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An efficient, one-pot synthesis of 2-amino-4H-chromenes catalyzed by (α-Fe_2O_3)-MCM-41-supported dual acidic ionic liquid as a novel and recyclable magnetic nanocatalyst

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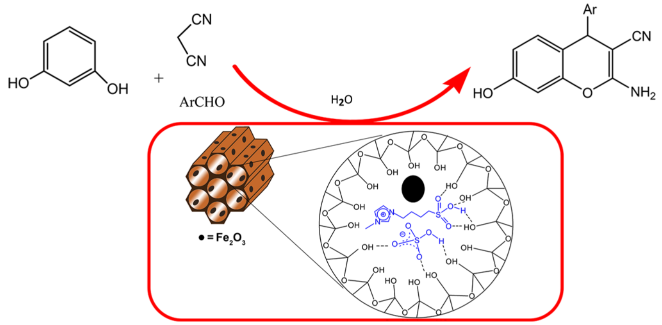
Abstract A novel (α-Fe_2O_3)-MCM-41-supported dual acidic ionic liquid (DAIL) catalytic system was prepared. This catalyst was characterized by XRD, TEM, SEM, IR spectra and N_2 adsorption–desorption measurements, TGA and acid–base titration. 1-Methyl-3-(4-sulfobutyl) imidazolium hydrogen sulfate as the DAIL inside the mesochannels of α-Fe_2O_3-MCM-41 led to preparation of a new material which was an efficient magnetic heterogeneous nanocatalyst for one-pot three-component synthesis of some new and known 2-amino-4H-chromenes in water medium conditions. This catalyst was more efficient than other reported catalysts for the synthesis of 2-amino-3-cyano-7-hydroxy-4H-chromenes. High product yields, short reaction times, use of non-corrosives, a robust and reusable magnetic nanocatalyst and water as a green solvent are the most important features of the present work.

Graphical Abstract



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Keywords Magnetically reusable nanocatalyst · (α-Fe_2O_3)-MCM-41-DAIL · Dual acidic ionic liquid · 2-Amino-4H-chromene · One-pot three-component reaction

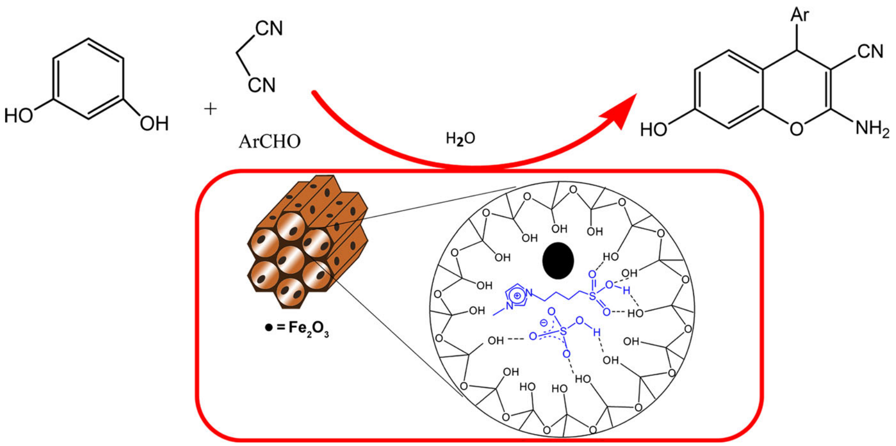
Introduction

In DAILs, the existence of both Brønsted acidic functional groups and acidic counter anions can enhance their acidities [1, 2]. Therefore, these types of DAILs have recently been prepared by several groups of researchers and applied in various organic reactions as catalysts [3]. However, the use of heterogeneous catalysts, due to conveniences in catalyst separation and recovery, is mostly preferred. On the other hand, most ionic liquids (ILs) are highly viscous, which often limits mass transfer during catalytic reactions. To overcome these problems, supported ILs have been developed [4]. Mesoporous materials such as MCMs (Mobil Composition of Matter) possess unique structural features such as high surface area, large pore volume, uniform porosity, and excellent thermal, chemical and mechanical stability. Therefore, they are ideal to use as support [5].

Chromenes are one of the most important classes of biologically active compounds, which have gained emerging interest in recent research areas due to their antidepressant, antiviral, antihypertensive, anti-tubulin and antioxidative activities [6, 7]. These compounds are also used as cosmetics, pigments [8] and potential biodegradable agrochemicals [9]. Despite the versatile applications of 2-amino-4H-chromenes, only a few methods have been reported for the preparation of these compounds from aldehydes, resorcinol and malononitrile using NEt_3 [10], 1,8-diazabicyclo[5.4.0]undec-7-ene (DBU) [11], piperazine [12], tungstic acid functionalized SBA-15 [13], glycine [14], trifluoroethanol [15] and nanocrystalline MgO [16]. Some of these methods are associated with the use of hazardous organic solvents, long reaction times, low product yields and use of toxic and/or corrosive non-reusable catalysts. Therefore, there is a need to develop a simple and efficient catalyst for the synthesis of 2-amino-4H-chromenes under mild and green conditions. As part of our continuous efforts into the design of magnetic nanocatalysts and application of them in organic reactions [5, 17–20], herein, we report preparation, characterization and catalytic application of (α-Fe_2O_3)-MCM-41-(1-methyl-3-(4-sulfobutyl) imidazolium hydrogen sulfate) as a novel and efficient nanocatalyst in one-pot three-component synthesis of 2-amino-4H chromenes in water medium conditions (Scheme 1).

Experimental

Chemicals were purchased from Sigma and Merck chemical companies. Melting points were determined using an IA 8103 melting point apparatus. FT-IR spectra were obtained with KBr pellets in the range of 400–4000 cm^{-1} with a Nicolet-860 spectrometer. ^{1}H-NMR and ^{13}C-NMR spectra were recorded in deuterated dimethyl sulfoxide (DMSO-d6) solvent using a DRX-300 spectrometer at 300 and 75 MHz, respectively, and trimethylsilane (TMS) as an internal standard. Elemental analysis for C, H, and N were performed using an Heraeus CHN rapid analyzer. The pore structure of the prepared catalyst was verified by the nitrogen sorption isotherm



Scheme 1 Synthesis of 2-amino-4H chromenes in the presence of (α-Fe_2O_3)-MCM-41-DAIL catalyst

([5.0.0.3] Belsorp, BEL Japan, Inc.). X-ray powder diffraction (XRD) was carried out on a Philips X'Pert diffractometer with CoKα radiation. The structure and morphology of the products were characterized by transmission electron microscopy (TEM) recorded on a Philips CM-10 instrument with an accelerating voltage of 100 kV, and scanning electron microscopy (SEM) analysis was performed using a Philips XL-30 scanning electron microscope (Poland) operating at an accelerating voltage of 17 kV.

Catalyst preparation

Synthesis of 1-methyl-3-(4-sulfobutyl) imidazolium hydrogen sulfate (DAIL)

In a 100-mL round bottom flask, a mixture of 1-methyl imidazole (0.1 mol, 8.21 g) and 1,4-butane sultone (0.1 mol, 11 mL) in toluene (30 mL) was refluxed for 24 h. The obtained white solid was washed with toluene (2 × 10 mL) and dried under reduced pressure. Then, a stoichiometric amount of concentrated sulfuric acid (98 %) was added to the solid and the mixture was stirred for 24 h at 70 °C to form the DAIL. It was then washed with diethyl ether (2 × 10 mL) and dried under vacuum.

Synthesis of (α-Fe_2O_3)-MCM-41

To prepare Fe_3O_4 nanoparticles, a solution with a molar composition of 3.2 $FeCl_3$:1.6 $FeCl_2$:39 NH_4OH:2300 H_2O was used. 2 g of iron (III) chloride ($FeCl_3$6-H_2O) and 0.8 g of iron (II) chloride ($FeCl_2.4H_2O$) were dissolved in 10 mL of distilled water under N_2 atmosphere. The resulting solution was added drop wise to a 100-mL solution of 1.0-M NH_4OH. The magnetic MCM-41 was prepared by adding 20 mL of the magnetic colloid to a 1-L solution with a molar composition of 292 NH_4OH:1 CTABr:2773 H_2O under vigorous mixing and sonication. Then, sodium silicate (16 mL) was added, and the mixture was allowed to react at room temperature for 24 h under well-mixed conditions. The template was then removed by calcination at 450 °C for 5 h to give the (α-Fe_2O_3)-MCM-41.

Preparation of (α-Fe_2O_3)-MCM-41-DAIL

1 mmol of the synthesized DAIL was dissolved in 10 mL of ethanol at room temperature. Then, 1 g (α-Fe_2O_3)-MCM-41 was added to the solution with vigorous stirring. The mixture was then stirred for 24 h at room temperature. The solvent was removed with a rotary evaporator to afford (α-Fe_2O_3)-MCM-41-DAIL. Then, it was washed with acetone (2 × 10 mL) and dried in a vacuum oven.

General procedure for the synthesis of 2-amino-4H-chromenes

The catalytic activity of (α-Fe_2O_3)-MCM-41-DAIL was examined in the synthesis of 2-amino-4H-chromenes. For this reason, a mixture of resorcinol (1 mmol), aldehyde (1 mmol), malononitrile (1 mmol), and catalyst (0.1 g) in 5 mL of H_2O was stirred at 80 °C for an appropriate time. After completion of the reaction (monitored via thin layer chromatography), the catalyst was separated by an external magnet. The remaining solution was dried and the product was crystallized from EtOH to afford the pure product. All of the known products were characterized by comparison of their physical and spectral data with the reported data.

Physical, spectral and analytical data of the newly synthesized products

2-amino-3-cyano-7-hydroxy-4-(2-bromophenyl)-4H-chromene (**4b**) yellow powder. m.p.: 205–206 °C. IR (KBr, cm^{-1}): $\upsilon_{max} = 3475, 3341, 3251, 2191, 1646, 1585, 1406, 1109$. ^1H NMR (300 MHz, DMSO-d6): δ 5.13 (s, 1H, H-4), 6.41 (s, 1H, H–Ar), 6.47 (d, 1H, $J = 8.1$ Hz, H–Ar), 6.72 (d, 1H, $J = 8.1$ Hz, H–Ar), 6.93 (s, 2H, NH_2), 7.12–7.24 (m, 2H, H–Ar), 7.30–7.34 (m, 1H, H–Ar), 7.57 (d, 1H, $J = 7.5$ Hz, H–Ar), 9.82 (s, 1H, OH). ^{13}C NMR (75 MHz, DMSO-d6): δ 55.11, 102.29, 103.29, 112.55, 120.22, 122.29, 128.48, 128.84, 129.13, 131.00, 132.84, 144.58, 148.87, 157.39, 160.40. Anal. Calcd. for $C_{16}H_{11}O_2N_2Br$: C, 55.97; H, 3.21; N, 8.16 %. Found: C, 55.95; H, 3.24; N, 8.17 %.

2-amino-3-cyano-7-hydroxy-4-(4-cyanophenyl)-4H-chromene (**4h**) yellow powder. m.p.: 180–181 °C. IR (KBr, cm^{-1}): $\upsilon_{max} = 3465, 3337, 3198, 2233, 2190, 1651, 1408, 1508, 1106$. ^1H NMR (300 MHz, DMSO-d6): δ 4.76 (s, 1H, H-4), 6.42 (s, 1H, H–Ar), 6.48 (d, 1H, $J = 8.2$ Hz, H–Ar), 6.78 (d, 1H, $J = 8.2$ Hz, H–Ar), 6.98 (s, 2H, NH_2), 7.35 (d, 2H, $J = 7.6$ Hz, H–Ar), 7.77 (d, 2H, $J = 7.6$ Hz, H–Ar), 9.40 (s, 1H, OH). ^{13}C NMR (75 MHz, DMSO-d6): δ 55.26, 102.42, 109.57, 112.49, 112.68, 118.83, 120.42, 128.51, 129.94, 132.76, 148.95, 151.82, 157.55, 160.50. Anal. Calcd. for $C_{17}H_{11}O_2N_3$: C, 70.59; H, 3.80; N, 14.53 %. Found: C, 70.56; H, 3.82; N, 14.51 %.

2-amino-3-cyano-7-hydroxy-4-(4-pyridine)-4H-chromene (**4i**) yellow powder. m.p.: 214–215 °C. IR (KBr, cm^{-1}): $\upsilon_{max} = 3416$, 3209, 2190, 1652, 1510, 1411, 1068. ^{1}H NMR (300 MHz, DMSO-d6): δ 4.67 (s, 1H, H-4), 6.42 (s, 1H, H–Ar), 6.49 (d, 1H, $J = 8.4$ Hz, H–Ar), 6.82 (d, 1H, $J = 8.4$ Hz, H–Ar), 7.00 (s, 2H, NH_2), 7.17 (d, 2H, $J = 4.8$ Hz, H-pyridine), 8.48 (d, 2H, $J = 4.8$ Hz, H-pyridine), 9.82 (s, 1H, OH). ^{13}C NMR (75 MHz, DMSO-d6): δ 54.69, 102.35, 112.14, 112.57, 120.34, 122.39, 122.61, 129.81, 148.95, 149.95, 154.48, 157.48, 160.57. Anal. Calcd. for $C_{15}H_{11}O_2N_3$: C, 67.92; H, 4.15; N, 15.85 %. Found: C, 67.93; H, 4.16; N, 15.87 %.

Results and discussion

The prepared catalyst [(α-Fe_2O_3)-MCM-41-DAIL] was characterized with IR, XRD, SEM, TEM, nitrogen physisorption measurements, thermal gravimetric analysis (TGA) and acid–base titration. The acid–base titration showed that the amount of H^+ in the catalyst is 1.45 mmol g^{-1}. The TGA thermogram was used to determine the amount of DAIL supported on (α-Fe_2O_3)-MCM-41 (Fig. 1). As shown in Fig. 1, the supported DAIL decomposes at about 341.79 °C. This figure also indicates that about 21.46 wt% is attributed to the DAIL in the (α-Fe_2O_3)-MCM-41-DAIL catalyst. The result obtained by TGA showed good agreement with that of acid–base titration.

The FT-IR spectra of DAIL, (α-Fe_2O_3)-MCM-41 and (α-Fe_2O_3)-MCM-41-DAIL are shown in Fig. 2. As this Figure shows, pure IL exhibited characteristic peaks of alkyl sulfonate and hydrogen sulfate at 1226, 1165, 1026, 959, 605 cm^{-1} [21]. In this spectrum, the bands observed at 1570 and 1627 cm^{-1} can be assigned to C–N and C–C vibrations in the imidazole ring [22]. Additional bands at 3162, 3085, 2931 and 1466 cm^{-1} are due to an imidazole moiety and alkyl chain [22]. In the FT-IR spectrum of (α-Fe_2O_3)-MCM-41 (Fig. 2b), the band due to Si–O stretching was observed at 1077 and 3419 cm^{-1} can be assigned to the Si–O–H groups [22] and the bands in the region of 400–800 cm^{-1} attributed to the stretching vibrations of the

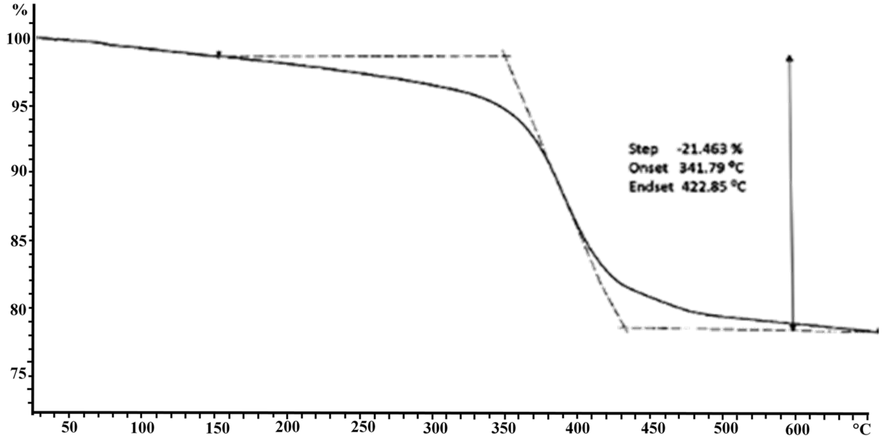


Fig. 1 Thermogram of (α-Fe_2O_3)-MCM-41-DAIL

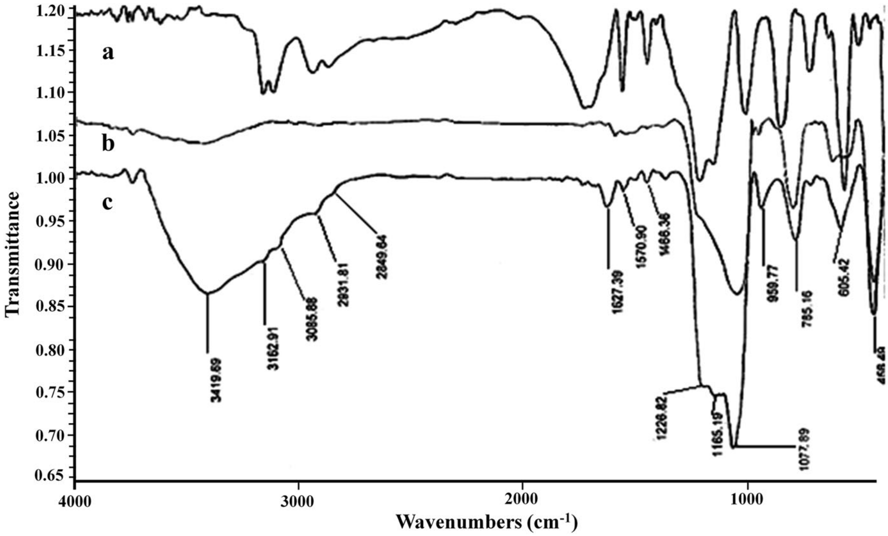


Fig. 2 The FT-IR spectra of (*a*) DAIL; (*b*) (α-Fe_2O_3)-MCM-41; (*c*) (α-Fe_2O_3)-MCM-41-DAIL

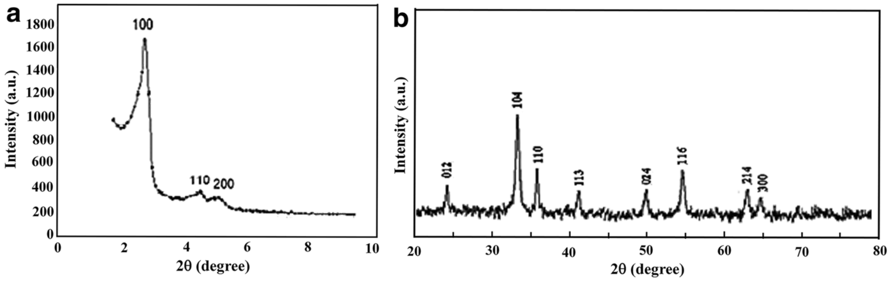


Fig. 3 XRD patterns of (α-Fe_2O_3)-MCM-41-DAIL in the region of **a** 2.0° (2*θ*) to 10.0° (2*θ*); and **b** 20.0° (2*θ*) to 80.0° (2*θ*)

Fe–O band. In the FT-IR spectrum of (α-Fe_2O_3)-MCM-41-DAIL (Fig. 2c), apart from the main peaks of (α-Fe_2O_3)-MCM-41, additional peaks attributed to the functional groups of DAIL were also observed. This indicates the successful supporting of the DAIL into the (α-Fe_2O_3)-MCM-41.

XRD patterns of the synthesized catalyst at the low angle region (Fig. 3a) indicates a high degree of pore symmetry of MCM-41. Also the XRD spectra of the catalyst at 20.0° (2*θ*) to 80.0° (2*θ*) (Fig. 3b) are in good agreement with the standard XRD pattern of α-Fe_2O_3 [23].

The TEM image clearly shows the ordered mesopore system of (α-Fe_2O_3)-MCM-41-DAIL catalyst and the particles with an average diameter of about 100 nm (Fig. 4a). The SEM image also shows that the diameter of these spherical particles is about 100 nm (Fig. 4b).

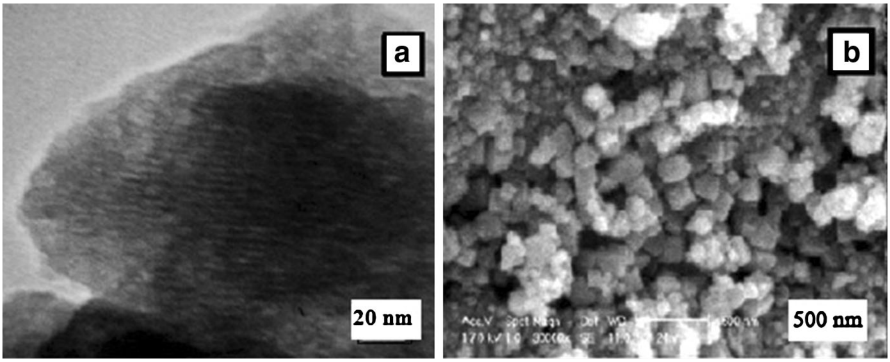


Fig. 4 **a** TEM image; and **b** SEM image of (α-Fe_2O_3)-MCM-41-DAIL

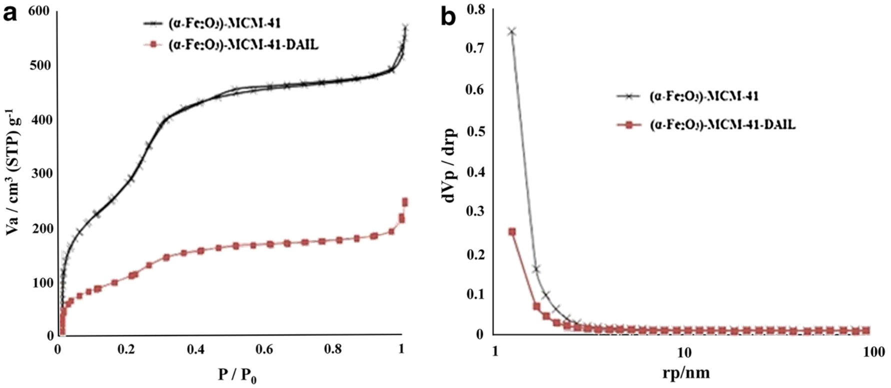


Fig. 5 **a** Nitrogen adsorption/desorption; and **b** BJH analysis of (α-Fe_2O_3)-MCM-41 and (α-Fe_2O_3)-MCM-41-DAIL

The N_2 adsorption/desorption isotherms of (α-Fe_2O_3)-MCM-41 and (α-Fe_2O_3)-MCM-41-DAIL (Fig. 5a) show hysteresis loops resembling the type IV isotherm with an H1-type, which is characteristic of highly ordered, two-dimensional hexagonal mesostructure materials with cylindrical channels. Comparison of these two isotherms shows that in (α-Fe_2O_3)-MCM-41-DAIL, the N_2 was adsorbed at lower values than in (α-Fe_2O_3)-MCM-41 and this indicates that the pores were occupied with DAIL. The specific surface area and total pore volume obtained by N_2 adsorption isotherms and calculated by the Brunauer–Emmet–Teller (BET) method [24], were 1075 $m^2\ g^{-1}$ and 0.87 $cm^3\ g^{-1}$, respectively, before DAIL loading, and 430 $m^2\ g^{-1}$ and 0.39 $cm^3\ g^{-1}$, respectively, after loading DAIL into the catalyst. These data also indicate that DAIL was successfully introduced inside the nanochannels of (α-Fe_2O_3)-MCM-41. The (α-Fe_2O_3)-MCM-41-DAIL Barrett-Joyner-Halenda (BJH) analysis, with a very narrow pore size distribution and pore diameter of 2.4 nm, indicates the highly ordered structure of this catalyst (Fig. 5b).

The efficiency of the prepared catalyst was tested in the synthesis of 2-amino-4H chromenes. In order to optimize the reaction conditions, the reaction between malononitrile, resorcinol and 4-chlorobenzaldehyde was studied as a model. The results are summarized in Table 1. As this table shows, the highest yield of the product was obtained in the presence of 0.1 g of the catalyst per each mmol of the reactants, using water as a solvent and a reaction temperature of 80 °C.

In order to show the unique catalytic behavior of (α-Fe_2O_3)-MCM-41-DAIL and compare its efficiency with other reported catalysts and its building components, the reaction for synthesis of 2-amino-3-cyano-7-hydroxy-4-(phenyl)-4H-chromene (**4d**) was performed using different catalysts (Table 2). The data showed that (α-Fe_2O_3)-

Table 1 Optimization of reaction conditions for the synthesis of 2-amino-3-cyano-7-hydroxy-4-(4-chlorophenyl)-4H chromene **(4a)**

Entry	Solvent	Temperature (°C)	Amount of catalyst (g)	Yield (%)
1	H_2O	r.t.	0.1	90
2	H_2O	60	0.1	90
3	H_2O	70	0.1	93
4	H_2O	80	0.1	95
5	H_2O	90	0.1	92
6	H_2O	80	0.07	89
7	H_2O	80	0.15	95
8	EtOH	78	0.1	88
9	MeOH	65	0.1	85
10	H_2O/EtOH	80	0.1	90
11	H_2O/MeOH	80	0.1	87
12	Solvent-free	80	0.1	75

1 mmol of the reactants were used in all experiments. r.t. room temperature

Table 2 Effect of different catalysts in the synthesis of 2-amino-3-cyano-7-hydroxy-4-(phenyl)-4H-chromene **(4d)**

Entry	Catalyst	Time (min)	Yield (%)	Refs.
1	No catalyst	180	Trace	This work
2	DAIL	180	64	This work
3	MCM-41	180	Trace	This work
4	(α-Fe_2O_3)-MCM-41	180	32	This work
5	MCM-41-DAIL	8	90	This work
6	(α-Fe_2O_3)-MCM-41-DAIL	8	95	This work
7	DBU[a]	40	76	[11]
8	Trifluoroethanol	Not reported	90	[15]
9	Glycine/sonic condition	9	94	[14]
10	SBA-15-tugestic acid	720	86	[13]
11	Piperazine/microwave irradiation	7	90	[12]

[a] DBU = 1,8-Diazabicyclo[5.4.0]undec-7-ene

Table 3 Synthesis of 2-amino-4-H-chromenes over the (α-Fe_2O_3)-MCM-41-DAIL in water medium

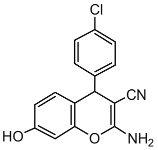
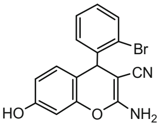
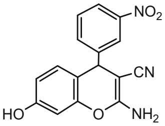
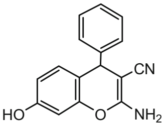
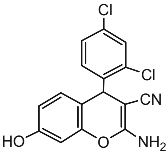
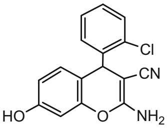
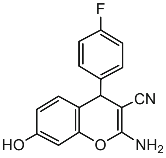
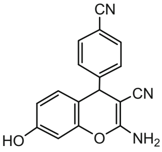
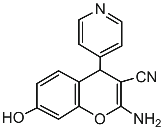
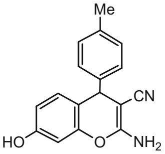
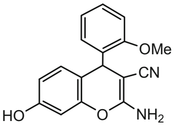
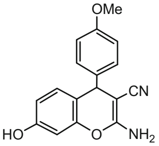
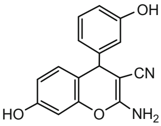
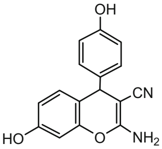
Entry	Product	Time (min)	Yield^a (%)	Measured m.p. (°C)/Lit. m.p. (°C) [Refs.]
1	 **4a**	10	95	162–163/161–162 [12]
2	 **4b**	10	91	205–206/New
3	 **4c**	12	96	170–171/169–170 [15]
4	 **4d**	8	95	232–233/231–233 [11]
5	 **4e**	12	90	256–258/256–258 [16]
6	 **4f**	15	90	188–189/188–190 [12]
7	 **4g**	12	95	186–187/186–188 [11]
8	 **4h**	12	96	180–181/New

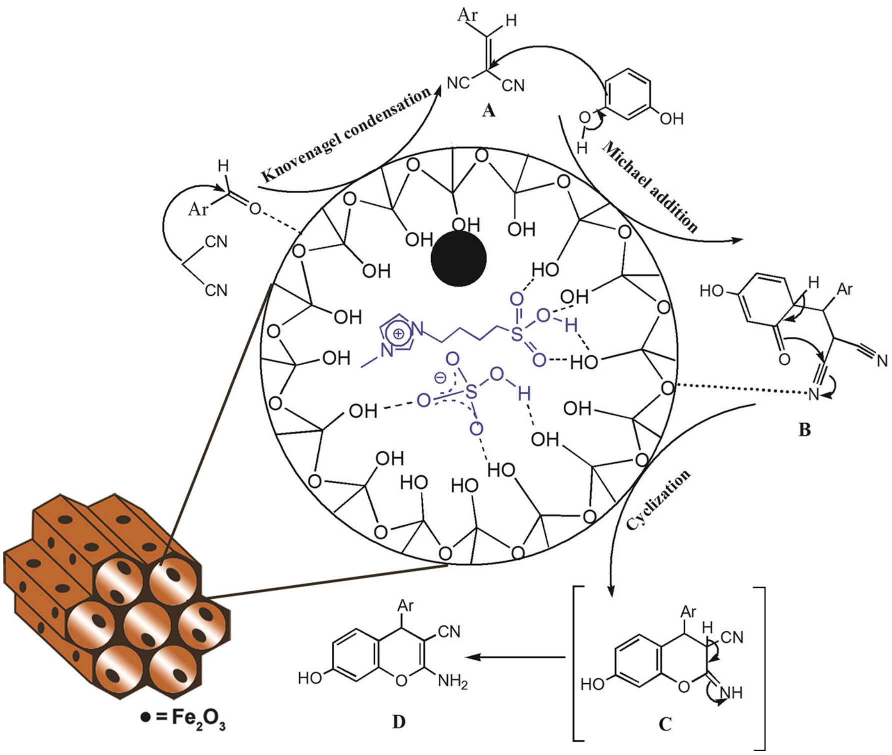
Table 3 continued

Entry	Product	Time (min)	Yield[a] (%)	Measured m.p. (°C)/Lit. m.p. (°C) [Refs.]
9	 **4i**	8	95	214–215/New
10	 **4j**	15	90	182–183/184–186 [11]
11	 **4k**	15	85	220–222/222–224 [16]
12	 **4l**	15	89	111–112/110–112 [11]
13	 **4m**	15	85	214–216/215–217 [16]
14	 **4n**	15	88	214–216/213–215 [16]

[a] Isolated yield

MCM-41-DAIL is the most efficient catalyst for this reaction (Table 2, entry 6). This shows that the acidic character (DAIL) or high surface area [(α-Fe_2O_3)-MCM-41 and MCM-41] alone is not sufficient for the reaction. Comparison the data in the entries 5 and 6 of Table 2 also shows that the presence of ferric oxide as Lewis acids resulted in higher yields of the product.

To examine the scope and generality of the process, various 2-amino-4H-chromenes were then synthesized under optimized reaction conditions. The results summarized in Table 3 indicate that use of only 0.1 g of (α-Fe_2O_3)-MCM-41-DAIL shows a marked efficiency for all of the reactions tested.



Scheme 2 A plausible mechanism for the synthesis of 2-amino-4H chromenes in the presence of (α-Fe_2O_3)-MCM-41-DAIL catalyst

A plausible mechanism for the formation of 2-amino-4H chromenes is shown in Scheme 2. First, the reactants absorb into the mesochannels of the high surface area catalyst. The dual Brønsted acidity of the IL and the Lewis acidity of Fe^{3+} all activate the aldehyde. The Knovenagel condensation reaction between aldehyde and malononitrile leads to the formation of intermediate **A**. Then, Michael addition takes place between intermediate **A** and resorcinol to form intermediate **B**, which is followed by intra-molecular cyclization to yield the desired product (**D**). In these reactions, the catalyst increases the electrophilicity of aldehyde carbon atoms, followed by dehydration to give intermediate **A**. Again, this catalyst increases the electrophilicity of the CN group so that Michael addition and cyclization steps can easily occur to yield the product [14–16].

It is important to note that the presence of ferric oxide in the catalyst facilitates its recovery from the reaction mixture during the work-up procedure by use of an external magnet in less than 1 min. Thus, the catalyst was simply collected and the recovered catalyst was used in subsequent runs without significant loss of its activity even after six runs (Scheme 3).

The FT-IR spectra of the fresh and recycled catalyst after 6 circles are respectively presented in Fig. 6a, b. This figure shows the presence of the IL in the catalyst after six runs.

Scheme 3 Catalyst recovery for the model reaction

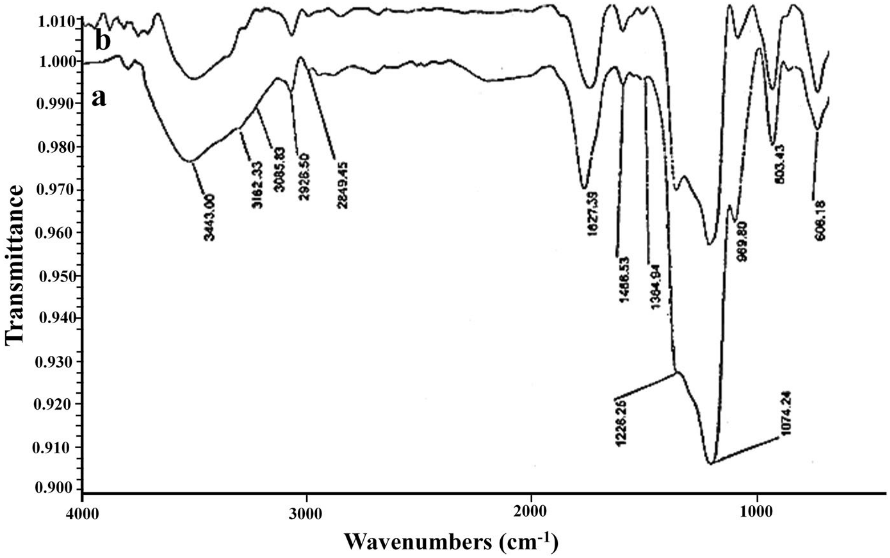


Fig. 6 The FT-IR spectra of (*a*) fresh; and (*b*) recycled catalyst after 6 *circles*

Conclusion

In summary, we have developed a novel magnetic (α-Fe_2O_3)-MCM-41-DAIL nanocatalyst that shows remarkable catalytic activity in one-pot three-component synthesis of 2-amino-3-cyano-7-hydroxy-4-H-chromenes. The Brønsted dual acidic property of the IL together with the Lewis acidity of ferric oxide and high surface area of MCM-41 have increased the catalytic activity of the catalyst. The results of our studies showed that this catalyst is more efficient than other reported catalysts for the synthesis of 2-amino-3-cyano-7-hydroxy-4H-chromenes. High product yields, short reaction times, use of non-corrosives, a robust and reusable magnetic nanocatalyst and water as a green solvent are the most important features of the present work.

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