A green approach to the Rapid Synthesis of 2-Phenyl Benzoxazole and its derivatives using Magnetically Separable $Ag@Fe_2O_3$ Core-Shell Nanoparticles

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Abstract
This study aims to develop $Ag@Fe_2O_3$ core-shell nanocatalyst and investigates its catalytic efficacy towards the synthesis of 2- phenyl benzoxazole and its derivatives via one-pot condensation of substituted aromatic aldehydes and 2-aminophenol. The catalyst manifests superior catalytic performance in the synthesis of benzoxazole derivatives at room temperature. All the products were isolated with a high yield of about 88-97%. The chemical structures were confirmed by FTIR and ^{1}H-NMR spectroscopy.

This synthetic strategy offers an environment-friendly pathway by using non-toxic and biodegradable solvent, heterogeneous, magnetically recoverable and reusable catalyst and reduction of hazardous waste. The nanocatalyst was characterized by XRD, TEM, BET surface area and zeta potential.

Keywords: $Ag@Fe_2O_3$, core-shell nanoparticles, magnetically recoverable, benzoxazole, green synthesis.

Introduction

The heterocyclic compounds gained a lot of interest by the community of researchers due to its attractive features such as a broad spectrum of biological activity, a vital part of many natural products and synthetic drugs and precursor or intermediates for many of the economically important organic preparations. The scientists are making extensive efforts for facile, efficient and green methods for the preparation of a variety of heterocyclic compounds.

However, the development of a green methodology for synthetic transformations is a major challenge of chemistry. Various techniques/protocols employing easily recoverable heterogeneous catalyst, microwave, solvent-free conditions, non-toxic and biodegradable chemicals are widely used for synthesis. However, the focus is always to build a clean and efficient catalytic methodology aiming at high process performances and environmental sustainability.

In this context, nano catalysis is an emerging technique which provides the advantage of high speed, selectivity, easy separation etc. Size, shape, high surface volume ratio, morphology, composition and magnetic properties of nanoparticles improve their catalytic performance, magnetic separation and reusability[1].

Benzoxazole is one of the useful heterocyclic moieties with a variety of useful biological and therapeutic characteristics such as antibacterial[2], anticancer[3], antimicrobial[4], anti-inflammatory[5], antiparkinsons[6], antioxidants[7] and anti-allergy activities[8]. Therefore, various significant approaches were reported for the synthesis of these important building blocks.

Conventionally it is synthesized by the condensation of 2-aminophenols with aldehydes[9] orthoesters[10] carboxylic acids[11] amides[12] acyl chlorides[13]. However, most of these approaches suffer from many disadvantages such as low yield, expensive chemicals/catalyst, long time, complicated operations, harsh reaction conditions such as strong acidic conditions, excess reagents, use of toxic and hazardous solvents and co-occurrence of several side products. Hence, it is a challenge to develop the catalyst which is simple, reusable, non-toxic, economical acceptable and environment-friendly.

In recent years, remarkable attention has been given to heterogeneous catalysts for the synthesis of organic moiety due to their reactivity, economical and ecological reasons. Researchers have developed large number catalysts which can be recovered from reaction mixtures by simple methods[14]. Among that, magnetically recoverable nanomaterials are prevalently used as catalysts in various organic transformations because of the ease of separation, low toxicity, simple methodology and low-cost[15].

Various magnetic catalysts such as cobalt ferrite,[16] SO_3H functionalised $CoFe_2O_3$[17], $NiFe_2O_4$[18], $CuFe_2O_4$[19], Fe_3O_4-SO_3H[20], ferrite-chitosan[21] etc. have been explored well for the efficient synthesis of various heterocyclic scaffolds.

Here an attempt is made to develop a facile and efficient protocol for the synthesis of substituted 2- aryl benzoxazoles by using inexpensive, magnetically separable $Ag@Fe_2O_3$ core-shell nanoparticles as a catalyst via one-pot condensation of substituted aromatic aldehydes and 2-aminophenol at room temperature.

Material and Methods

All the reagents and chemicals used for the present work were of AR grade and purchased from Sigma Aldrich. All were used without any further purification.

Synthesis of catalyst: The catalyst was prepared by sol-gel auto combustion method.[22] The stoichiometric amount of ferric nitrate nonahydrate, silver nitrate and citric acid were

dissolved in a minimum quantity of deionised water and stirred for 10 min. NH_3 was added slowly with stirring till the solution became alkaline. The solution was then heated at 60ºC for one hour under magnetic stirring. The temperature was raised to 100ºC to form a gel which on further heating undergoes auto-combustion to form $Ag@Fe_2O_3$ nanoparticles.

Characterisation of catalyst: Formation of the nanoparticles was confirmed from X-ray diffraction pattern recorded using monochromatized Cu-Kα radiation on a Philips X-ray diffractometer, X'pert PRO. An external standard used is silicon for correction due to instrumental broadening. The average crystallite size was determined by using a Scherer formula. Core-shell morphology was confirmed from TEM micrographs recorded on JEOL 123 TEM fitted with a GATAN ORIUS CCD Camera.

Zeta potential was determined by Malvern Zeta Sizer. The surface area was determined by single point dynamic N_2 BET method at -150ºC with N_2 purging for one hour on SMART SORB 93 surface area analyser.

General procedure for the synthesis of benzoxazole derivatives: A reaction mixture was prepared by adding 20 mg of $Ag@Fe_2O_3$ nanoparticles in the mixture of 2-aminophenol (1.5 mmol), benzaldehyde (1.5 mmol) in 6 ml dispersion of water: ethanol (5:1). The mixture was stirred at room temperature for the given time (Table 2).

The progress of the reaction was monitored by TLC using petroleum Ether: EtOAc (4:1). After completion of the reaction, EtOAc was added to the mixture and the product was extracted into EtOAc, the organic phase was washed with H_2O and dried with $MgSO_4$. Evaporation of the EtOAc gave the crude product. The product was recrystallized using ethanol (96%, 2 ml) to get pure 2-phenyl benzoxazole derivatives. The catalyst was separated by using an external magnet. The separated catalyst was repeatedly washed with chloroform and reused for seven more cycles. $Ag@Fe_2O_3$ nanocatalyst was found to be highly efficient catalyst for the synthesis of benzoxazole derivatives with high yields (Table 1, Entries 2b -10b).

The melting points of synthesized compounds were determined by open capillary methodology and were reported without any further correction. The chemical structures of the obtained product were confirmed from Fourier Transform Infrared (FT-IR) spectra recorded on Shimadzu IR spectrophotometer and ^1H NMR spectra performed in $CDCl_3$ using TMS as a internal standard on Brucker AVANCE II 300 MHz instrument. All Proton chemical shifts (δ) are relative to tetramethyl silane (TMS, δ = 0.00) as an internal standard and coupling constants (J) are given in Hz.

2-phenyl Benz[d]oxazole (1^b): IR (KBr)/υ (cm^{-1}) 1683 (C=N), 1548, 1468 (Ar, C=C), 1249 (C-O-C); ^1H NMR ($CDCl_3$, 300 MHz, ppm) δ 8.29 (d, 2H, J = 7.6 Hz), 7.81 (d, 1H, J = 3.3 Hz), 7.61 (d, 1H, J = 3.4 Hz), 7.62-7.54 (m, 3H), 7.38 (d, 2H, J = 40 6.0 Hz)

2-(4-nitrophenyl) Benz[d]oxazole (4^b): IR (KBr)/υ (cm^{-1}) 1684 (C=N), 1587 (N-O), 1507, 1480 (Ar, C=C), 1286 (C-O-C), 1103 (C-N); ^1H NMR ($CDCl_3$, 300MHz)/δ (ppm): 8.31 (d, 2H, J = 7.6 Hz), 7.89 (d, 1H, J = 3.3 Hz), 7.70 (d, 1H, J = 3.4 Hz), 7.68-7.52(m, 3H), 7.31 (d, 2H, J = 40 6.0 Hz)

2-(2-hydroxyphenyl) Benz[d]oxazole (6^b): IR (KBr)/υ (cm^{-1}) 3063 (Ar, OH), 1608 (C=N), 1526, 1486 (Ar, C=C), 1219 (C-O-C); ^1H NMR ($CDCl_3$, 300MHz)/δ (ppm): 11.50 (1H, s, OH), 8.04 (1H, dd J= 7.63, 1.4 Hz), 7.75 (1H, m), 7.63 (1H, m), 7.40-7.47 (3H, m), 7.14-7.12 (1H, m), 7.04-7.00 (1H, m)

2-(4-methoxyphenyl) Benz[d]oxazole (9^b): IR (KBr)/υ (cm^{-1}) 1684 (C=N), 1510, 1482 (Ar, C=C), 1382 (C-H), 1246 (C-O-C); ^1H NMR ($CDCl_3$, 300MHz)/δ (ppm): 8.22-8.20 (2H, dt, J= 8.40, 2.4, 2.4 Hz), 7.74 (1H, m), 7.58 (1H, m), 7.35-7.32 (2H, m), 7.04 (2H, dt, J= 8.40, 2.5, 2.45), 3.90 (3H, s)

Results and Discussion

Morphology and structural characterization of catalyst: XRD pattern (figure 2) shows well-specified diffraction peaks corresponding to face centred cubic Ag (JCPDS 65-2871) and Fe_2O_3 (JCPDS 39-1346). The crystallite size for each peak ranged between 83.6 nm to 118.5 nm. TEM image (Figure 3) revealed the formation of core-shell nanocubes. Size of the nanocubes was found in agreement with XRD data.

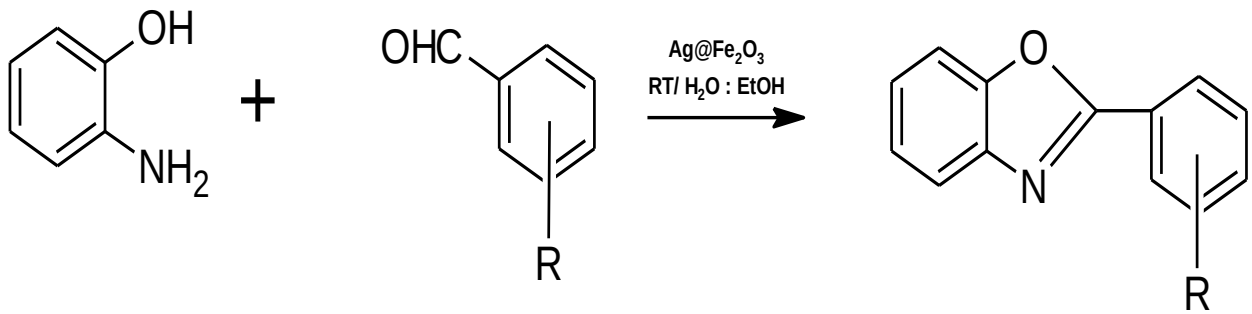
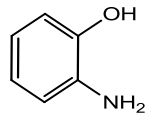
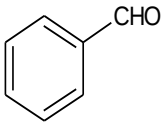
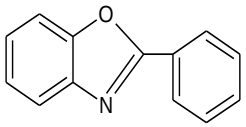
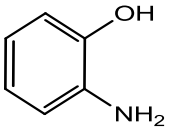
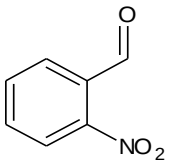
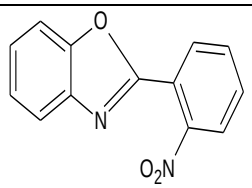
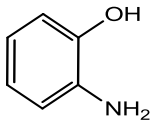
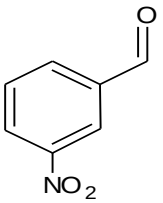
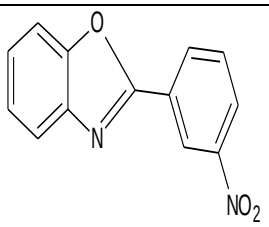
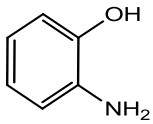
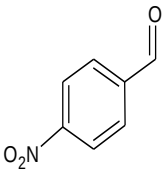
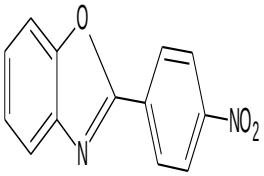
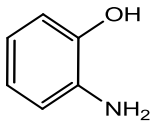
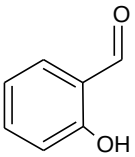
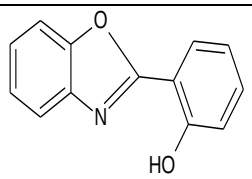
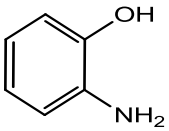
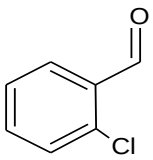
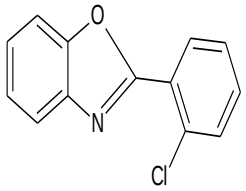
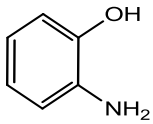
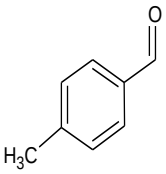
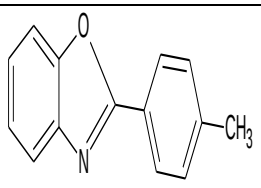
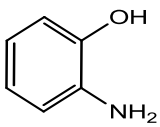
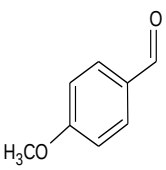
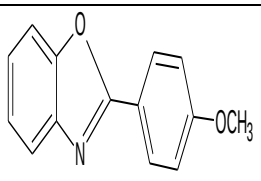
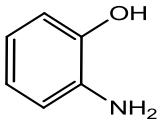
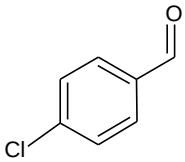
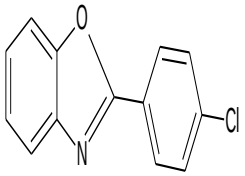
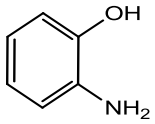
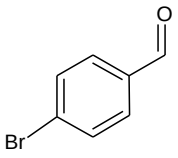
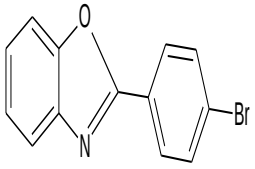
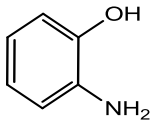
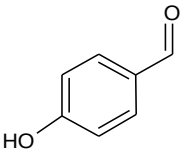
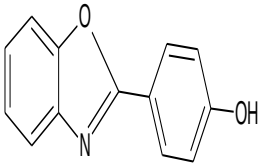


Figure 1: Synthetic Pathways for the synthesis of 2-Aryl Benzoxazoles

Table 1
Reaction between 2-aminophenol and aromatic substituted aldehydes by using $Ag@Fe_2O_3$ nanostructures as a catalyst in H_2O: EtOH solvent system at room temperature.

Entry	2-Aminophenol	Aromatic aldehyde[a]	Product[b]	$Ag@Fe_2O_3$ Nano catalyst		
				Time (Min)	Yield[c] (%)	M.P (^{O}C)
1				7	92 %	98-100
2				9	88 %	101-102
3				5	93 %	211-213
4				4	97 %	264-266
5				12	88 %	122-124
6				10	89 %	70-72
7				9	93 %	114-116
8				11	90 %	100-102

9				6	92 %	148-150
10				7	90 %	156-157
11				--	--	Sticky product

[a]The substrate was treated with aromatic aldehydes (1.5 mmol) in presence of 20 mg of nanocatalyst in C_2H_5OH: H_2O at room temperature
[b]All products were identified by comparison with authentic samples
[c]Isolated yields

Table 2
Effect of amount of catalyst on the synthesis of 2- phenyl Benzoxazole (Table 1 entry 1[b])

Entry	Amount of catalyst (mg)	Time (min)	Yield[a] %
1	5 mg	5 min	71 %
2	5 mg	10 min	71 %
3	5 mg	15 min	71 %
4	10 mg	10 min	78 %
5	15 mg	10 min	85 %
6	20 mg	7 min	92 %
7	20 mg	10 min	92 %
8	20 mg	15 min	92 %
9	25 mg	10 min	92 %
10	30 mg	10 min	92%

[a]Isolated yields

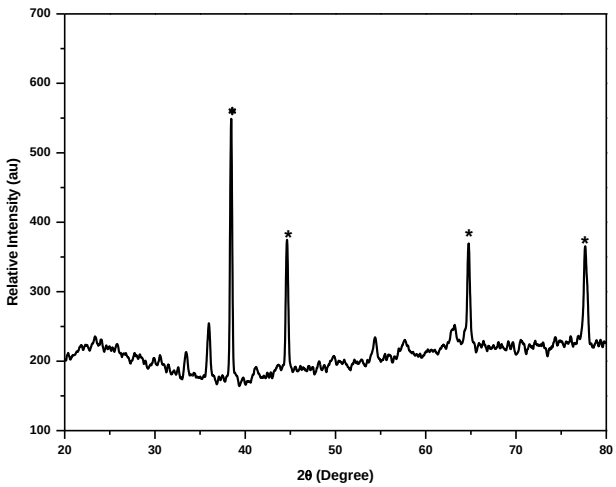


Figure 2: XRD patterns of $Ag@Fe_2O_3$ (Asterisk show the diffraction peaks of Ag)

The BET surface area analysis showed the surface area of the particles 6.93 m^2/gm. Zeta potential was found to be – 31.4mV which indicates the colloidal stability of nanocatalyst in heterogeneous catalytic reactions.

Catalytic performance of $Ag@Fe_2O_3$ core-shell nanoparticles in 2- aryl benzoxazole synthesis: Catalytic efficiency of $Ag@Fe_2O_3$ core-shell nanoparticles was studied at the various operational variables such as the amount of catalyst, reaction time, temperature and solvents. The reaction parameters were optimized by using 2-aminophenol and benzaldehyde as model starting materials (Table 1 entry 1). The catalytic amount of 20 mg was found sufficient to carry out the reaction with an excellent yield of 2-phenyl benzoxazole in minimum reaction time of 7 min. (Table 2 entry 6).

Increase in the amount of catalyst does not show any improvement in the yield. The main advantage of using nanoparticles as a catalyst that very little amount is sufficient to complete the reaction with high yield as compared to their bulk counterparts. This improved catalytic activity may attribute to the high surface area of nanoparticles.

Various organic solvents and temperature were assessed to achieve mild reaction conditions. It is observed that the catalyst performed well in all the solvents used. However, the mixture of ethanol (EtOH): water in 1:4 proportion was appeared to be the best solvent with the improved performance of reaction at room temperature (RT).

As far as heterogeneous catalytic reactions are concerned, reusability of catalysts is considered as one of the most important parameters. As $Ag@Fe_2O_3$ is a magnetic material, it can be easily isolated from the reaction mixture by using an external magnet and can be reused for the next setup. The catalyst was washed with EtOAC after every use followed by the next run with fresh reaction setup. The catalyst was consecutively reused for seven more times without any significant loss of activity (Table 4) which indicates that $Ag@Fe_2O_3$ catalyst has very good stability and recyclability

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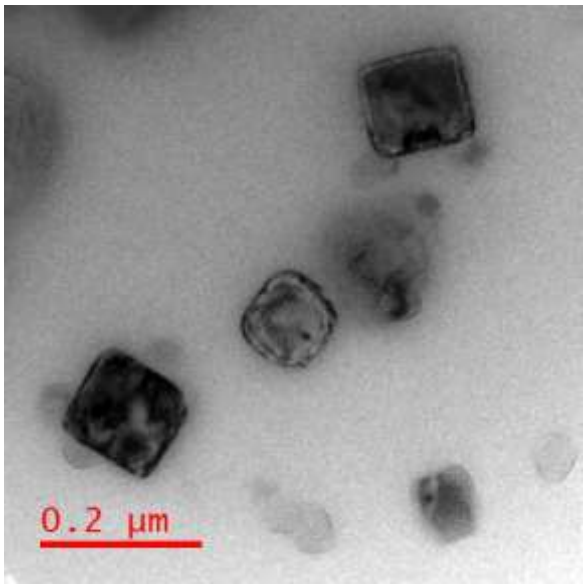

Figure 3: TEM image of $Ag@Fe_2O_3$

Table 3
Effect of various solvent and temperature on synthesis of 2- phenyl Benzoxazole (Table 1 entry 1[b])

Entry	Solvent	Time (min)	Temperature (°C)	Yield[a] (%)
1	$CHCl_3$	22	RT	92
2	CH_3CN	15	RT	91
3	CH_2Cl_2	14	RT	89
4	H_2O	14	RT	72
5	H_2O	14	60^0C	72
6	EtOH	9	RT	90
7	EtOH: H_2O (1:5)	07	RT	92
8		10	60^0C	92
9		15	60^0C	92
10		20	60^0C	92

[a]Isolated yields

Basis on the results mentioned, a possible mechanism for the synthesis of 2- substituted benzoxazole is illustrated in fig. 4.

Comparison of catalytic activity of $Ag@Fe_2O_3$ with other reported nanocatalysts: The catalytic activity of magnetic $Ag@Fe_2O_3$ nanoparticles was compared with some of the earlier reported nanocatalysts (Table 5). It was found that $Ag@Fe_2O_3$ core-shell nanoparticles are excellent with respect to the yield and the reaction conditions. Facile and efficient synthesis using benign solvent at room temperature and a catalytic amount of $Ag@Fe_2O_3$ catalyst make this approach green and environment friendly.

Table 4
Study of $Ag@Fe_2O_3$ catalyst reusability model reaction

Entry	Reaction time	Yield[a]
1	07 min	92 %
2	07 min	92 %
3	07 min	90 %
4	07 min	89 %
5	07 min	89 %
6	07 min	87 %
7	07 min	87 %

9.[a]Isolated yields

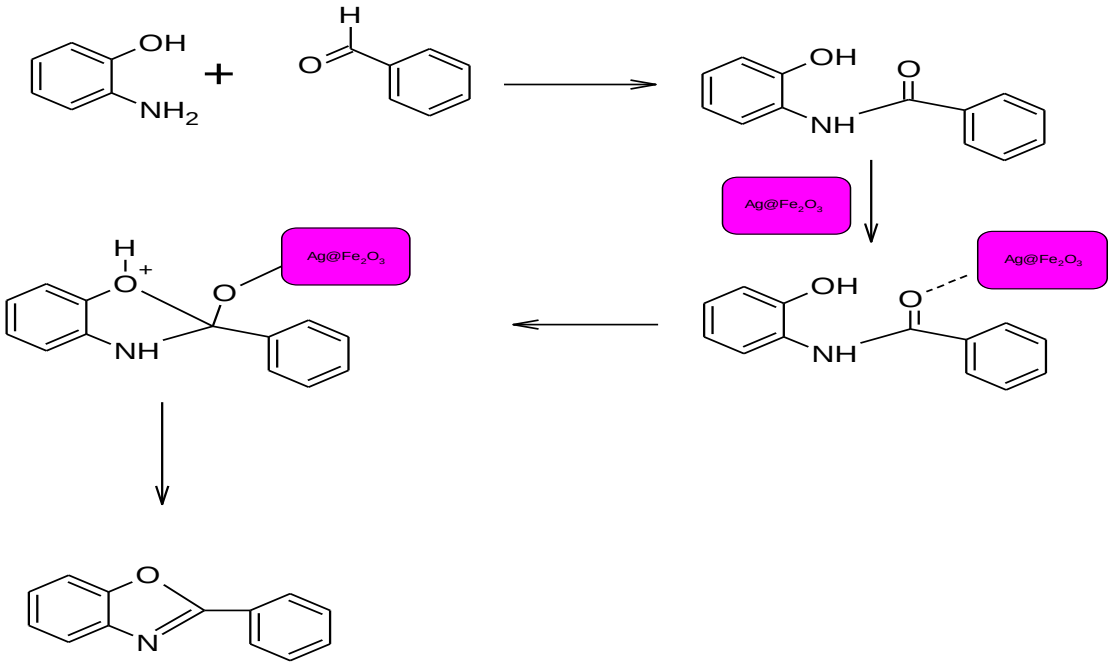

Figure 4: Plausible mechanism for synthesis of Benzoxazole by using $Ag@Fe_2O_3$

Table 5
Comparative data for the synthesis of 2-aryl Benzoxazole derivatives in presence of various nano catalysts

S.N.	Catalyst	Catalyst loading	Solvent	Reaction conditions	Time	Yield %
1	KCN/MWCNT[23]	0.02mg	DMF	ultrasonication	10s	95%
2	Schiff base/SBA-15[24]	0.01mg	H_2O	60^{O}C Reflux	3h	92%
3	CuO/SiO_2[25]	10 mol %	MeOH	RT	4h	93%
4	CuNiFe[26]	0.025mg	Toulene / KOH	130 ^{O}C	24h	94%
5	$CuFe_2O_4$[18]	20 mol %	Toulene	110 ^{O}C	2h	94%
6	Nano CeO_2[27]	10 mol %	H_2O	RT	20min	96%
7	$Ag@Fe_2O_3$*	0.02 mg	H_2O/EtOH (5:1)	RT	7min	92 %

*Present work

Conclusion

This study proposes an effective, facile, inexpensive and green approach for the synthesis of 2- substituted benzoxazoles by condensation between 2-amino phenols with various aromatic aldehydes in water: EtOH dispersion at room temperature under mild conditions. The remarkable feature of this protocol is an easy magnetic recovery of catalyst, reusability more than seven runs which is favourable from the point of view of environmental protection.

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