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REGULAR ARTICLE

Preparation and application of a magnetic organic-inorganic hybrid nanocatalyst for the synthesis of α-aminonitriles

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Abstract. This article is the first report of the catalytic application of copper ferrite-coated chitosan in organic reactions as a bio-nanocomposite. $CuFe_2O_4$/chitosan was used as a hybrid nanocatalyst for the multicomponent Strecker synthesis of α-aminonitriles by using aryl aldehydes, trimethylsilyl cyanide (TMSCN) and aromatic amines at room temperature in ethanol as a green solvent. The catalyst was characterized by Fourier transform infrared spectroscopy (FT-IR), thermogravimetric analysis (TGA), field-emission scanning electron microscopy (FE-SEM) and energy-dispersive X-ray spectroscopy (EDX) analyses. The nanocatalyst was recovered and reused several times without significant loss of catalytic activity. The organic products were obtained easily without need for column chromatography in good-to-excellent yields.

Keywords. Bionanocomposite; magnetic nanocatalyst; chitosan; α-aminonitriles; green chemistry.

1. Introduction

Due to noteworthy role of catalysis in chemical processes, either in industry or science, nanocatalysts are highly important in this field.[1] Multicomponent reactions (MCRs) are one of the most effective strategy in the field of green chemistry; which is the utilization of a set of principles that reduces or eliminates the use of or generation of hazardous substances in the design, manufacture and application of chemical products.[2] Therefore, by application of the nanocatalysts in MCRs, chemical synthesis can approach aims of green and sustainable chemistry. Magnetic nanocatalysts as a subdivision of catalyst are an interesting category to design new strategy in green chemistry. On the other hand, chitosan as a natural polysaccharide-based biopolymer has attracted considerable recent attention in various chemical, medicinal and industrial applications because of diverse properties such as biodegradability, biocompatibility, non-toxicity and so on.[3,4] It is used in many organic syntheses as a catalyst.[5–8] Supporting chitosan with a magnetic nanocatalyst enhances its properties to yield green catalysts known as bionanostructures.[9]

α-Amino nitriles are an interesting and useful class of intermediates for the synthesis of various nitrogen-containing heterocycles, α-amino acids, amides and diamines.[10,11] The Strecker reaction as the first MCR between an aldehyde an amine and hydrogen cyanide is widely regarded in organic chemistry for the synthesis of α-aminonitriles.[12] They are very useful precursors for the synthesis of α-amino acids,[13,14] thiadiazoles and imidazoles[15,16] and other biologically useful molecules.[17] Recently, to improve the synthetic procedure, many catalysts have been introduced for the synthesis of α-aminonitriles such as lanthanum(III)-binaphthyl disulfonate,[18] nanocrystalline magnesium oxide,[19] BINOL-phosphoric acid,[20,21] N-heterocyclic carbene (NHC)-amidate palladium(II) complex,[22] $Yb(OTf)_3$-pybox,[23] K_2PdCl_4,[24] N,N-dimethylcyclohexylamine,[25] superparamagnetic iron oxide[26] and ionic liquid[27] under various conditions. However, most of these methods require expensive reagents, long reaction times, harsh reaction conditions and tedious work-up procedures and give unsatisfactory yields. Considering these facts, there is still a need to introduce new, efficient, eco-friendly and inexpensive catalysts for this reaction.

In continuation of our interest in the application of new catalysts in organic synthesis *via* MCRs,[28] herein, an efficient and selective synthesis of α-aminonitrile derivatives **4a–l** were carried out by using various aromatic aldehydes **1**, amines **2** and trimethylsilyl cyanide **3** in the presence of a catalytic amount of magnetically recoverable chitosan-supported $CuFe_2O_4$ nanoparticles ($CuFe_2O_4$/chitosan) in ethanol at ambient temperature in high yields (Scheme 1). The nanocatalyst can be recovered easily and reused without any significant loss of the catalytic activity. To the best of our

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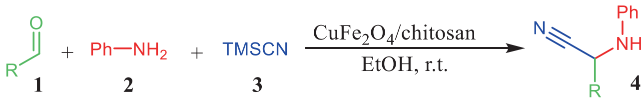

Scheme 1. $CuFe_2O_4$/chitosan-catalyzed synthesis of α-aminonitrile derivatives **4a–l**.

knowledge, this is the first report on the application of $CuFe_2O_4$/chitosan as a catalyst in organic reactions.[29]

2. Experimental

2.1 *General*

All the solvents, chemicals and reagents were purchased from Merck, Fluka and Aldrich. Melting points were measured on an Electrothermal 9100 apparatus and are uncorrected. IR spectra were recorded on a Shimadzu IR-470 spectrometer by the method of KBr pellets. ^1H NMR (500 MHz) and ^{13}C NMR (125 MHz) spectra were recorded in $CDCl_3$ solution with a Bruker DRX-500 Avance spectrometer. The magnetic property was measured on VSM-AGFM (Meghnatis Daghigh Kavir Co., Iran) vibrating sample magnetometer at room temperature. FE-SEM images were obtained with a Seron AIS 2100. EDX spectra were recorded with a Numerix DXP–X10P. The products are known and were identified by comparison of their spectroscopic and analytical data with those of authentic samples.

2.2 *Preparation of $CuFe_2O_4$/chitosan nanocomposite*

$CuFe_2O_4$/chitosan nanocomposite was prepared by a two-step process. At first, $CuFe_2O_4$ magnetic nanoparticles were synthesized *via* reported method described in the literature.[30] Briefly, $CuFe_2O_4$ nanoparticles were prepared by thermal decomposition of $Cu(NO_3)_2$ and $Fe(NO_3)_3$ in water in the presence of sodium hydroxide. $Fe(NO_3)_3.9H_2O$ (3.34 g, 8.2 mmol) and $Cu(NO_3)_2.3H_2O$ (1 g, 4.1 mmol) were dissolved in 75 mL of distilled water. Then, 3 g (75 mmol) of NaOH dissolved in 15 mL of water was added to metal ion solution at room temperature during 10 min, meanwhile a reddish-black precipitate was formed. Then, the reaction mixture was warmed to 90°C and stirred under ultrasonic irradiation for 2 h and then it was cooled to room temperature. The obtained magnetic nanoparticles were separated by an external magnet. It was washed with distilled water (3 × 30 mL) and it was kept in air oven overnight at 80°C then it was ground in a mortar-pestle and kept in a furnace at 700°C for 5 h (step up temperature at 2°C/min) and then cooled to room temperature. At the next step, an aqueous solution of 1.5% chitosan and 1% acetic acid was employed. Next, the synthesized ferrite nanopowder (20 wt%) was added gradually to a chitosan polymer solution and stirred at room temperature for 6 h. After that, ammonia solution was added dropwise to neutralize the solution. The resulted gel separated from the reaction mixture by a permanent magnet, washed several times with distilled water, and vacuum dried at 50°C during 12 h to obtain the chitosan-supported magnetite nanoparticles.

2.3 *General procedure for the synthesis of 2-(N-anilino)-2-(4-chlorophenyl) acetonitrile **4a***

A mixture of 4-chlorobenzaldehyde (0.140 g, 1 mmol), aniline (0.093 g, 1 mmol) and TMSCN (0.120 g, 1.2 mmol) in 3 mL of EtOH was stirred for 15 min at ambient temperature in the presence of $CuFe_2O_4$/chitosan nanocomposite (0.020 g). After completion of the reaction, as indicated by TLC (ethyl acetate–n-hexane, 1:3), the catalyst was removed easily by adsorbing on to the magnetic stirring bar when the stirring was stopped. Then, the solution was filtered off and the filtrate was evaporated under reduced pressure to afford the pure product. Further purification was followed by crystallization from ethanol to give pale yellow crystalline product.

3. Results and Discussion

3.1 *Characterization of the nanocatalyst*

3.1a *FT-IR Analysis*: The FT-IR spectrum of the synthesized nanoparticles is shown in Figure S1 in Supplementary Information. The spectrum of $CuFe_2O_4$ shows absorption at 580 cm^{-1} for Fe–O band, the intensive broad absorption at 3400 cm^{-1} represents the stretching mode of H_2O molecules and OH groups. The absorption band at 1630 cm^{-1} refers to the vibration of remainder H_2O in the sample and less intense absorption band at 1062 cm^{-1} indicates strong hydrogen bridges. FT-IR spectrum of $CuFe_2O_4$/chitosan, broad absorption band at 3100–3450 refers to stretching band of OH and NH groups of chitosan that bind to copper ferrite nanoparticles. It is also due to O–H of nanoparticles. The absorption bands around 1076 cm^{-1} refer to the stretch vibration of C–O bond and the 599 cm^{-1} band represents the Fe–O group of $CuFe_2O_4$.

3.1b *EDX analysis*: EDX analysis was performed for determination of the elements constitutes catalyst (see Figure S2 in Supplementary Information). As it can be seen in the figure, there is mainly C, Fe, Cu and O atoms in the nanocomposite.

3.1c *FE-SEM analysis*: FE-SEM images are used to investigate the surface structure of the nanocomposite. As seen in Figure 1, the distribution of the nanoparticles on the chitosan surface is obvious.

3.1d *Thermal stability analysis*: Thermogravimetric (TG) curve of $CuFe_2O_4$/chitosan shows first weight loss at about 100°C due to evaporation of adsorbed water in the sample (Figure 2). TG curve shows no weight loss up to about 300°C. Nanocomposite is thermally stable and suitable for organic reactions.

3.2 *Application of the nanocatalyst in organic synthetic reaction*

In this work, α-aminonitrile derivatives were synthesized by the reaction of different aromatic aldehydes with aniline and trimethylsilyl cyanides in the presence of a catalytic amount of magnetically recoverable chitosan supported $CuFe_2O_4$ nanoparticles ($CuFe_2O_4$/chitosan) in ethanol at ambient temperature. The work-up procedure of the product was easy as the nanocatalyst can be separated simply by an external magnet. To optimize the reaction conditions, a series of experiments was performed with variation of different reaction parameters such as solvent and amount of catalyst for a representative condensation of 4-chlorobenzaldehyde (1 mmol), aniline (1 mmol) and TMSCN (1.2 mmol). The results are summarized in Table 1. The best results were obtained by carrying out the reaction at room temperature in ethanol for 15 min using 20 mg of $CuFe_2O_4$/chitosan as the catalyst.

After determining the optimum reaction conditions, the investigation proceeded by performing the reaction between a series of electron rich and deficient aromatic aldehydes, aniline and trimethylsilyl cyanides. To show the common applicability of this method, numerous aldehydes were competently reacted under similar conditions. The results of this study are summarized in Table 2. It was indicated that both electron deficient and electron rich aromatic aldehydes yielded a wide range of α-aminonitrile derivatives in good to excellent yields.

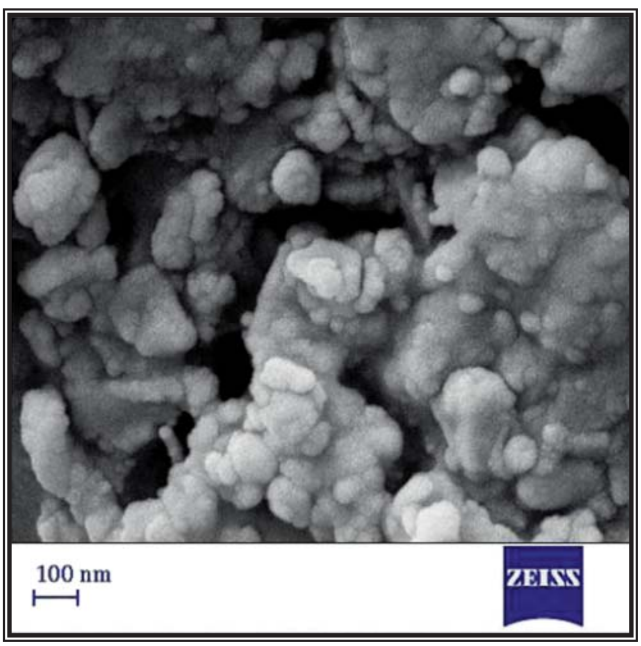

Figure 1. FE-SEM image of $CuFe_2O_4$/chitosan nanocomposite.

Table 1. Optimization of reaction conditions in the reaction of 4-chlorobenzaldehyde, aniline and TMSCN at room temperature.

Entry	Catalyst (g)	Solvent	Time (min)	Yield (%)[a]
1	–	EtOH	60	Trace
2	0.004	EtOH	15	78
3	0.008	EtOH	15	82
4	0.01	EtOH	15	90
5	0.02	EtOH	15	94
6	0.03	EtOH	15	94
7	0.02	H_2O	15	50
8	0.02	CH_3CN	15	80
9	0.02	–	15	75

[a] Isolated yield.

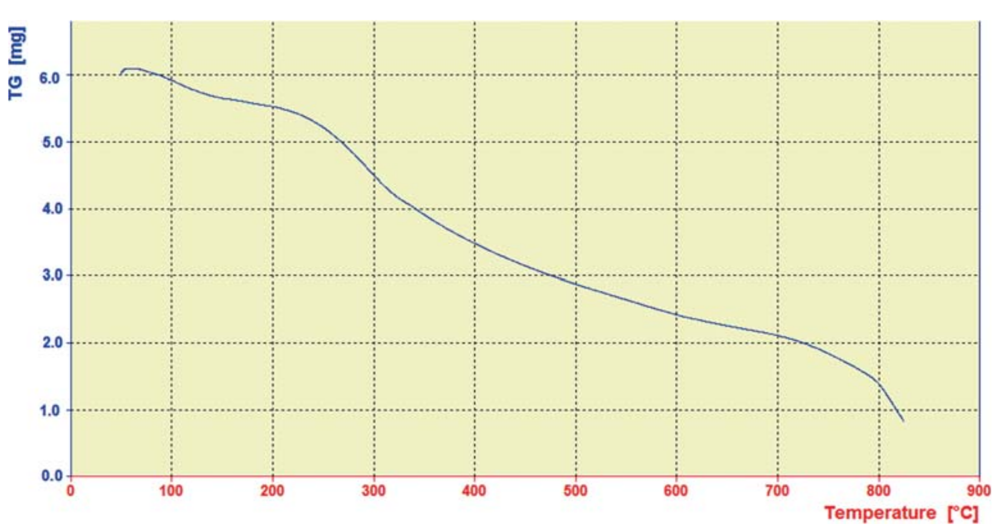

Figure 2. TGA analysis of $CuFe_2O_4$/chitosan nanocomposite.

Table 2. Synthesis of α-aminonitriles **4a–l** by using $CuFe_2O_4$/chitosan at room temperature.

Entry	R	Product	Time (min)	Yield[a] (%)	M.p. (°C) Observed	M.p. (°C) Literature
1	4-Chlorophenyl	4a	15	94	111–112	108–110[31]
2	4-Bromophenyl	4b	15	94	100–101	101–103[32]
3	Phenyl	4c	15	85	73–75	75–76[31]
4	4-Methoxylphenyl	4d	15	91	90–91	92–94[32]
5	3-Nitrophenyl	4e	15	93	89–90	90–91[33]
6	4-Hydroxyphenyl	4f	15	88	124–125	120–122[34]
7	2-Chlorophenyl	4g	15	90	70–72	63–66[35]
8	4-Methylphenyl	4h	15	92	70–71	72–74[35]
9	Cyclohexyl	4i	15	86	75–76	74–76[36]
10	2-Nitrophenyl	4j	15	91	135–137	132–133[36]
11	3-Hydroxyphenyl	4k	15	87	Oil	Oil[36]
12	4-Nitrophenyl	4l	15	94	86–88	90–92[37]

[a] Isolated yield.

3.3 *Catalyst recycling*

The possibility of recycling the catalyst for several times was examined under optimized conditions. It has been shown that $CuFe_2O_4$/chitosan could be recovered and reused several times in subsequent runs using the same recovered catalyst without a considerable loss of catalytic activity. The isolated yields in five subsequent runs were 94, 91, 89, 87 and 87%, respectively (see Figure S5 in Supplementary Information).

4. Conclusions

In summary, for the first time, we have introduced copper ferrite-coated chitosan as a magnetic organic-inorganic hybrid nanocatalyst for the multicomponent synthesis of organic compounds. Then, efficient and selective synthesis of α-aminonitrile derivatives were carried out by using a variety of aromatic aldehydes, aniline and trimethylsilyl cyanides in the presence of a catalytic amount of the magnetically recoverable chitosan-supported $CuFe_2O_4$ nanoparticles in ethanol at room temperature in high yields.

Supplementary Information (SI)

Additional experimental data and spectroscopic characterization data are given in the Supplementary Information which is available at www.ias.ac.in/chemsci.

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