 (m, 14H). Anal. calcd. for $C_{21}H_{17}N_2Br$: C 85.85, H 5.72, N 9.43; Found: C 85.78, H 5.69, N 9.43. ^{13}C NMR ($CDCl_3$): δ 42.35, 60.47, 113.35, 119.50, 124.23, 127.94, 128.05, 129.06, 129.16, 129.35, 130.59, 132.41, 133.18, 139.66, 145.25, 147.14. IR (KBr, cm^{-1}): ν_{max} 1126, 1502, 1598.

5-(4-Methoxyphenyl)-1,3-diphenyl-2-pyrazoline (**6i**). ^{1}H NMR ($CDCl_3$): δ 3.12 (dd, J = 7.1, 17.1 Hz, 1H), 3.82 (s, 3H, OCH_3), 3.87 (dd, J = 12.1, 16.9 Hz, 1H), 5.24 (dd, J = 7.2, 12 Hz, 1H), 6.75–7.85 (m, 14H). Anal. calcd. for $C_{22}H_{20}N_2O$: C 84.62, H 6.41, N 8.97; Found: C 84.56, H 6.40, N 8.93. ^{13}C NMR ($CDCl_3$): δ 44.08, 55.64, 64.14, 113.72, 114.46, 119.54, 126.42, 127.91, 128.47, 128.89, 129.19, 130.56, 133.21, 135.03, 145.34, 147.17. IR (KBr, cm^{-1}): ν_{max} 1120, 1261, 1512, 1597.

3-(4-Chlorophenyl)-1,5-diphenyl-2-pyrazoline (**6j**). ^{1}H NMR (DMSO): δ 3.15 (dd, J = 7.1, 17.0 Hz, 1H), 3.87 (dd, J = 12.2, 17.1 Hz, 1H), 5.33 (dd, J = 7.3, 12.4 Hz, 1H), 6.83–7.67 (m, 14H). Anal. calcd. for $C_{21}H_{17}N_2Cl$: C 84.85, H 5.72, N 9.43; Found: C 84.81, H 5.77, N 9.47. ^{13}C NMR (DMSO): δ 42.39, 61.31, 113.53, 117.26, 126.83, 126.94, 128.64, 129.16, 129.39, 130.55, 132.17, 132.19, 136.05, 139.61, 143.73, 147.39. IR (KBr, cm^{-1}): ν_{max} 1121, 1509, 1599.

2 Results and discussion

2.1 Physicochemical characterization

Figure 1(a) shows the obtained XRD patterns of pure PA. Peaks that correspond to an amorphous polymer are present. Figure 1(b) shows the XRD pattern of a 35 wt% PW_{12} impregnated PA that was dried at 110 °C. We observed that the impregnated products are crystalline. Some peaks of PW_{12} overlapped with that of PA. This may result from the interaction of PW_{12} with the support.

FT-IR is a powerful technique for the study of surface interactions between HPA and organic or inorganic supports. Figure 2(1) shows the FT-IR spectra of pure PA. The FT-IR spectrum of the 35 wt% PW_{12}/PA shows four bands in the range 1250–500 cm^{-1} (Fig. 2(2)). For the 110 °C dried samples,

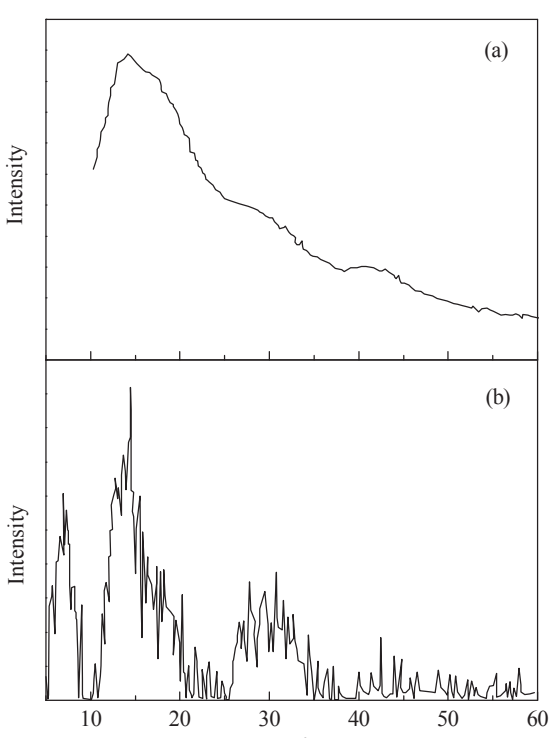


Fig. 1. XRD patterns of PA (a) and PW_{12}/PA (b).

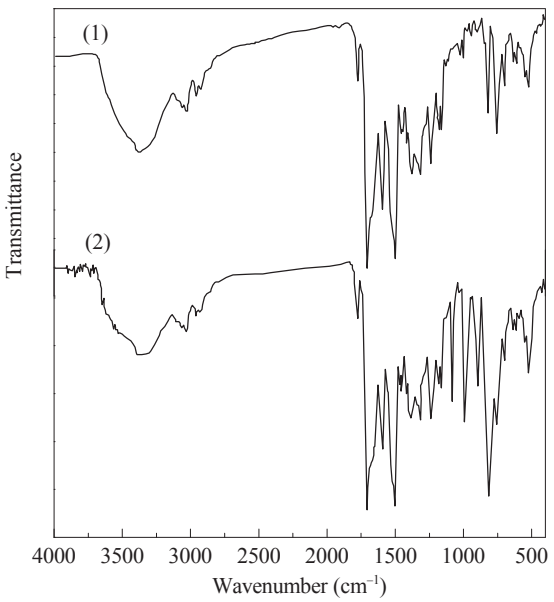


Fig. 2. FT-IR spectra of PA (1) and PW_{12}/PA (2).

Keggin bands are observed at 1079, 979, 816, and 634 cm^{-1} for PW_{12}/PA.

2.2 Choice of reaction medium and the effect of the catalyst and HPA loading on the synthesis of the 1,3,5-triaryl-2-pyrazolines derivatives

The condensation reaction between chalcone and phenylhydrazine was initially performed in the presence of a catalytic amount of several supported polyoxometalates (Fig. 3). To determine the most appropriate medium in this heterocyclization reaction, the synthesis of 1,3,5-triaryl-2-pyrazolines from the condensation of chalcone and phenylhydrazine in the presence of a catalytic amount of PW_{12}/PA was selected as a model reaction.

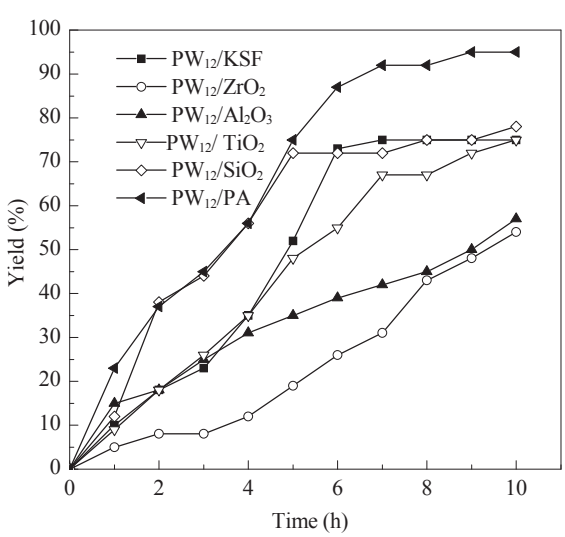


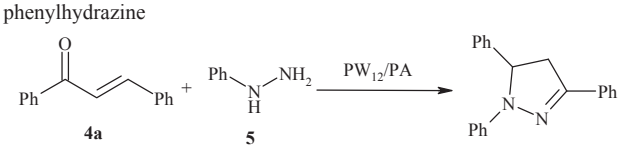
Fig. 3. Effect of the different supported-PW_{12} compounds in the reaction of chalcone with phenylhydrazine. Reaction conditions: chalcone 1 mmol, phenylhydrazine 1 mmol, and supported-PW_{12} (2 mol%) in solvent (5 ml) after 10 h at 45 °C.

To establish the optimal conditions, a series of experiments was carried out by varying the amount of catalyst, the stoichiometry of the reaction, and the temperature. Alcoholic solvents (methanol and ethanol), CH_2Cl_2, CH_3CN, and $CHCl_3$ gave good conversions (Table 1, entries 1–4). Clearly, ethanol stands out as the solvent of choice as it gave a fast conversion, a high yield, and was easily removed. The effect of different PW_{12} loading on the catalytic activity for the synthesis of the 1,3,5-triaryl-2-pyrazoline derivatives (after the catalyst was dried at 150 °C) was investigated and the results are shown in Table 1, entries 4–7. Hardly any reaction with PA occurred. When PW_{12} was supported on PA, the catalytic activity increased remarkably with an increase in the PW_{12} loading but when the loading exceeded 35%, the catalytic activity decreased. The support does not always have a mere mechanical role as it can also modify the catalytic properties of the deposit and favor the growth of particular structures, and this includes weak and strong interactions. The true role of the support in this work is not clear but it appears that the nature of the support and the amount of deposit may influence the catalytic properties of the supported catalysts [36]. These conditions were applied to a series of substituted aromatic aldehydes.

2.3 Synthesis of 1,3,5-triaryl-2-pyrazoline derivatives in the presence of catalytic amounts of PW_{12}/PA

On the basis of the above results, we extended the scope and generality of this method by cyclizing several structurally diverse chalcones and phenylhydrazines to give 1,3,5-triaryl pyrazoline using PW_{12}/PA. The results are listed in Table 2. The reactions proceeded well for all the substrates but those

Table 1 Effect of different conditions in the reaction of chalcone with phenylhydrazine



4a 5 6a

Entry	Solvent	PW_{12} loading (%)	Yield[a] (%)
1	MeOH	25	56
2	CH_3CN	25	60
3	CH_2Cl_2	25	38
4	EtOH	25	87
5	EtOH	30	90
6	EtOH	35	95
7	EtOH	40	95
8	EtOH	35	70[b]

Reaction conditions: chalcone 1 mmol, phenylhydrazine 1 mmol, and PW_{12}/PA (3 mol%) in solvent (5 ml) after 5 h at 45 °C.
[a]Isolated yield. [b]r.t.

Table 2 Synthesis of 1,3,5-triaryl-2-pyrazoline derivatives in the presence of PW_{12}/PA

Product	Ar_1	Ar_2	Time (h)	Yield[a] (%)	TOF	Melting point (°C)	
						Found	Reported
6a	C_6H_5	C_6H_5	3	> 98	1.09	132–134	134–135 [33]
6b	C_6H_5	4-NO_2-C_6H_4	4.25	90	0.72	137–139	—
6c	C_6H_5	4-Me-C_6H_4	2.75	> 98	1.33	129–131	128–130 [31]
6d	C_6H_5	4-Cl-C_6H_4	5.25	90	0.58	132–134	133–134 [32]
6e	C_6H_5	3-Cl-C_6H_4	3.5	90	0.91	130–132	134–136 [31]
6f	C_6H_5	2-Cl-C_6H_4	4.5	87	0.67	132–134	134–135 [32]
6g	C_6H_5	2,4-Cl_2-C_6H_4	5	87	0.58	128–130	—
6h	C_6H_5	3-Br-C_6H_4	4	92	0.77	139–141	141–143 [31]
6i	C_6H_5	4-MeO-C_6H_4	2	> 98	1.64	110–112	110–112 [31]
6j	4-Cl-C_6H_4	C_6H_5	4	92	0.77	141–143	143–145 [31]
6k	4-Br-C_6H_4	C_6H_5	5.5	90	0.55	143–145	—

Reaction conditions: chalcone 1 mmol, phenylhydrazine 1 mmol, and PW_{12}/PA (3 mol%) in EtOH (5 ml) after 3 h at 45 °C.

[a]Isolated yield.

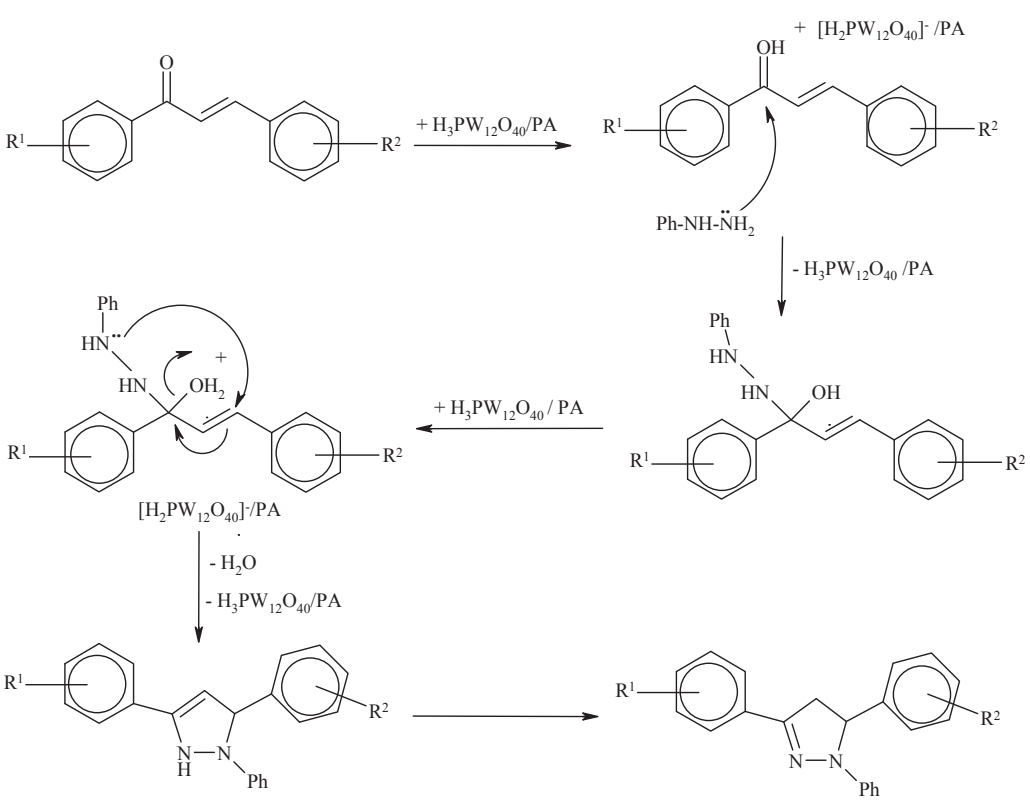
with electron-donating groups were generally more reactive than those with electron-withdrawing groups.

A reasonable pathway for the reaction between chalcone and phenylhydrazine in the presence of PW_{12}/PA is given in Scheme 3.

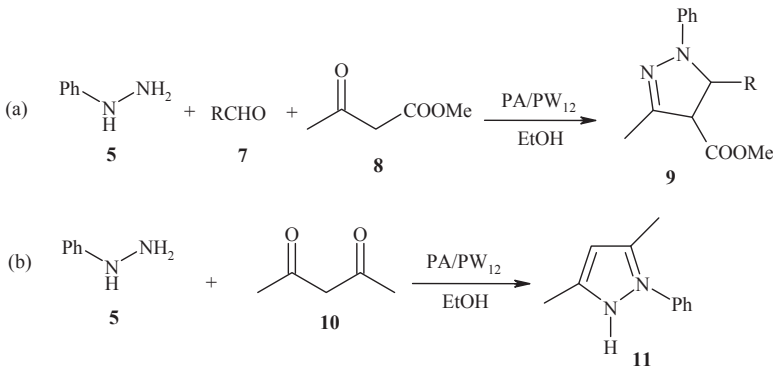
We then attempted to prepare other types of 2-pyrazolines using the above-mentioned optimum conditions. When aldehyde **7**, methyl acetoacetate **8**, and phenylhydrazine **5** (Scheme 4(a)) or acetyl acetonate **10** (1,3-diketone) and phenylhydrazine **5** (Scheme 4(b)) were used, the corresponding 2-pyrazolines (**9**, **11**) were obtained in 87% or 82% yields, respectively (Scheme 4).

2.4 Catalyst recovery

The recovery and reusability of the PW_{12}/PA catalyst was also investigated. We noticed that after the addition of CH_2Cl_2 to the reaction mixture, these catalysts are easily recovered quantitatively by simple filtration. The wet catalysts were recycled (the nature of the recovered catalysts has previously been determined by NAA, XRD, and FT-IR spectra) and no appreciable change in activity was noticed after two cycles. The obtained products were of the same purity as that in the first and second runs, but the yields gradually decreased in the runs carried out using the recycled catalyst. For example, the



Scheme 3. Suggested mechanism for the reaction of chalcone with phenylhydrazine in the presence of PW_{12}/PA.



Scheme 4. Preparation of other types of 2-pyrazolines in the presence of PW_{12}/PA.

Table 3 Recycling of 35% PW_{12}/PA for the synthesis of the 1,3,5-triaryl-2-pyrazoline derivatives

Run	Yield[a] (%)
1	98
2	95
3	67
4	43

Reaction conditions: chalcone (**4a**) 1 mmol, phenylhydrazine 1 mmol, and PW_{12}/PA (3 mol%) in EtOH (5 ml) after 3 h at 45 °C. The recovered catalyst was washed with ethanol, dried at 100 °C for 6 h, and calcined at 400 °C for 3 h. [a]Isolated yield.

reaction between chalcone and phenylhydrazine with PW_{12}/PA afforded the corresponding 1,3,5-triaryl-2-pyrazoline in 98%, 95%, 67%, and 43% yields over four cycles in EtOH (Table 3). HPA dissolves in polar media and, therefore, the leaching of HPA from the HPA/support system was significant.

2.5 Comparison between the catalytic activity of bulk PW_{12} and supported PW_{12}/PA

To show the effect of the polymer-support on the catalytic activity of PW_{12} in the synthesis of the 1,3,5-triaryl-2-pyrazoline derivatives, we repeated all the reactions under the same reaction conditions using the same amounts of catalyst and substrates in EtOH. The obtained results show that the turnover frequencies of the synthesis of the 1,3,5-triaryl-2-pyrazoline derivatives catalyzed by PW_{12}/PA were higher than that of PW_{12}. One explanation for this behavior is that bulk HPAs have low specific structure areas (1–10 m^2/g). It is important to increase the surface area or, even better, increase the number of accessible acid sites on the HPAs. This can be achieved by dispersing the HPAs on a solid support with a high surface area [35].

3 Conclusions

We developed an alternative and simple procedure for the synthesis of 1,3,5-triaryl pyrazoline using solid HPA supported on a thermally stable and highly organosoluble polymer (PW_{12}/PA). This is an eco-friendly, inexpensive, and efficient catalyst. The advantages of this catalytic system are mild reaction conditions, short reaction times, high product yields, easy catalyst preparation, non-toxicity of the catalysts, and simple and clean work-up of the desired products. In addition, the catalysts can be reused several times, although activity will be reduced.

Acknowledgements

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