

A mild, three-component one-pot synthesis of 2,4,5-trisubstituted imidazoles using Mo(IV) salen complex in homogeneous catalytic system and Mo(IV) salen complex nanoparticles onto silica as a highly active, efficient, and reusable heterogeneous nanocatalyst

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Abstract Mo(IV) salen complex (2.5 mol%) was found to be a highly efficient catalyst for the one-pot synthesis of 2,4,5-triarylimidazoles via a three-component reaction using benzil or benzoin, aryl aldehydes, and ammonium acetate as a nitrogen source under mild conditions. In order to recover and the reuse of the catalyst, a new Mo(IV) salen–silica nanoparticle as heterogeneous catalyst was prepared by simple and successful immobilization of the catalyst onto silica (3-aminopropyl functionalized silica gel). This procedure can be applied to large-scale conditions with high efficiency. Experimental evidence showed that the catalyst is stable and can be easily recovered and reused for at least five times without significant loss of activity. The nanocatalyst was characterized using FT-IR spectroscopy, scanning electron microscopy, atomic force microscopy, powder X-ray diffraction, transmission electron microscopy, thermogravimetric instrument for analysis of nitrogen adsorption, and inductively coupled plasma spectrometer.

Keywords Mo(IV) salen complex · Heterogeneous catalyst · Nanoparticle · 2, 4, 5-Trisubstituted imidazoles · MCRs · Multicomponent reaction

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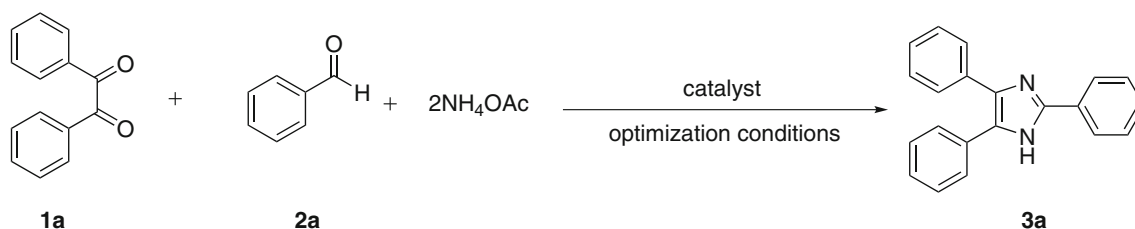
Introduction

Multicomponent reactions (MCRs) have attracted great interest in modern organic synthesis and the pharmaceutical industry because they are one of the best synthesis tools available for the preparation of molecular libraries in a cost and time effective way [1].

Compounds bearing imidazole ring systems are well known for their diverse biological activities and are the core fragment of different natural products [1–3]. Not only they have many beneficial pharmaceutical properties [4, 5], but are also used in photography as photosensitizer compounds [3].

A number of synthetic procedures have been developed for the synthesis of 2,4,5-trisubstituted imidazoles via multicomponent reactions. Generally, these imidazoles have been made via the multicomponent reaction of a 1,2-diketone, α -hydroxy ketone, or α -ketomoxime [6] with ammonium acetate and an aldehyde in the presence of L-proline [1], scolecite [3], polymer-supported zinc chloride [5], $\text{InCl}_3 \cdot 3\text{H}_2\text{O}$ [7], ZrCl_4 [8], heteropoly acid supported on silica [9], $\text{NiCl}_2 \cdot 6\text{H}_2\text{O}/\text{Al}_2\text{O}_3$ [10], NaHSO_3 [11], sulfated tin oxide [12], iodine [13, 14], ceric ammonium nitrate (CAN) [15], $\text{UO}_2(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ [16], ionic liquids [17], and DABCO [18]. The major, most common drawbacks of these approaches include the use of elevated temperatures, long reaction times, harsh reaction conditions, and low yields. Therefore, continuing efforts are invested in the search for better catalysts (operational simplicity, economic viability, and reusability) for the synthesis of 2,4,5-trisubstituted imidazoles.

Heterogeneous catalysts are of great importance in green synthesis due to their easy recovery and reusability [19]. In this regard, in recent years significant efforts in catalysis research have been devoted to the identification and application of effective and safe heterogeneous catalysts [5].



Scheme 1 Model reaction of synthesis of 2,4,5-triarylimidazoles derivatives

Table 1 Catalyst effects on the model reaction

Entry	Catalyst	Time (h)	Temperature (°C)	Yield (%) ^a
1	–	3	50	–
2	Salen (2.5 mol%)	3	50	–
3	Salen-Cr(Cl) (2.5 mol%)	3	50	> 10
4	Salen-Fe(Cl) (2.5 mol%)	3	50	> 10
5	Salen-V(O) (2.5 mol%)	3	50	55
6	Salen-Cu (2.5 mol%)	3	50	15
7	Salen-Mn(Cl) (2.5 mol%)	3	50	25
8	Salen-Co(OAc) (2.5 mol%)	3	50	> 10
9	Salen-Mo(O) (2.5 mol%)	1	50	94
10	Salen-Mo(O) (5 mol%)	1	50	94
11	Salen-Mo(O) (10 mol%)	1	50	94
12	Salen-Mo(O) (1 mol%)	1	50	72
13	Salen-Mo(O) (2.5 mol%)	10	25	45

^a Isolated yield

In continuation of our previous study in the synthesis and applications of heterogeneous catalysts for the development of heterocyclic compounds [20–25], in this article we report a simple, mild, and efficient one-pot multicomponent method for the synthesis of 2,4,5-triarylimidazoles using Mo-salen complex as a homogeneous catalyst and Mo-salen complex nanoparticles onto silica as a heterogeneous nanocatalyst (2.5 mol%) in EtOH at 50 °C. Our methodology offers several advantages including substrate generality, catalyst recyclability, mild reaction conditions, large-scale synthesis, and high yields. This combination of attributes makes our methodology an attractive approach for the synthesis of 2,4,5-triarylimidazoles.

Results and discussion

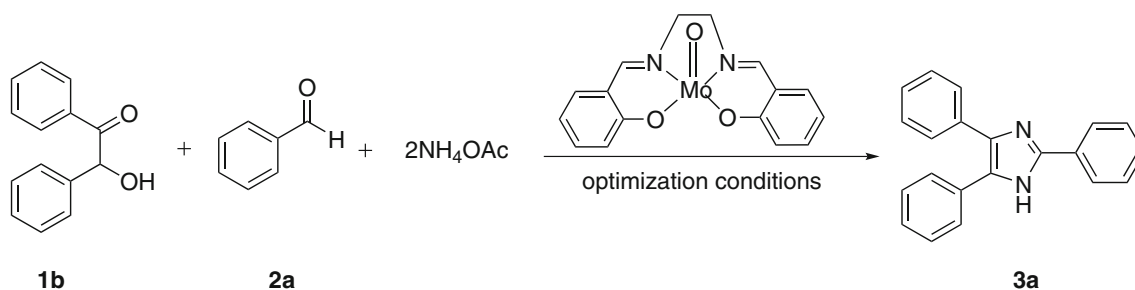
Catalytic role of metal-salen complexes for the synthesis of 2,4,5-trisubstituted imidazoles

As a part of our continuing efforts in the synthesis of organic compounds using efficient and novel catalysts [20–24], we explored the synthesis of 2,4,5-trisubstituted imidazoles using metallosalens as catalyst. We found that these complexes effectively catalyze the condensation reaction of

benzil and ammonium acetate with different aryl aldehydes to form 2,4,5-triarylimidazoles in ethanol under mild conditions (Scheme 1). The condensation of benzil, ammonium acetate, and benzaldehyde was chosen as our model reaction.

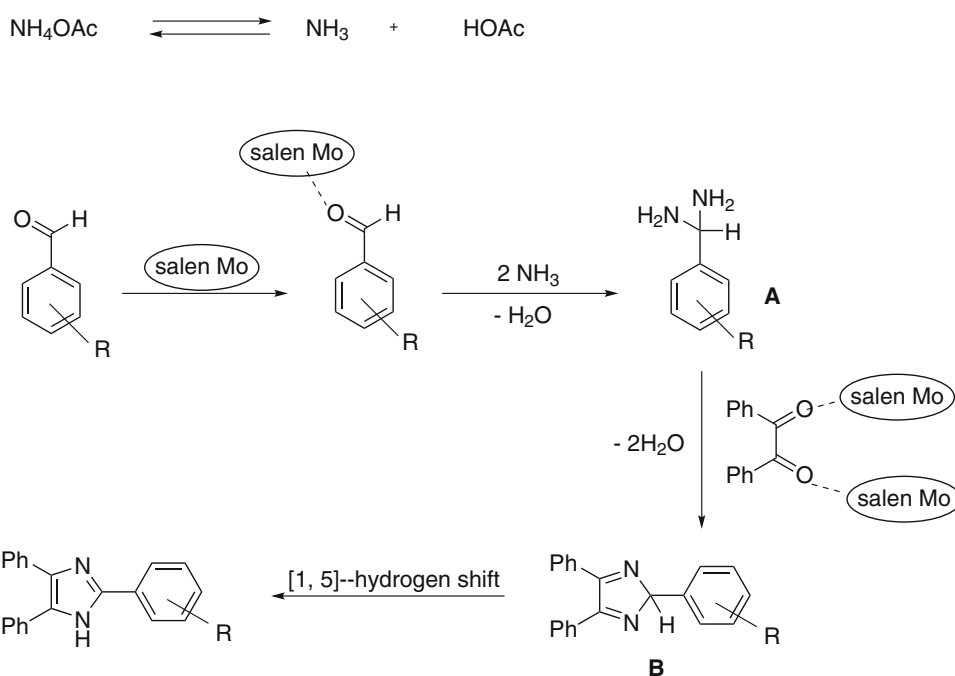
The impact of the different metallic ions in metallosalen on the performance of the reaction was examined. The reaction times and yields using various metal-salen complexes are summarized in Table 1 (entries 3–9). It was determined that metal-salen complexes containing Co, Cu, Mn, Fe, V, and Cr ions gave low to moderate yields, and using larger amounts did not improve the output of the reaction. On the other hand, among the metal-incorporated tested, Mo-salen complex was found to improve greatly both reaction rate and yield.

Then, we studied the impact of using different catalytic amounts of Mo-salen in the synthesis of 2,4,5-triphenyl-1H-imidazole using of benzil (1.0 mmol), ammonium acetate (2.0 mmol), and benzaldehyde (1.0 mmol). The best results were obtained when 2.5 mol% of Mo-salen complex was used since lower catalytic amounts led to lower yields, whereas higher amounts did not improve appreciably both yield and reaction time (Table 1, entries 9–12). To examine the essential role of Mo-salen complex, the reactions were carried out in the absence or presence of ligand (Table 1, entries 1 and 2). In both cases, only traces of the desired product were detected indicating the crucial role the catalyst has in the reaction.



Scheme 2 Synthesis of 2,4,5-triarylimidazoles

Scheme 3 Proposed mechanism for the 2,4,5-trisubstituted imidazole formation using of benzil, ammonium acetate, and aryl aldehydes



We also studied the temperature effect on the reaction (Scheme 1). The best results were obtained at 50 °C when using ethanol for 1 h in the presence of Mo-salen complex (2.5 mol%) as a catalyst (Table 1).

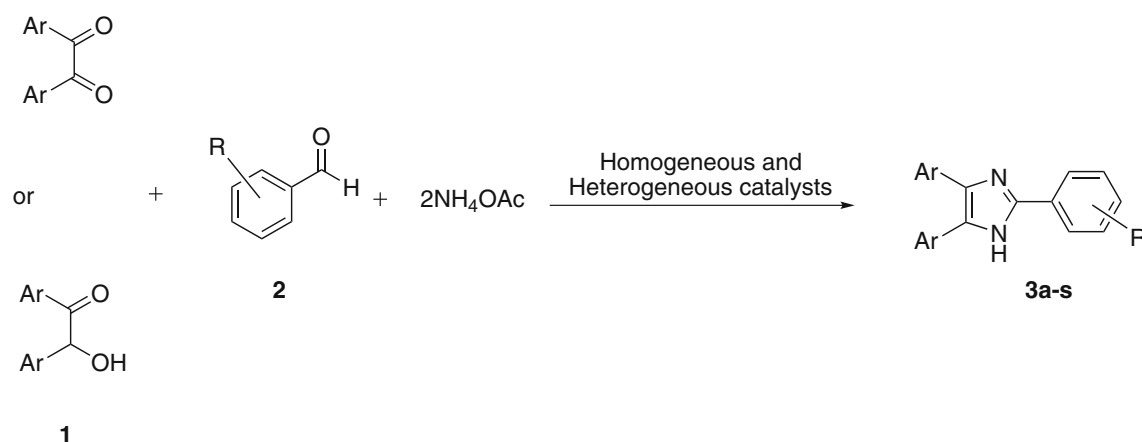
The choice of solvent is also a very important factor for multicomponent reactions. During our optimization studies, the solvent effect was also examined. The model reaction was carried out using a variety of solvents such as EtOH, MeOH, CH₃CN, DMF, CH₂Cl₂, CHCl₃, THF, 1,4-dioxane, and toluene. From these, ethanol was selected because it contributed to faster reaction rates, excellent product yield, and its environmental acceptability.

Using our optimized reaction conditions, we pursued the preparation of 2,4,5-triphenyl-1*H*-imidazole this time using benzoin instead of benzil as a starting material (Scheme 2). We found that while 2,4,5-triphenyl-1*H*-imidazole was obtained in excellent yield (90 %), the reaction time increased (1.5 h).

A plausible mechanism for the Mo-salen complex catalyzed synthesis of 2,4,5-trisubstituted imidazoles has been proposed [13–18] (Scheme 3). Firstly, ammonium acetate

dissociates into ammonia which is required for the initial condensation. Upon formation of diamine intermediate **A** by the reaction of an aryl aldehyde and ammonia in the presence of Mo-salen complex catalyst. The Mo-salen complex facilitates the formation of a diamine intermediate **A** which could be attributed to an increase in the electrophilicity of the carbonyl group. The intermediate **A**, in the presence of Mo-salen complex reacts with the benzil followed by dehydration to afford the imino intermediate **B**, which in turn rearranges to the 2,4,5-trisubstituted imidazole by a [1,5]-hydrogen shift [7].

We also propose a mechanism for the Mo-salen complex catalyzed reaction for the preparation of 2,4,5-trisubstituted imidazoles when the benzoin was used instead of benzil as a starting material, similar to the reported experiment for the synthesis of 2,4,5-triaryl imidazoles by an ionic liquid [17] at 100 °C and I₂ at reflux condition [14]. A solution of benzoin in the presence of Mo-salen complex was stirred in EtOH at 50 °C for several hours in an atmosphere of air. Benzoin was converted to benzil in excellent yield. The formed benzil then reacts with intermediate **A** followed by dehydration to



Scheme 4 Synthesis of 2,4,5-triarylimidazoles in the presence of Mo(IV) salen complex and Mo(IV) salen complex nanoparticles onto silica

yield the imino intermediate **B**, which in turn rearranges to the product by a [1,5]-hydrogen shift (Scheme 3).

The most significant advantages of heterogeneous catalysts are the possibility of catalyst recycling, ease of separation, and reuse of the catalyst. Since Mo-salen complex was proven to be the best catalyst among the other metal-salen complexes tested, we aimed at preparing a new heterogeneous catalyst via a simple impregnation of Mo-salen complex onto 3-aminopropyl functionalized silica gel.

In industry the reusability of a catalyst is important particularly for large-scale operations. In this regard, the recovery and reusability of the catalyst was tested. The advantage of recovery of catalyst for different uses has been provided by utilizing the Mo-salen complex nanoparticles supported onto silica. The recovered Mo-salen complex onto silica was dried and reused several times without observing a decrease in catalytic performance.

To determine if leaching of the catalyst takes place, the heterogeneous catalyst was stirred in EtOH at 50 °C for 120 min. The catalyst was recovered and the filtrate was used for the preparation of 2,4,5-triphenyl-1*H*-imidazole under the optimized conditions for 3 h. Since no product was formed, it was concluded that no leaching took place. Moreover, the ICP results were obtained for the Mo-salen complex after reusing the catalyst for at least five times.

To test this method on a preparative scale, we carried out the synthesis of 2,4,5-triphenyl-1*H*-imidazole under the optimized conditions at a 50 mmol scale and found that the desired imidazole product was obtained in excellent yield after 70 min.

Synthesis of different 2,4,5-trisubstituted imidazoles using homogeneous and heterogeneous catalysts

In the next step, we used the optimized conditions using different aromatic aldehydes in the presence of Mo(IV) salen

complex and Mo(IV) salen complex nanoparticles onto silica (2.5 mol%) to synthesize different 2,4,5-trisubstituted imidazoles (Scheme 4).

We found in all cases the reactions were clean taking place between 40 and 170 min. Heteroaryl aldehydes, such as 2-thiophenecarboxaldehyde, 2-pyridinylcarboxaldehyde, and 4-pyridinylcarboxaldehyde, (Table 2, entries 9–11), were also well tolerated under these mild conditions. We found that our method is compatible with functional groups such as Me, OMe, Cl, NO₂, and OH.

In the presence of Lewis acids, such as InCl₃ · 3H₂O [7] and ZrCl₄ [8], the reaction yields and times are comparable to our method, but the reaction either needs higher temperature or a more toxic solvent (Table 3).

Conclusion

We have developed a simple, mild, and efficient one-pot three-component methodology for the preparation of 2,4,5-trisubstituted imidazoles in the presence of Mo-salen complex and Mo-salen complex nanoparticles onto silica (2.5 mol%) in ethanol at 50°C. The catalyst is stable and can be easily recovered and reused at least five times without significant loss of performance. These attributes make our procedure convenient, mild, economic, green, and efficient for the synthesis of 2,4,5-trisubstituted imidazoles.

Experimental section

Instrumentation, analysis, and starting material

Chemical materials and solvents were purchased from Fluka, Aldrich and Merck. The used aminopropyl silica gel was also purchased from Fluka. NMR spectra were recorded on

Table 2 Synthesis of 2,4,5-trisubstituted imidazoles

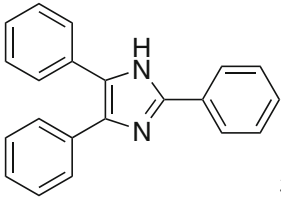
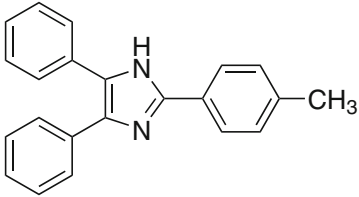
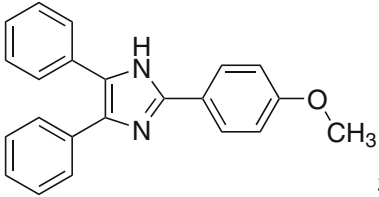
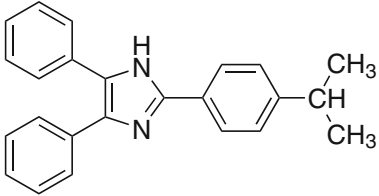
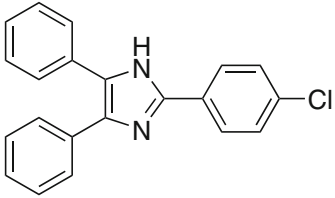
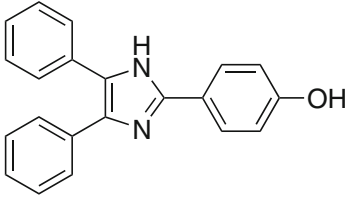
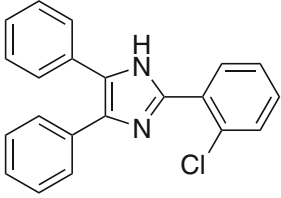
Entry	Product	Homogeneous catalyst				Heterogeneous catalyst			
		Time (min)		Yield (%) ^a		Time (min)		Yield (%) ^a	
		Benzil	Benzoin	Benzil	Benzoin	Benzil	Benzoin	Benzil	Benzoin
1		60	90	92	90	50	80	93	90
2		45	60	94	91	40	50	94	92
3		90	110	94	91	80	100	95	91
4		60	75	96	92	50	65	96	91
5		120	145	94	90	100	140	95	90
6		120	150	93	89	100	140	94	90
7		90	110	96	90	80	95	96	91

Table 2 continued

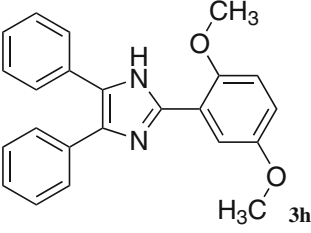
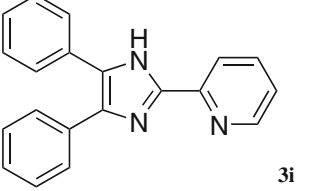
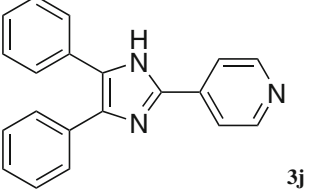
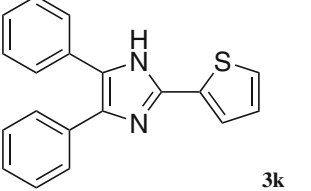
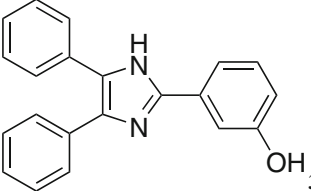
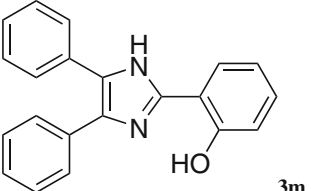
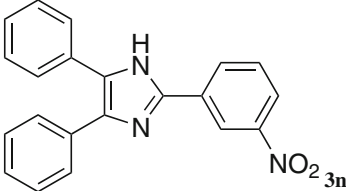
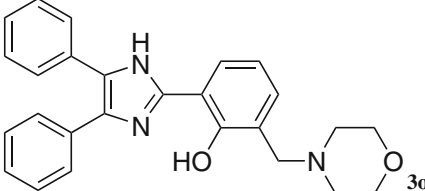
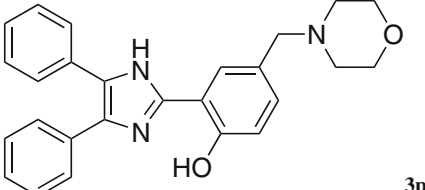
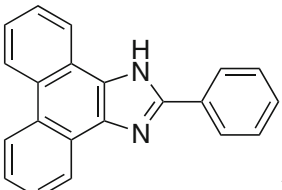
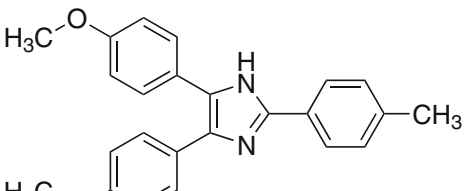
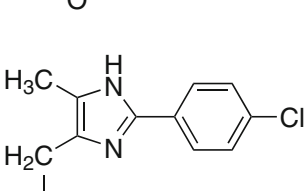
Entry	Product	Homogeneous catalyst				Heterogeneous catalyst			
		Time (min)		Yield (%) ^a		Time (min)		Yield (%) ^a	
		Benzil	Benzoin	Benzil	Benzoin	Benzil	Benzoin	Benzil	Benzoin
8		70	90	95	91	60	80	96	90
9		120	150	91	88	100	140	93	88
10		150	180	92	88	130	170	92	87
11		150	170	90	87	130	160	91	88
12		90	110	94	91	80	100	95	90
13		120	140	93	90	110	125	93	89

Table 2 continued

Entry	Product	Homogeneous catalyst				Heterogeneous catalyst				
		Time (min)		Yield (%) ^a		Time (min)		Yield (%) ^a		
		Benzil	Benzoin	Benzil	Benzoin	Benzil	Benzoin	Benzil	Benzoin	
14										
		150	170	90	87	130	150	91	87	
15										
		120	150	92	89	110	135	93	88	
16										
		120	140	92	88	115	130	92	88	
17										
		120	-	89	-	110	-	90	-	
18										
		-	120	-	88	-	115	-	90	
19										
		180	-	55	-	170	-	55	-	

^a Isolated yield

Table 3 Comparison of the efficiency of various catalysts in the synthesis of 2,4,5-triphenyl-1*H*-imidazole

Entry	Benzil/Benzoin	Conditions	Catalyst	Time (min)	Yield (%)	Ref.
1	Benzil	EtOH/50 °C	Mo(IV) salen complex	50	93	This study
	Benzoin		Nanoparticles onto silica	80	90	
2	Benzil	MeOH/60 °C	L-proline	540	90	[1]
3	Benzil	EtOH/reflux	Scolecite	35	96	[3]
	Benzoin			60	94	
4	Benzil	EtOH/reflux	Polymer-supported zinc chloride	90	96	[5]
5	Benzil	MeOH/RT	InCl ₃ · 3H ₂ O	498	82	[7]
6	Benzil	CH ₃ CN/RT	ZrCl ₄	45	95	[8]
7	Benzil	Solvent-free/140 °C	Heteropolyacid supported on silica	120	85	[9]
		Solvent-free/ MW		10	90	
8	Benzil	EtOH/reflux	NiCl ₂ · 6H ₂ O/Al ₂ O ₃	25	94	[10]
9	Benzil	Ethanol–water/reflux	NaHSO ₃	30	98	[11]
	Benzoin			40	96	
10	Benzil	Ethanol–water/	sulfated tin oxide	30	92	[12]
	Benzoin	Reflux		30–55	85–98	
11	Benzil	Ethanol-water/65 °C	Ceric ammonium nitrate (CAN)	50	98	[15]
	Benzoin			70	90	
12	Benzil	EtOH/reflux	UO ₂ (NO ₃) ₂ · 6H ₂ O	30	93	[16]
13	Benzil	[Hbim]BF ₄ /100 °C	Ionic liquids	60	95	[17]
	Benzoin			60	95	
14	Benzil	<i>t</i> -BuOH/ 60–65 °C	DABCO	720	92	[18]

a Bruker Avance DPX-250 (¹H-NMR 250 MHz and ¹³C NMR 62.9 MHz) spectrometer using TMS as the internal standard in pure deuterated chloroform and dimethyl sulfoxide. The following NMR spectra abbreviations are used: s = singlet, d = doublet, t = triplet, q = quartet, m = multiplet, and dd = doublet of doublets. IR spectra were obtained by a Shimadzu FT-IR 8300 spectrophotometer. SEM instrumentation is used for Scanning electron micrographs (SEM, XL-30 FEG SEM, Philips, at 20 KV). AFM images are also obtained by an atomic force microscope (AFM, DME-SPM, version 2.0.0.9). Mass spectra were determined at 70 or 20 eV on a Shimadzu GCMS-QP 1000 EX instruments. Melting points were obtained in open capillary tubes using a Büchi-535 circulating oil melting point apparatus (melting points are uncorrected). Catalysts were characterized using X-ray diffraction (XRD, D8, Advance, Bruker, axs). Thermogravimetry analysis (TG) was carried out using a lab-made TGA instrument. ICP analysis was conducted using a Varian Vista-pro analyzer. The purity of substrates and reaction monitoring were determined by TLC on silica gel Poly-Gram SILG/UV 254 plates. Column chromatography was performed in glass columns on short columns of silica gel 60 (70–230 mesh). Schiff-base complexes and Schiff-base *N*, *N'*-(salicylaldehyde)ethylenediamine (Salen) were prepared according to the literature [20,21,25]. Mo-salen complex was synthesized according to the literature [25].

Immobilization of Mo-salen complexes onto silica

3-Aminopropyl functionalized silica gel (1.0 g) and Mo-salen complex (0.05 g) in 50 mL of dry toluene were mixed. The mixture was stirred under reflux for 4 h. The solid was separated by centrifugation, washed several times with toluene and ethanol, and then dried in vacuum at 60 °C.

General procedure for synthesis of 2,4,5-triarylimidazole derivatives in the presence of a homogeneous catalyst

A mixture of benzil or benzoin (1.0 mmol), an aromatic aldehyde (1.0 mmol), ammonium acetate (2.0 mmol), and Mo-salen complex (2.5 mol %) was stirred in EtOH (96 %, 10 mL) at 50 °C for the time specified in Table 2. The reaction was monitored by TLC. After reaction completion, the solvent was evaporated to give the crude product, which was purified by silica gel column chromatography employing *n*-hexane/ethyl acetate (7:1) as eluent.

General procedure for synthesis of 2,4,5-triarylimidazole derivatives in the presence of a heterogeneous catalyst

For each reaction, benzil or benzoin (1.0 mmol), an aryl aldehyde (1.0 mmol), and ammonium acetate (2.0 mmol) was stirred in 10.0 mL of EtOH (96 %) in the presence of

heterogeneous catalyst (2.5 mol%) at 50 °C. The reactions were monitored by TLC using *n*-hexane/ethyl acetate (7:1). After the completion of the reaction, the reaction mixture was centrifuged and rinsed with EtOH (3 × 15 mL). The recovered catalyst was stored for future reaction. The combined filtrate was concentrated to give crude product, which was purified by silica gel column chromatography employing *n*-hexane / ethyl acetate (7:1) as eluent.

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