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Synthesis of 2,4,5-Tri substituted Imidazoles Using Nano-[Zn-2BSMP]Cl₂ as a Schiff Base Complex and Catalyst

H. Goudarziafshar*, A.R. Moosavi-Zare* and Z. Jalilian

Sayyed Jamaledin Asadabadi University, Asadabad, 6541861841, I. R. Iran

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Nano-Zn-[2-boromophenyl-salicylaldehyde-methylpyranopyrazole]Cl₂ (nano-[Zn-2BSMP]Cl₂) as a Schiff base complex and catalyst was used for the preparation of 2,4,5-tri substituted imidazoles by the one-pot multi-component condensation reaction of benzil with various aromatic aldehydes and ammonium acetate at 80 °C under solvent-free conditions.

Keywords: Multi-component reaction, Nano-[Zn-2BSMP]Cl₂, Schiff base complex, Solvent-free, 2,4,5-Tri substituted imidazole

INTRODUCTION

Imidazole derivatives are one of the most significant classes of five-membered nitrogen heterocycle compounds which are of interest for chemists due to their location in the structure of many biological and active compounds including histidine, histamine, mercaptopurine and biotin [1]. Imidazole moiety is also found in many significant drugs including losartan, olmesartan, eprosartan, miconazole, clotrimazole and trifluoromethyl [2]. Imidazoles have been considered as greener solvents in ionic liquids [3] and also in the precursors for the synthesis of stable *N*-heterocyclic carbenes in organometallic chemistry [4-7].

2,4,5-Tri substituted imidazoles were prepared through the one-pot multi-component condensation reaction of benzil with various aldehydes and ammonium acetate using various catalysts including nano copper ferrite [8] MoO₃/SiO₂ [9], trichloro isocyanuric acid (TCCA) [10], magnetic Fe₃O₄ nano particles [11], zinc(II) (tetra(4-methylphenyl)) porphyrin [12], and sulfamic acid functionalized magnetic nano particles SA-MNP [13].

Multi-component condensation reactions display a notable pattern in combinatorial chemistry due to their ability to prepare the related products with high efficiency and atomic economy by the production of structural complexity in one step from three or more starting materials [14-21].

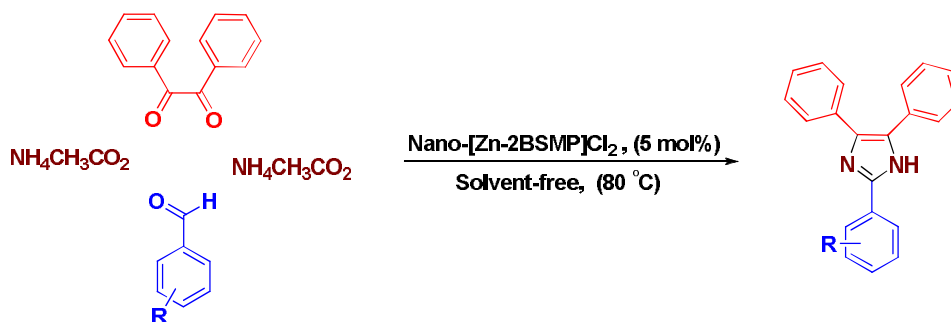
Schiff base compounds are widely reported as one of the significant classes of ligands due to their easy and convenient protocols for the preparation and their interesting coordination chemistry [22]. We have recently prepared and identified a new category of Schiff base complexes by the reaction of pyranopyrazole derivatives with salicylaldehyde and different metal salts [23-26]. In continuing our previous investigations on the synthesis of Schiff base complexes and their catalytic applications in organic transformations, we have used nano-Zn-[2-boromophenyl-salicylaldehyde-methylpyranopyrazole]Cl₂ (nano-[Zn-2BSMP]Cl₂) as an efficient complex and effective catalyst for the preparation of 2,4,5-tri substituted imidazoles (Scheme 1).

EXPERIMENTAL

General

All chemicals were purchased from Merck or Fluka Chemical Companies. The known products were identified by comparison of their melting points and spectral data with

*Corresponding authors. E-mail: moosavizare@yahoo.com; hamid_gafshar@yahoo.com



Scheme 1. The preparation of 2,4,5-tri substituted imidazoles using nano-[Zn-2BSMP]Cl₂

those reported in the literature. Progress of the reactions was monitored by TLC using silica gel SIL G/UV 254 plates.

Procedure for the Synthesis of Nano-Zn-[2-bromophenyl-salicylaldimine-methylpyrano-pyrazole]Cl₂ (Nano-[Zn-2BSMP]Cl₂)

A mixture of 2-bromobenzaldehyde (0.185 g, 1 mmol) malononitrile (0.066 g, 1 mmol), ethyl acetoacetate (0.13 g, 1 mmol) hydrazine hydrate (0.062 g, 1.25 mmol) and isonicotinic acid (0.0123 g, 0.1 mmol, 10 mol%) was added into a 25 ml round-bottomed flask connected to a reflux condenser, and stirred at 100 °C. After completion of the reaction, as monitored by TLC, the reaction mixture was cooled to room temperature. Water was added to the reaction mixture to dissolve isonicotinic acid and the aqueous layer was separated from the reaction mixture. Then, the solid residue (crude product) was triturated by a mixture of ethanol and water (19/1) to furnish 6-amino-4-(2-bromophenyl)-3-methyl-2,4-dihydropyrano[2,3-c]pyrazole-5-carbonitrile as an amine and product [27]. The prepared amine (1 mmol) and ZnCl₂ (1 mmol) were pulverized in a mortar for 10 min and then, transferred to a 25 ml round-bottomed flask containing salicylaldehyde (1.5 mmol), connected to a reflux condenser and stirred at 120 °C for 72 h. After this time, the reaction mixture was washed by ethylacetate and hexane (9/1) for three times to purify nano-[Zn-2BSMP]Cl₂ from excess salicylaldehyde (Scheme 2) [28].

General Procedure for the Synthesis of 2,4,5-Tri Substituted Imidazoles Using Nano-[Zn-2BSMP]Cl₂

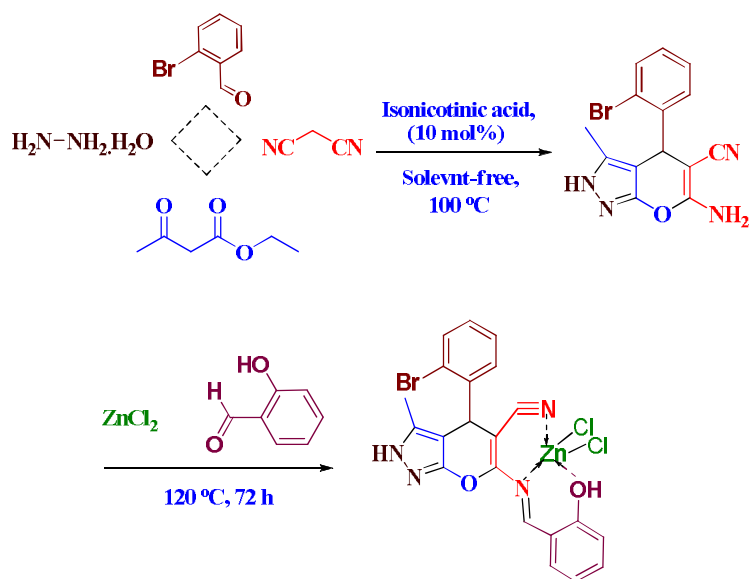
A mixture of benzyl (1 mmol), various aldehyde

(1 mmol), ammonium acetate (2 mmol) and nano-[Zn-2BSMP]Cl₂ (5 mol%) in a 25 ml round-bottomed flask connected to a reflux condenser was stirred in an oil-bath at 80 °C. After completion of the reaction, as monitored by TLC, the reaction mixture was cooled to room temperature. Then, warm ethanol (10 ml) was added in the reaction mixture and stirred for 5 min. The reaction mixture was soluble in warm absolute ethanol and separated from the catalyst by simple filtration (the catalyst was insoluble in ethanol). Finally, the desired product was purified by recrystallization of the reaction mixture from ethanol (95%).

Selected Spectral Data

2-(2-Methoxyphenyl)-4,5-diphenyl-1H-imidazole (1). White solid; m. p.: 209 °C; Yield: 92%; IR (KBr cm⁻¹): 3430, 3064, 1601, 1504, 1446, 1303, 1124, 1100, 973, 853, 764, 747, 696, 613; ¹H NMR (400 MHz, DMSO-*d*₆): δppm; 3.93 (s, 3H, -CH₃), 7.08 (t, 1H, *J* = 6.80 Hz), 7.17-7.24 (m, 2H), 7.30 (t, 2H, *J* = 7.20 Hz), 7.40-7.48 (m, 6H), 7.54 (d, 2H, *J* = 7.20 Hz), 8.05 (d, 1H, *J* = 7.60 Hz), 11.92 (s, 1H); ¹³C NMR (100 MHz, DMSO-*d*₆): δppm: 56.0, 112.0, 119.3, 121.0, 126.8, 127.5, 127.9, 128.1, 128.6, 129.0, 129.1, 129.3, 130.2, 131.7, 135.7, 136.8, 143.6, 156.4.

2-(2,5-Dimethoxyphenyl)-4,5-diphenyl-1H-imidazole (2). White solid; m. p.: 185-186 °C; Yield: 95%; IR (KBr cm⁻¹): 3422, 3354, 3061, 1604, 1525, 1443, 1310, 1122, 1070, 968, 869, 764, 743, 697, 618; ¹H NMR (400 MHz, DMSO-*d*₆): δppm: 3.80 (s, 3H), 3.89 (s, 3H), 6.97 (dd, 1H, *J* = 6.00, 3.20 Hz), 7.11 (d, 1H, *J* = 9.20 Hz), 7.23 (t, 1H, *J* = 7.20 Hz), 7.31 (t, 2H, *J* = 7.20 Hz), 7.39 (d, 1H, *J* = 7.20 Hz), 7.38-7.49 (m, 3H), 7.33-7.55 (m, 3H), 7.61 (d, 1H, *J* = 3.20 Hz), 11.91 (s, 1H); ¹³C NMR (100 MHz,



Scheme 2. The proposed structure of nano-[Zn-2BSMP]Cl₂

DMSO-*d*₆): δppm: 55.9, 56.4, 113.2, 113.8, 115.5, 119.8, 126.9, 127.6, 128.1, 128.6, 129.0, 129.1, 131.6, 135.6, 136.9, 143.4, 150.7, 153.5.

2-(2-Fluorophenyl)-4,5-diphenyl-1H-imidazole (3).

White solid; m. p.: 209 °C; Yield: 90%; IR (KBr cm⁻¹): 3153, 3061, 1603, 1503, 1441, 1324, 1126, 1073, 968, 835, 765, 739, 698, 608; ¹H NMR (400 MHz, DMSO-*d*₆): δppm: 7.24 (t, 1H, *J* = 7.20 Hz), 7.31 (s, 1H), 7.35 (s, 1H), 7.37-7.39 (m, 2H), 7.41 (s, 1H), 7.43 (s, 1H), 7.46 (d, 2H, *J* = 7.20 Hz), 7.50 (d, 2H, *J* = 6.80 Hz), 7.55 (d, 3H, *J* = 7.20 Hz), 7.99-8.03 (m, 1H), 12.57 (s, 1H); ¹³C NMR (100 MHz, DMSO-*d*₆): δppm: 116.6, 116.8, 119.0, 125.2, 127.0, 127.6, 128.3, 128.6, 129.0, 130.1, 130.8, 131.3, 135.4, 137.6, 141.3, 158.0, 160.5.

2-(3-Chlorophenyl)-4,5-diphenyl-1H-imidazole (4).

White solid; m. p.: 304 °C; Yield: 95%; IR (KBr cm⁻¹): 3312, 3065, 1604, 1503, 1443, 1323, 1073, 974, 778, 766, 698, 610; ¹H NMR (400 MHz, DMSO-*d*₆): δppm: 7.25 (t, 1H, *J* = 7.20 Hz), 7.33 (t, 2H, *J* = 7.20 Hz), 7.42-7.47 (m, 5H), 7.52-7.53 (m, 3H), 7.56 (d, 2H, *J* = 7.20 Hz), 8.06 (d, 1H, *J* = 7.60 Hz), 8.17 (t, 1H, *J* = 2.00 Hz), 12.85 (s, 1H); ¹³C NMR (100 MHz, DMSO-*d*₆): δppm: 124.1, 125.1, 127.1, 127.5, 128.3, 128.7, 128.8, 129.1, 129.2, 131.1, 131.2, 132.7, 134.0, 135.3, 137.8, 144.3, 144.4.

2-(2-Bromophenyl)-4,5-diphenyl-1H-imidazole (5).

White solid; m. p.: 206-207 °C; Yield: 96%; IR

(KBr cm⁻¹): 3421, 3064, 1602, 1503, 1446, 1322, 1071, 974, 762, 732, 695, 605; ¹H NMR (400 MHz, DMSO-*d*₆): δppm 7.24 (t, 1H, *J* = 7.20 Hz), 7.32 (t, 2H, *J* = 7.20 Hz), 7.38 (d, 1H, *J* = 7.20 Hz), 7.43-7.46 (m, 3H), 7.50-7.53 (m, 3H), 7.55-7.57 (m, 2H), 7.74 (dd, 1H, *J* = 6.00, 1.60 Hz), 7.80 (dd, 1H, *J* = 6.80, 1.20 Hz), 12.66 (s, 1H); ¹³C NMR (100 MHz, DMSO-*d*₆): δppm: 122.1, 127.0, 127.6, 128.1, 128.2, 128.5, 128.6, 129.1, 130.9, 131.4, 132.3, 132.6, 133.8, 135.6, 137.1, 144.9, 145.1.

2-(4-(Phenoxy)methyl)phenyl)-4,5-diphenyl-1H-imidazole (6).

White solid; m. p.: 241 °C; Yield: 95%; IR (KBr cm⁻¹): 3402, 3056, 1609, 1440, 1300, 1325, 1126, 1070, 970, 833, 765, 738, 696, 603; ¹H NMR (400 MHz, DMSO-*d*₆): δppm: 5.18 (s, 2H), 7.14 (d, 2H, *J* = 8.80 Hz), 7.23 (t, 1H, *J* = 7.20 Hz), 7.31 (t, 2H, *J* = 7.20 Hz), 7.36-7.39 (m, 2H), 7.43-7.47 (m, 4H), 7.49-7.52 (m, 4H), 7.56 (d, 2H, *J* = 7.20 Hz), 8.03 (d, 2H, *J* = 8.80 Hz), 12.53 (s, 1H); ¹³C NMR (100 MHz, DMSO-*d*₆): δppm: 69.7, 115.4, 123.8, 126.9, 127.2, 127.5, 128.1, 128.2, 128.3, 128.4, 128.6, 128.8, 128.9, 129.1, 131.6, 131.7, 135.8, 137.2, 137.4, 145.9, 146.0, 159.0.

RESULTS AND DISCUSSION

Nano-Zn-[2-bromophenyl-salicylaldehyde-methyl-pyranopyrazole]Cl₂ (nano-[Zn-2BSMP]Cl₂) was

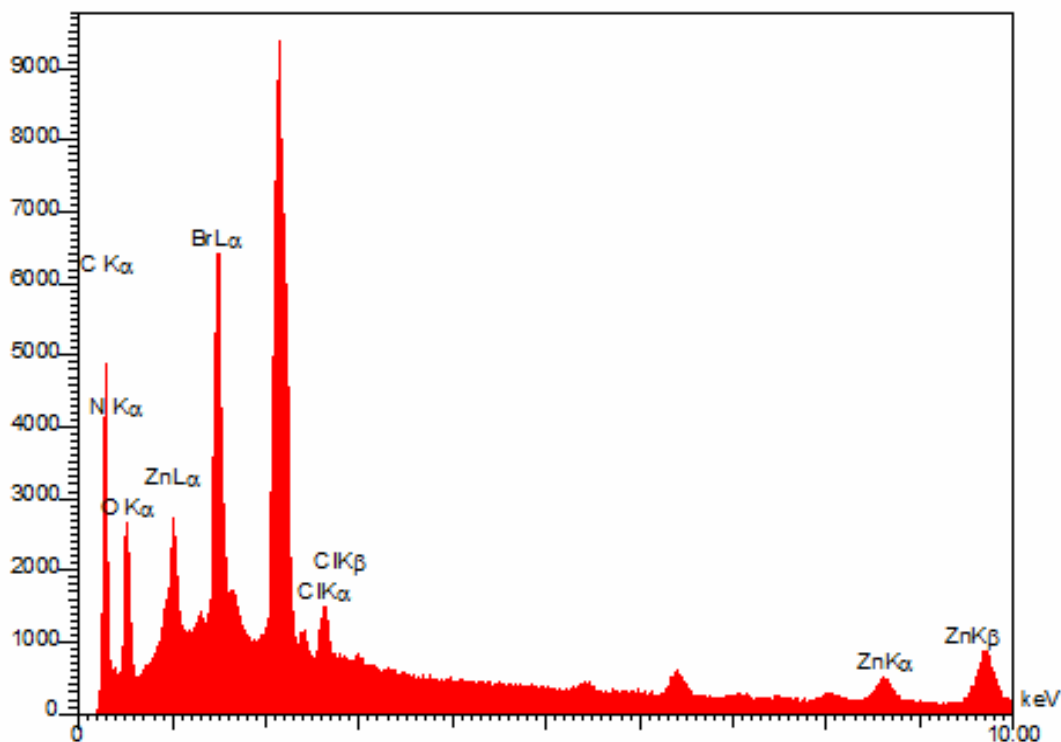


Fig. 1. Energy-dispersive X-ray spectroscopy (EDX) of nano-[Zn-2BSMP]Cl₂.

synthesized by the reaction of 2-bromobenzaldehyde with ethyl acetoacetate, malononitrile and hydrazine hydrate to furnish the corresponded pyranopyrazole according to previous literature [27]. The prepared pyranopyrazole reacted with salicylaldehyde and ZnCl₂ to obtain nano-Zn-[2-bromophenyl-salicylalimine-methylpyranopyrazole] Cl₂ (nano-[Zn-2BSMP]Cl₂) as a nano-Schiff base complex (Scheme 2) [28,29].

Based on the scanning electron microscope (SEM) micrographs of nano-[Zn-2BSMP]Cl₂, the size of particles obtained was less than 100 nanometers (Fig. 1).

Energy-dispersive X-ray spectroscopy (EDX) of nano-[Zn-2BSMP]Cl₂ displayed the presence of the desired elements, including carbon, oxygen, nitrogen, bromine, zinc and chlorine, in the structure of the catalyst. Thus, the structure of nano-[Zn-2BSMP]Cl₂ was completely supported by EDX analysis (Fig. 2).

To confirm the nanostructure of nano-[Zn-2BSMP]Cl₂, TEM measurements were performed as displayed in Fig. 3. TEM micrograph confirms the presence of particles in nano

size.

Nitrogen adsorption-desorption isotherm (BET) of nano-[Zn-2BSMP]Cl₂ is shown in Fig. 3. According to this figure, BET surface area and total pore volume of the catalyst were found to be 1.5701 m² g⁻¹ and 0.016151 cm³ g⁻¹, respectively (Fig. 4).

To optimize the reaction conditions, the reaction of benzil (1 mmol), 4-chlorobenzaldehyde (1 mmol), and ammonium acetate (2 mmol) was considered as model reaction and tested in the presence of different amounts of nano-[Zn-2BSMP]Cl₂ in a domain of 25 to 100 °C under solvent-free conditions. As shown in Table 1, the best results are achieved when the reaction is performed under 5 mol% of nano-[Zn-2BSMP]Cl₂ at 80 °C (Table 1). The model reaction was also tested in the presence of some various solvents such as H₂O, CH₃CN, toluene, dichloromethane and ethanol which has no superiority to the solvent-free conditions (Table 1).

The scope and efficiency of the reaction condition was investigated under the optimized reaction conditions. For

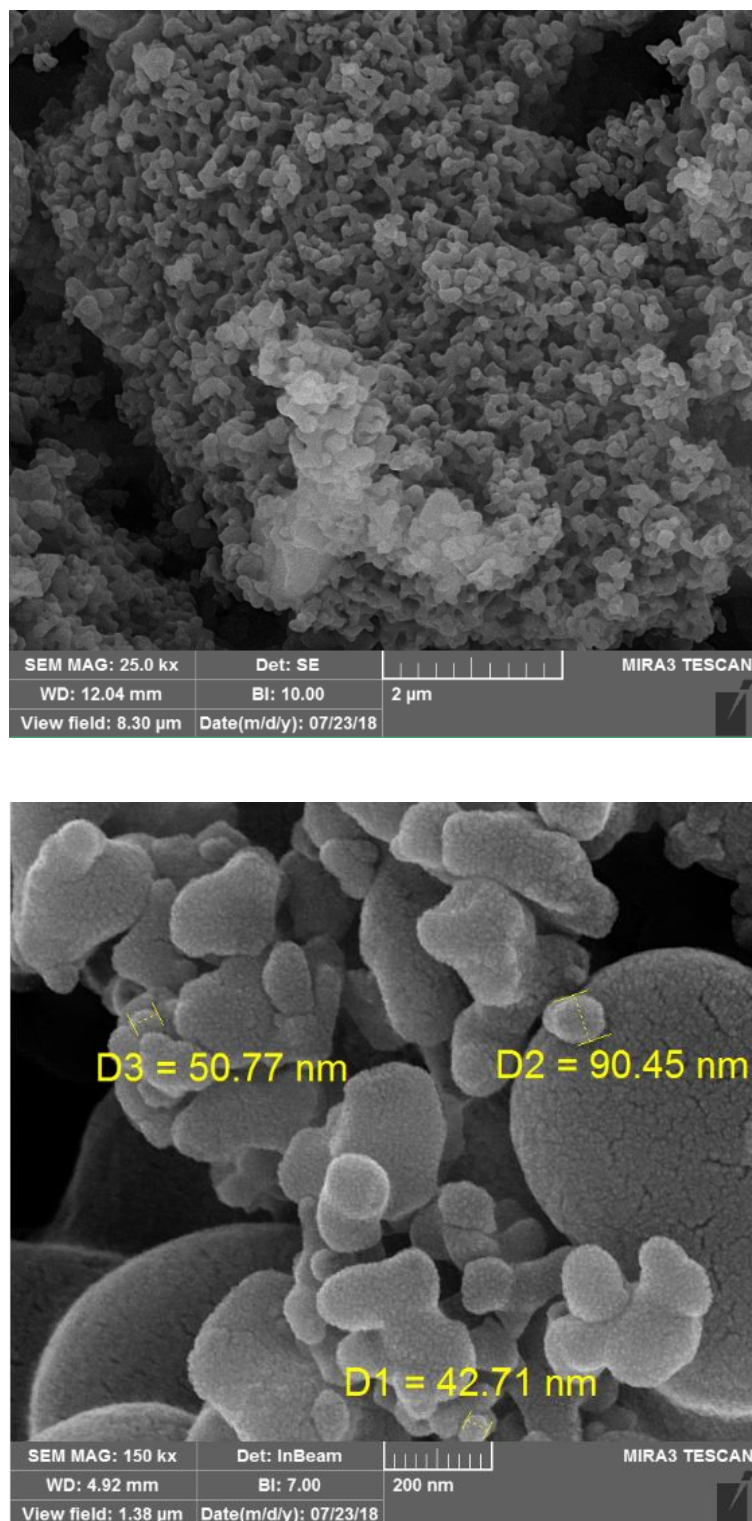


Fig. 2. SEM micrograph of nano-[Zn-2BSMP]Cl₂.

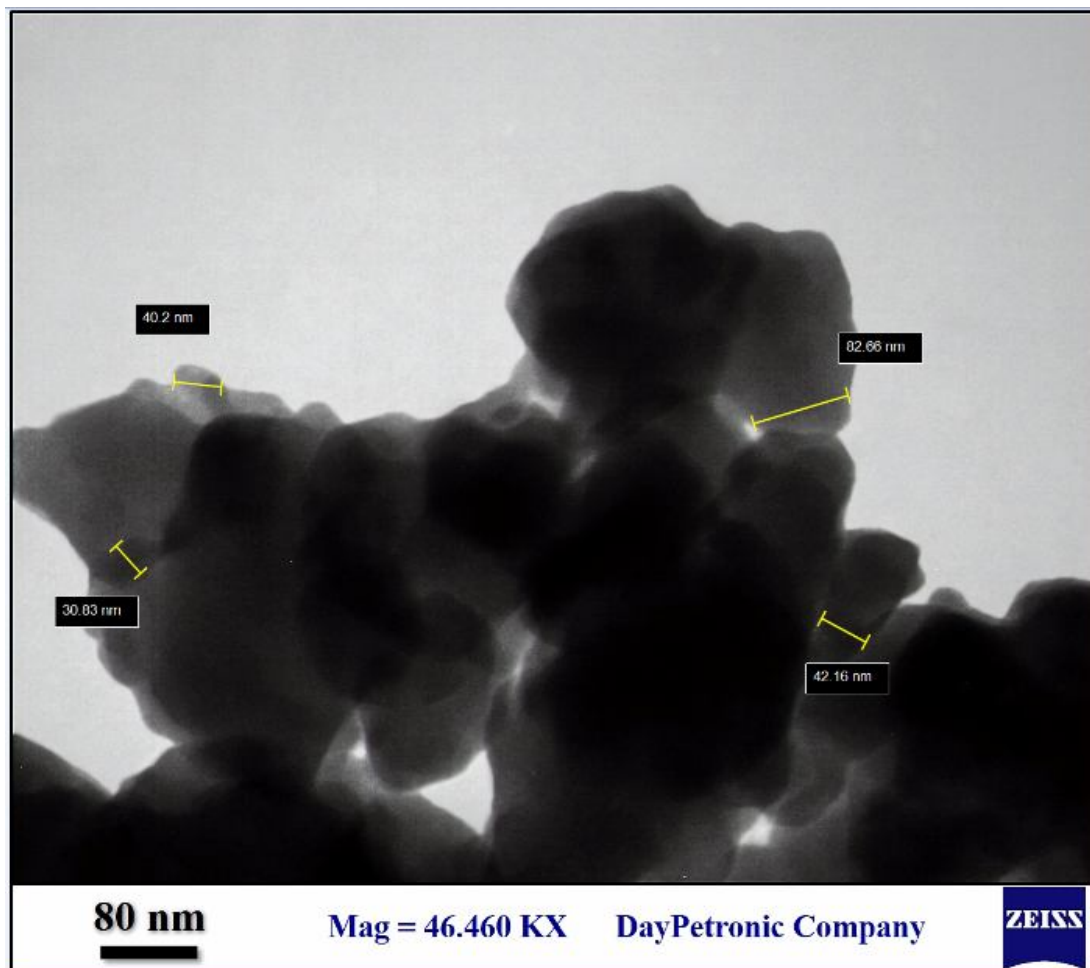


Fig. 3. Transmission electron micrographs (TEM) of nano-[Zn-2BSMP]Cl₂.

doing so, a wide range of aromatic aldehydes were successfully reacted with benzil and ammonium acetate to give the corresponded products at 80 °C. As shown in Table 2, all products were prepared with high to excellent yields in short reaction times.

In a suggested mechanism supported by previous literature [30-41], an aldehyde activated by the catalyst reacts with ammonia, resulted from ammonium acetate, to give an imine. The reaction of this imine with another molecule of ammonia results in intermediate I. In the next step, I as a diamine compound reacts with benzil to furnish II, and finally, the desired product is prepared after removing two molecules of water (Scheme 3).

Reusability of nano-[Zn-2BSMP]Cl₂ was investigated by

the reaction of 4-chlorobenzaldehyde (1 mmol), benzil (1 mmol) and ammonium acetate (2 mmol). After completion of the reaction, as monitored by TLC, warm ethanol (10 ml) was added to the reaction mixture and stirred for 5 min. The reaction mixture was soluble in warm absolute ethanol and separated from the catalyst by simple filtration (the catalyst was insoluble in ethanol). The catalyst was washed with acetone and dried to reuse it for another reaction. We observed that the catalytic activity of the catalyst was restored within the limits of the experimental errors for two successive runs (Fig. 5).

To compare the applicability and the efficiency of nano-[Zn-2BSMP]Cl₂ with some previously reported ones for the synthesis of 2,4,5-tri substituted

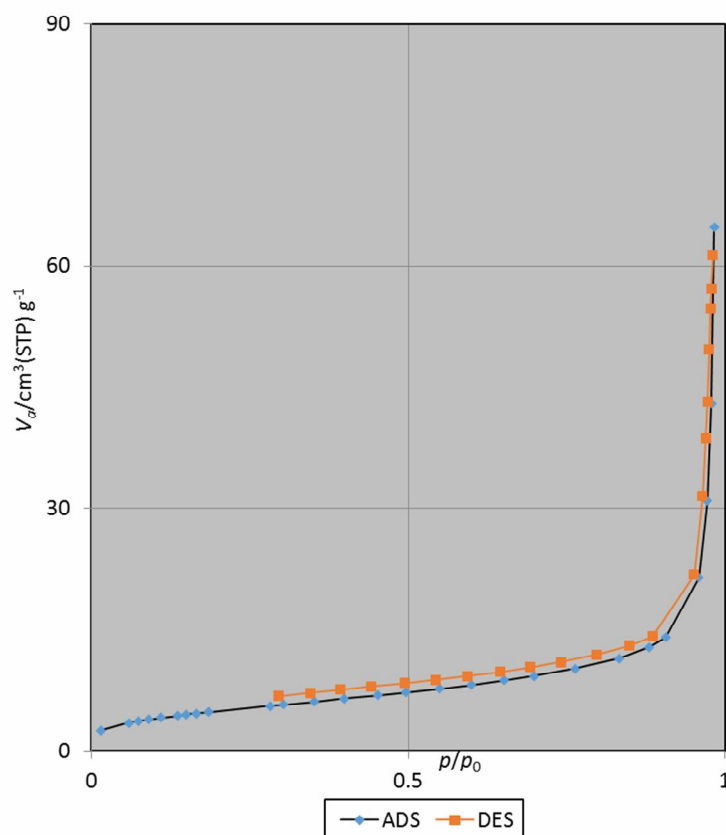

Fig. 4. N₂ adsorption/desorption isotherm of nano-[Zn-2BSMP]Cl₂.

Table 1. The Effect of Different amounts of Catalyst, Types of Solvents and Various Temperatures on the Reaction of Benzil (1 mmol), 4-Chlorobenzaldehyde (1 mmol), and Ammonium Acetate (2 mmol)

Entry	Catalyst (mol%)	Solvent	Temperature (°C)	Time (min)	Yield (%) ^a
1	-	-	80	120	25
2	3	-	80	25	80
3	5	-	80	10	95
4	8	-	80	10	80
5	10	-	80	10	82
6	5	-	25	30	40
7	5	-	100	5	78
8	5	H ₂ O	Reflux	90	-
9	5	CH ₃ CN	Reflux	40	50
10	5	Toluene	Reflux	100	10
11	5	Dichloromethane	Reflux	25	69
12	5	Ethanol	Reflux	20	95

^aIsolated yield.

Table 2. The Synthesis of 2,4,5-Tri substituted Imidazoles

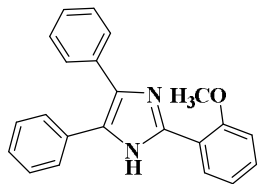
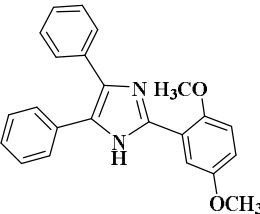
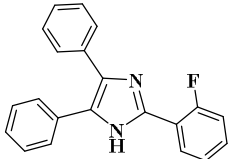
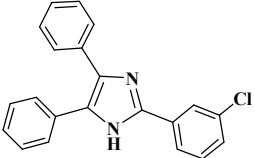
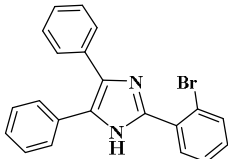
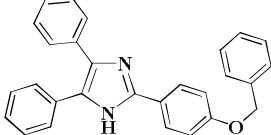
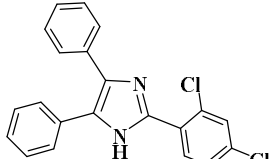
Entry	Product	Time (min)	Yield (%) ^a	M.p. (°C)	M.p. (°C) (Lit.)
1		15	92	209	[208-210] ³⁰
2		13	95	185-186	-
3		15	90	209	-
4		17	95	304	[286-288] ³¹
5		13	96	206-207	[200-202] ³¹
6		13	95	241	-
7		20	70	179-181	[178-180] ³⁰

Table 2. Continued

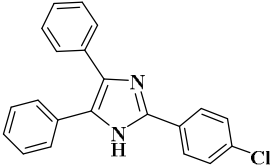
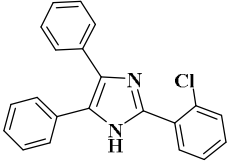
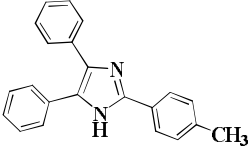
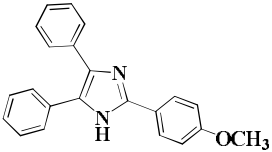
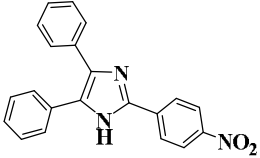
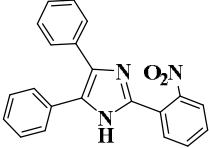
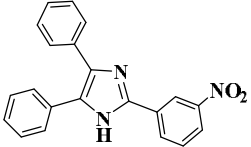
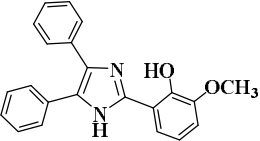
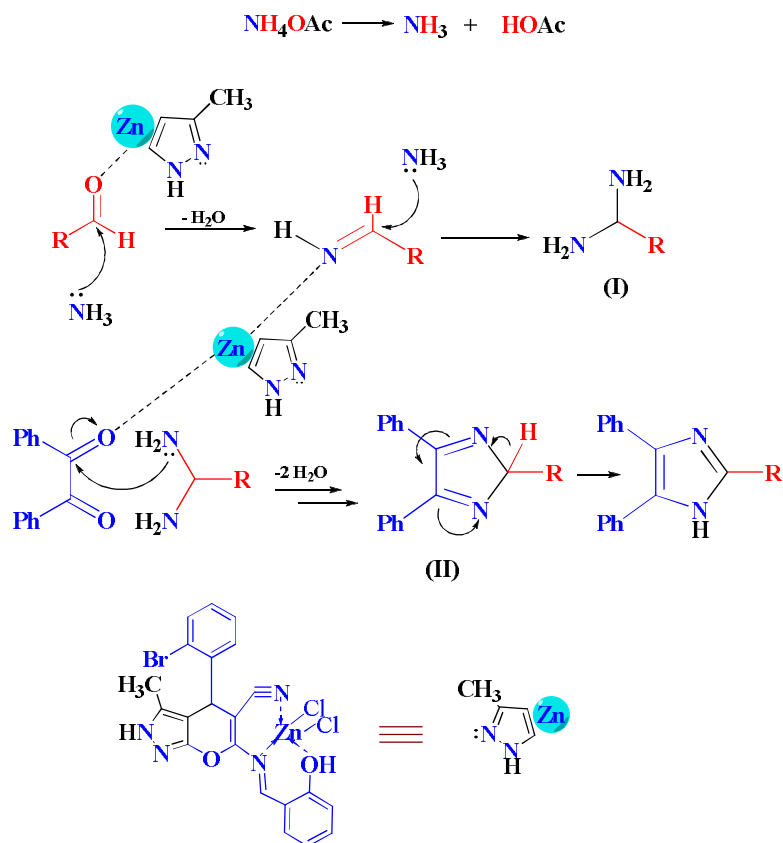
8		10	96	260-263	[261-263] ³¹
9		12	95	197-198	[195-198] ³¹
10		15	78	226-229	[231-233] ³¹
11		23	90	230-233	[229-231] ³¹
12		27	87	239-241	[240-242] ³⁰
13		30	86	227-229	[228-230] ³³
14		32	87	300-303	[301-302] ³²
15		13	80	169-170	[168-170] ³⁴

Table 2. Continued

15		13	80	169-170	[168-170] ³⁴
16		20	84	255-257	[259-260] ³⁶
17		16	93	230-233	[232-233] ³²
18		13	90	249-250	[250-252] ³²
19		13	80	245-246	[248-250] ³¹
20		20	87	198-201	[201-202] ³⁵
21		28	65	234-235	[234-236] ³⁵
22		25	70	293-295	[290-291] ³¹

^aIsolated yield.



Scheme 3. The proposed mechanism for the synthesis of 2,4,5-tri substituted imidazoles using nano-[Zn-2BSMP]Cl₂

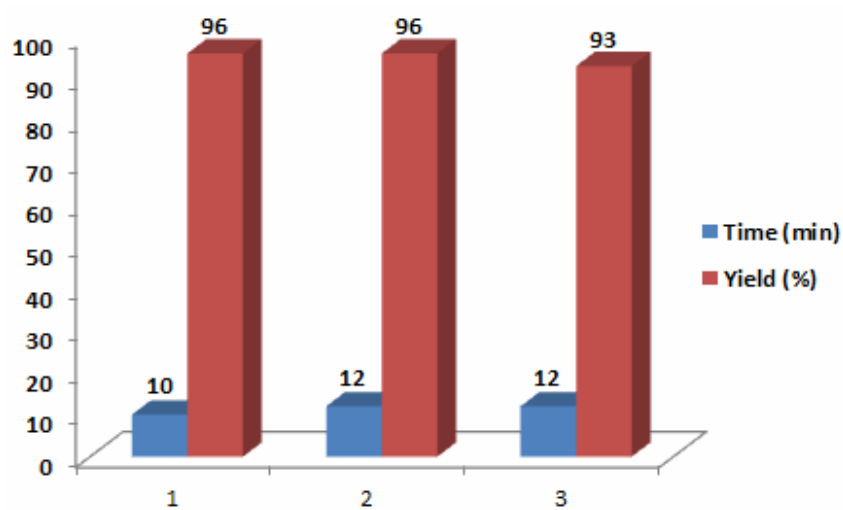


Fig. 5. The synthesis of 2-(4-chlorophenyl)-4,5-diphenyl-1H-imidazole in the presence of reused nano-[Zn-2BSMP]Cl₂ at 80 °C under solvent-free conditions.

Table 3. Comparison of the Results on the Reaction of Benzil, 4-Chlorobenzaldehyde, and Ammonium Acetate Catalyzed by Nano-[Zn-2BSMP]Cl₂ with those Obtained by the Recently Reported Catalysts

Reaction Conditions	Catalyst loading	Time (min)	Yield (%) ^a	TOF (min ⁻¹) ^b	Ref.
MgAl ₂ O ₄ , solvent-free, 120 °C	2 g	30	92	-	[37]
HNO ₃ @nano SiO ₂ , solvent-free, 100 °C	0.012 g	207	91	-	[38]
ZnO NRs, H ₂ O, reflux	20 mol%	87	83	0.047	[39]
SiO ₂ :SnO ₂ , solvent-free, 80 °C	50 mol%	20	91	0.091	[40]
(4-SB)T(4-SPh)PHSO ₄ , solvent-free, 120 °C.	15 mol%	10	98	0.653	[41]
Nano-[Zn-2BSMP]Cl ₂ , solvent-free, 80 °C	5 mol%	10	95	1.900	This work

^aIsolated yield. ^bTurn-over frequency.

imidazoles, the results of this catalyst are shown in Table 3. The results indicate that nano-[Zn-2BSMP]Cl₂ remarkably improves the preparation of 2,4,5-tri substituted imidazoles in turnover frequency.

CONCLUSIONS

In summary, we have introduced nano-[Zn-2BSMP]Cl₂ as a Schiff base complex and catalyst for the preparation of 2,4,5-tri substituted imidazoles by the one-pot multi-component condensation reaction of benzil with various aldehydes and ammonium acetate at 80 °C under solvent-free conditions.

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