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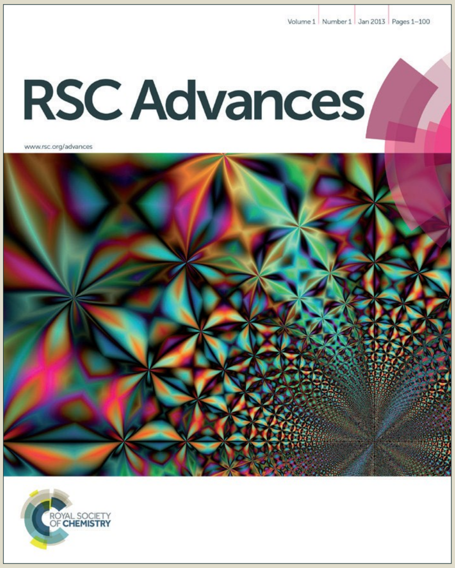

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Mild, efficient, and environmentally friendly synthesis of symmetrical N, N′-alkylidenebisamides under solvent-free conditions catalyzed by $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$

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Abstract

A series of symmetrical N, N'-alkylidenebisamides were successfully synthesized via a one-pot three-component green and environmentally friendly condensation of aliphatic and/or aromatic aldehydes with benzamide by the mediation of 2.5 mol% $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ as *Lewis/Bronsted* acid catalyst with good to excellent yields in moderately short reaction times. This developed methodology is effective for diverse substituted aldehydes; thus, it constitutes a general synthetic route for the preparation of symmetrical alkylidenebisamides. Findings demonstrated that aromatic aldehydes containing electron-withdrawing groups are more effective than the electron-donating counterparts.

Keywords: N, N'-alkylidenebisamides, $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$, catalytic, heteropolyacid, multi-component.

Introduction

The development of simple, effectual, and clean methodologies for the economic preparation of extensively used pharmacophores by using readily available reagents under catalytic conditions are the major challenges for chemists which attempt to improve the quality of the environment.[1,2] Compounds bearing amide and bisamide groups are important intermediates in organic synthesis and can be easily transformed into other pharmacological and peptidomimetic materials.[3] Bisamides are key fragments for the introduction of *gem*-diaminoalkyl residues in retro-inverse pseudo-peptide derivatives.[4,5] Prevalence of amide

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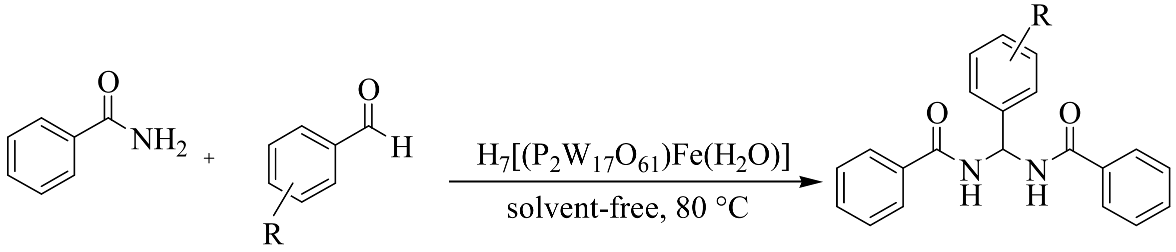
moieties in natural products and pharmacologically active compounds has established a diverse array of synthetic approaches to these compounds; therefore, preparation of amides has been attracted considerable attention.[6,7]

Symmetrical alkylidenebisamides are usually prepared by the direct reaction of an aldehyde with the corresponding amide. So far, various *Bronsted* acids, such as phosphotungstic acid,[8] sulfamic acid,[9] triflic acid,[10] *p*-toluene sulfonic acid,[11] SiO_2-$MgCl_2$,[12] $B(HSO_4)_3$,[13] and silica supported polyphosphoric acid (SiO_2-PPA)[14] have been employed to catalyze the reaction. Although, strong *Bronsted* acids showed high catalytic activity, but these expensive materials are usually corrosive, toxic, and rare; hence, they are not beneficial to industrial applications. Moreover, most of the introduced methods were not gratifying and need certain restrictions with regards to scope and reaction conditions.

Due to the above comments, investigation of suitable *Bronsted/Lewis* acids as efficient catalysts under mild conditions is required for the target transformation. During the course of our investigations directed towards the development of environmentally friendly procedures for several important organic transformations,[15-19] we introduce capability of the *Wells-Dawson* $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ as an efficient acid catalyst for the synthesis of symmetrical N, N′-alkylidenebisamides through condensation of aryl aldehydes with benzamide under solvent free conditions at 80 °C (Scheme 1). This catalyst could be easily prepared and reused for several times without considerable loss of activity.



Scheme 1. General formulation for the preparation of symmetrical N, N'-alkylidenebisamides.

2. Experimental

2.1. Materials and methods

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All starting materials and solvents were obtained from commercial sources and used as received. $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$,[20] $K_6[P_2W_{18}O_{62}]$,[21] $H_{14}[NaP_5W_{30}O_{40}]$,[22] and $H_3[PMo_{12}O_{40}]$,[23] were prepared according to the reported methods. Fourier transform infrared (FT-IR) spectra were recorded on a 8700 Shimadzu Fourier Transform spectrophotometer in the region of 400 to 4000 cm^{-1} using KBr pellets. Progress of the reactions was monitored by thin layer chromatography (TLC). Melting points were recorded on a Bamstead electrothermal type 9200 melting point apparatus. ^1H NMR spectra were recorded on Bruker AVANCE 100 and 400 MHz instruments using TMS as internal reference. All products were identified by comparison of their spectral and physical data with those previously reported.[24-26]

2.2. Synthesis of α_2-$K_{10}P_2W_{17}O_{61}.20H_2O$

α_2-$K_{10}P_2W_{17}O_{61}.20H_2O$ was prepared according to the literature procedure.[27] K_6 [α- or β-$P_2W_{18}0_{62}$].XH $_2$O (80 g, 1.15 mol) was dissolved in 200 ml water. Then, 20 g (200 mmol) of potassium hydrogen carbonate in 200 ml water was added while stirring. The reaction was completed after 1 h and the white precipitate was filtered on a coarse sintered glass frit. The solid product was dried under suction; then, re-dissolved in 500 ml hot water (95 °C). The snow-like crystals appeared on cooling (after 3 h) were filtered and dried under suction for 5 h. The final solid product (57g, 70%) was air-dried for 2 to 3 days.

2.3. Preparation of K_7-α_2-$Fe(OH_2)(P_2W_{17}O_{61}).19H_2O$

A sample containing 2.43 g (6 mmol) of $Fe(NO_3)_3.9H_2O$ was dissolved in 65 ml water, and 23 g (4.7 mmol) of α_2-$K_{10}P_2W_{17}O_{61}.20H_2O$ was added with complete stirring. The whole dissolution achieved at 30 °C, and the solution was left at 6 °C overnight. The appeared yellow crystals were filtered and dried in air. Anal. Calcd for $K_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)].19H_2O$: K, 5.6; P, 1.28; Fe,1.15; W, 64.4; H_2O, 7.42. Found: K, 5.46; P, 1.29; Fe, 1.16; W, 65.2;H_2O, 7.41.

2.4. Preparation of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ *acid catalyst*

Wells-Dawson $H_7[(p_2W_{17}O_{61})Fe(H_2O)]$ acid catalyst (HPA) was prepared according to the previously reported method.[28] The iron–substituted hetropolyanion, *K_7-α_2-*

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$Fe(OH_2)(P_2W_{17}O_{61}).19H_2O$ was converted into the corresponding acid by passing it through a column of Dowex-50w-X8 ion exchange resin. A sample of K_7-α_2-$Fe(OH_2)(P_2W_{17}O_{61}).19H_2O$ (2 g) was dissolved in 50 ml water; then, the solution passed through the resin column in the H^+ form. This process was repeated for two times. Slow evaporation of the final solution gave the pure $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$.

2.5. General procedure for the preparation of N, N′-alkylidenebisamides

To a mixture of aldehyde (2 mmol) and amide (4 mmol) was added 0.01 mmol of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$. Then, the mixture was stirred at 80 °C in an oil bath and the reaction was followed by TLC analysis (n-hexane/ethyl acetate, 3:2). After completion of the reaction, the mass was cooled to 25 °C, and the mixture was dissolved in boiling ethanol. The liquor involving catalyst and starting materials was concentrated and the solid product was purified by re-crystallization in ethanol 40% as the appropriate solvent. All products were characterized by IR and ^1H NMR as obtained in the supporting information.

3. Results and Discussion

To investigate easy, one-pot, and environmentally benign synthetic routes to prepare different organic aggregates, the condensation reaction of aromatic aldehydes and benzamide was studied in the presence of a catalytic amount of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ under solvent free conditions.

In order to investigate the appropriate amount of catalyst, a model reaction was carried out by applying 0-10 mol% of HPA for the preparation of N, N′-(phenylmethylene)dibenzamide (Table 1). First, the reaction was non-productive to give the correlative product in the absence of heteropolyacid and led to only 20% of product after 2 h (entry 1) which indicated that presence of catalyst plays an important role in the reaction progress. It was found that 1 mol% of catalyst attained 50% yield after 20 min (entry 2). This observation established that the HPA catalyst exhibited high catalytic activity in the desired transformation. Then, the reaction of benzaldehyde with benzamide was conducted in the presence of a catalytic amount of HPA (2.5 mol%). After completion of the reaction followed by standard work-up and purification, the corresponding N, N′-alkylidenebisamide was obtained in 85% yield (Table 1, entry 3).

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As is envisaged, yield% was increased with enhancing catalyst concentration and the maximum yield of 85% was achieved with 2.5 mol% of catalyst after 20 min. However, higher amounts of catalyst lowered yield% to 75 at the same time (entry 5). Therefore, 2.5 mol% of catalyst was chosen as the optimal quantity to push the reaction forward.

Table 1. Effect of catalyst amount on the condensation of benzaldehyde with benzamide.

Entry	$H_7[(p_2W_{17}O_{61})Fe(H_2O)]$ (mol%)	Time (min)	Yield (%)
1	0	120	20
2	1	20	50
3	2.5	20	85
4	5	20	77
5	10	20	75

The desired amount of $H_7[(p_2W_{17}O_{61})Fe(H_2O)]$ was added to a mixture of benzaldehyde (2 mmol) and benzamide (4 mmol). Then, the mixture was stirred at 80 °C in an oil bath and the reaction was followed by TLC. After completion of the reaction, the crude product was purified as described in the experimental section.

Organic solvents are on the top of the list of damaging chemicals, because they are employed in enormous amounts and are usually volatile liquids that are difficult to be stored. Search for alternative reaction media to replace volatile, flammable, and often toxic solvents commonly used in organic synthesis is an important objective in the development of green chemical processes. Therefore, efficacy of the solvent-free condition was compared with the solvent condition by carrying out condensation of benzaldehyde with benzamide in different solvents (Table 2). Fortunately, the best yield was achieved under solvent-free case with less reaction time and found as optimum condition. However, a considerable amount of the desired product was formed in methanol, acetonitrile, and chloroform with 35, 30, and 20% yields, respectively.

Table 2. Effect of different solvents on the condensation of benzaldehyde with benzamide.

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Entry	solvents	Time (h)	Yield (%)
1	acetonitrile	17	30
2	n-hexane	17	10
3	methanol	17	35
4	chloroform	17	20
5	solvent free	20 min.	85

Reaction condition is described below Table 1.

To improve the yield% and optimize the reaction conditions, effect of temperature was studied on the condensation reaction of benzaldehyde with benzamide (Fig. 1). At 80 °C, the reaction proceeded smoothly and almost complete conversion of the product was observed after 20 min. Further increase in temperature didn't improve yield%; therefore, the reaction temperature 80 ºC was kept for all runs. Reactions at different conditions and at various molar ratios of substrates in the presence of the heterogeneous $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ revealed that the best condition was solvent-free at 80 ºC in the presence of 2.5 mol% catalyst.

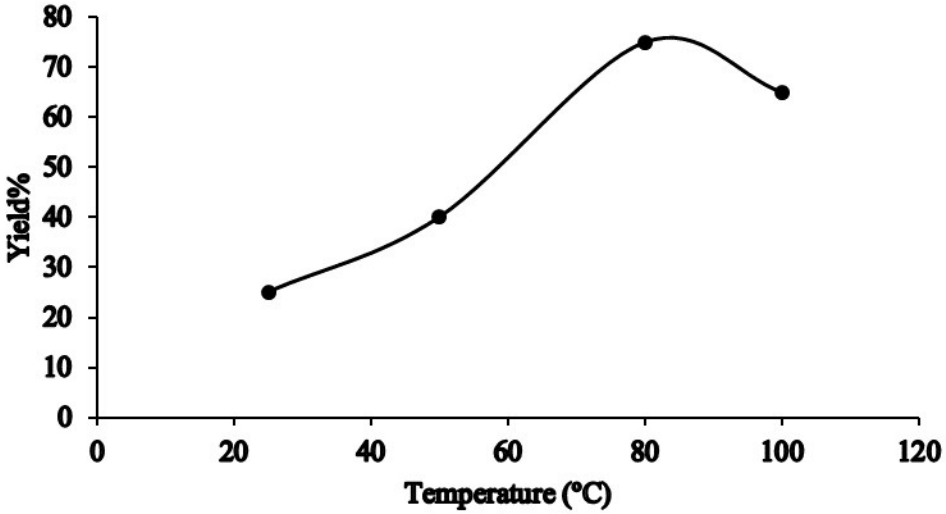

Fig. 1. Effect of reaction temperature on the condensation of benzaldehyde with benzamide.

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Effect of reaction time was also investigated to explore minimum time required to obtain the maximum yield in the preparation of N, N′-alkylidenebisamide. As illustrated in Fig. 2, 20 min is sufficient to get 85% yield. No important increase in yield% was detected after a prolonged reaction time.

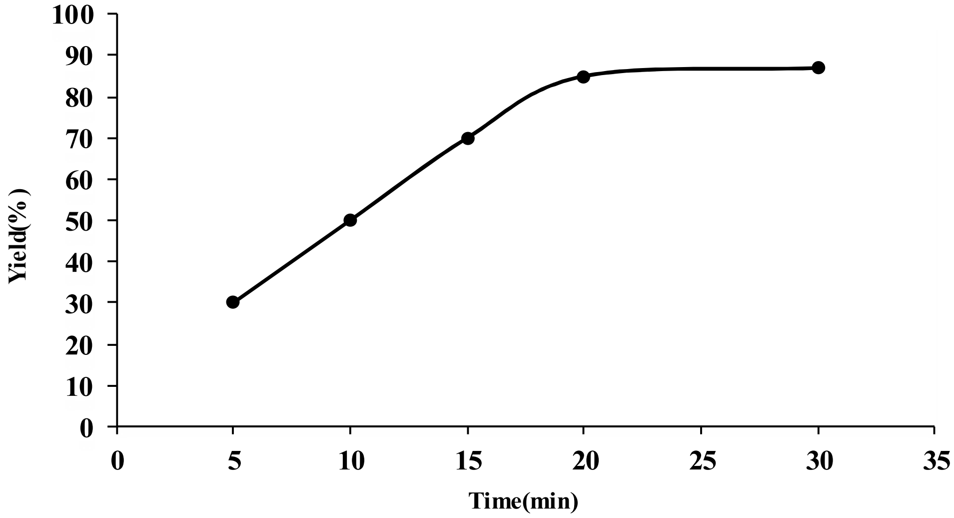

Fig.2. Effect of reaction time on the condensation of benzaldehyde with benzamide.

Effect of the kind of the heteropolyacid was investigated on the efficacy of the catalytic system. Therefore, catalytic activity of$H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ was compared with other indicative familiar heteropolyacids in the preparation of N, N′-(phenylmethylene) dibenzamide (Table 3). Almost, all the introduced *Keggin* and *Wells-Dawson* heteropolyacids behaved as good catalysts in the respective transformation. Although, *Keggin* and *Wells-Dawson* heteropolyacids have different structures, however, both types behaved similar in the preparation of N, N′-alkylidenebisamide derivatives. Among different HPA's, the *Preyssler* type showed less catalytic activity and resulted in 40% yield after 20 min. Many factors such as acidity of the heteropolyacid, negative charge density smeared over oxygen atoms, structural composition and distortions, and approach of the substrate molecule into the bulk of the heteropolyacid would contribute to the catalytic efficiency of the heteropolyacids under the reaction conditions.[29]

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Table 3. Effect of the heteropolyacid nature and structure on the efficacy of the preparation of *N, N′*-alkylidenebisamide.

Catalyst	**Structure type**	**Time (min)**	**Yield (%)**
$H_3PMo_{12}O_{40}$	*Keggin*	20	68
$H_3PW_{12}O_{40}$	*Keggin*	20	77
$H_6P_2W_{18}O_{62}$	*Wells-Dawson*	20	81
$H_4SiW_{12}O_{40}$	*Si-substituted Keggin*	20	65
$H_7[(p_2W_{17}O_{61})Fe(H_2O)]$	*Wells-Dawson*	20	85
$H_5PW_{10}V_2O_{40}$	*Keggin*	20	79
$H_{14}NaP_5W_{30}O_{110}$	*Preyssler*	20	40

2.5 mol% of catalyst was used in all cases. Yield% refers to the isolated yield.

With the obtained optimal conditions and to show the generality of the reaction, the reaction of a series of aromatic aldehydes bearing different substituents was studied with benzamide to afford the corresponding symmetrical N, N′-alkylidenebisamides (Table 4). It seems that substitution at the *para*-position of the aromatic ring produced no significant effects; aldehydes possessing electron-donating groups caused slightly lower reaction yields. Whereas, aromatic aldehydes substituted with electron-withdrawing groups reacted well to give the corresponding products in good to excellent yields under the reaction conditions. However, the reaction of benzaldehyde with acetamide did not occur and only starting material was recovered (Scheme 2). Moreover, in order to synthesize the unsymmetrical *N, N′*-alkylidenebisamides, the reaction of benzaldehyde with one equivalent of benzamide and one equivalent of acetamide was carried out. The reaction provided only symmetrical N, N′-alkylidenebisamide derived from benzamide.

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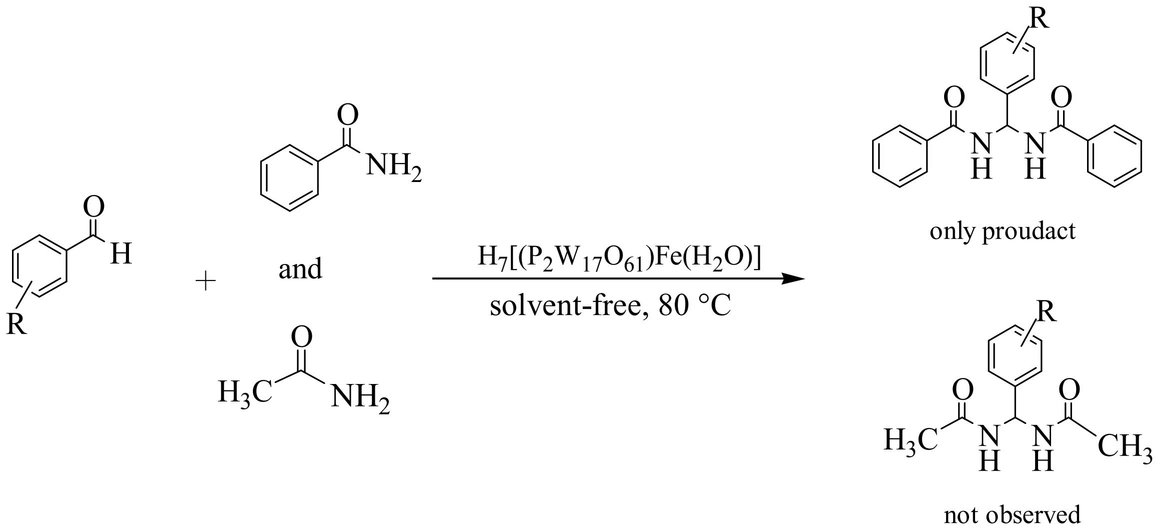
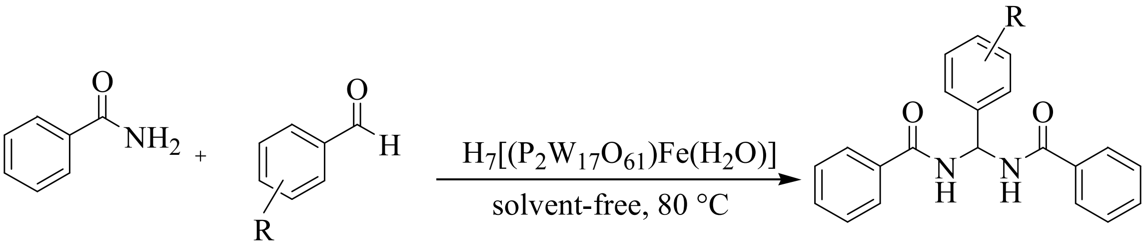
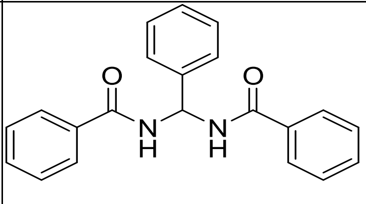


Scheme 2. reaction of benzaldehyde with one equivalent of benzamide and one equivalent of acetamide.

Table 4. Synthesis of different *N, N′*-alkylidenebisamides in the presence of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ under solvent-free conditions.



Aldehyde	m.p.(°C)	lit. m.p. (°C)	Time (min)	Yield (%)	Product
Benzaldehyde	238-239	237-238	20	85	

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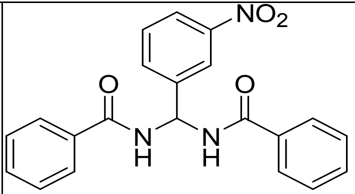
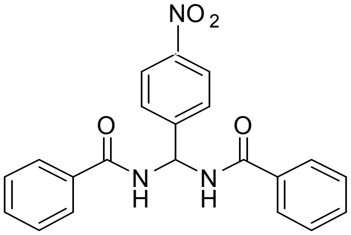
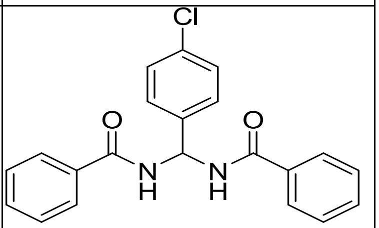
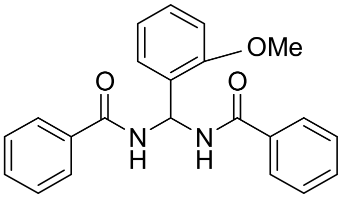
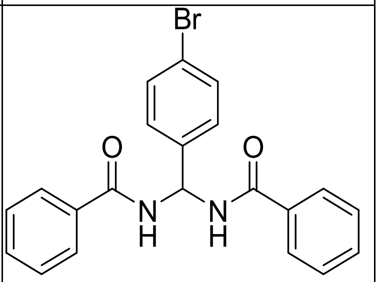
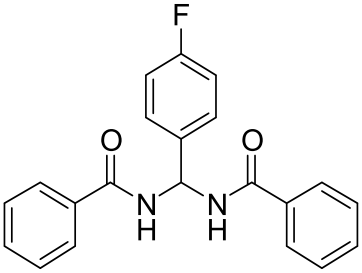
3-NO_2-benzaldehyde	234-236	236-237	20	97	
4-NO_2-benzaldehyde	240-242	241-243	15	98	
4-Cl-benzaldehyde	231-233	232-235	20	93	
2-MeO-benzaldehyde	230-232	229-230	20	77	
4-Br-benzaldehyde	261-263	252-254	20	98	
4-F-benzaldehyde	225-227	226-228	40	76	

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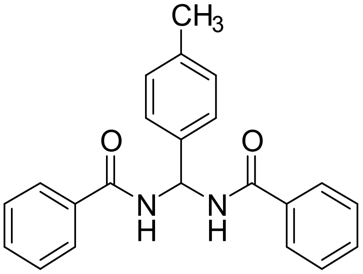
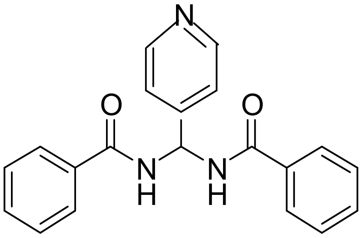
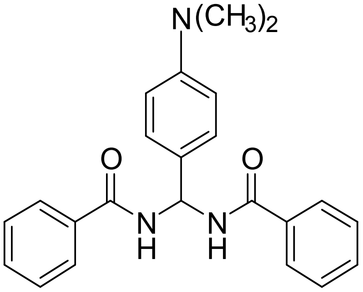
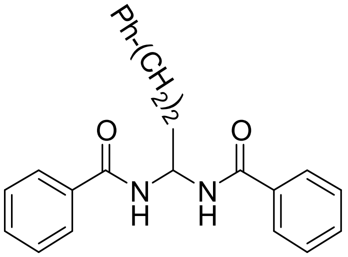
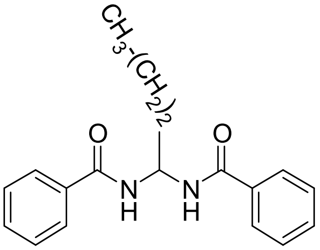
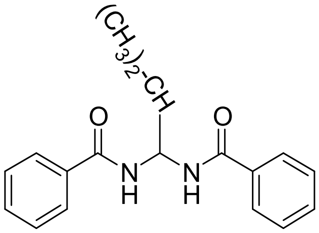
4-Me-benzaldehyde	224-225	224-226	30	82	
4-pyridylaldehyde	208-213	208-212	75	73	
4-N(CH$_3$)$_2$-benzaldehyde	208-210	210-212	75	64	
Ph-(CH$_2$)$_2$-COH	249-251	248-249	75	67	
CH$_3$-(CH$_2$)$_2$-COH	171-173	172-173	60	77	

$(CH_3)_2$-CH-COH	126-128	126-127	75	68	

Yields refer to the isolated pure products. The desired pure products were characterized by comparison of their physical data with those of known compounds. Melting points refer to the desired products.

Comparison of the efficacy of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$catalyst with some reported catalysts is presented in Table 5. The model reaction of benzamide and benzaldehyde was considered as a representative example and the comparison was in terms of mol% or amount of the catalyst, temperature, reaction time, and percentage yields. Evidently, $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ was more effective than most of the conventional catalysts listed in Table 5. The present methodology employed a very low amount of the catalyst under solvent-free conditions. Although, some of the introduced additives catalyzed the reaction, even though at a lower temperature, they required toxic and expensive solvents, higher mol% of catalyst, and longer reaction times. The present catalytic system conveys distinct advantages in competition with the previously reported methods, in terms of yield%, mild reaction conditions, simple procedure and work-up, and lack of toxicity.

Table 5. Comparison of the catalytic efficiency of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ with some reported catalysts.

Entry	Catalyst and Conditions	Time (min)	Yield (%)	Ref.
1	$ZnCl_2$, Solvent, 110 ºC, 20 mol%	180	91	30
2	SiO_2-$BaCl_2$, Solvent free, 100 ºC, 0.025 g	120	74	31
3	SiO_2-$MgCl_2$, Solvent-free , 100 °C, 0.025 g	180	73	12
4	Phosphotungstic acid, Toluene/reflux, 0.3 mmol	1200	70	8
5	$B(OH)_3$, MW, 0.3 mmol	40	80	24
6	$BF_3.SiO_2$, $CHCl_3$/reflux , 0.03 g	270	81	25

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7	SiO_2-PPA,Solvent free, 100 ºC, 0.025 g	70	60	32
8	Boric acid,toluene,120 ºC, 0.3 mmol	960	92	24
9	$H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$, 80 ºC, 2.5 mol%	20	85	-

Reusability and reproducibility of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ was approved in the synthesis of N, N′-(phenylmethylene)dibenzamide, as shown in Fig. 3. The catalyst was recovered from the reaction mixture via eluting with ethyl acetate after each run, washed thoroughly with ethanol, dried in an oven at 90 ºC for 4 h and tested in the subsequent run. The catalyst was tested for six runs. It was observed that the catalyst displayed very good reusability. Moreover, to ensure reproducibility of the transformation, repeated typical experiments were carried out under identical reaction conditions. The obtained yields were found to be reproducible within ±3% variation.

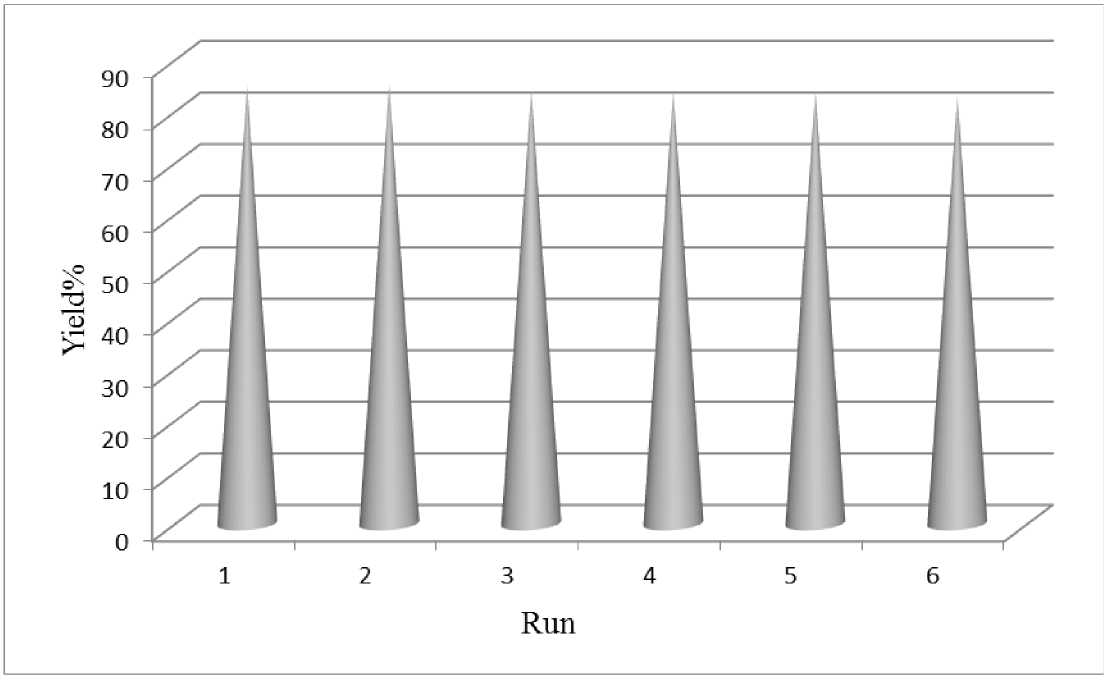

Fig. 3. Yield% as a function of reusability of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$.

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To confirm that the catalytic activity was generated from the heterpolyacid catalyst, and not from the decomposed components of the heteropolyacid in the reaction mixture, a hot filtration test was planned. In this technique, to a mixture of aldehyde (2 mmol) and amide (4 mmol) was added 0.01 mmol of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ and the reaction was continued as described in the experimental section. At this stage, the yield of the product was 75%. Then, the catalyst was filtered off under hot conditions and with the filtrate obtained after separation of the catalyst, the reaction was continued for another 15 min at the same reaction temperature. However, no corresponding increase in yield beyond 75% was attained. This result clearly confirmed the stability of the heteropolyacid in this condensation reaction and no degradation of the heteropolyacid occurred during the course of the reaction. This result was also confirmed by reusability and reproducibility studies of the catalyst as mentioned above. Moreover, ^{31}P NMR spectrum of the re-used catalyst was very similar to the fresh one and no change was occurred in the structure of the heteropolyacid after six re-using.

Finally, a plausible reaction pathway for the preparation of *N, N′*-alkylidenebisamides in the presence of $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ is proposed in Scheme 3. It is conceivable that H^+ from the heteropolyacid first bound to the carbonyl oxygen of the aromatic aldehyde and increased its Lewis acidity. Then, benzamide attaches to the carbonyl group of aldehyde and led to the generation of (I) and followed by loss of H_2O affords (II). As reported in the literature, the *Knoevenagel* type coupling of benzamide with an active carbonyl compound such as benzaldehyde gives N, N′-alkylidenebisamide.[33]

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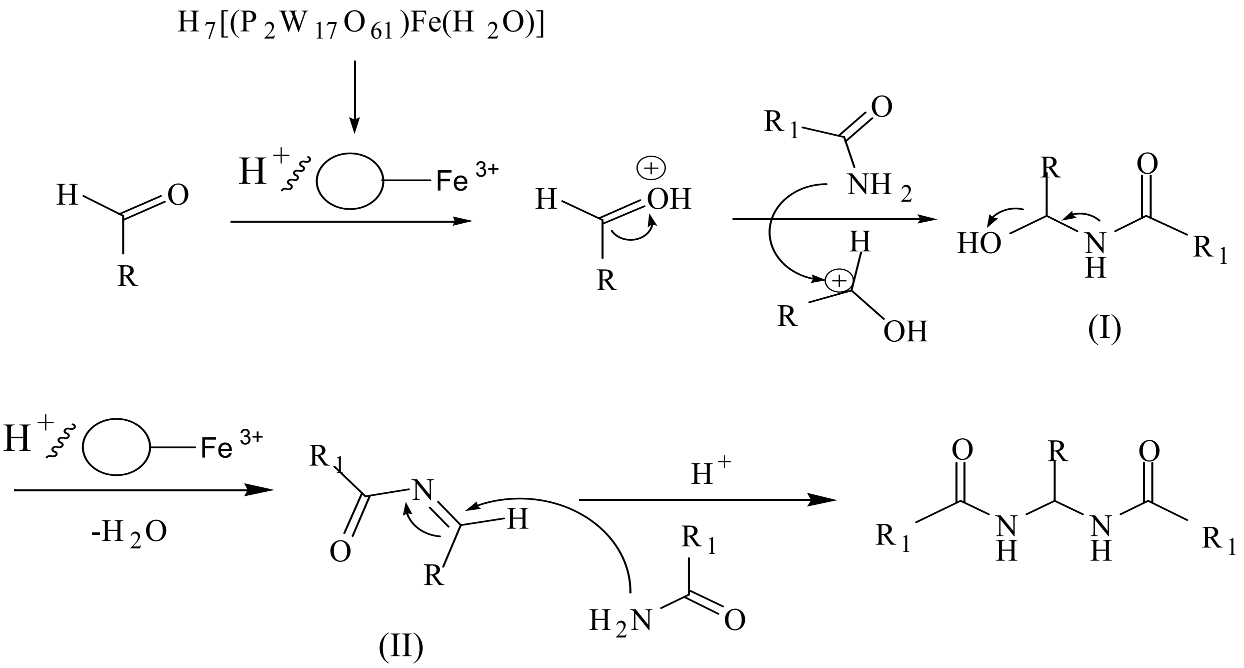

Scheme 3. Proposed mechanism for the synthesis of *N,N′*-alkylidenebisamide.

Conclusion

A green and easily available $H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ was used as a strong, effective, and selective *Lewis/Bronsted* acid catalyst for the preparation of symmetrical N, N′-alkylidenebisamides under solvent-free conditions. The present protocol has some advantages over the previously reported methods, such as good to excellent product yields, short reaction times, low toxicity, low cost, and simple scale-up. The work-up procedure is very clear-cut; that is, the products were isolated and purified by simple filtration and re-crystallization from aqueous ethanol.

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Graphical Abstract

$H_7[(P_2W_{17}O_{61})Fe^{III}(H_2O)]$ is introduced as an effective catalyst for the one-pot synthesis of N, N′-alkylidenebisamides. The catalyst could be easily separated without noticeable reduction in activity.

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