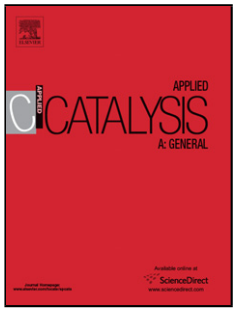

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MCM-41-*N*-propylsulfamic acid: An efficient catalyst for one-pot synthesis of 1-amidoalkyl-2-naphtols

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ABSTRACT

An efficient, one-pot three-component condensation of aromatic aldehydes, acetamide and 2-naphtol in the presence of catalytic amounts of MCM-41-*N*-propylsulfamic acid under thermal solvent-free conditions is described for the preparation of 1-amidoalkyl-2-naphtols in good to excellent yields. In this work, MCM-41-*N*-propylsulfamic acid was prepared by reaction of propylamine functionalized MCM-41 and chlorosulfonic acid and characterized by FTIR, XRD and TGA.

Keywords: 1-Amidoalkyl-2-naphtols, MCM-41, MCM-41-*N*-propylsulfamic acid, Solvent-free, Multicomponent reaction.

1. Introduction

One of the principal problems of organic chemistry in recent years is the creation of structurally complex organic compounds from simple substrates [1]. The first and most important method used for this purpose is multicomponent reaction (MCR). Multicomponent reactions are performed without need to isolate any intermediate during the reaction, which reduces the amount of time and energy used [2]. There have been tremendous developments in three or four component reactions. Condensation of 2-naphtol, aldehydes and amides in the presence of various catalysts for the synthesis of 1-amidoalkyl-2-naphtol is an example of MCR. 1-Amidoalkyl-2-naphthol derivatives have attracted considerable interest because of their biological and pharmacological activities [3]. Also these compounds can be converted into important biological active 1-aminoalkyl-2-naphthol derivatives by amide hydrolysis [3,4]. In addition, 1-amidoalkyl-2-naphthols can be converted to 1,3-oxazines [5]. 1,3-Oxazines have analgesic [6] and antirheumatic properties [7]. Preparation of 1-amidoalkyl-2-naphtols carried out in the presence of Lewis or Bronsted acid catalysts. Therefore, some methods using various homogeneous or heterogeneous catalysts including Montmorillonite K10 [8], iodine [9], sulphamic acid [10], silica supported perchloric acid [11], silica sulfuric acid [12], polymer supported sulfunic acid [13], ionic liquids [14], graphite supported perchloric acid [15], tris(triphenylphosphine) ruthenium(II) dichloride [4], zinc benzenesulfonate [16], cation exchanged resins [17], Silica gel–supported polyphosphoric acid [18] and Trityl Chloride [19] have been reported. However, many of these reported methods suffer from one or more drawbacks such as long reaction times, poor yields, the use of toxic organic solvents, the use of expensive or toxic metal salts as catalysts, and tedious work-up procedures. Therefore, search for finding an efficient, general and environmentally gentle method and milder catalysts for the synthesis of 1-amidoalkyl-2-naphthols have been under permanent attention. In recent years, mesoporous materials such as MCM-41 have attracted the attention of many scientists [20]. They enable to carrying high dosages of a variety of drugs in their mesopores, called drug delivery vehicles [21]. Furthermore mesoporous silica containing specific functional groups have high specific surface areas and porosities that opened a wide field of applications in removal of toxic metal ions from waste-water and natural-waters [22,23]. Thus they provide a suitable approach to the development of green chemistry principles. Because of the availability of a large number of free silanol

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groups on the surface of MCM-41, modification by reaction of the surface silanol groups can occur and inorganic-organic hybrid mesoporous materials have been developed [24]. These modified mesoporous could be applied as catalysts for different types of reactions such as nitration of phenols [25], epoxidation of styrene [26] and reduction of p-nitrophenol to p-aminophenol [27] or could be used for environmental purification.

The present investigation reports the synthesis, characterization and catalytic properties of functionalized MCM-41 (MCM-41-*N*-propylsulfamic acid) in the multicomponent synthesis of 1-amidoalkyl-2-naphtols. In this study, one-pot condensation reaction of aromatic aldehydes, 2-naphtol and acetamide in the presence of catalytic amounts of MCM-41-*N*-propylsulfamic acid under solvent-free conditions at 130 ^{0}C carried out to afford 1-amidoalkyl-2-naphtol derivatives.

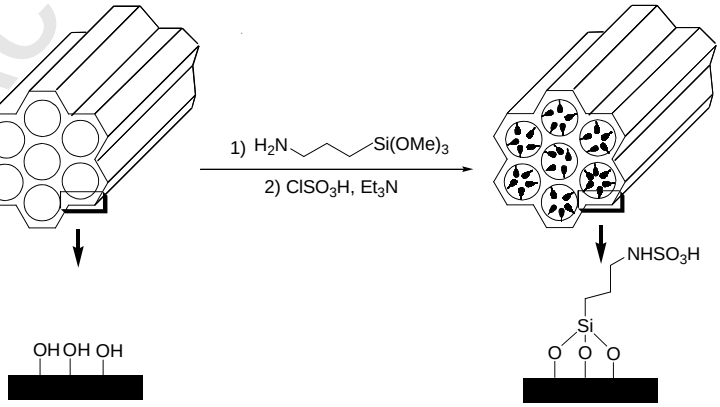
2. Experimental

2.1. Chemicals

Analytical grade hexadecyltrimethylammonium bromide (CTAB), acetic acid, hydrochloric acid (37%), sodium hydroxide (NaOH), aldehydes, 2-naphtol and acetamide were all purchased from Aldrich and Merck and used without further purification. Deionized distilled water was used in the preparation of all solutions. The known products were characterized by comparison of their spectral (1H NMR, and 13CNMR, Bruker NMR-Spectrometer FX 400Q) and physical data with those of authentic samples. Unknown compounds were identified by their 1H and 13CNMR spectra and elemental analysis.

2.2. Synthesis of MCM-41-N-propylsulfamic acid

MCM-41 was synthesized according to the literature method [28] by the reaction of tetraethyl orthosilicate (TEOS) as the source of silica (1 mmol) and cetyltrimethylammonium bromide (CTAB, 0.1 mmol), in basic solution of 0.3 mmol NaOH: 60 mL H_2O. The gel mixture was then crystallized under hydrothermall treatment at 110 ˚C for 60 h in a Teflon lined autoclave. After cooling to room temperature the resultant solid was recovered by filtration, washed with deionized water and dried in air. The collected product was calcined at 550 ˚C for 12 h to remove the polymeric surfactants and CTAB. This mesoporous material is designated as MCM-41. In the next step, according to the literature [29] 27 mmol of 3-Aminopropyl trimethoxysilane (APTMS) was stirred with MCM-41 (4.8 g) in n-hexane (96 mL) at 80 ˚C for 24 h under nitrogen atmosphere. The resulting solid NH_2-$(CH_2)_3$-MCM-41 was filtered, washed several times with n-hexane and dried in vacuum. Finally for synthesis of MCM-41-*N*-propylsulfamic acid, to a solution of propylamine functionalized MCM-41 (2 g) in n-hexane (10 mL), Et_3N (0.83 mL) was added. The mixture was stirred for 10 min. Chlorosulfonic acid (0.266 mL) was added dropwise. HCl gas evolved from the reaction vessel immediately. The mixture was stirred for next 3 h for completing the reaction. Afterwards, the residue was filtered and washed with n-hexane. Then the resulting product was washed with ethanol and then water to afford crystalline MCM-41-*N*-propylsulfamic acid (scheme 1).



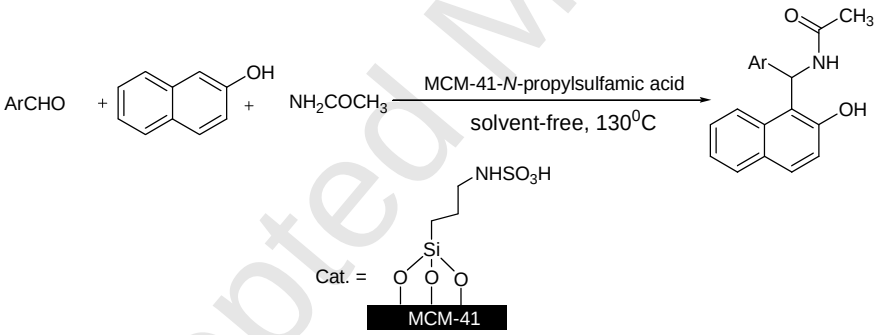
Scheme 1. Synthesis of MCM-41-*N*-propylsulfamic acid

2.3. Catalyst characterization

The crystalline structure of synthesized samples was examined by X-ray diffraction using GBC-Difftech MMA diffractometer. The nickel filtered Cu Kα (λ= 1.54A˚) radiation was used at acceleration voltage of 35 kV and current of 34.2 mA. The diffraction angle was scanned from 1˚ to 10˚, 2θ at a rate of 1˚/min. In order to determine the textural properties, the nitrogen adsorption–desorption isotherms were measured using a BEL sorp-mini II volumetric adsorption analyzer in order to determine the textural properties. All of the samples were degassed at 100 ˚C under an argon gas flow for 3 h before analysis. The specific surface area of the synthesized materials was evaluated using the BET method, and the pore size distribution was calculated by the BJH method. Fourier transform infrared spectroscopy (FTIR) analyses were carried out on FTIR, spectrophotometer (Bruker, Germany) Vertex 70 in the range of 400–4000 cm^{-1}. A thermogravimetric analysis (TGA) was carried out (PerkinElmer Pyris Diamond, U.K.) from an ambient temperature to 840 ˚C, using a ramp rate of 10˚C/min.

2.4. General procedure for the synthesis of 1-amidoalkyl-2-naphtols

In a typical reaction, to a mixture of 2-naphthol (1 mmol), aldehydes (1 mmol) and acetamide (1.5mmol), catalyst (0.1 gr) was added. The mixture was stirred at 125 ˚C in an oil bath as indicated by thin-layer chromatography (TLC) for a complete reaction. After the completion of the reaction, the mixture was cooled to room temperature, then the solid residue was solved in boiling EtOH and the mixture was stirred for 10 minutes. Catalyst was filtered, and the solution was concentrated under reduced pressure to obtain the product. The product was crystallized from ethanol (Scheme 2). The products were characterized by IR, ^{13}C NMR and ^{1}H NMR, and values were compared with the literature data of known products.



Scheme 2. One-pot synthesis of 1-amidoalkyl-2-naphthols

N-[(4-chloro-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide (entry 2, table 3):

mp: 235-237 °C; ^{1}H NMR (400 MHz, DMSO-d6): δ_H= 2.02 (s, 3H), 7.14 (d, *J* = 8, 1H), 7.19 (d, *J* = 8.4 1H), 7.29 (m, 3H), 7.4 (t, *J* = 7.2, 1H), 7.81 (m, 3H), 7.71 (d, *J* = 8.8, 1H), 8.51 (d, *J* = 8, 1H), 10.1 (s, 1H) (ppm). ^{13}C-NMR (100 MHz, DMSO): δ_C= 23.1, 56.6, 118.9, 119, 123, 123.6, 127, 128.4, 128.9, 129.1, 130, 131.1, 132.7, 142.3, 153.7, 170 (ppm).

N-[(4-hydroxy-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide (entry 6, table 3):

mp: 233-234 °C; ^{1}H NMR (400 MHz, DMSO-d6): δ_H= 1.98 (s, 3H), 6.67 (d, *J* = 8.4, 2H), 6.99 (d, *J* = 8, 2H), 7.06 (d, *J* = 8.4, 1H), 7.38 (t, *J* = 6.4, 1H), 7.78 (m, 2H), 7.88 (s, 1H), 8.42 (d, *J* = 8.4, 1H), 9.24 (s, 1H), 9.99 (s, 1H) (ppm). ^{13}C-NMR (100 MHz, DMSO): δ_C= 23.2, 48, 115.3, 119, 119.6, 122.8, 123.8, 126.6, 127.8, 129, 129.5, 132.8, 133, 153.5, 156.2, 169.5 (ppm).

N-[(3-hydroxy-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide (entry 7, table 3):

mp: 227-228 °C; ^{1}H NMR (400 MHz, DMSO-d6): δ_H= 1.99 (s, 3H), 6.56 (s, 1H), 6.58 (d, *J* = 8, 1H), 6.61 (d, *J* = 8.4, 1H), 6.63 (d, *J* = 6.4, 1H), 7.1 (m, 1H), 7.22 (m, 1H), 7.28 (d, 1H), 7.77 (d, 1H), 7.81 (d, *J* = 8.4,1H), 8.39 (s, 1H), 8.42 (s, 1H), 9.99 (s, 1H) (ppm). ^{13}C-NMR (100 MHz, DMSO): δ_C= 22.6, 40.1, 47.6, 113.1, 116.7, 118.4, 118.9, 122.3, 123.3, 123.4, 126.2, 128.4, 128.9, 129.1, 132.4, 144.1, 153.1, 157.1, 169.2

(ppm).

N-[(4-ethoxy-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide (entry 9, table 3):

mp: 213-217 °C; ^{1}H NMR (400 MHz, DMSO-d6): δ_H= 1.29 (t, *J* = 6.8, 3H), 2 (s, 3H), 3.94 (q, *J* = 6.8, 2H), 6.82 (d, *J* = 8.8, 2H), 7.11 (t, *J* = 8.4, 3H), 7.27 (t, *J* = 8, 2H), 7.38 (m, 2H), 7.89 (s, 1H), 8.47 (d, *J* = 8.4, 1H), 10.04 (s, 1H) (ppm). ^{13}C-NMR (100 MHz, DMSO): δ_C= 23.2, 48, 56.6, 63.4, 114.4, 119, 119.5, 122.8, 123.8, 126.7, 127.7, 129, 129.6, 132.8, 134.7, 153.6, 157.4, 169.6 (ppm).

N-[(3-nitro-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide (entry 10, table 3):

mp: 251-253 °C; ^{1}H NMR (400 MHz, DMSO-d6): δ_H= 2.04 (s, 3H), 7.27 (m, 3H), 7.43 (t, *J* = 7.2, 1H), 7.57 (m, 2H), 7.86 (m, 3H), 8.03 (s, 1H) 8.06 (m, 1H), 8.65 (d, *J* = 8, 1H), 10.17 (s, 1H) (ppm). ^{13}C-NMR (100 MHz, DMSO): δ_C= 23, 48.1, 118.3, 118.9, 120.9, 121.7, 123.1, 123.3, 127.3, 128.9, 129.2, 130.1, 130.4, 132.6, 133.3, 145.9, 148.2, 153.8, 170.2 (ppm).

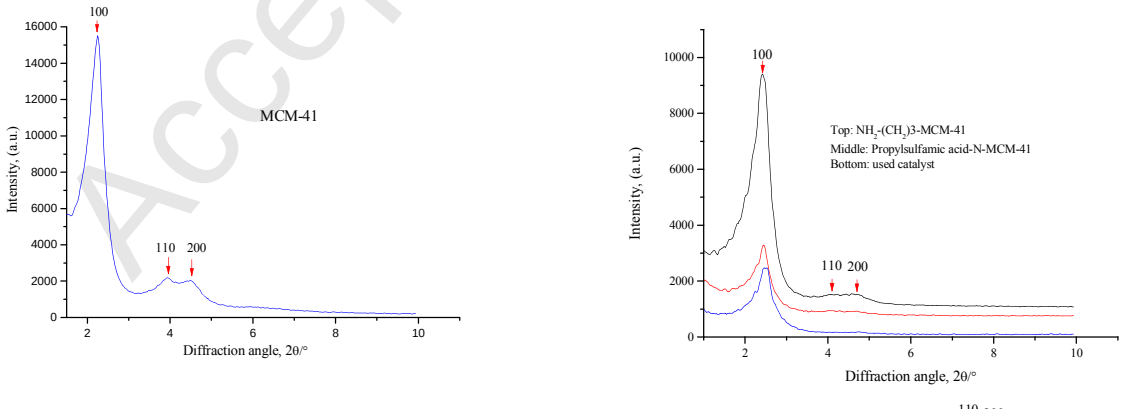
3. Results and discussion

3.1. Structural features of catalyst

The XRD pattern of MCM-41 and functionalized catalyst (NH_2-$(CH_2)_3$-MCM-41 and MCM-41-*N*-propylsulfamic acid) is presented in Fig 1. These spectra (Figure1.a) revealed that one-dimensional hexagonal mesoporous structure of MCM-41 was obtained. As shown, the MCM-41 pattern exhibits very sharp (1 0 0) diffraction peak at 2.24 and two additional high order peaks (1 1 0 and 2 0 0) with lower intensities at 3.91 and 4.5, respectively [30]. The values of *d*-spacing for these XRD peaks appear at 39.38, 22.58 and 19.60 ˚A, respectively. A unit cell parameter, a_0, of 45.50 ˚A was obtained using the following equation [31].

$$a_0=2d_{100}/\sqrt{3} \quad (1)$$

For NH_2-$(CH_2)_3$-MCM-41 and MCM-41-*N*-propylsulfamic acid, a considerable decrease in the XRD peaks intensity was observed (Figure 1.b). It should be noted that the intensity decreases in the (1 0 0) peak for functionalized MCM-41 providing further evidence of functionalization occurring mainly inside the mesopore channels. Collectively, the XRD pattern of the functionalized MCM-41 also suggest not only a significant degree of short range ordering of the structure and well-formed hexagonal pore arrays of the samples, but also the maintenance of the structural order of the synthesized materials after functionalization. Also, used catalyst XRD diagram revealed that mesopore channels during catalytic reaction was stable as physical structure damage has not been observed.



(a) (b)

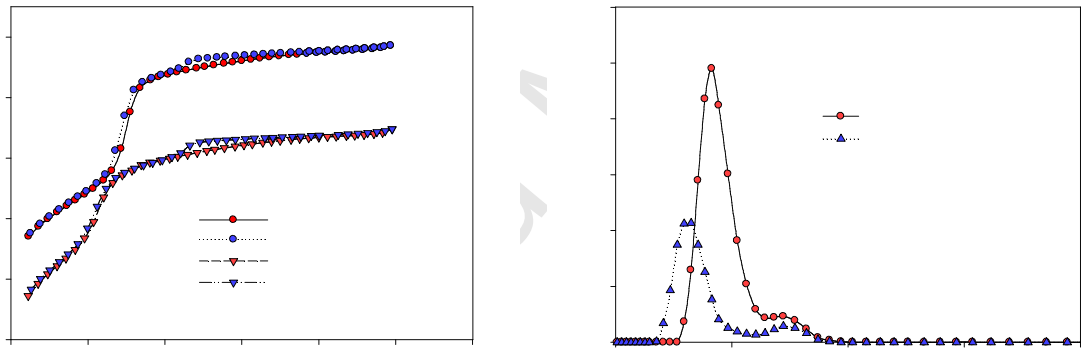
Fig. 1. Low-angle XRD patterns of: a) MCM-41. b) NH_2-$(CH_2)_3$-MCM-41, MCM-41-*N*-propylsulfamic acid and Used catalyst

Physical parameters of nitrogen isotherms for the Barret-Joyner-Halenda average pore diameter (D_{BJH}), the Brunauer-Emmett-Teller surface area (S_{BET}) and the total pore volumes (V_{total}) of the calcined MCM-41 and NH_2-$(CH_2)_3$-MCM-41 are summarized in table 1.

Table 1. Textural properties of MCM-41and NH_2-$(CH_2)_3$-MCM-41.

Sample	**S_{BET} ($m^2.g^{-1}$)**	**D_{BJH} (nm)**	**V_{total} ($cm^3.g^{-1}$)**
MCM-41	986.16	3.65	0.711
NH_2-$(CH_2)_3$-MCM-41	694.98	3.30	0.340

Nitrogen adsorption/desorption isotherms of on MCM-41 show the typical type IV isotherm based on the IUPAC nomenclature for MCM-41 (Figure 2.a). The isotherms indicate that, at a relative pressure (P/P^0) between 0.047-0.26, nitrogen adsorption takes place as a thin layer on the walls (monolayer coverage). A sharp inflection at 0.26–0.33 is related to the capillary condensation and confirms the existence of uniform pores [31]. Pore size distribution (ADS) of MCM-41 according to BJH methods is presented in Fig.2.b. After functionalization, a decrease in the S_{BET} and D_{BJH} average pore diameter was observed that can be easily interpreted due to the fact that the presence of pendant group on the surface that partially blocks the adsorption of nitrogen molecules.



(a) (b)

Fig. 2. Nitrogen adsorption-desorption isotherms and pore size distribution (ADS) of MCM-41 and NH_2-$(CH_2)_3$-MCM-41.

FTIR spectroscopy was used to detect the presence of binding groups in the MCM-41-*N*-propylsulfamic acid, NH_2-$(CH_2)_3$-MCM-41 and used catalyst (Figure 3). The vibration signals around 1055, 793 and 458 cm^{-1} are typical Si-O-Si bands attributed to the asymmetric stretching, symmetric stretching and bending, respectively [32,33]. The spectra showed a broad band around 3000-3600 cm^{-1} for MCM-41-*N*-propylsulfamic acid and used catalyst, which is due to acidic O-H vibration signal of SO_3H. Presence of the peak around 1635 cm^{-1} mainly from the NH_2 symmetric bending vibration, indicates the successful grafting of organic amine onto the surface [34, 35]. The absorbance of the C-N stretching vibration is normally observed around 1000-1300 cm^{-1}. However this peak cannot be resolved due to its overlap with the absorbance of Si–O–Si stretch in the 970-1350 cm^{-1}. The spectrum of the recovered MCM-41-*N*-propylsulfamic acid (Black line) shows that the catalyst is stable during the reaction.

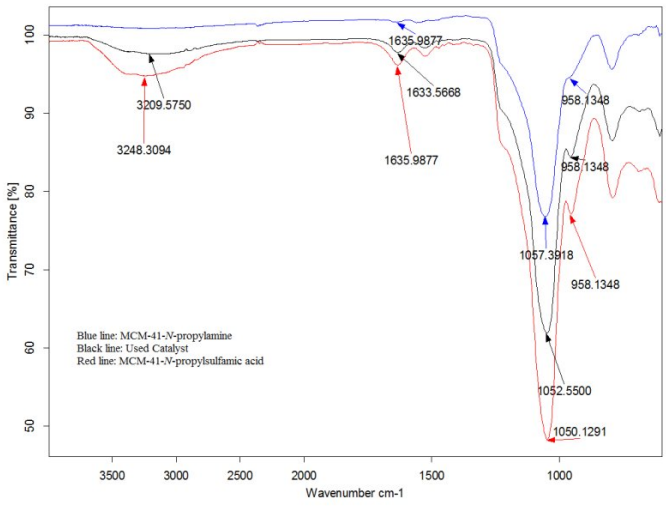


Fig. 3. FT-IR spectra of NH_2-$(CH_2)_3$-MCM-41, MCM-41-*N*-propylsulfamic acid and used catalyst.

The thermograms of the synthesized mesoporous silica are presented in Figure 4. The thermogram of the calcined MCM-41 shows two steps of weight loss. The weight loss is 4.16% in the first step from 25 ˚C to 120 ˚C that is associated with desorption of water [36] and in the second step (120-840 ˚C), the weight loss (1.67%) is mainly due to water loss formed by the condensation of silanol groups [37]. The thermogram of the amino-functionalized MCM-41 shows a gradual weight loss up to 840 ˚C. The TGA curve of the NH_2-$(CH_2)_3$-MCM-41 shows four steps of weight loss (25-120 ˚C, 120-340 ˚C, 340 ˚C -600 ˚C and >600 ˚C). The weight loss is 7.9% in the first step and is due to desorption of physisorbed water held in the pores. The weight losses in the second and third steps (10.47%) are mainly associated with oxidative decomposition of organic functional groups. In the final step, the weight loss (1.67%) is due to the dehydroxylation of the silicate networks. On the basis of elemental analysis, the amount of functional groups of APTMS was calculated to be 0.58 mmol.g^{-1} of MCM-41. The TGA curve of the Propylsulfamic acid-N-MCM-41 shows four steps of weight loss (25-135 ˚C, 135-370 ˚C, 370 ˚C -640 ˚C and >640 ˚C). The weight loss is 5.2% in the first step and is due to desorption of water. The weight losses in the second and third steps (20.82%) are mainly associated with oxidative decomposition of organic functional groups. So, the amount of functional groups of Propylsulfamic acid was calculated to be 0.090 mmol.g^{-1} of MCM-41. In the final step, the weight loss (0.26%) is due to the dehydroxylation of the silicate networks.

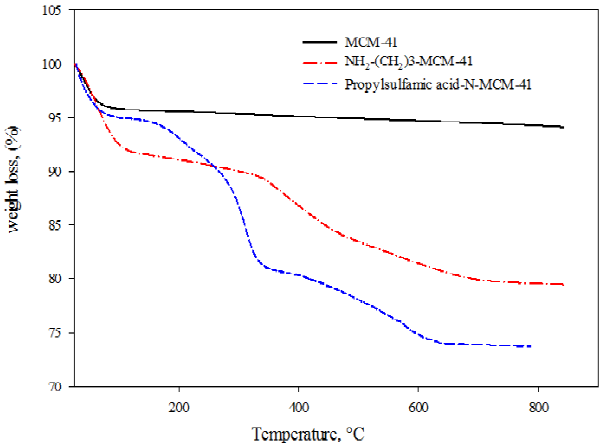


Fig. 4. Thermograms of MCM-41,NH_2-$(CH_2)_3$-MCM-41 and MCM-41-*N*-propylsulfamic acid.

3.2. Reactions

Herein, we wish to report a novel protocol for the synthesis of a variety of biologically significant 1-amidoalkyl-2-naphthols using a catalytic amount of MCM-41-*N*-propylsulfamic acid under solvent-free conditions. To find the optimum conditions, a model reaction of 4-chlorobenzaldehyde (1 mmol), 2-naphthol (1 mmol), and acetamide (1.5 mmol) in the presence of different amounts of MCM-41-*N*-propylsulfamic acid was performed under solvent-free conditions at 130 ˚C (Table 2). In the absence of any catalyst, desirable product was obtained in very low yield (entry 1), whereas best result was obtained in the presence of 0.1 g of catalyst (entry 5). A slight excess of the acetamide was found to be advantageous for yields of 1-amidoalkyl-2-naphthols and hence the molar ratio of aromatic aldehydes, 2-naphthol and acetamide was kept at 1:1:1.5.

Table 2. Optimization of MCM-41-*N*-propylsulfamic acid as catalyst for synthesis of N-[(4-chloro-phenyl)-(2-hydroxy-napthalen-1-yl)-methyl]-acetamide at 130 °C [a]

Entry	Catalyst (g)	Time (hr)	Yield (%)[b]
1	-	24	17
2	0.02	8	33
3	0.05	5	45
4	0.08	3	63
5	0.1	1.75	95

[a] Reaction conditions: 4-Chlorobenzaldehyde (1 mmol), 2-naphtol (1 mmol), acetamide (1.5 mmol). [b] Crude yields.

Furthermore, during the optimization of the reaction condition, the relation between the yields of the model reaction with temperature was also studied. We carried out the reaction at temperatures ranging from 25 ˚C to 140 ˚C (Figure 5). As seen, the yield of the product increased with increasing of temperature from 25 ˚C until it reaches a maximum at 130 ˚C, and decreases again after that. Therefore, the best suited reaction conditions for this three-component reaction were obtained in the presence of 0.1 gr of MCM-41-*N*-propylsulfamic acid at 130 ˚C under solvent-free condition.

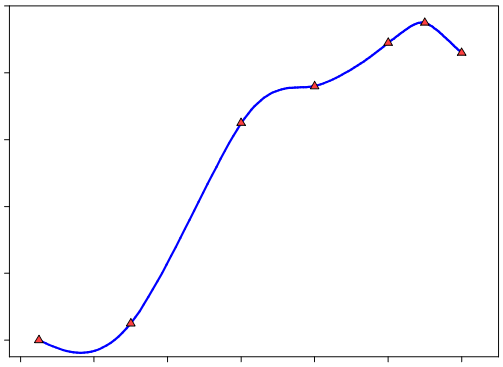


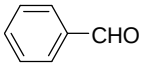
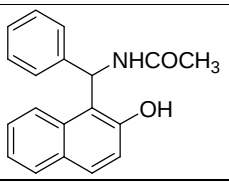
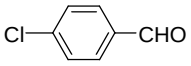
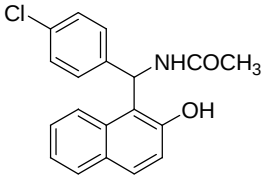
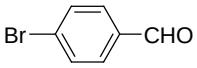
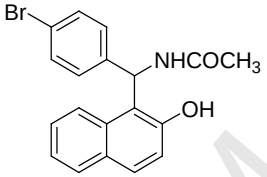
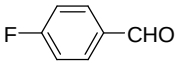
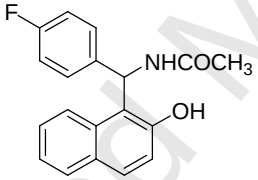
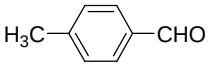
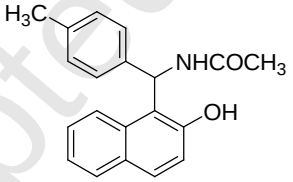
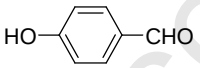
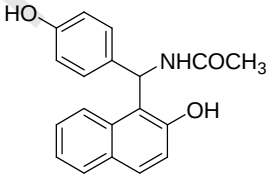
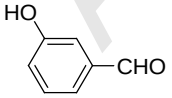
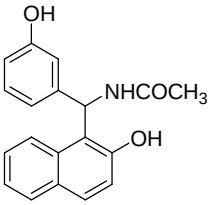
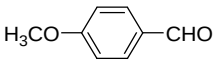
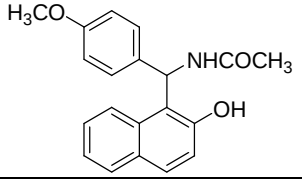
Fig. 5. Effect of temperature on the reaction of 4-Chlorobenzaldehyde, 2-naphtol and acetamide

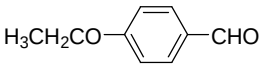
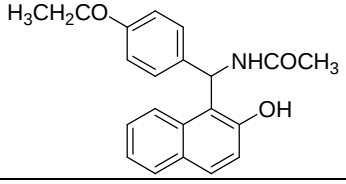
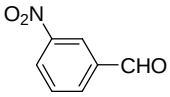
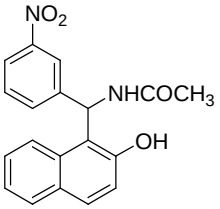
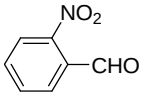
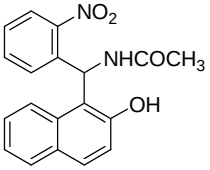
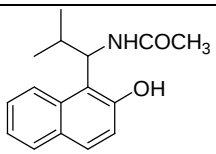
The one-pot three-component condensation of various aromatic aldehydes with acetamide and 2-naphthol catalyzed by MCM-41-*N*-propylsulfamic acid was then explored under the optimized reaction conditions described above. The results are summarized in Table 3.

As shown in table 3 in all cases, the reaction proceeded smoothly to give the corresponding 1-amidoalkyl-2- naphthol derivatives in satisfactory yields. Above and beyond all other consideration, aromatic aldehydes with substituents bearing either electron-donating (such as methyl, methoxy

or ethoxy) or electron-withdrawing groups (such as nitro or halide) reacted successfully in the presence of MCM-41-*N*-propylsulfamic acid as catalyst.

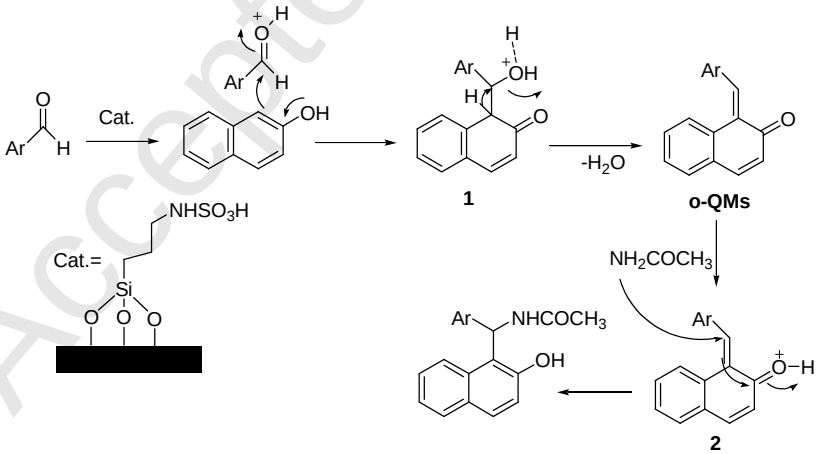
Table 3. MCM-41-*N*-propylsulfamic acid -catalyzed synthesis of 1-amidoalkyl-2-naphtols[a]

Entry	Aldehyde	Product	Time (min)	Yield (%)[b]	Mp (lit. mp)	Ref.
1			120	97	238-240 (242-244)	[38]
2			105(300)[c]	95(80)[c]	235-237 (229-230)	[14a]
3			190	87	228-230 (229-231)	[14a]
4			150	86	226-228 (230-232)	[11]
5			180	95	218-222 (221-223)	[38]
6			90	98	233-234	-
7			105	98	227-228	-
8			240	86	178-180 (181-183)	[38]

9			240	98	213-217	-
10			160	56	251-253 (241-242)	[38]
11			210	93	216-218 (218-219)	[39]
12			270	35	198-201 (181-183)	[40]

[a] Reaction conditions: Aldehydes (1 mmol), 2-naphtol (1 mmol), acetamide (1.5 mmol), MCM-41-*N*-propylsulfamic acid (0.1 g), 130 °C [b] Crude yields. [c] Reaction performed in the presence of recovered catalyst.

The proposed mechanism for this acid catalysis condensation reaction via in situ generation of ortho-quinone methides (**o-QMs**) is presented in Scheme 3. First nucleophilic addition of 2-naphtol to the activated aldehydes by catalyst have been occurred to afford intermediate **1**. After dehydration of **1,** the resultant **o-QMs** have been reacted with acetamide via conjugate addition to form intermediate **2**. Finally this intermediate will aromatize to produce 1-amidoalkyl-2-naphtols as desirable products.



Scheme 3. Proposed mechanism for synthesis of 1-amidoalkyl-2-naphthols

To show the merit of MCM-41-*N*-propylsulfamic acid in comparison with other reported catalysts, we summarize several results for the preparation of N-[phenyl (2-hydroxynaphthalen-1-yl) methyl] acetamide from benzaldehyde, 2-naphthol, and acetamide in table 4. It is obvious that MCM-41-*N*-propylsulfamic acid showed good reaction time and higher yield than other catalysts used in literature.

Table 4. Comparison results of MCM-41-*N*-propylsulfamic acid with other catalysts for the synthesis of N-[phenyl-(2-hydroxy-napthalen-1-yl)-methyl]

Entry	Catalyst (mol %)	Time	Yield (%)	Ref.
1	Montmorillonite K10 clay (0.1 g)	1.5 h	89	[8]
2	I_2 (5)	4.5 h	87	[9]
3	Silica sulfuric acid (0.02 g)	2 h	85	[12]
4	Graphite-$HClO_4$ (7.5)	2 h	81	[15]
5	SiO_2- $HClO_4$ (0.6)	40 min	89	[11]
6	Polymer supported sulphonic acid (0.17 g)	5 h	96	[13]
7	Indion-130 resin (0.25 g)	20 min	81	[17]
8	PPA-SiO_2 (0.03 g)	7 min	84	[18]
9	MCM-41-*N*-propylsulfamic acid (0.1 g)	2h	97	This work

4. Conclusion

Briefly, this is the first time that application of MCM-41-*N*-propylsulfamic acid as an efficient and heterogeneous catalyst for the multicomponent one-pot synthesis of a variety of 1-amidoalkyl-2-naphthols under solvent-free conditions have reported. This simple procedure reported here offers several significant advantages, such as high yields, easy handling, good reaction times, operational simplicity and easy workup. The XRD pattern of the MCM-41-*N*-propylsulfamic acid also suggests not only a well-formed hexagonal pore arrays of the samples, but also the maintenance of the structural order of the synthesized materials after functionalization. Also, used catalyst XRD diagram and IR revealed that mesopore channels were stable during catalytic reaction.

Acknowledgments

Financial support to this work by the Ilam University, Ilam, Iran is gratefully acknowledged.

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Graphical abstract:

MCM-41-*N*-propylsulfamic acid: An efficient catalyst for one-pot synthesis of 1-amidoalkyl-2-naphtols

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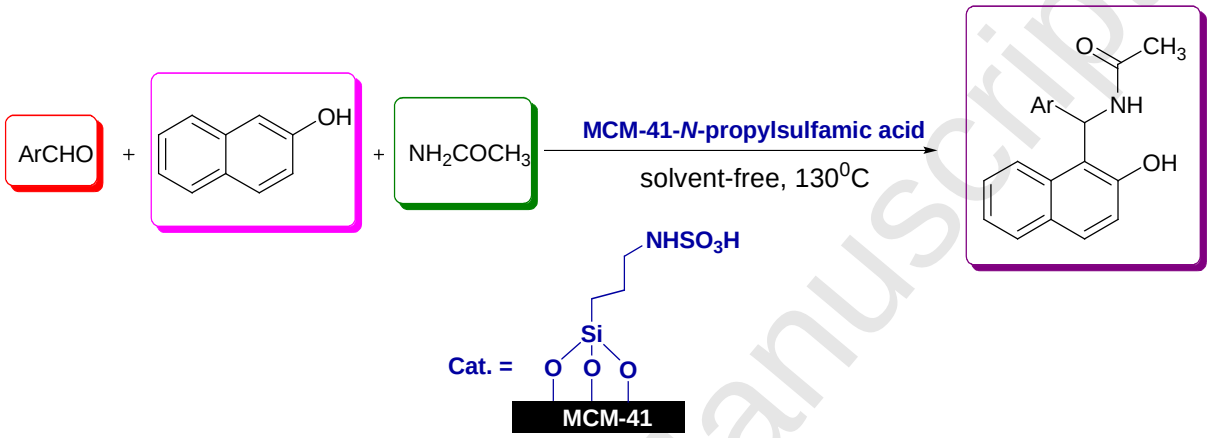

An efficient, one-pot three-component condensation of aromatic aldehydes, acetamide and 2-naphtol in the presence of catalytic amounts of MCM-41-*N*-propylsulfamic acid under thermal solvent-free conditions is described for the preparation of 1-amidoalkyl-2-naphtols.

Highlights

- MCM-41-*N*-propylsulfamic acid was synthesized and characterized.
- Synthesis of 1-amidoalkyl-2-naphthols catalyzed by MCM-41-*N*-propylsulfamic acid.
- Used catalyst XRD and IR revealed mesopore channels was stable during reaction.