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ORIGINAL PAPER

$Ag–TiO_2$ nanocomposite-catalyzed one-pot synthesis of 1,2,4,5-tetrasubstituted imidazoles: a green and benign approach

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Abstract

$Ag–TiO_2$ nanocomposite as an efficient catalyst was synthesized and characterized by AFM analysis. This nanocomposite acts as a heterogeneous and recyclable catalyst for the room temperature synthesis of 1,2,4,5-tetrasubstituted imidazoles from aldehydes, benzil, ammonium acetate and several anilines or aliphatic amines via one-pot four-component condensation reaction in H_2O as a green solvent. $Ag–TiO_2$ nanocomposite was simply recovered by filtration and was reused up to three times.

Keywords $Ag–TiO_2$ nanocomposite · Four-component condensation reaction · 1,2,4,5-tetrasubstituted imidazoles

Introduction

Heterogeneously catalyzed approaches have useful rule in generalization of the procedure, easy separation, shrinking of waste production and recycling of the catalysts [1–4]. It is important for the green chemistry to build up efficient and eco-friendly mild catalytic methods which have become one of the chief themes of present synthetic chemistry [5, 6]. One of the most substantial field in terms of its efficacy is nano-catalysis [7]. These catalysts are identified as effective techniques for increasing recyclable catalysts because of their effortlessness of separation [8–11]. The nanoparticles improve the revealed surface area of the active component of the catalyst, thus improving the contact between substrates and catalyst [12–16].

Multi-component reactions (MCRs) are important 'medicinal toolkit' reactions approving fast assembly of three or further components in a one-pot sequence to afford a target product [17–20]. This influential synthetic tool gives an informal and suitable access to archives of organic products with various substitution frameworks [21, 22].

Among the numerous *N*-containing skeletons, imidazole derivatives are main building blocks for the preparation of biologically active molecules and pharmacologically active compounds [23, 24]. Imidazole rings have a varied range of effective biological activities such as antifungal, anti-inflammatory, antibiotic, anti-allergenic and antitumor [25–27]. Numerous approaches and catalysts have been described to produce 1,2,4,5-tetrasubstituted imidazoles such as nano-sized Cu/Zn-modified MCM-41 [28], isocyanurate-based periodic mesoporous organosilica [29], sodium lauryl sulfate [30], $\{[2,6\text{-DMPyH}]C(NO_2)_3\}$ [31], Fe_3O_4-polyethylene glycol-Cu nanocomposite [32], $NiFe_2O_4$ nanoparticle supported phosphomolybdic acid [33], Wells–Dawson heteropolyacid supported on silica [34], copper(I) complex of 1,3-dimethylbarbituric acid-modified SBA-15 [35], peptide nanofiber-templated zinc oxide nanostructures [36] and nanoaluminum nitride [37].

In continuation of our previous investigation on the application of nano-$Ag–TiO_2$ catalyst and other nano-metal oxide catalysts in organic synthesis and multi-component reactions [38–45], we herein report an efficient approach to give 1,2,4,5-tetrasubstituted imidazoles with the aid of a recyclable heterogeneous $Ag–TiO_2$ nanocomposite catalyst starting from aldehydes, benzil, ammonium acetate and several anilines or aliphatic amines in H_2O as a green solvent at room temperature (Scheme 1).

Experimental

General methods

$Ag–TiO_2$ nanocomposite was purchased from commercial centers and then characterized by atomic force microscope

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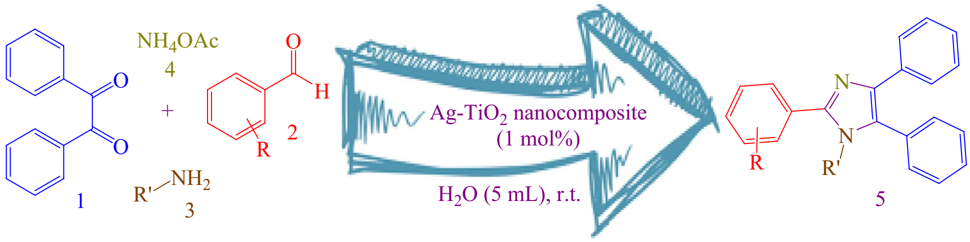
Scheme 1 Synthesis of 1,2,4,5-tetrasubstituted imidazoles by using Ag–TiO_2 nanocomposite catalyst



(AFM) and transmission electron microscopy (TEM) techniques [38–40]. All reagents were purchased from Sigma-Aldrich or Merck and were applied without purification. Solvents were dried, distilled and stored over molecular sieves. All the reactions and the purity of the products were observed by TLC by using TLC aluminum sheets silica gel F_{254}. All melting points measured on a Thermo Scientific apparatus and also are uncorrected. The infrared spectra were recorded in KBr disks on a FT-IR Bruker (WQF-510) instrument. The ^{1}H NMR and ^{13}C NMR spectra were recorded on a Bruker DRX-400 MHz instrument, using TMS as an internal standard in DMSO-d_6. Chemical shifts are measured in ppm. The surface morphologies were described by AFM.

General procedure for the synthesis of 1,2,4,5-tetrasubstituted imidazoles

To a solution of aldehydes (1 mmol), benzil (1 mmol; 0.210 g), ammonium acetate (1 mmol; 0.077 g) and several anilines or aliphatic amines (1 mmol) in H_2O (5 mL) at room temperature with continuous stirring, Ag–TiO_2 nanocomposite (1 mol%) was added and reacted for appropriate time (Table 2). After completion of the reaction monitored by TLC (*n*-hexane/ethyl acetate; 4:1), the precipitate was collected by filtration. The filtrate was washed with ethanol and dried to obtain desired pure products.

Selected spectral data analysis for desired product

***1,2-Bis(4-chlorophenyl)-4,5-diphenyl-1H-imidazole* (Table 2, *entry 25*):** White solid, M.p.: 187–189 °C; Yield: 85%; FT-IR (KBr)(υ_{max}, cm^{-1}): 3050, 1486, 1089, 835; ^{1}H NMR (400 MHz, DMSO-d_6): $\delta = 7.18$–7.20 (d, 2H, $J = 8.0$ Hz, Ar–H), 7.23–7.27 (dd, 4H, $J = 8.0$ Hz and $J = 4.0$ Hz, Ar–H), 7.30–7.34 (dd, 4H, $J = 8.0$ Hz and $J = 4.0$ Hz, Ar–H), 7.40–7.43(dd, 6H, $J = 6.0$ Hz and $J = 4.0$ Hz, Ar–H), 7.48–7.50 (d, 2H, $J = 8.0$ Hz, Ar–H); ^{13}C NMR (100 MHz, DMSO-d_6): 126.8, 127.1, 128.7, 128.9, 129.1, 129.2, 129.3, 129.5, 129.8, 130.5, 131.0, 131.6, 131.9, 133.8, 133.9, 134.5, 135.8, 137.5, 145.4.

Results and discussion

Characterization of Ag–TiO_2 nanocomposite

AFM analysis was also performed to confirm the structure morphology and topography of Ag–TiO_2 nanocomposite (Figure S1) and transmission electron micrograph showing the size of nanocomposite (Figure S2) [38–40].

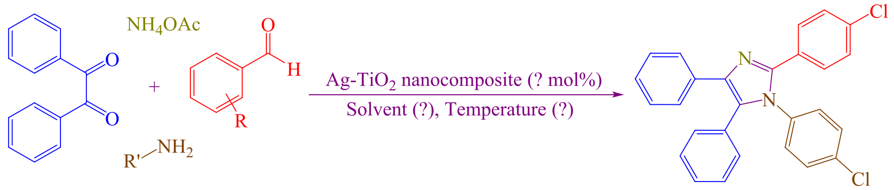
Synthesis of 1,2,4,5-tetrasubstituted imidazoles by using Ag–TiO_2 nanocomposite

Firstly, we studied the condensation of 4-chlorobenzaldehyde, benzil, ammonium acetate and 4-chloroaniline as a model of reaction in the presence of Ag–TiO_2 nanocomposite in numerous conditions. As revealed in Table 1, we used several solvents containing H_2O, C_2H_5OH, CH_3CN, CH_2Cl_2, ethyl acetate, benzene and toluene (Table 1, entries 1–7) at room temperature. According to reaction time and yield of desired product, the suitable result was attained by using 1 mol% catalytic amount of Ag–TiO_2 nanocomposite in H_2O as a solvent with respect to possessing a green nature and clean workup process (Table 1, entry 1). Also, various amounts of Ag–TiO_2 nanocomposite (Table 1, entries 1 and 8–11) were used and the results showed that 1 mol% of catalyst is appropriate (Table 1, entry1).

Moreover, in the absence of any catalyst, the reaction did not progress and no desired product was attained even when the reaction was heated under reflux conditions in H_2O for 120 min, and the substrates were recovered together (Table 1, entry 12), therefore suggesting the need of catalyst. The results showed that on increasing the temperature from room temperature to reflux condition the desired product was achieved in similar isolated yield. Additionally, the reaction was carried out in the presence of several quantities of starting materials. The best result was attained with a 1:1:1:1 molar ratio for 4-chlorobenzaldehyde, benzil, ammonium acetate and 4-chloroaniline, respectively.

Based on the above optimized conditions, we have taken several aldehydes having different electron-withdrawing

Table 1 Optimization of reaction condition for the synthesis of 1,2,4,5-tetrasubstituted imidazoles[a]



Entry	Solvent	Catalyst loading (mol%)	Time (min)	Yield (%)[b]
1	H_2O	1	10	85
2	C_2H_5OH	1	10	85
3	CH_3CN	1	10	78
4	CH_2Cl_2	1	20	75
5	Ethyl acetate	1	35	70
6	Benzene	1	120	45
7	Toluene	1	60	49
8	H_2O	0.5	25	75
9	H_2O	2	10	85
10	H_2O	5	10	85
11	H_2O	10	10	80
12	H_2O	–	120	–

[a] Reaction condition: 4-chlorobenzaldehyde (1 mmol; 0140 g), benzil (1 mmol; 0.210 g), ammonium acetate (1 mmol; 0.077 g), 4-chloroaniline (1 mmol; 0.127 g), r.t.; [b] Isolated yield

and releasing groups in the *ortho*, *meta* and *para* positions and hetero-aromatic aldehydes for the synthesis of 1,2,4,5-tetrasubstituted imidazoles (Table 2). As summarized in Table 2, the substituents of the aldehydes shown displayed that the reaction time for the aromatic aldehydes bearing electron-withdrawing substituents is rather faster and with slightly higher yield than those for electron-donating substituents. Furthermore, aniline derivatives and aliphatic amines afforded appropriate yields of the desired products.

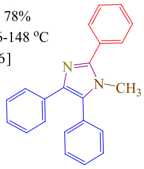
In another investigation, reusability of Ag–TiO$_2$ nanocomposite was studied in model reaction. Subsequent to the completion of the reaction, hot ethanol was added to the reaction mixture and the resulted solid (recycled catalyst) was filtered. The recycled catalyst was washed three times with hot ethanol (3×5 mL) and dried at 80 °C and reused at least six times without any loss of its catalytic activity (Fig. 1). The structure of recycled catalyst was investigated by AFM analysis, and the results showed that the catalyst is stable (Figure S3).

A suggested mechanism for the synthesis of 1,2,4,5-trisubstituted imidazoles is shown in Scheme 2 [31]. The Ag–TiO$_2$ nanocomposite catalyst activates the carbonyl functional group of aldehyde **2** for the nucleophilic addition of anilines or aliphatic amines **3** on the intermediate **2′** to give intermediates **6**. The synthesis of intermediate **7** occurs via elimination of water. Ammonia is formed in situ by decomposition of ammonium acetate **4** and then nucleophilic attack of ammonia to intermediate **7** to afford aminal intermediate **8**. In the next step, aminal intermediate **8** is condensed with activated benzyl **1′** to provide intermediate **9** which rearranges via elimination of water to the 1,2,4,5-trisubstituted imidazoles target molecule **5**.

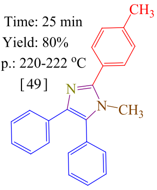
To show the significant properties of our study, we have compared our result with the identified data from the other works for the synthesis of 1,2,4,5-tetraphenyl-1*H*-imidazole (Table 2, entry 7). For comparison, this target molecule was chosen, as revealed in Table 3. No use of hazardous solvent, short reaction time and high yields of products make this method as a valuable approach for the synthesis of library of 1,2,4,5-trisubstituted imidazoles.

So as to assess the effectiveness of Ag–TiO$_2$ nanocomposite catalyst on the synthesis of 1,2,4,5-trisubstituted imidazoles, turn over frequency (TOF) value was calculated via the equation: TOF = yield of product (%)/[reaction time (min) × amount of catalyst (mol%)], TOF value is min^{-1}. Also, turn over number (TON) was calculated *via* the equation: TON = [TOF × reaction time (min)], and TON is a useless value (Table 4) [63–66].

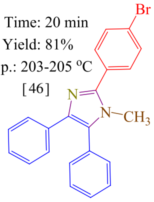
Table 2 Synthesis of 1,2,4,5-tetrasubstituted imidazoles in the presence of Ag-TiO_2nanocomposite.[a,b]



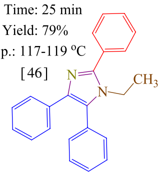
1; Time: 25 min
Yield: 78%
M.p.: 146-148 °C
[46]



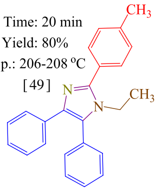
2; Time: 25 min
Yield: 80%
M.p.: 220-222 °C
[49]



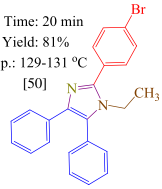
3; Time: 20 min
Yield: 81%
M.p.: 203-205 °C
[46]



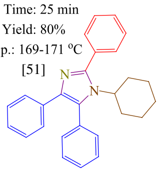
4; Time: 25 min
Yield: 79%
M.p.: 117-119 °C
[46]



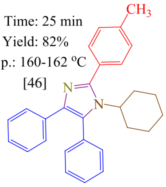
5; Time: 20 min
Yield: 80%
M.p.: 206-208 °C
[49]



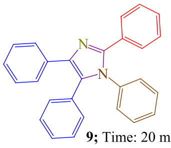
6; Time: 20 min
Yield: 81%
M.p.: 129-131 °C
[50]



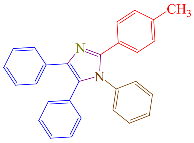
7; Time: 25 min
Yield: 80%
M.p.: 169-171 °C
[51]



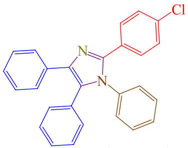
8; Time: 25 min
Yield: 82%
M.p.: 160-162 °C
[46]



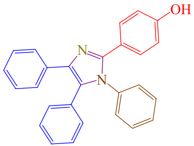
9; Time: 20 min
Yield: 81%
M.p.: 218-220 °C
[46]



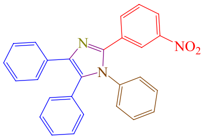
10; Time: 15 min
Yield: 83%
M.p.: 181-183 °C
[46]



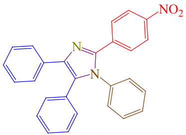
11; Time: 15 min
Yield: 84%
M.p.: 157-159 °C
[46]



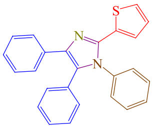
12; Time: 20 min
Yield: 83%
M.p.: 193-195 °C
[46]



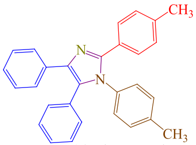
13; Time: 15 min
Yield: 83%
M.p.: 241-243 °C
[52]



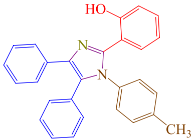
14; Time: 15 min
Yield: 86%
M.p.: 167-169 °C
[48]



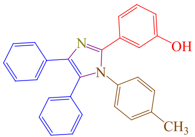
15; Time: 20 min
Yield: 81%
M.p.: 284-286 °C
[52]



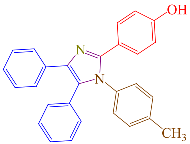
16; Time: 15 min
Yield: 84%
M.p.: 187-189 °C
[46]



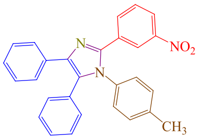
17; Time: 20 min
Yield: 81%
M.p.: 224-226 °C
[46]



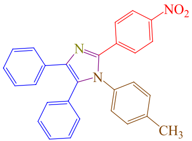
18; Time: 20 min
Yield: 83%
M.p.: 229-231 °C
[46]



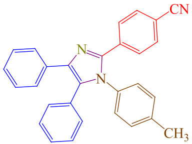
19; Time: 15 min
Yield: 84%
M.p.: 273-275 °C
[47]



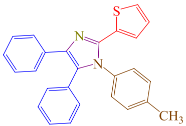
20; Time: 15 min
Yield: 84%
M.p.: 217-219 °C
[46]



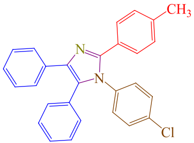
21; Time: 10 min
Yield: 86%
M.p.: 151-153 °C
[46]



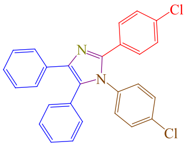
22; Time: 10 min
Yield: 81%
M.p.: 198-200 °C
[46]



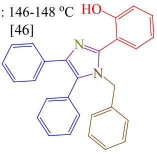
23; Time: 15 min
Yield: 82%
M.p.: 201-203 °C
[46]



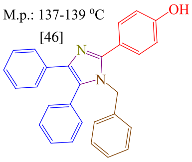
24; Time: 10 min
Yield: 83%
M.p.: 167-169 °C
[46]



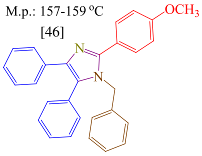
25; Time: 10 min
Yield: 85%
M.p.: 187-189 °C
[46]



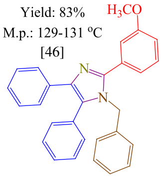
26; Time: 25 min
Yield: 81%
M.p.: 146-148 °C
[46]



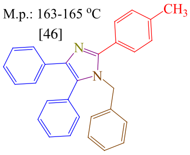
27; Time: 20 min
Yield: 83%
M.p.: 137-139 °C
[46]



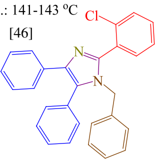
28; Time: 20 min
Yield: 84%
M.p.: 157-159 °C
[46]



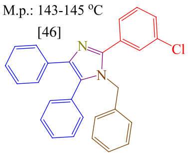
29; Time: 20 min
Yield: 83%
M.p.: 129-131 °C
[46]



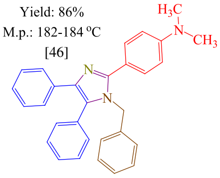
30; Time: 20 min
Yield: 84%
M.p.: 163-165 °C
[46]



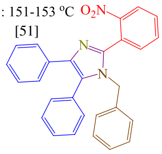
31; Time: 20 min
Yield: 82%
M.p.: 141-143 °C
[46]



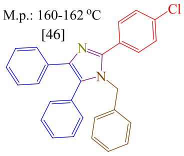
32; Time: 15 min
Yield: 84%
M.p.: 143-145 °C
[46]



33; Time: 15 min
Yield: 86%
M.p.: 182-184 °C
[46]



34; Time: 20 min
Yield: 83%
M.p.: 151-153 °C
[51]



35; Time: 15 min
Yield: 85%
M.p.: 160-162 °C
[46]

[a] Reaction condition: aldehyde (1 mmol), benzil (1 mmol; 0.210 g), ammonium acetate (1 mmol; 0.077 g), several anilines or aliphatic amines (1 mmol), Ag–TiO_2 nanocomposite (1 mol%); H_2O (5 mL), room temperature; [b] Isolated yield

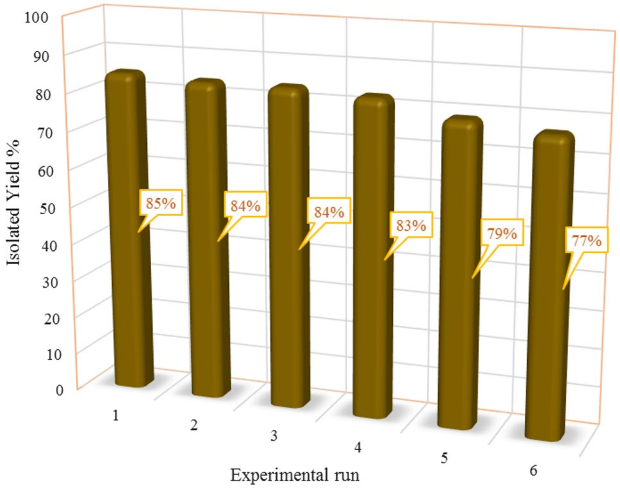


Fig. 1 Recyclability studies of Ag–TiO_2 nanocomposite

Conclusion

In summary, we have revealed an efficient and benign approach for one-pot, four-component synthesis of 1,2,4,5-trisubstituted imidazoles utilizing Ag–TiO_2 nanocomposite catalyst in H_2O as a green solvent at room temperature. The combination of easy experimental processes and effortlessness of recovery and reuse of catalyst as well as the reaction medium is expected to provide to the progress of a green approach for the synthesis of target molecules. High isolated yields, improved reaction rates, eco-friendly and no requirement of column chromatography for separation are also the significant advantages of this procedure.

Scheme 2 Proposed mechanism for the synthesis of 1,2,4,5-trisubstituted imidazoles by using Ag–TiO_2 nanocomposite

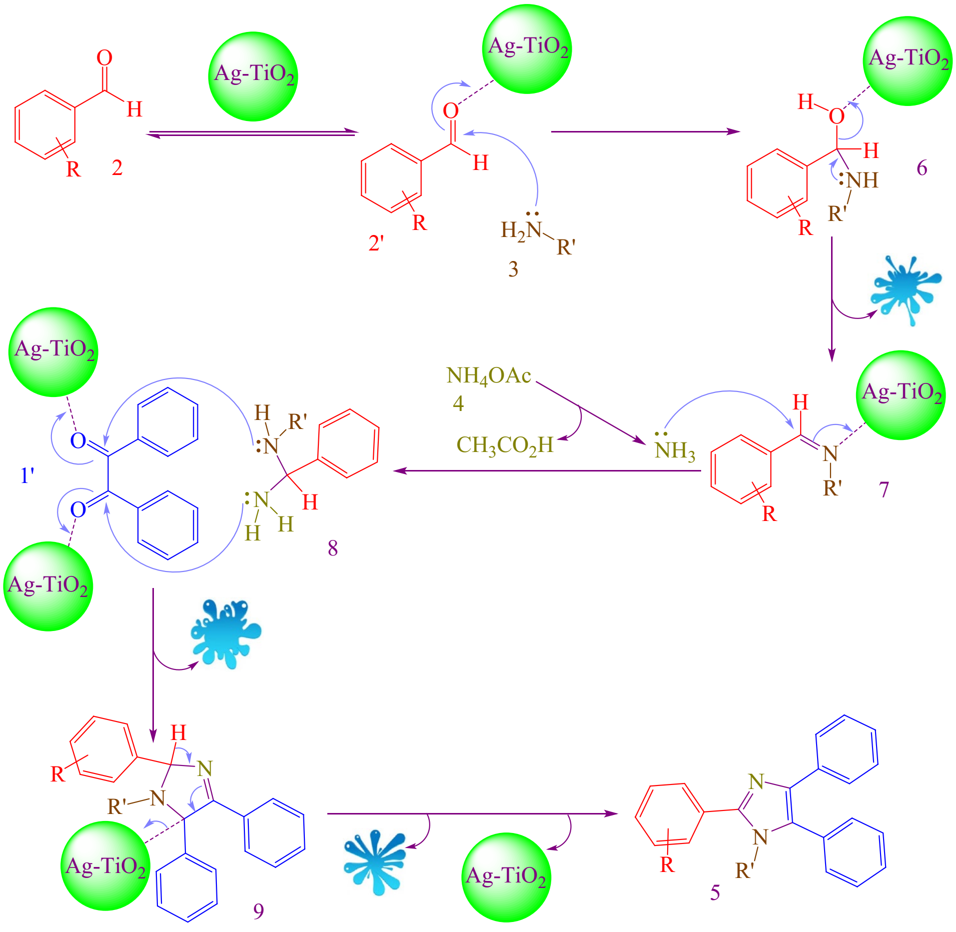
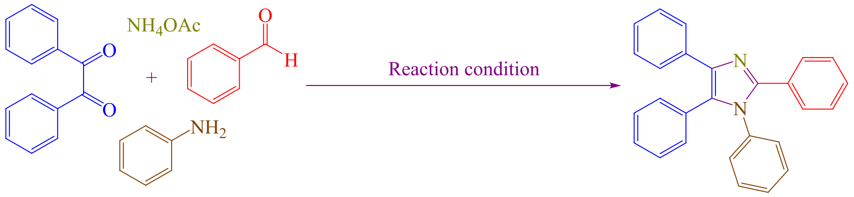


Table 3 Comparison of this procedure with some other procedures for the synthesis of 1,2,4,5-tetraphenyl-1*H*-imidazole



Entry	Reaction condition	Time (min)	Yield (%)[a]	Reported reference
1	Ag–TiO_2 nanocomposite (1 mol%), H_2O, r.t	20	81	This work
2	TFA (20 mol%), solvent free, MW (150 W)	4	88	[53]
3	*n*-Bu_4NBr (10 mol%), *t*-BuOH, 80 °C	480	87	[54]
4	Pivalic acid (1 equiv), DMSO/H_2O (1:1), 140 °C, air	24–48 h	88	[55]
5	DBSA (20 mol%), H_2O, reflux	240	81	[56]
6	$CuFe_2O_4$ (0.05 g), solvent free, 90 °C	80	77	[57]
7	SLS (0.02 g), H_2O, 90 °C	60	90	[33]
8	Zeolite H-ZSM 22 (0.08 g), C_2H_5OH, 140 °C	15–30	81	[58]
9	Nano-crystalline $MgAl_2O_4$ (0.05 g), C_2H_5OH, 60 °C	12–25	91	[59]
10	Fe_2O_3@TiO_2-EG-CuII (2.5 mol%), solvent free, 100 °C	25	98	[60]
11	Fe_3O_4@SiO_2@TiO_2 (10 mol%), solvent free, 110 °C	15	82	[61]
12	Nano-TiO_2@SiO_2 (0.04 g), CH_3OH, r.t	180	78	[62]

[a] Isolated yield

Table 4 TON and TOF of products **1** to **35**

1; TOF=3.12 min^{-1} TON=78	**2**; TOF=3.20 min^{-1} TON=80	**3**; TOF=4.05 min^{-1} TON=81	**4**; TOF=3.16 min^{-1} TON=79	**5**; TOF=4.00 min^{-1} TON=80
6; TOF=4.05 min^{-1} TON=81	**7**; TOF=3.20 min^{-1} TON=80	**8**; TOF=3.28 min^{-1} TON=82	**9**; TOF=4.05 min^{-1} TON=81	**10**; TOF=5.53 min^{-1} TON=83
11; TOF=5.60 min^{-1} TON=84	**12**; TOF=4.15 min^{-1} TON=83	**13**; TOF=5.53 min^{-1} TON=83	**14**; TOF=5.73 min^{-1} TON=86	**15**; TOF=4.05 min^{-1} TON=81
16; TOF=5.60 min^{-1} TON=84	**17**; TOF=4.05 min^{-1} TON=81	**18**; TOF=4.15 min^{-1} TON=83	**19**; TOF=5.60 min^{-1} TON=84	**20**; TOF=5.60 min^{-1} TON=84
21; TOF=8.60 min^{-1} TON=86	**22**; TOF=8.10 min^{-1} TON=81	**23**; TOF=5.47 min^{-1} TON=82	**24**; TOF=8.30 min^{-1} TON=83	**25**; TOF=8.50 min^{-1} TON=85
26; TOF=3.24 min^{-1} TON=81	**27**; TOF=4.15 min^{-1} TON=83	**28**; TOF=4.20 min^{-1} TON=84	**29**; TOF=4.15 min^{-1} TON=83	**30**; TOF=4.20 min^{-1} TON=84
31; TOF=4.10 min^{-1} TON=82	**32** TOF=5.60 min^{-1} TON=84	**33** TOF=5.73 min^{-1} TON=86	**34**; TOF=4.15 min^{-1} TON=83	**35**; TOF=5.67 min^{-1} TON=85

Supporting Information

The supporting information includes AFM and TEM analysis and spectral images of FT-IR, ^1H NMR and ^{13}C NMR of selected product.

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Compliance with ethical standards

Conflict of interest The authors declare no conflict of interest.

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