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Yttrium Aluminum Garnet (YAG: $Al_5Y_3O_{12}$) as an Efficient Catalyst for the Synthesis of Benzimidazole and Benzoxazole Derivatives

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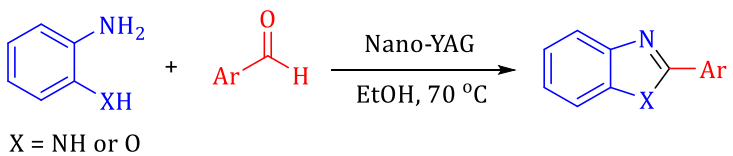
ABSTRACT

Yttrium aluminum garnet (YAG) was used to efficiently catalyzed and as an eco-friendly method and efficient catalyst for the synthesis of benzimidazole and benzoxazole derivatives by through the one-pot cyclocondensation of various aldehydes with *o*-phenylenediamines and *o*-aminophenol in ethanol at 70 °C. The present method revealed several advantages such as high yields, easy purification, mild reaction conditions, easy work-up, and short reaction times. Also, the nanoparticles (YAG) were found to be easily synthesized, cheap, air and moisture stable, heterogenic, and green catalyst.

Graphical Abstract



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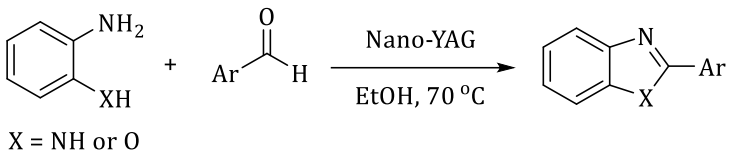
Introduction

Benzoxazole, benzimidazoles, and their derivatives are the important classes of in several fields of organic chemistry [1]. In particular, they are common heterocyclic scaffold in biologically active and medicinally significant compounds and are found in a large variety of natural products [2]. In recent years, many studies conducted on the benzimidazole derivatives because many of them have shown various spectrum of pharmacological activities including vitamin B_{12} [3], anti-ulcer, anti-tumour and anti-viral [4], anti-microbial [5], anti-cancer [6], anti-helminthic [7], anti-hypertensive [8], anti-oxidant [9], anti-tubercular [10], anti-inflammatory [11], anti-malarial [12], and selective inhibition of the platelet-derived growth factor receptor [13]. The most prominent benzimidazole in nature is *N*-ribosyl-dimethyl benzimidazole, which serves as an axial ligand for cobalt in vitamin B_{12} [14], a proton pump inhibitors [15], omeprazole, pantoprazole, and lansoprazole [16]. In recent years, several methods have reported the synthesis of benzimidazoles using various catalysts including, $TiCl_3OTf$ [17], $VOSO_4$ [18], Fe_3O_4@SiO_2/collagen [19], $Zn_3(BTC)_2$ [20], and Fe_3O_4 MNPs [21]. Usually, the condensation of *o*-phenylenediamines with aldehydes at the presence of acid [22], base or metal catalyst [23] produces the benzimidazoles. Recently, a flow chemistry protocol has been developed to synthesize benzimidazoles by condensation of *o*-phenylenediamines with aldehydes [24].

Also, benzoxazole derivatives have shown various biological and pharmacological activities including anti-tumour [25], anti-viral [26], anti-microbial [27], as non-nucleoside topoisomerase 1 poison, HIV-1 reverse transcriptase and/or DNA gyrase inhibitors [28], anti-cancer [29], and anti-biotic [30]. Their use in the field of advanced materials is also worthy of note [31]. In recent years, several methods have been reported for the synthesis of benzoxazoles using various catalysts such as CuO [32], Ni-SiO_2 [33], and PEG-SO_3H [34].

Due to the importance of the benzimidazoles, benzoxazoles, and the catalytic ability of nano-yttrium aluminum garnet (YAG) in organic reactions, we wish now report a new method for the synthesis of benzimidazole and benzoxazole derivatives at the presence of catalytic amounts of nano-yttrium aluminum garnet in EtOH at 70 °C (as shown in Scheme 1).



Scheme1. Synthesis of benzimidazole and benzoxazole derivatives

Experimental

Material and methods

All the materials were purchased from Fluka, Aldrich, and Merck and used without further purification. The products were characterized by comparison of their physical properties and spectroscopic data with those reported in the literature. Infrared (IR) spectra were recorded on KBr pellets by using a Shimadzu IR presting-21 spectrophotometer at the range of 4000–400 cm^{-1}. NMR spectra were recorded by DMSO-d_6 on a Bruker advanced DPX 400 MHz spectrometer using TMS as an internal reference. Melting points were obtained in open capillary tubes and were measured using a Buchi melting point B-540 B.V.CHI apparatus.

Synthesis of yttrium aluminum garnet (YAG)

Ppy/YAG nanocomposite was synthesized electrochemically by cyclic voltammetry in 0.1 M KCl solution containing Py monomer (0.1 M), YAG (0.5% wt) and sodium dodecyl sulfate (0.005 M) that dispersed in solution by sonication. Ppy electrode was synthesized in same solution without YAG. Electropolymerizations were conducted by 10 consecutive cycles at the sweep rate of 50 $mV.s^{-1}$ and the potentials of 0.0-1 V. The mass of Ppy films was approximated assuming a current efficiency for the electropolymerization process of 100% using Faraday's law.

General procedure for synthesis of benzimidazoles

A mixture of aldehydes (1 mmol), *o*-phenylenediamines (1 mmol) and nano-yttrium aluminum garnet (0.028 g) was heated in EtOH at 70 °C for appropriate time that indicated in Table 5. The progress of the reaction was monitored by TLC (*n*-hexane: ethyl acetate 1:2). After completion of the reaction, the mixture was washed with cold ethanol and the crude product was recrystallized by ethanol to obtain the pure benzimidazole derivatives in 82-98% yields.

General procedure for synthesis of benzoxazoles

The catalyst, nano-yttrium aluminum garnet (0.028 g), was added to a mixture of aldehydes (1 mmol), and *o*-aminophenol (1 mmol) in ethanol and and nano-yttrium aluminum garnet (0.028 g) was heated in EtOH up to at 70 °C for appropriate time as shown in (Table 6). The progress of the reaction was monitored by TLC (*n*-hexane: ethylacetate 1:2). After completion of the reaction, the mixture was washed with cold ethanol and the crude product was recrystallized by ethanol to obtain the pure benzoxazole derivatives in 84-98% yields.

The selected spectral data

2-(4-nitrophenyl)-1H-benzo[d]imidazole

M.p.: 99-101 °C, IR (KBr, υ_{max} cm^{-1}) 3347, 1635, 1540, 1448, 1367, 1225. ^{1}HNMR (400 MHz, DMSO-d_6), δ: 5.00 (brs, 1H, NH), 7.26-8.25 (m, 8H, aromatic), ^{13}CNMR (100 MHz, DMSO-d_6), δ: 115.1, 117.1, 121.6, 123.2, 128.7, 136.3, 139.3, 148.0, 152.2, 158.1, 164.3, 168.3 (Table 5, Entry 3).

4-(1H-benzo[d]imidazole-2-yl)-*N*-methylbenzenamide

M.p.: 193-195 °C, IR (KBr, υ_{max} cm^{-1}) 3371, 3325, 1749, 1542, 1500, 1150. ^{1}HNMR (400 MHz, DMSO-d_6), δ: 2.78 (s, 3H, CH_3), 4.01 (brs, 1H, NH), 6.49-7.70 (m, 8H, aromatic), ^{13}CNMR (100 MHz, DMSO-d_6), δ: 29.5, 113, 2, 114.7, 118.8, 119.1, 122.2, 123.3, 126.4, 128.1, 137.8, 147.6, 148.3, 151.8, 157.2 (Table 5, Entry 4).

2-(1H-benzo[d]imidazole-2-yl)-6-methoxyphenol

M.p.: 95-97 °C, IR (KBr, υ_{max} cm^{-1}) 3357, 3300, 1679, 1538. ^{1}HNMR (400 MHz, DMSO-d_6), δ: 3.71 (s, 3H, OCH_3), 5.01 (brs, 1H, NH), 5.08 (brs, 1H, OH), 6.56-7.70 (m, 7H, aromatic). ^{13}CNMR (100 MHz, DMSO-d_6), δ: 56.2 (OCH_3), 113.7, 114.9, 117.5, 121.3, 122.4, 123.5, 128.2, 128.5, 138.1, 153.4, 162.3, 171.0 (Table 5, Entry 5).

4-(1H-benzo[d]imidazole-2-yl)phenol

M.p.: 180-182 °C, IR (KBr, υ_{max} cm^{-1}) 3334, 3320, 1720, 1672, 1621. ^{1}HNMR (400 MHz, DMSO-d_6), δ: 5.21 (brs, 1H, OH), 5.25 (brs, 1H, NH), 6.79-7.85 (m, 8H, aromatic), ^{13}CNMR (100 MHz, DMSO-d_6), δ: 115.4, 116.8, 123.1, 127.3, 128.5, 136.4, 138.2, 151.2, 153.4, 163.5, 168.7 (Table 5, Entry 6).

2-(3,4-dimethoxyphenyl)-1H-benzo[d]imidazole

M.p.: 136-138 °C, IR (KBr, υ_{max} cm^{-1}) 3334, 1620, 1692, 1633. ^{1}HNMR (400 MHz, DMSO-d_6), δ: 3.21 (d, 6H, OCH_3), 5.03 (brs, 1H, NH), 6.72-7.80 (m, 7H, aromatic), ^{13}CNMR (100 MHz, DMSO-d_6), δ: 56.1, 56.4, 113.7, 114.9, 116.8, 123.2, 138.2, 149.1, 150.5, 153.4, 163.5, 168.7, 170.1, 172.3 (Table 5, Entry 7).

2-(4-nitrophenyl)benzo[d]oxazol

M.p.: 156-158 °C, IR (KBr, υ_{max} cm^{-1}) 1635, 1448, 1420, 1550. ^{1}HNMR (400 MHz, DMSO-d_6), δ: 7.5-8.70 (m, 8H, aromatic), ^{13}CNMR (100 MHz, DMSO-d_6), δ: 110.2, 119.1, 121.3, 122.6, 123.8, 128.5, 132.3, 141.7, 148.0, 150.7, 162.6, 173.2 (Table 6, Entry 3).

4-(benzo[d]oxazole-2-yl)-*N*-methylbenzenamide

M.p.: 79-81 °C, IR (KBr, υ_{max} cm^{-1}) 3371, 1749, 1542, 1500, 1150. ^{1}HNMR (400 MHz, DMSO-d_6), δ: 2.60 (s, 3H, CH_3), 6.44-7.20 (m, 8H, aromatic), ^{13}CNMR (100 MHz, DMSO-d_6), δ: 29.7, 112.2, 118.1, 120.3, 122.6, 123.7, 124.3, 128.5. 141.6, 150.8, 162.8, 169.2, 173.1 (Table 6, Entry 4).

2-(benzo[d]oxazole-2-yl)-6-methoxyphenol

M.p.: 192-194 °C, IR (KBr, υ_{max} cm^{-1}) 3347, 1550, 1679, 1538. ^{1}HNMR (400 MHz, DMSO-d_6), δ: 3.73 (s, 3H, OCH_3), 5.01 (brs, 1H, OH), 6.43-7.36 (m, 7H, aromatic). ^{13}CNMR (100 MHz, DMSO-d_6), δ: 55.2, 111.2, 113.1, 115.9, 117.5, 120.1, 134.3, 137.7, 141.2, 150.4, 162.0, 163.1, 168.3, 172.1 (Table 6, Entry 5).

4-(benzo[d]oxazole-2-yl)phenol

Oil, IR (KBr, υ_{max} cm^{-1}) 3343, 1620, 1635, 1623. ^{1}HNMR (400 MHz, DMSO-d_6), δ: 5.07 (brs, 1H, OH), 6.69-7.31 (m, 8H aromatic), ^{13}CNMR (100 MHz, DMSO-d_6), δ: 110.4, 112.2, 114.1, 115.9, 116.5, 118.1, 119.5, 128.2, 133.9, 158.4, 162.1, 168.9 (Table 6, Entry 6).

2-(3,4-dimethoxyphenyl)-1H-benzo[d]oxazole

Oil, IR (KBr, υ_{max} cm^{-1}) 1540, 1630, 1672, 1683. ^{1}HNMR (400 MHz DMSO-d_6), δ: 3.73 (d, 6H OCH_3), 6.88-7.36 (m, 7H, aromatic), ^{13}CNMR (100 MHz, DMSO-d_6), δ: 56.3, 111.8, 112.2, 136.5, 139.1, 141.3, 148.2, 149.1, 150.5, 158.2, 159.3, 163.4, 164.1, 171.2 (Table 6, Entry 7).

Results and discussion

In this work, we report a comprehensive study of the reactions between *o*-phenylenediamines and *o*-aminophenol with various aldehydes. To find the optimum reaction conditions, the reaction of 2-nitrobenzaldehyde (1 mmol) with 4-methyl-1,2-phenylenediamine or *o*-aminophenol (1 mmol) was performed under various conditions and different quantities of nano-yttrium aluminum garnet.

To establish the better catalytic activity of nano-yttrium aluminium garnet, we compared the reaction at the presence of nano-YAG in ethanol at 70 °C with the reaction by other catalysts (Table 1). The problems in the reported protocols such as prolonged reaction time and poor yields motivated us to develop a new rapid method offering excellent yields using a solid-phase basic green catalyst for the synthesis of the benzimidazoles and benzoxazoles.

To determine the optimum quantity of the nano-YAG, the model reaction was carried out in EtOH at 70 °C, using different quantities of nano-yttrium aluminum garnet (Table 2). The results showed revealed that 0.028 g of catalyst provided an excellent yield of product (Table 2, Entry 4).

Table 1. Evaluation of the activity of different catalysts for the synthesis of benzimidazoles and benzoxazoles

Entry	Catalyst	Benzimidazole		Benzoxazole		Ref.
		Time (min)	Yield (%)	Time (min)	Yield (%)	
1	-	180	20	180	20	-
2	Rose Bengal	120	60	-	-	[17]
3	*N,N*-Dimethylaniline/graphite	150	78	-	-	[18]
4	NH_4Cl	180	80	-	-	[19]
5	CuO	-	-	900	86	[12]
6	PEG-SO_3H	-	-	420	78	[11]
7	Ni-SiO_2	-	-	90	70	[13]
8	$Al_5Y_3O_{12}$	10	98	10	98	-

Table 2. Optimization amount of nano-yttrium aluminum garnet at 70 °C for 10 minutes

Entry	Catalyst (g)	Yield (%)
1	-	0
2	0.010	65
3	0.020	80
4	0.028	98
5	0.035	90

The above reaction was also evaluated in various solvents (Table 3), and the results indicated that, different solvents affected the efficiency of the reaction. Most of these solvents required a longer time and gave moderate yields, and the best results were obtained in ethanol (Table 3, Entry 5).

Table 3. Effect of the solvent on the synthesis of benzimidazoles and benzoxazoles using nano-YAG at 70 °C in 10 minutes

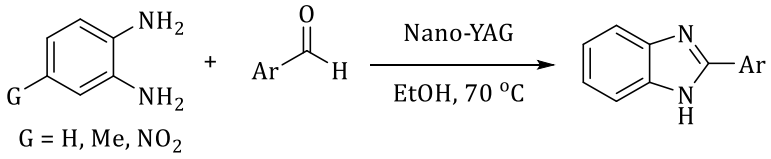
Entry	Solvent	Yield (%)	
		Benzimidazole	Benzoxazole
1	H_2O	0	0
2	EtOAc	55	30
3	$CHCl_3$	60	25
4	CH_2Cl_2	65	20
5	EtOH	98	98

To optimize the temperature in the aforementioned reaction, we have carried out a model reaction using 0.028 g of catalyst in ethanol at various temperatures (Table 4). The results clearly demonstrated that the reaction at 70 °C offered the higher yields of desired products (Table 4, Entry 4).

Table 4. Optimization of temperature using nano-YAG catalyst in EtOH

Entry	Temperature (°C)	Time (min)	Yield (%)	
			Benzimidazole	Benzoxazole
1	25	90	15	20
2	40	60	70	70
3	60	50	80	80
4	70	10	98	98

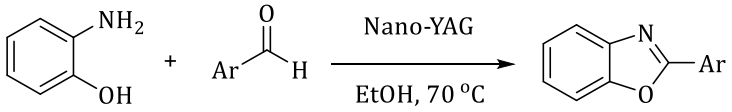
After optimization of the reaction conditions, the generality and synthetic scope of this protocol for the cyclocondensation of various aryl aldehydes with *o*-phenylenediamines and *o*-aminophenol were demonstrated under the optimal conditions (Schemes 2 and 3, Tables 5 and 6).



Scheme 2. Synthesis of benzimidazole derivatives using nano-YAG

Table 5. Reaction between aldehydes and different o-phenylenediamines catalyzed by nano-YAG

Entry	Ar	G	Yields (%)	M.p. (°C)
1	2-$NO_2C_6H_4$	H	90	99-102
		CH_3	98	188-190
		NO_2	87	133-136
2	3-$NO_2C_6H_4$	H	89	115-117
		CH_3	96	108-110
		NO_2	87	170-172
3	4-NO_2C6H_4	H	91	149-151
		CH_3	96	103-106
		NO_2	89	220-222
4	4-$NHCH_3C_6H_4$	H	85	193-195
		CH_3	91	137-139
		NO_2	84	Oil
5	2-OH-3-$CH_3OC_6H_3$	H	83	95-97
		CH_3	90	112-115
		NO_2	82	Oil
6	4-OHC_6H_4	H	87	180-182
		CH_3	94	143-145
		NO_2	91	145-147
7	3,4-$(CH_3O)_2C_6H_3$	H	88	136-138
		CH_3	92	Oil
		NO_2	83	Oil



Scheme 3. Synthesis of benzoxazole derivatives using nano-YAG

Finally, the recyclability and reusability of the catalyst was investigated for the reaction of 2-nitrobenzaldehyde with 4-methyl-1,2-phenylenediamine. After the reaction completion, the reaction mixture was filtered and the precipitate was washed with ethanol. The catalyst was recycled and washed with ethanol. After becoming air dried, the recycled catalyst could be reused as such in

subsequent experiments (up to four cycles) under the similar conditions. The yields of the product remained comparable in all experiments after 10 min (98, 96, 95, and 92 in cycles 1-4, respectively), indicating that the catalyst can be recycled at least four times with no considerable loss in its activity.

Table 6. Reaction between aldehydes and o-aminophenol catalyzed by nano-YAG

Entry	Ar	Yields (%)	M.p. (°C)
1	2-$NO_2C_6H_4$	98	92-94
2	3-$NO_2C_6H_4$	91	130-132
3	4-$NO_2C_6H_4$	90	156-158
4	4-$NHCH_3C_6H_4$	88	79-81
5	2-OH-3-$CH_3OC_6H_3$	89	192-194
6	4-OHC_6H_4	88	Oil
7	3,4-$(CH_3O)_2C_6H_3$	84	Oil

Conclusions

We have developed an efficient procedure for the synthesis of benzimidazole and benzoxazole derivatives using nano-yttrium aluminum garnet in ethanol at 70 °C. The important features of this procedure are mild reaction condition, easy work-up, high yield, green aspects such as avoiding hazardous organic solvents, toxic catalysts and waste, ease of recovery and reuse of the catalyst. The catalyst showed a good performance and can be easily retrieved from the reaction mixture and reused several times without any significant loss in its catalytic activity.

Acknowledgement

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Conflict of Interest

We have no conflicts of interest to disclose.

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