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Nanoporous MCM-41-SO$_3$H as an efficient catalyst for chalcones and dibenzylidene alkanones synthesis via solvent-free aldol condensation reaction

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KEYWORDS
MCM-41-SO$_3$H;
Heterogeneous catalyst;
The aldol condensation;
Solvent-free grinding.

Abstract. A one-pot and simple method for the solvent-free synthesis of chalcones and dibenzylidene alkanones, catalyzed by the heterogeneous and nanoporous MCM-41-SO$_3$H catalyst, is reported. The products were afforded, in most cases, with excellent yields.

1. Introduction

The aldol condensation reaction is one of the most important C-C bond formation reactions in classic organic chemistry [1]. Formation of the C-C bond and intramolecular condensation to prepare 5-, 6-, and 7-membered rings are some of the noteworthy advantages of the aldol condensation reaction. Chalcones are synthetic products formed from the aldol condensation reaction with various pharmaceutical properties. The aldol condensation process using microwave irradiation in the presence of various catalysts, such as TiO$_2$-P25-SO$_4^{-2}$ [2a], bis(4-methoxyphenyl)telluroxide (BMPTO) [2b], sulfated zirconia [2c], or by ultrasonic irradiation with catalysts such as acid clay [2d] has been reported. Some Lewis acids such as Yb(OTf)$_3$ [3a], TiCl$_3$(SO$_3$CF$_3$) [3b], Y(TFA)$_3$ [3c], silica sulfuric acid [3d], and Mg(HSO$_4$)$_2$ [3e] have been also used to catalyze the aforementioned reaction. Some few examples of using basic reagents such as NaOH [3f] and [BMIm]Cl/NaOH [3g] have also been reported. In addition, mesoporous materials such as semicrystalline zeolitic-mesoporous material UL-ZSM-5 [4a], diamino-functionalized MCM-41 [4b], magnesium-aluminum hydrotalcite [4c], and Pd/cobalt aluminate [4d] have also successfully catalyzed the aldol condensation reaction. Most of these methods are time-consuming; require harsh conditions, huge amounts of organic solvents, and microwave or ultrasound irradiation; and sometimes suffer from incomplete dehydration of the intermediates.

In continuation of our interest in studying the applicability of nanoporous MCM-41-SO$_3$H catalyst in organic reactions [5a], and in continuation of studying the usefulness of solvent-free grinding in organic reactions, here we report MCM-41-SO$_3$H as a heterogeneous catalyst for the synthesis of chalcones and dibenzylidene alkanones via the aldol condensation reaction by grinding in a solvent-free and efficient method without formation of any by-product (Scheme 1).

2. Experimental

2.1. Materials

MCM-41 [5a], M-MCM-41s (M= B, Al, Fe or Zn), and MCM-41-SO$_3$H [5b] were prepared according to the

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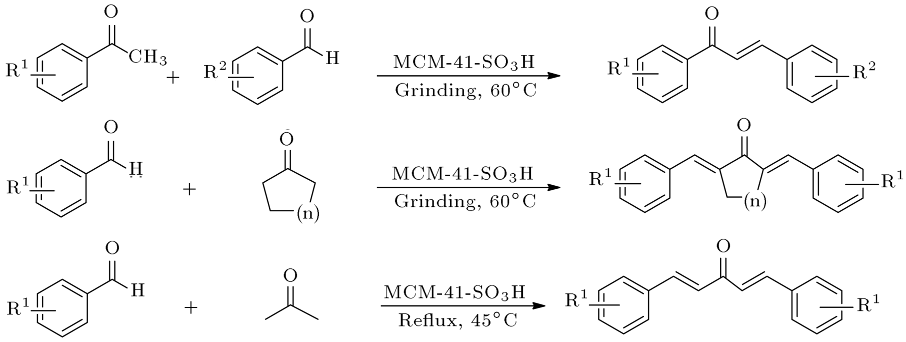


Scheme 1. The aldol condensation reaction catalyzed by MCM-41-SO_3H.

literature. All other chemicals were purchased from Merck and Aldrich and were used as received, except for benzaldehyde that was freshly distilled prior to use. The TLC plates used were Merck 0.2 mm silica gel 60 F-254 aluminum plates.

2.2. Characterization of the nanocatalyst MCM-41-SO_3H

The MCM-41-SO_3H was characterized by SEM, BJH, FT-IR spectroscopy, and acid-base titration, as reported previously [5a]. The O-H stretching bond in the region of 3200-3400 cm^{-1} is assigned to the vibration of hydroxyl groups. The symmetric and asymmetric stretching vibrations of S=O of the sulfonic acid group appear at 1286 cm^{-1} and 1321 cm^{-1}. In addition, the Si-O-Si asymmetric and symmetric stretching vibrations appear as strong bands at 1174 cm^{-1} and 850 cm^{-1}, respectively. The nanopores has proven to have an average width of 3.4 nm based on BJH adsorption test. The acidity of modified nanocatalyst MCM-41-SO_3H was measured by titration of 1.0 g MCM-41-SO_3H by 1.2 mL NaOH 0.01 with a factor of 0.94 N and was calculated as 0.011*10^{-3} mmol H^+/g.

2.3. General procedure for chalcone synthesis

1.0 mmol of an aldehyde, 1.0 mmol of a ketone, and 10 mg MCM-41-SO_3H were mixed and grinded together in a dry mortar. The mortar was then covered by aluminum folio and heated in an ordinary oven at 60°C. The mixture was grinded from time to time and progress of the reaction was monitored by TLC. The grinding process was repeated until the reaction was complete. The reaction mixture was then extracted with dichloromethane and filtered by a nano-sized filter paper. The solvent was removed in vacuum and the obtained solid was recrystallized from hot ethanol, if necessary.

2.4. General procedure for the synthesis of bis(benzylidene) cycloalkanones

An aldehyde (2.0 mmol) was mixed with 1.0 mmol of cyclohexanone (**3d**) or cyclopentanone (**3e**) and 10 mg MCM-41-SO_3H. The mixture was grinded in a dry mortar by a pestle. The remaining process was just like the chalcone synthesis.

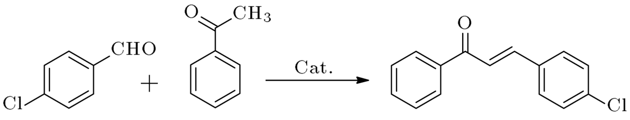
2.5. General procedure for synthesis of diarylidene acetone

1.0 mmol of an aldehyde and 10 mg MCM-41-SO_3H were mixed with an excess amount of acetone and the mixture was heated at 45°C in an oil bath. The reaction progress was monitored by TLC. The remaining process was just like the chalcone synthesis.

3. Results and discussions

To find the optimized reaction conditions for chalcone synthesis, the reaction of 1.0 mmol of 4-chlorobenzaldehyde and 1.0 mmol of benzophenone at 60°C was chosen as the model reaction (Scheme 2). This reaction was carried out with various catalysts. The results are shown in Table 1.

As shown, non-functionalized MCM-41 did not



2b **3a** **4b**

Scheme 2. The model reaction for the synthesis of the chalcone **4b**.

Table 1. Effects of various catalysts and catalyst loading in synthesis of chalcone **4b** at 60°C.

Entry	Catalyst	Catalyst loading (mg)	Time (min)	Yield (%)[a]
1	MCM-41	10	90	-
2	Al-MCM-41	10	90	25
3	B-MCM-41	10	90	20
4	MCM-41-SO_3H	5	90	70
5	MCM-41-SO_3H	10	15	98
6	MCM-41-SO_3H	15	20	95
7	MCM-41-SO_3H	20	15	96
8	Chitosan	10	90	-

[a] Conversion yield.

catalyze the reaction, whereas acidic MCM-41s afforded the product with 20-98% yield. Obviously, the reaction was preceded with high yield and short reaction time with MCM-41-SO_3H as a Brönsted acid catalyst (Table 1, entry 5). In comparison to the previously reported similar Brönsted acid for this reaction [3d], the higher yield and shorter reaction time by MCM-41-SO_3H is due to the high surface area of such mesoporous materials, whereas milder acidic property of MCM-41-SO_3H does not yield any byproduct.

To choose the optimum temperature for carrying out the model reaction, the aforementioned reaction was done in various temperatures in the presence of MCM-41-SO_3H catalyst (Table 2).

Table 2. Effect of temperature in model reaction in the presence of 10 mg MCM-41-SO_3H as catalyst.

Entry	Temperature (°C)	Time (min)	Yield (%)[a]
1	rt	60	-
2	45	60	35
3	60	15	98
4	80	15	98

[a] Conversion yield.

According to the results shown in Tables 1 and 2, the model reaction catalyzed by MCM-41-SO_3H at 60°C was chosen as the optimized model for chalcone synthesis via the aldol condensation reaction. The synthesis of chalcones under optimized conditions was performed with various aldehydes and ketones (Table 3). As shown, the presence of electron donor groups in aldehydes like -OMe, -Me, and -OH caused reaction time reduction in some derivatives such as **4e**, **4f**, **4g**, **4j**, **4p**, **4q**, **4t**, and **4u**. These reactions have been completed in 10, 15, 10, 15, 20, 25, 15, and 10 min, respectively. On the other hand, the presence of electron withdrawing groups like $-NO_2$ increased the reaction time in the derivatives including **4i**, **4o**, **4y**, and **4dd**, which were carried out in 30, 20, 30, and 30 min, respectively.

A similar protocol was used to prepare some bis(arylidene)cycloalkanones from the reaction of aromatic aldehydes with cyclopentanone (**3d**) or cyclohexanone (**3e**). The results have been tabulated in Table 4. As shown, the presence of electron donating groups will accelerate the reaction, whereas electron-withdrawing groups such as NO_2 will slow down it.

Among the various catalysts previously reported for carrying out such reactions, silica sulfuric acid has

Table 3. Synthesis of chalcone derivatives catalyzed by MCM-41-SO_3H via the aldol condensation reaction.

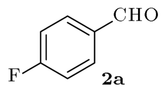
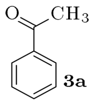
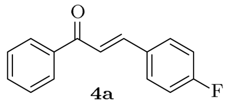
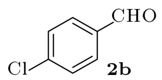
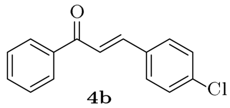
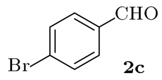
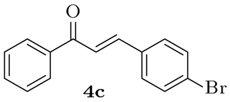
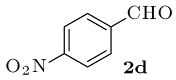
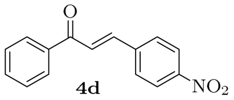
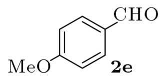
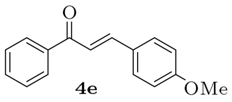
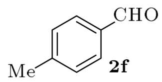
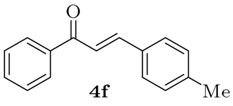
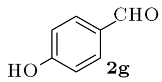
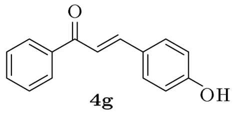
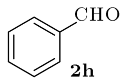
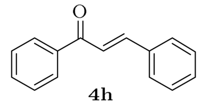
Entry	Aldehyde	Ketone	Product	Time (min)	Yield (%)[a]	mp (°C)	Lit. mp [ref]
1	 2a	 3a	 4a	10	100	86-88	87-88 [6]
2	 2b	3a	 4b	15	98	109-111	110-112 [6]
3	 2c	3a	 4c	15	99	122-123	121-123 [6]
4	 2d	3a	 4d	20	98	156-159	158-159 [7]
5	 2e	3a	 4e	10	100	73-74	72-74 [3d]
6	 2f	3a	 4f	15	97	97-98	97-98 [6]
7	 2g	3a	 4g	10	100	123-124	122-123 [3d]
8	 2h	3a	 4h	45	95	56-57	54-56 [8]

Table 3. Synthesis of chalcone derivatives catalyzed by MCM-41-SO_3H via the aldol condensation reaction (continued).

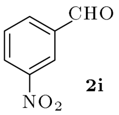
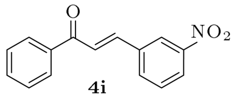
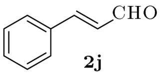
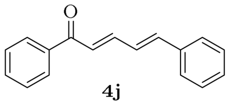
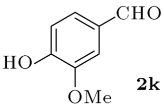
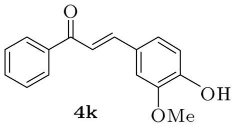
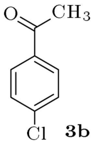
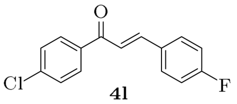
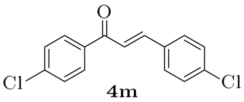
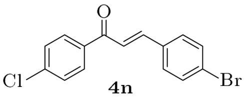
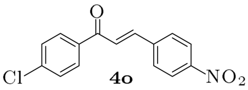
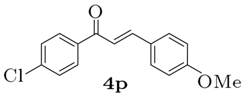
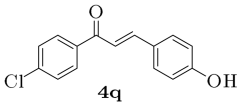
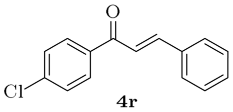
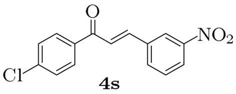
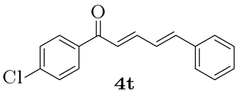
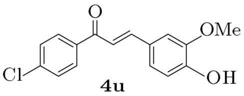
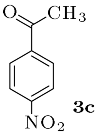
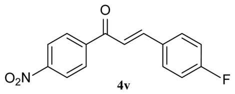
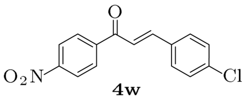
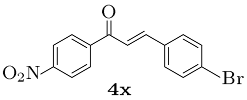
Entry	Aldehyde	Ketone	Product	Time (min)	Yield (%)[a]	mp (°C)	Lit. mp [ref]
9	 **2i**	**3a**	 **4i**	30	99	145-146	146-148 [9]
10	 **2j**	**3a**	 **4j**	15	98	86	86 [10]
11	 **2k**	**3a**	 **4k**	20	99	94-95	91-95 [11]
12	**2a**	 **3b**	 **4l**	10	100	133-134	135 [12]
13	**2b**	**3b**	 **4m**	15	97	110-111	108-111 [13]
14	**2c**	**3b**	 **4n**	15	97	169-170	166-170 [14]
15	**2d**	**3b**	 **4o**	20	98	125-126	124-126 [15]
16	**2e**	**3b**	 **4p**	25	96	122-123	120-122 [11]
17	**2g**	**3b**	 **4q**	10	100	173-175	175-176 [11]
18	**2h**	**3b**	 **4r**	20	95	91-92	90-92 [15]
19	**2i**	**3b**	 **4s**	15	98	151-152	156 [16]
20	**2j**	**3b**	 **4t**	45	99	144-145	143-144 [17]
21	**2k**	**3b**	 **4u**	10	99	118-119	118-120 [18]
22	**2a**	 **3c**	 **4v**	15	100	208-209	210 [19]
23	**2b**	**3c**	 **4w**	15	98	159-160	158-161 [13]
24	**2c**	**3c**	 **4x**	20	98	154-155	152-154 [20]

Table 3. Synthesis of chalcone derivatives catalyzed by MCM-41-SO_3H via the aldol condensation reaction (continued).

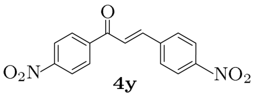
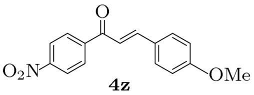
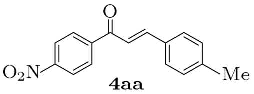
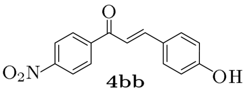
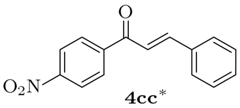
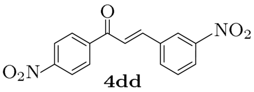
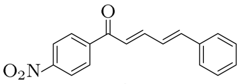
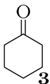
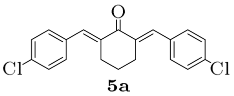
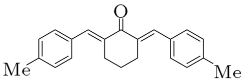
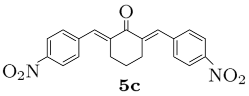
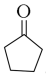
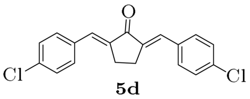
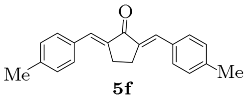
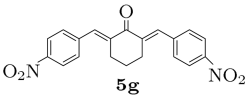
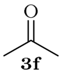
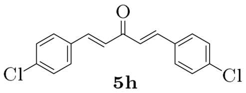
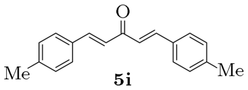
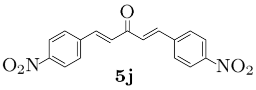
Entry	Aldehyde	Ketone	Product	Time (min)	Yield (%)[a]	mp (°C)	Lit. mp [ref]
25	**2d**	**3c**	 **4y**	30	96	204-206	193 [21]
26	**2e**	**3c**	 **4z**	15	99	195-197	192-194 [22]
27	**2f**	**3c**	 **4aa**	35	98	155-156	163-164 [9]
28	**2g**	**3c**	 **4bb**	45	96	206-207	205-206 [23]
29	**2h**	**3c**	 **4cc***	40	95	119-120	115-116 [24]
30	**2i**	**3c**	 **4dd**	30	99	214-215	213 [25]
31	**2j**	**3c**	 **4ee**	35	100	173-174	172-173 [26]

Table 4. Synthesis of bis(substituted benzylidene)cycloalkanones catalyzed by MCM-41-SO_3H via the aldol condensation reaction.

Entry	Aldehyde	Ketone	Product	Time (min)	Yield (%)[a]	mp (°C)	Lit. mp (°C) [ref]
1	**2b**	 **3d**	 **5a**	15	96	148-149	147-148 [6]
2	**2f**	**3d**	 **5b**	10	97	170-171	170-171 [6]
3	**2d**	**3d**	 **5c**	20	93	163-164	162-163 [3d]
4	**2b**	 **3e**	 **5d**	15	97	227-228	229-230 [3d]
5	**2f**	**3e**	 **5f**	10	97	243-244	242-243 [3d]
6	**2d**	**3e**	 **5g**	30	97	230-231	229-230 [3d]
7[b]	**2b**	 **3f**	 **5h**	90	70	190-192	192-193 [6]
8[b]	**2f**	**3f**	 **5i**	10	80	175-176	174-176 [6]
9[b]	**2d**	**3f**	 **5j**	85	75	247-249	250 [27]

[a] Conversion yields; [b] The reaction performed under reflux conditions at 45°C.

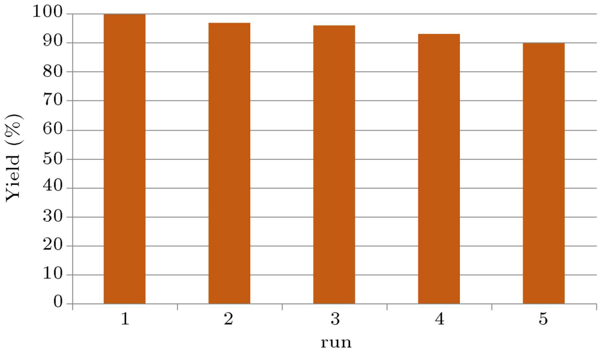


Figure 1. Recyclability of nano-ordered MCM-41-SO_3H catalyst in five consecutive runs.

the most similar structure to that of the MCM-41-SO_3H. Compared with silica sulfuric acid, MCM-41-SO_3H has very shorter reaction times, presumably due to the nanoporous sites in MCM-41-SO_3H which can act as nanoreactors for the reaction. For example, the synthesis of derivatives **5a**, **5b**, **5c**, **5f**, and **5g** have been done in 3, 3.5, 3, 2.5, and 3 h catalyzed by silica sulfuric acid, compared to 15, 10, 20, 10, and 30 min, respectively in our method.

3.1. Catalyst reusability and stability

Reusability of the catalyst was investigated in five consecutive runs for the model reaction. After each run, the raw product was dissolved in dichloromethane and the catalyst was filtered and washed with dichloromethane and then was dried. The recycled catalyst was then used in the consecutive run for the model reaction. As can be seen in Figure 1, the MCM-41-SO_3H catalyst promotes the reaction with remarkable retaining catalytic activity each time.

4. Conclusion

In summary, MCM-41-SO_3H was found to be a suitable nanoporous catalyst for the solvent-free synthesis of 41 chalcones and substituted dibenzylidene cycloalkanones via the aldol condensation reaction. Solvent-free reaction conditions, short reaction times, low catalyst loading, easy work-up, and high yields are some advantages of the present method. No by-product from incomplete dehydration of the β-hydroxycarbonyl intermediates was observed.

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