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Nano TiO_2@SiO_2 as an efficient and reusable catalyst for the synthesis of multi-substituted imidazoles

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ABSTRACT

Nano TiO_2 supported on SiO_2 (Nano TiO_2@SiO_2) as a solid Lewis acid, was described to be an effective and reusable catalyst for one-pot three-component reaction of benzil, aryl aldehydes and ammonium acetate for the synthesis 2-aryl-4,5-diphenyl-1*H*-imdazoles synthesis. To explore the high efficacy of the catalytic system the four-component cyclization of benzil, aryl aldehydes, ammonium acetate and anilines has also been accomplished successfully to obtain their 1,2-diaryl-4,5-diphenyl-1*H*-imidazole adducts. A plausible mechanism has also been expressed. The catalyst has been recovered and its reusability confirmed in 4 runs. High yields, simple operation, easy-work procedure, mild reaction conditions in addition with the reusability are some advantages of this protocol.

Keywords: *Multi-substituted imidazoles; One-pot reaction; Benzil; Nano TiO_2.*

1. Introduction

Imidazoles are key motifs present in the structure of many natural products which possess wide-range of biological activities containing anti-inflamatory [1], anti-allergic [2], and analgesic [3]. They are also the structural scaffolds in many therapeutic agents. Omeprazole [4], losartan, prosartan, and trifenagrel [5] are some of the considerable drugs in market with various functionalization around the imidazole motif. In addition to the biologically activity, the imidazole core has passkey usage in some advanced areas of technology such as fluorescence labeling agents [6], Biological imaging [7], and chromophores for non-linear optic systems [8]. In view of the Multifarious biological, pharmacological, and material properties of this five-membered heterocycle, developing of new synthetic protocols for imidazoles is still a matter of interest. In recent years numerous methods have been developed for the synthesis of highly substituted imidazoles using various catalytic systems including $SiO_2/NaHSO_4$ [9], molecular iodine [10], $K_5CoW_{12}O_{40}\cdot3H_2O$ [11], L-proline [12], $FeCl_3.6H_2O$ [13], 1,4-diazabicyclo[2.2.2]octane (DABCO) [14], Tetrabutylammonium bromide (TBAB) [15], $H_6P_2W_{18}O_{62}\cdot24H_2O$ supported on silica (WD/ SiO_2) [16], L-cysteine [17], nanocrystalline $MgAlO_4$ [18], Nano SiO_2/H_3PO_4 [19], N-Bromosuccinimide [20], silica-supported $SbCl_3$ [21], and SBA-15/2,2,2-trifluoroethanol (SBA/TFE) [22]. However, some of the reported methods suffer from disadvantages including the use of expensive, moisture-sensitive reagents, long reaction times, tedious work-up procedures, and large amount of catalyst loadings which leads in generation of huge amount of wastes into the environment.

Nano-TiO_2 has been devoted as good catalyst in organic chemistry because of its high activity, non-toxicity, easy availability, reusability, strong oxidizing power, Lewis acidity and long-term stability [23, 24]. Some catalytic activity of titania nanoparticles has been reported in the preparation of Hantzsch esters and polyhydroquinoline derivatives [25], Friedel-Crafts alkylation of indoels with nitroalkanes [26], xanthene derivative synthesis [27], α-acetamido ketones [28] and bis(indolyl)methanes synthesis [29], and preparation of imines [30].

Binary supporting catalysts are great urgent necessity for organic chemists due to their expanding surface area. SiO_2 (silica) is a common support for this

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purpose. Nano TiO_2 supported on silica has recently been used for photodegradation of new Fuchsin (C.I. 42520) and Amaranth (C.I. 16185) [31] and Photocatalytic oxidation of trinitro toluene (TNT) [32]. Herein we report the catalytic activity of Nano TiO_2/SiO_2 as an impressive, inexpensive and easily handling solid Lewis acid in the synthesis of 2-aryl-4,5-diphenylimidazoles and 1,2-diaryl-4,5-dipenyl imidazoles from one-pot multi-component reactions (MCRs) of benzil, aryl aldehydes, ammonium acetate and anilines at room temperature in CH_3OH (Scheme 1).

2. Experimental

2.1. General

Chemicals and solvents were purchased from Merck and Aldrich and used without further purifications. Anilines used in their distilled form. Commercial Nano TiO_2 (APS:10-25 nm, Purity:99%, SSA: 200-240 m^2/gr and Bulk density: 0.24 g/cm^3) in anatase phase were purchased from Nanosany corporation. Melting points were determined using a Stuart Scientific apparatus and are uncorrected. Fourier transform infrared (FT-IR) spectra (KBr discs, 500-4000 cm^{-1}) were recorded using a Bruker FTIR model Tensor 27 spectrometer. ^{1}H-NMR spectra were recorded in $CDCl_3$ solvent on a Bruker 400 MHz spectrometer. Preparative layer chromatography (PLC) carried out on 20×20 cm^2 plates, coated with a 1 mm layer of Merck silica gel PF254, prepared by applying the silica as slurry and drying in air. The scanning electron microscope (SEM, model ∑-IJMA) was used to characterize the nano structures.

All the products were characterized by comparison of their spectroscopic data (IR and ^{1}H-NMR) with those of the authentic samples in the literature.

Yields refer to isolated products.

2.2. Prepration of nano $TiO_2@SiO_2$

To a mixture of SiO_2 60 (for column chromatography, mesh 70-230, 0.06-0.2 mm, 2 g) in 50 mL $CHCl_3$, nano TiO_2 (Anatase phase, 10-25 nm, 2 g) was added. The mixture was stirred at room temperature for 90 min. The solvent was evaporated at room temperature overnight to obtain a white solid of 50% (W/W) nano $TiO_2@SiO_2$. The catalyst was characterized by IR spectra (Fig. 1) and SEM image (Fig. 2).

2.3. General procedure for the synthesis of 2-aryl-4,5-dipenyl-1H-imidazoles

To a mixture of benzil 1 (1 mmol), aryl aldehydes 2a-p (1 mmol) and ammonium acetate 3 (1 mmol) in CH_3OH (5 mL) at room temperature, 0.04 g of Nano $TiO_2@SiO_2$ was added. After completion of the reaction monitored by TLC (Table 2), the mixture was filtered and washed with further CH_3OH to separate the catalyst. The filtrate was evaporated and the residue was purified utilizing plate chromatography (PLC) (petroleum ether/EtOAc: 9/2) to obtain the pure products (5a-p).

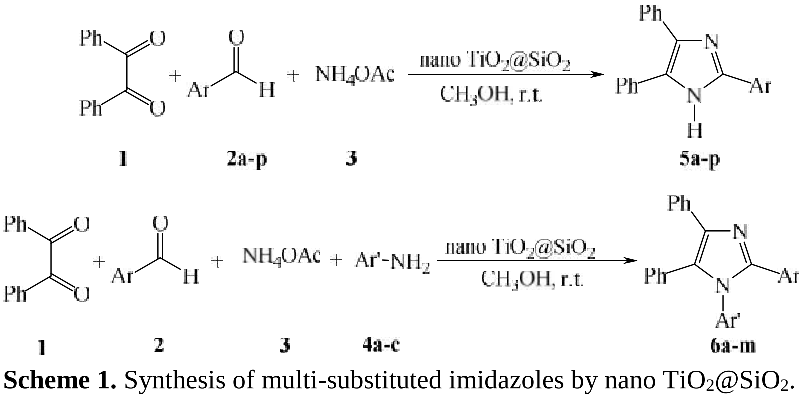
2.4. General procedure for the synthesis of 1,2,4,5-tetraaryl-1H-imidazoles

A mixture of benzil 1 (1 mmol), appropriate aryl aldehydes 2 (1 mmol), NH_4OAc 3 (4 mmol) and aryl amines 4a-c (4 mmol) in CH_3OH (5 mL) in the presence of 0.04 g of Nano $TiO_2@SiO_2$ stirred at room temperature. The progress of the reaction was monitored by TLC (Table 3). After completion, the mixture was filtered and washed with methanol (2×5 mL). The solvent was evaporated and the residue purified by PLC (petroleum ether/EtOAc: 9/2) to afford the pure products 6a-m. The spectral data of selected compounds is given below.

Selected spectral data

*Table 2, entry 2 (**5b**):*

m.p.= 268-270 °C [21]. IR (KBr): $\bar{\nu}$ = 3453, 3067, 1636, 1323 cm^{-1}. ^{1}HNMR (400 MHz, $CDCl_3$): δ= 12.75 (s, 1H), 8.11 (d, *J*= 8.1 Hz, 2H), 7.01 (t, *J*= 7.5 Hz, 2H), 7.18 (t, *J*= 6.75, 2H), 7.36 (t, *J*= 7.25 Hz, 4H), 7.51 (d, *J*= 7.97 Hz, 2H), 7.79 (d, *J*= 7.88 Hz, 2H) ppm.



Scheme 1. Synthesis of multi-substituted imidazoles by nano $TiO_2@SiO_2$.

*Table 3, entry 6 (**6f**):*

m.p.: 174-177 °C [18]. IR (KBr): $\bar{\nu}=$ 3045, 1618, 1578, 1158 cm^{-1}. ^{1}HNMR (400 MHz, $CDCl_3$): δ= 7.48 (d, *J*= 7.2 Hz, 1H), 7.15-7.34 (m, 15 H), 6.85 (d, *J*= 8.8 Hz, 2H), 3.6 (s, 6H) ppm.

3. Results and discussion

Firstly, nano $TiO_2@SiO_2$ has been characterized by FT-IR spectra and SEM technique. From FT-IR spectra spectrum (Fig. 1) nano TiO_2/SiO_2 formation confirmed by Si-O strong vibration at 1099 cm^{-1}, Ti-O-Ti stretching vibration at 706 cm^{-1} and weak Si-O-Ti vibration at 963 cm^{-1} [33] The SEM image (Fig. 2) revealed nano TiO_2/SiO_2 formation. Secondly, the condensation of 4-chlorobenzaldehyde 2b, benzil 1 and ammonium acetate 3was chosen as a model to obtain the optimized reaction conditions (Table 1). Performing the reaction under solvent-free conditions both at room temperature or in the presence of conventional heating did not obtained satisfactory results (entries 1, 2). Utilizing CH_3CN or H_2O as the solvent media in comparison to CH_3OH, at the same conditions, yielded to the weaker results (entries 3, 5). The results of the reaction progress in the presence of nano TiO_2 or SiO_2 lonely confirmed the high catalytic efficacy of the manufactured binary nano $TiO_2@SiO_2$ (entries 6, 7). Verifying the catalyst amount was not a suitable choice (entries 8, 9). Decreasing NH_4OAc amount to 2 mmol affected the reaction progress diminution (entry 10). Finally the best results were gained in the presence of Nano $TiO_2@SiO_2$ (0.04 g) at room temperature in CH_3OH media (entry 4).

In the next step, the general applicability of catalyst for varoius deriatives of 2-aryl-4,5-diphenyl-1*H*-imidazoles was investigated (Table 2). It was observed that benzaldehyde and its activated derivatives as well as deactivated aromatic aldehydes gave their corresponding tri-substituted imidazoles 4a-m succesfully.

It must be mentioned that electron-defiecnt substituents yielded better results in comparison to their electron-donating correspondances. The reaction time of 3- and 2-substituted arylaldehydes is a bit longer in comparison to their 4- substituted analogous. It could be due indirect inductive and/or resonance effects on the reaction center. The steric hindrance in 2-substituted derivatives is another interceptor operative for this sluggish response. Furfural and 1-naphthaldehyde also reacted successfully (4n-o). The specialty of the catalyst has been also perceived in the reaction of unsaturated aldehydes such as 3-phenyl-2-propenal. In this case no by-products has been obtained (5q). Next we focused on the synthesis of tetra substituted imidazoles. Aniline 4a has been chosen as a candidate to achieve the tetrasubstituted imidazoles (Table 3). According to Table 3 different aryl aldehydes with variant anilines alternated the four-component cyclization prosperously (6a-m).

The data of Table 3 affirm the high efficacy of the synthesized nano catalyst to gain the multi-substituted imidazoles. In the next step, the reusability of the recovered catalyst has been studied as another efficient and important aspect of this protocol. For this reason, the Nano $TiO_2@SiO_2$ was recovered from the reaction mixture of 2-(4-Chlorophenyl)-1,4,5-triphenyl-1*H*-imidazole (5b) by filtration and washing the solid residue with further CH_3OH (2 × 10 mL).

Drying overnight at room temperature and heating at 70 °C for 2 h leaded the recovered catalyst respectively. The obtained solid acid, reused and recycled with in four runs with almost no activity decrease. As could be seen in Table 2, the recovered Nano $TiO_2@SiO_2$ catalyzed the one-pot three-component reaction to obtain tri-substituted imidazole 5b within 90, 90, 88 and 85 % yield during 4 alternate separation and further usage (Fig. 3). In order to show the efficacy of nano $TiO_2@SiO_2$ with other catalysts, a

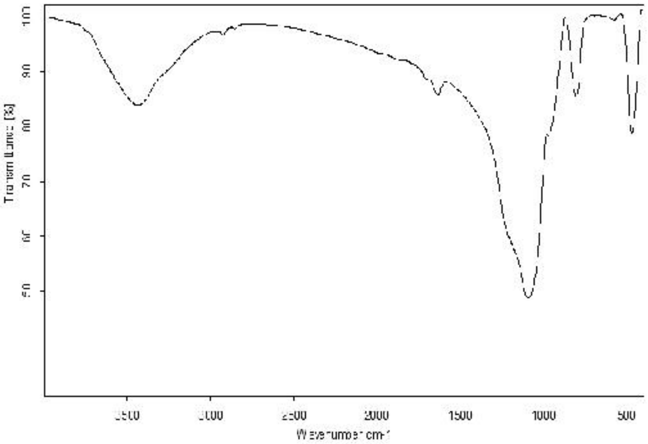


Fig. 1. IR-spectra of nano $TiO_2@SiO_2$.

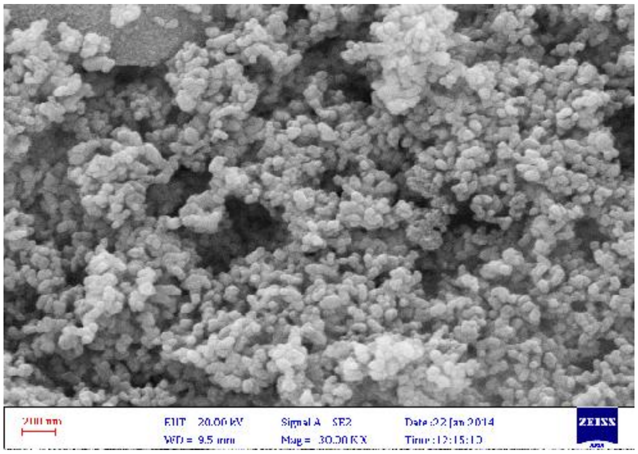


Fig. 2. The SEM image of synthesized nano TiO2@SiO_2.

Table 1. Screening optimization conditions in the condensation of benzil, 4-chlorobenzaldehyde and NH_4OAc in the presence of Nano $TiO_2@SiO_2$[a].

Entry	Condition	Time (h)	Yield[b] (%)
1	Nano $TiO_2@SiO_2$ (0.04 g)/ 80 °C/ solvent-free	2	-[c]
2	Nano $TiO_2@SiO_2$ (0.04 g)/ r.t./ solvent-free	5	70
3	Nano $TiO_2@SiO_2$ (0.04 g)/ r.t./ CH_3CN [d]	2.5	81
4	Nano $TiO_2@SiO_2$ (0.04 g)/ r.t./ CH_3OH	2	90
5	Nano $TiO_2@SiO_2$ (0.04 g)/ r.t./ H_2O	3	65
6	Nano TiO_2 (0.04 g)/ r.t./ CH_3OH	3	71
7	SiO_2 (0.04 g)/ r.t./ CH_3OH	4	70
8	Nano $TiO_2@SiO_2$ (0.03 g)/ r.t./ CH_3OH	2.5	82
9	Nano $TiO_2@SiO_2$ (0.05 g)/ r.t./ CH_3OH	2	90
10	Nano $TiO_2@SiO_2$ (0.04 g)/ NH_4OAc (2 mmol)/ r.t./ CH_3OH	3	74

[a] Benzil (1 mmol), 4-chlorobenzaldehyde (1 mmol) and NH_4OAc (4 mmol) have been used.
[b] Isolated yields.
[c] A mixture of products have been obtained.
[d] 5 mL of each solvent has been used.

Table 2. One-pot multi-component reaction of benzyl, different aldehydes and NH_4OAc in the presence of Nano $TiO_2@SiO_2$ at room temperature in CH_3OH for 2-aryl-4,5-diphenyl-1*H*-imidazoles synthesis.[a]

Entry	Ar	Product	Time (h)	Yield (%)	m.p. (°C)		[Ref.]
					Found	Reported	
1	C_6H_5	5a	2.75	88	264-266	266-267	[17]
2	4-ClC_6H_4	5b	2	90	268-270	272-273	[21]
3	4-BrC_6H_4	5c	2	92	198-201	203-205	[21]
4	$N(CH_3)_2C_6H_4$	5d	3.5	78	260-261	256-259	[12]
5	4-MeC_6H_4	5e	3	83	227-229	231-232	[17]
6	3-$NO_2C_6H_4$	5f	1.5	92	292-294	299	[15]
7	3-ClC_6H_4	5g	1.15	93	205-207	207-209	[21]
8	3-$MeOC_6H_4$	5h	2.5	85	217-219	220-221	[17]
9	2-$NO_2C_6H_4$	5i	2	81	230-232	230-231	[12]
10	2-OHC_6H_4	5j	3.5	78	280-282	282-284	[21]
11	2-$MeOC_6H_4$	5k	4	70	209-211	210-212	[17]
12	3,5-$(OMe)_2C_6H_3$	5l	4.5	78	260-263	269-271	[21]
13	2-OH-6-$NO_2C_6H_3$	5m	3.75	80	215-217	219-221	[17]
14	1-Naphthyl	5n	4	78	210-213	215	[17]
15	2-Furyl	5o	4	79	210-212	213-214	[17]
16	$C_6H_5CH=CH$	5p	3.45	81	194-196	198-199	[17]

[a] Benzil (1 mmol), aldehydes (1 mmol), NH_4OAc (4 mmol) have been used in the presence of 0.04 g of $TiO_2@SiO_2$ in CH_3OH (5 mL).

Table 3. One-pot condensation of benzyl, aryl aldehydes, anilines and ammonium acetate in the presence of Nano $TiO_2@SiO_2$ at room temperature in CH_3OH for 1,2-diaryl-4,5-diphenyl-1*H*-imidazoles synthesis.[a]

Entry	Ar	Ar'	Product	Time (h)	Yield (%)	m.p.(˚C)		[Ref.]
						Found	Reported	
1	C_6H_5	C_6H_5	6a	3	78	214-216	216-218	[21]
2	$4\text{-}ClC_6H_4$	C_6H_5	6b	2	81	150-153	152-154	[18]
3	$4\text{-}BrC_6H_4$	C_6H_5	6c	2	82	158-160	165-168	[18]
4	$4\text{-}MeC_6H_4$	C_6H_5	6d	4.5	83	182-184	186-188	[21]
5	$4\text{-}MeOC_6H_4$	C_6H_5	6e	5	75	250-251	253-254	[18]
6	$3,4\text{-}(MeO)_2C_6H_3$	C_6H_5	6f	4	83	174-177	178-180	[18]
7	$2\text{-}OHC_6H_4$	C_6H_5	6g	5.5	72	247-250	252-254	[21]
8	$2\text{-}MeC_6H_4$	C_6H_5	6h	5	74	185-187	187-189	[12]
9	$4\text{-}MeC_6H_4$	$4\text{-}ClC_6H_4$	6i	3	81	166-168	167-169	[11]
10	$4\text{-}ClC_6H_4$	$4\text{-}ClC_6H_4$	6j	2	88	185-187	187-189	[9]
11	$4\text{-}MeC_6H_4$	$4\text{-}MeC_6H_4$	6k	3	81	186-188	188-191	[11]
12	$3\text{-}NO_2C_6H_4$	$4\text{-}MeC_6H_4$	6l	2.5	83	146-147	149-151	[11]
13	$2\text{-}OHC_6H_4$	$4\text{-}MeC_6H_4$	6m	3	80	221-224	226-228	[11]

[a] Benzil (1 mmol), aldehydes (1 mmol), anilines (1 mmol), and NH_4OAc (4 mmol) have been used in the presence of 0.04 g of $TiO_2@SiO_2$ in CH_3OH (5 mL).

comparison between the present study for the synthesis of 2-(4-Chlorophenyl)-1,4,5-triphenyl-1*H*-imidazole (5b) with some other reported results in the literature, have been shown in Table 4. Performing the reaction at room temperature, utilizing small amount of the catalyst, reusability within several runs, relative short reaction time and high yield, in addition with eco-friendly nature of nano $TiO_2@SiO_2$ are interesting advantages of this newly synthesized nano catalyst in the synthesis of multi-substituted imidazoles. Finally the proposed mechanism for 1,2,4,5-tetrasubstitiuted imidazoles synthesis has be outlined in Scheme 2.

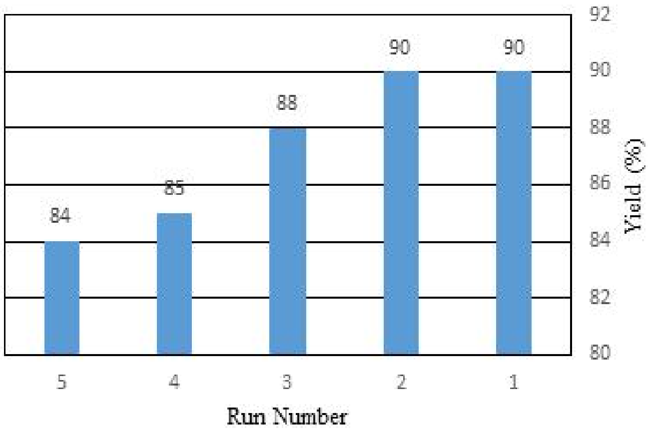


Fig. 3. The reusability of nano $TiO_2@SiO_2$ in the synthesis of 2-(4-Chlorophenyl)-1,4,5-triphenyl-1*H*-imidazole (**5b**).

As can be seen in Scheme 2, the catalyst activated the carbonyl group of aldehyde to form diamine intermediate [A]. Nucleophilic attack of [A] to activated benzil [B] yielded [C] which releases water to form [D]. Intramolecular nucleophilic attack in [D] leaded cyclization to [E]. The solid catalyst from this stage can be utilized for another cycle. Subsequently, dehydration of [E] gives the tetrasubstituted imidazole product.

4. Conclusions

In summary, Nano $TiO_2@SiO_2$ has been extinguished as an effective, eco-friendly and reusable solid acid catalyst which catalyzes the one-pot three-component reaction of benzil with aldehydes and ammonium acetate to form 2-aryl-4,5-diphenyl-1*H*-imidazoles in high yields. The applicability of the catalyst has also been confirmed in four component cyclo-condensation of benzil, aryl aldehydes, ammonium acetate and anilines to form their correspondence 1,2-diaryl-4,5-diphenyl-1H-imidazoles successfully. Simple reaction operation, easily work-up procedure associated with reusability and efficiency of the synthesized nano catalyst are the highlighted points of this work.

Acknowledgments

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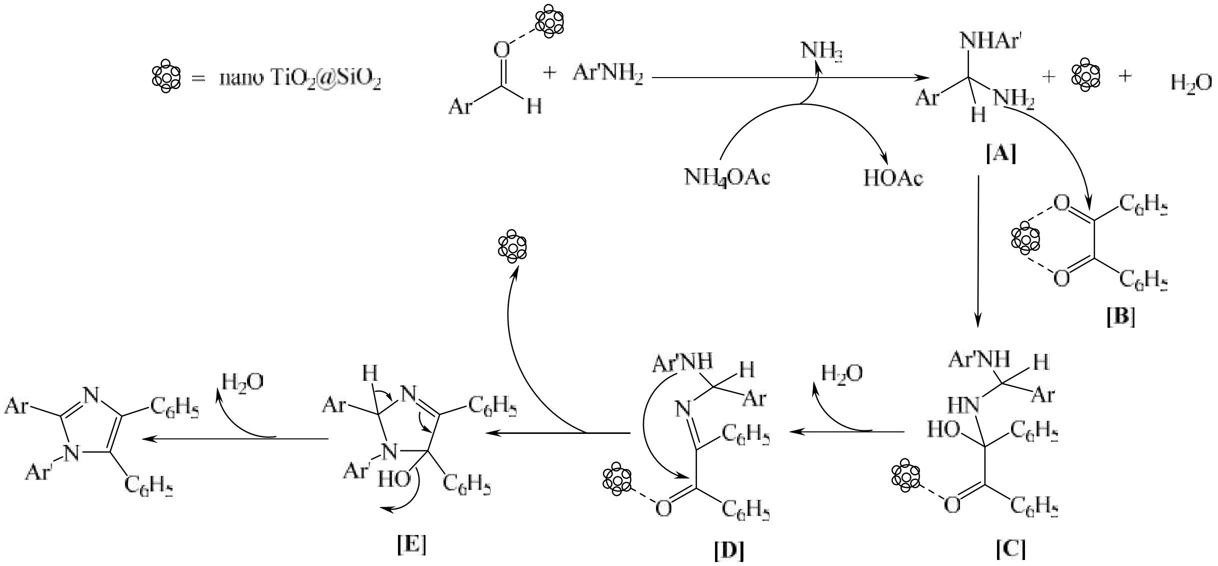
Table 4 Comparison of some reported methods with the present protocol for the synthesis of 2-(4-Chlorophenyl)-1,4,5-triphenyl-1*H*-imidazole (**5b**).

Entry	Condition	Time (h)	Yield (%)	[Ref.]
1	I_2 (5 mol%)/ 75 °C/ solvent -free	0.25	98	[10]
2	L-proline (15 mol%)/ 60 °C/ CH_3OH	9	88	[12]
3	(WD/ SiO_2) (0.6 g)/ r.t./ solvent -free	2	88	[16]
4	Nano SiO_2/H_3PO_4 (0.1 g)/ 140 °C / solvent -free	3	85	[19]
5	NBS (15 mol%)/ 120 °C/ solvent -free	0.75	92	[20]
6	SBA/TFE (0.1 g/3 mL)/ 90 °C/ solvent -free	3	92	[22]
7	Nano TiO_2@SiO_2 (0.04 g)/ r.t./ CH_3OH	2	90	This work



Scheme 2. The suggested mechanism of tetra-substituted imidazoles synthesis.

References

[1] J.G. Lombardino, E.H. Wiseman, J. Med. Chem. 17 (1974) 1182-1188.

[2] S. Wellert, H. Imhof, M. Dolle, H.J. Altmann, A. Richardt, T. Hellweg, Colloid Polym. Sci. 286 (2008) 417–426.

[3] U. Ucucu, N.G. Karaburun, I. Isikdag, Farmaco 56 (2001) 285-290.

[4] P. Lindberg, P. Nordberg, T. Alminger, A. Brandstorm, B. Wallmark, J. Med. Chem. 29 (1986) 1327-1329.

[5] R. Mannhold, Drugs Future 10 (1985) 570-577.

[6] H.J. Zhu, J.S. Wang, K.S. Patrick, J.L. Donovan, C.L. DeVane, J.S.J. Markowitz, Chromatogr. B: Anal. Technol. Biomed. Life Sci. 858 (2007) 91-97.

[7] Y.F. Sun, W. Huang, C.G. Lu, Y.P. Cui, Dyes Pigments 81 (2009) 10-17.

[8] M. Staehelin, D.M. Burland, M. Ebert, R.D. Miller, B.A. Smith, R.J. Twieg, W. Volksen, C.A. Walsh, Appl. Phys. Lett. 61 (1992) 1626-1628.

[9] A.R. Karimi, Z. Alimohammadi, J. Azizian, A.A. Mohammadi, M.R. Mohmmadizadeh, Catal. Commun. 7 (2006) 728-732.

[10] M. Kidwai, P. Mothsra, V. Bansal, R.K. Somvanshi, A.S. Ethayathulla, S. Dey, T.P. Singh, J. Mol. Catal. A: Chem. 265 (2007) 177-182.

[11] L. Nagarapu, S. Apuri, S. Kantevari, J. Mol. Catal. A: Chem. 266 (2007) 104-107

[12] S. Samai, G.C. Nandi, P. Singh, M.S. Singh, Tetrahedron 65 (2009) 10155-10161.

[13] M.M. Heravi, F. Derikvand, M. Haghighi, Monatsh. Chem. 139 (2008) 31-33.

[14] S.N. Murthy, B. Madhav, Y.V.D. Nageswar, Tetrahedron Lett. 51 (2010) 5252-5257.

[15] M.V. Chary, N.Ch. Keerthysri, S.V.N. Vupallapati, N. Lingaiah, S. Kantevari, Catal. Commun. 9 (2008) 2013-2017.

[16] A.R. Karimi, Z. Alimohammadi, M.M. Amini, Mol. Divers. 14 (2010) 635-641.

[17] H.N. Roy, M.M. Rahman, P.K. Pramanick, Indian J. Chem. B 52 (2013) 153-159.

[18] J. Safari, S. Gandomi Ravandi, Z. Akbari, J. Adv. Res. 4 (2013) 509-514.

[19] A. Bamoniri, B.F. Mirjalili, S. Nazemian, N.Y. Mahabadi, Bulg. Chem. Commun. 46 (2014) 79-84.

[20] B. Maleki, S. Sedigh Ashrafi, J. Mex. Chem. Soc. 58 (2014) 76-81.

[21] J. Safari, N. Naseh, Z. Zarnegar, Z. Akbari, J. Taibah Univ. Sci. (2014) doi.org/10.1016/j.jtusci.2014.01.007.

[22] S. Rostamnia, A. Zabardasti, J. Fluorine Chem. 144 (2012) 69-72.

[23] S. Yamazaki, Bull. Chem. Soc. Jpn. 69 (1996) 2955-2959.

[24] H.J. Reich, F. Chow, S.L. Peake, Synthesis 4 (1978) 299-301.

[25] M. Tajbakhsh, E. Alaee, H. Alinezhad, M. Khanian, F. Jahani, S. Khaksar, P. Rezaee, M. Tajbakhsh, Chin. J. Catal. 33 (2012) 1517-1522.

[26] M.L. Kantam, S. Laha, J. Yadav. P. Srinivas, Synth. Commun. 39 (2009) 4100-4108.

[27] B.F. Mirjalili, A. Bamoniri, A. Akbari, N. Taghavinia, J. Iran. Chem. Soc. 8 (2011) S129-S134.

[28] B.F. Mirjalili, A. Akbari, Z. Naturforsch. B. 64b (2009) 347-350.

[29] M. Rahimizadeh, Z. Bakhtiarpoor, H. Eshghi, M. Pordel, Gh. Rajabzadeh, Monatsh. Chem. 140 (2009) 1465-1469.

[30] M. Hosseini-Sarvari, Chin. J. Chem. 22 (2011) 547-550.

[31] A. Mahyar, M.A. Behnajady, N. Modirshahla, Indian J. Chem. Sect A. 49 (2010) 1593-1600.

[32] S.V. Ingale, P.B. Wagh, A.K. Tripathi, A.S. Dudwadkar, S.S. Gamre, P.T. Rao, I.K. Singh, S.C.Gupta, J. Sol-Gel Sci. Technol. 58 (2011) 682-688.

[33] X. Zhang, F. Zhang, K.Y. Chan, Appl. Catal. A: Gen. 284 (2005) 193-198.