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Synthesis of the biologically active henna based benzochromene derivatives using ionic liquid functionalized SBA-15 as a nanoreactor

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ABSTRACT

SBA-15 was prepared and then functionalized with *N*-methyl-*N*'-propyltrimethoxysilyl imidazolium chloride as ionic liquid moiety. The ionic liquid functionalized SBA-15 (SBA-IL) was characterized by different analytical techniques including FT-IR, N_2 adsorption-desorption, TGA and SEM image. According to the obtained results it was found that the organic groups were grafted onto the pores of SBA-15 because its pore size and BET surface dropped after modification step. Then, it was used as an efficient nanoreactor in the synthesis of biologically active henna based benzochromene derivatives under solvent-free conditions. Consequently, the catalyst acted efficiently under solvent free system and gave the products in high yields and short reaction times. Some of the obtained products exhibited antibacterial activities as well as tetracycline.

Keywords*: SBA-IL, 2-Hydroxy-1,4-naphtoquinone, Lawsone, Solvent-free reaction, Multicomponent reaction, Henna based benzochromenes.*

1. Introduction

Naphthoquinones structure is one of the most interesting classes of organic compounds due to their industrial applications, biological activities, and organic syntheses [1]. Lawsone is a famous natural naphthoquinone extracted from the leaves of *Lawsonia inermis*; it has been used as a hair dye for millennia [2]. Lawsone has different applications in varied fields of science; for example, it was used for the detection of fingerprint on the paper surface [3] and also for the detection of cyanide ions [4]. Its derivatives exhibited antimalarial [5], antibacterial and antifungal [6] activities. So far, lawsone has been used in various multi-component reactions (MCRs) to gain new naphthoquinones derivatives [7-9].

According to the recent progresses, MCRs play a key role in the synthesis of new heterocyclic compounds [10-12].

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The definition of MCRs is a process in which more than two different starting materials can react together in a same reaction vessel to yield ideally a major product [13]. MCRs have reached major significance as a tool to synthesize a wide variety of valuable materials, such as pharmaceuticals [14,15] and natural products [16].

Up to now, a large volume of studies has been published describing the role of heterogeneous catalysis in MCRs [17-20]. In recent years, application of ionic liquid (IL) catalysts has brought important advantages in organic syntheses [21-23]. However, heterogenization of ionic liquids on the solid support is extremely desirable due to their easy separation from the mixture [24,25].

Since the innovation of mesoporous silica compounds, a variation of ordered mesoporous materials has been synthesized using different methods. These mesoporous materials with large pore size and high surface area have practical applications in the area of heterogeneous organocatalysis through immobilizing organic functionalities onto their pores [26,27]. Among

the existing siliceous supports, Santa Barbara Amorphous (SBA-15) is the best choice for immobilization of the organocatalyst due to its controllable pore size, high surface area, and high thermal and hydrothermal stabilities [28,29].

Although, there is a large amount of published articles explaining the experimental aspects of synthesized benzochromenes [30-35], any study has not been performed on the use of ionic liquid functionalized SBA-15 (SBA-IL) as nano-catalyst in the synthesis of such compounds. On the other hand, high product yield, fast reaction time in a few minutes, easy workup and convenience in the separation of catalyst from the reaction mixture may not be reported by traditional previous studies. Regarding to our previous publications [36,37], in this paper we want to investigate the role of SBA-IL in the synthesis of benzochromenes derivatives based on lawsone.

2. Experimental

2.1. Materials and methods

All chemicals were purchased from Merck and Aldrich Company and used without further purification. Melting points were determined using an Electrothermal 9200 apparatus by capillary tube method. Fourier-transform Infrared (FT-IR) spectra were recorded from KBr disks applying a FT-IR Bruker Tensor 27 instrument. Nuclear magnetic resonance (NMR) spectra were obtained by a Bruker DPX at 250 MHz for ^{1}H NMR and 62.5 MHz for ^{13}C NMR. The NMR spectra were recorded in DMSO-d_6 as solvent and tetramethylsilane (TMS) was used as internal standard. The surface morphology of catalyst was observed by a field emission scanning electron microscope (FESEM, Hitachi S-4160 Japan). Weight change curve in nitrogen was measured on a TGA instrument of BAHR Thermo analyse STA 503 with the maximum heating rate of 10 °C/min.

2.2. Catalysts preparation

2.2.1. Synthesis of the mesoporous silica SBA-15

The mesoporous silica SBA-15 used in this reaction was synthesized through a common method reported before [38].

2.2.2. Synthesis of N-methyl-N′-propyltrimethoxy silylimidazolium chloride (10)

N-Methyl-*N′*-propyltrimethoxysilylimidazolium chloride was synthesized *via* the following the reported method by Coll and coworkers [39]. In this experiment, 1-methyl-1*H*-imidazole **8** (6.57 g, 80 mmol) and (3-chloropropyl)trimethoxysilane **9** (15.89 g, 80 mmol) were mixed well and then, heated at 70 °C for 48 h. Afterward, the mixture was cooled to room temperature, the yellow liquid product was extracted with ether.

2.2.3. Synthesis of imidazolium functionalized SBA-15

The calcined SBA-15 (1.0 g) was dried at 100 °C under vacuum for 2 h to remove the adsorbed water onto its pores. The activated SBA-15 was added to dried toluene (100 mL) and then, stirred for 30 min. Subsequently, *N*-methyl-*N*'-propyltrimethoxysilyl imidazolium chloride (5.61 g) was added to the mixture. The resultant solution mixture was stirred for 48 h under Argon atmosphere. The obtained imidazole functionalized SBA-15 was filtered, washed with dichloromethane using a Soxhlet apparatus for 48 h, and then, dried under vacuum.

2.3. General procedure for the synthesis of benzochromene derivatives 4a-l

A catalytic amount of activated SBA-IL (0.02 g) was added to a mixture of the malononitrile **1** (1 mmol) and aldehyde **2** (1 mmol), and heated at 130 °C under solvent-free conditions for about 2 min. Then, 2-hydroxy-1,4-naphtoquinone **3** was added to it and the reaction mixture was stirred at 130 °C for the specific time as shown in Table 2. After complication of the reaction, which was followed by TLC, the reaction mixture was dissolved in *N*,*N*-dimethylformamide (DMF). The SBA-IL was insoluble in DMF and separated by a simple filtration. The solvent was evaporated under reduced pressure and the pure obtained solid of pure product was recrystallized in ethanol. All the pure products were characterized by comparison of their physical data such as melting point with those of known compounds in literature. The new synthesized products were analyzed using FT-IR and NMR.

Spectral data for new synthesized products

*2-amino-4-(2-methoxyphenyl)-5,10-dioxo-5,10-dihydro-4H-benzo[g]chromene-3-carbonitrile (**4j**):*

Orange powder. FT-IR (KBr): $\bar{\nu}$= 3408, 3324 (NH_2), 3254 (C-H Aromatic), 2964 (C-H, OMe), 2196 (CN), 1667 (C=O), 1635 (C=O) cm^{-1}. ^{1}HNMR (250 MHz, DMSO-d_6): δ= 3.75 (3H, s, OMe), 4.89 (1H, s, CH), 6.80-8.03 (10H, m, arom and NH_2) ppm. ^{13}CNMR (62.5 MHz, DMSO-d_6): δ= 31.5 (CH), 56.18 (OCH_3), 56.92, 112.12, 119.8, 121.42, 122.35, 126.24, 126.50, 128.87, 129.55, 130.94, 131.46, 131.70, 134.55, 135.01, 149.98, 157.17, 159.29 (C-alkene and arom), 177.42, 182.97 (2C=O) ppm. MS: m/e= 358 [$M^{+\cdot}$], 341, 327, 251, 107, 77.

*2-amino-4-(2,3-dimethoxyphenyl)-5,10-dioxo-5,10-dihydro-4H-benzo[g]chromene-3-carbonitrile (**4k**):*

Orange powder. FT-IR (KBr): $\bar{\nu}$= 3446, 3343 (NH_2), 3216 (C-H Aromatic), 2970, 2830 (C-H, OMe), 2193 (CN), 1662 (C=O), 1629(C=O) cm^{-1}. ^{1}HNMR (250 MHz, DMSO-d_6): δ= 3.51 (3H, s, OMe), 3.76 (3H, s, OMe), 4.87 (1H, s, CH), 6.76-8.47 (10H, m, arom and NH_2) ppm. ^{13}CNMR (62.5 MHz, DMSO-d_6): δ= 31.74, 56.00, 57.29, 60.56, 112.21, 119.89, 121.42, 122.68, 124.37, 126.24, 126.50, 130.84, 131.40, 134.58, 135.05, 136.90, 146.61, 149.61 152.71, 159.00 (C-alkene and arom), 177.40, 182.97 (2C=O) ppm. MS: m/e= 388 [$M^{+\cdot}$], 373, 357, 251, 223, 137, 77.

2.4. Antibacterial tests

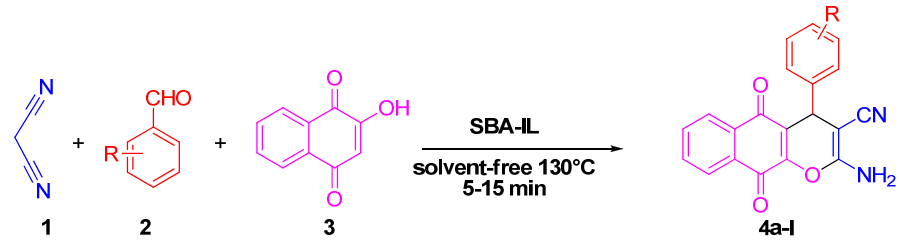
All compounds were dissolved in DMSO (200 µg/mL), and 25 µl of them was loaded on a 6-mm paper discs. One hundred µL of the microorganism's suspension (109 cell/mL) was spread on the sterile Mueller–Hinton agar plates, and the prepared discs were placed on the surface of culture plates.

3. Results and Discussion

For optimization of the reaction, three-component reaction of malononitrile **1**,4-chlorobenzaldehyde **2a** and lawsone **3** was firstly tested under reflux condition in water, EtOH, or solvent-free system using SBA-IL (0.02 g) as catalyst. The results are demonstrated in Table 1. As shown in entry 1, only a trace amount of product **4a** was obtained under reflux condition in water. Among the tested conditions, solvent-free system at 130 °C was the best condition and chosen for this reaction (entry 5). In addition, the lack of reaction development in the absence of the catalyst (entry 6, Table 1) highlights the important role of SBA-IL in progress of the reaction. To prove catalytic effect of IL loaded onto the pores of SBA-15, the optimized reaction was also repeated in the presence of pure SBA-15 (entry 7). Since no product was obtained using SBA-15, the reaction progress in the presence of SBA-IL is due to the loaded-IL groups onto the SBA-15 pores.

Moreover, to evaluate the adaptability and generality of this reaction, various aldehydes were used under the optimized conditions. The obtained results are shown in Table 2. Propionaldehyde as an aliphatic aldehyde produced trace amount of the desired product, thus, this reaction may not be applicable for aliphatic aldehydes.

The recyclability of SBA-IL was also investigated under the optimized conditions for the synthesis of benzochromene **4a**. For this aim, the first cycle of reaction was accomplished for three times to recover about 0.05 gr SBA-IL. Following, it was washed with hot EtOH and then reused. The catalytic activity of recovered SBA-IL drops slightly from the first cycle (95% of product's yield) to the second (87%) owing to leaching of some IL groups to the reaction mixture because they were not grafted well on the surface of SBA-15. Furthermore, no significant drop in catalytic activity of SBA-IL (84% of product's yield) was observed for the third cycle which confirms recyclability of the catalyst.



Scheme 1. Synthesis of benzochromene derivatives in the presence of SBA-IL.

Table 1. The optimization of reaction conditions for the synthesis benzochromene **4a** using SBA-IL.

Entry	Catalyst	Solvent	Condition	Time (min)	Yield (%)
1	SBA-IL	H_2O	Reflux	120	trace
2	SBA-IL	EtOH	Reflux	120	15
3	SBA-IL	-	r.t.	120	N.R.
4	SBA-IL	-	100 °C	30	30
5	SBA-IL	-	130 °C	5	95
6	-	-	130 °C	60	-
7	SBA-15	-	130 °C	60	-

Table 2. Synthesis of benzochromene derivatives in the presence of SBA-IL.

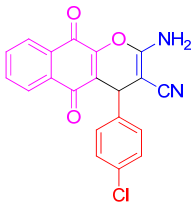
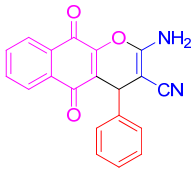
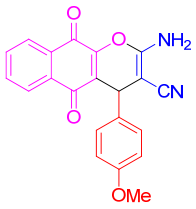
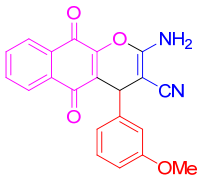
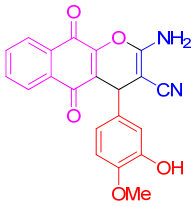
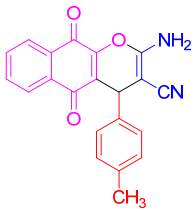
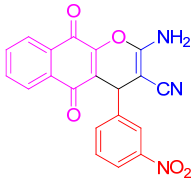
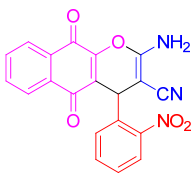
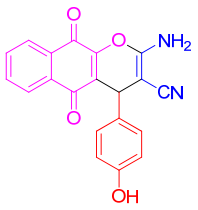
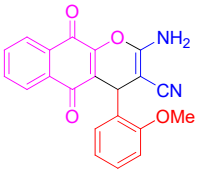
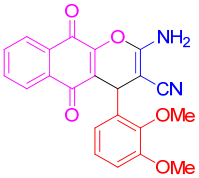
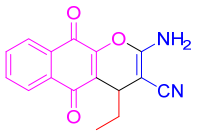
Entry	No.	Aldehyde	Product	Time (min)	Yield (%)	m.p. (°C)		Ref.
						Found	Reported	
1	**4a**	4-ClC_6H_4		5	95	250-251	250-252	[30]
2	**4b**	C_6H_5		5	93	260-263	261-262	[30]
3	**4c**	4-$OMeC_6H_4$		5	87	241-243	240-242	[30]
4	**4d**	3-$OMeC_6H_4$		5	94	246-248	247-248	[32]
5	**4e**	3-OH-4-$OMeC_6H_3$		10	95	243-245	243-245	[34]
6	**4f**	4-MeC_6H_4		5	88	244-247	246-248	[34]
7	**4g**	3-$NO_2C_6H_4$		12	95	248-249	247-249	[32]
8	**4h**	2-$NO_2C_6H_4$		12	93	243-244	242-244	[34]

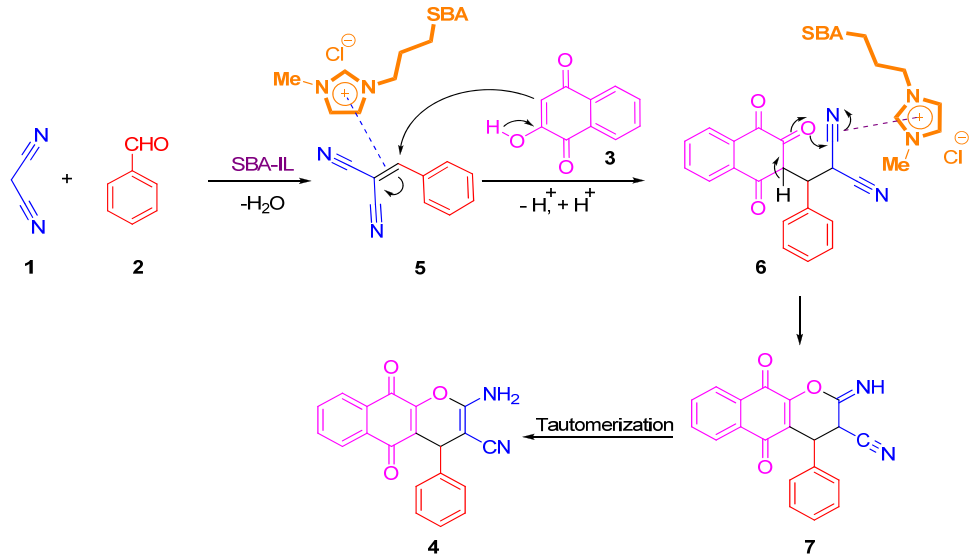
Table 2. (*Continued*).

9	**4i**	4-OHC$_6$H$_4$		15	83	256-260	258-260	[34]
10	**4j**	2-OMeC$_6$H$_4$		5	92	262-264	-	New
11	**4k**	2,3-(OMe)$_2$C$_6$H$_3$		5	94	262-265	-	New
12	**4l**	C$_2$H$_5$		60	trace	-	-	-

A possible mechanism has been proposed for the preparation of benzochromene derivatives in the presence of SBA-IL (Scheme 2).

The Knoevenagel condensation of malononitrile **1** and benzaldehyde **2** in the presence SBA-IL as catalyst affords 2-benzylidenemalononitrile **5**. Subsequently, the latter is attacked by lawsone **3** in the presence of SBA-IL to gain the intermediate **6**. The intramolecular nucleophilic addition of oxygen atom of carbonyl group to the carbon atom of nitrile group **6** causes cyclization and after tautomerization gives the final product **4**.

The efficiency of various catalysts in the synthesis of benzochromene derivatives has been compared in Table 3. The high yield and short reaction time are the reasons for the high efficiency of SBA-IL as a nanoreactor.



Scheme 2. The proposed mechanism.

Table 3. Comparison of different conditions to obtain product **4b**.

Entry	Catalyst	Solvent	Condition	Time (h)	Yield (%)	Ref.
1	Fe_3O_4–proline MNPs[a]	EtOH	r.t.	24	90	[30]
2	TEBA[b]	-	85 °C	4	92	[31]
3	[Bmim]OH[c]	EtOH	r.t.	1	91	[34]
4	DBU[d]	Water	reflux	1	87	[33]
5	Et_3N	CH_3CN	r.t.	24	82	[32]
6	SBA-IL	-	130 °C	5 min	93	This work

[a]MNPs: Magnetic nanoparticles.
[b]TEBA: Triethylbenzylammonium chloride.
[c][Bmim]OH: 1-butyl-3-methylimidazolium hydroxide.
[d]DBU: 1,8-Diazabicyclo[5.4.0]undec-7-ene.

According to these results, it was found that the synthesized SBA-IL is an efficient catalyst for the one-pot synthesis of a different benzochromene derivatives under solvent-free conditions compared to those conditions reported in literature.

All synthesized compounds were monitored for antimicrobial activity against *Subtilis bacillus* (*S. basillus*) and *Escherichia coli* (*E. coli*) as gram-positive and gram-negative bacteria, respectively, through the method of minimum inhibitory concentration (MIC) [40]. The MIC of the prepared benzochromenes was also determined by microdilution method and compared with the commercial tetracycline. As shown in Table 4, except **4b**, **4h** and **4j**, all compounds showed antibacterial activities; it means that the functionalized phenyl group is essential and 2-functionalized phenyl groups (such as **4h** and **4j**) have no antibacterial activities perhaps due to the steric hindrance. Compound **4c** showed excellent to good antibacterial activities against *E. coli* as same as tetracycline. Additionally, **4g** has very good activities against both tested bacteria, which were close to that of tetracycline.

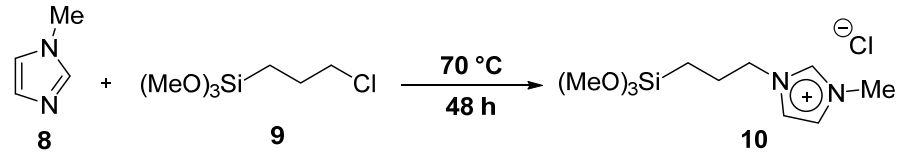
The proposed mechanism of action for the antibacterial activity of these compounds is probably like as that of for tetracycline due to similarity of their structures [41]. Accordingly, benzochromene derivative can diffuse onto the bacteria cell and binds to the 30S subunit of ribosomes. Subsequently, the attachment of aminoacyl-tRNA to the mRNA is blocked and protein synthesis is inhibited.

3.1. Preparation and characterization of the SBA-IL

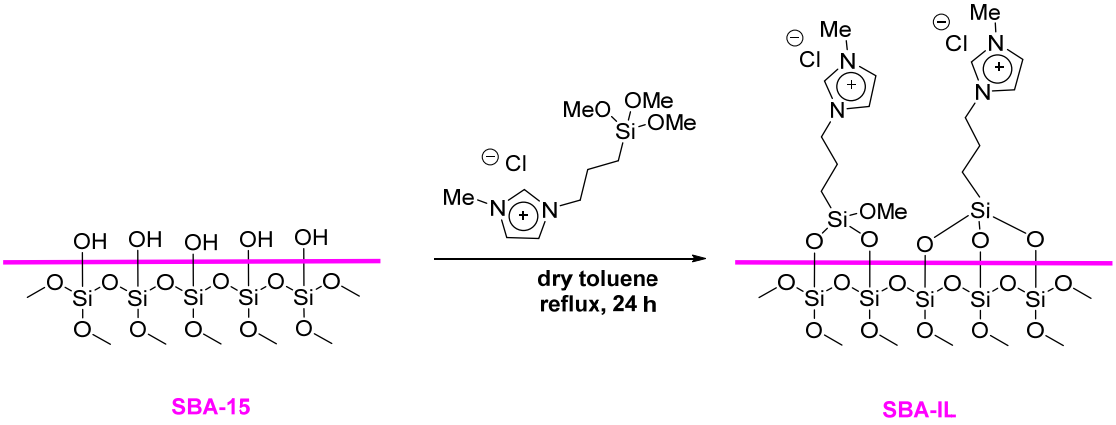
In order to functionalize the surface of SBA-15, trimethoxysilylimidazolium **10** was firstly prepared as shown in Scheme 3. Then, it was added to a mixture of SBA-15 in toluene and heated under reflux condition for about 24 h (Scheme 4). The obtained crude SBA-IL was washed well with CH_2Cl_2 and dried for further characterization.

Table 4. Antimicrobial activities of benzochromene derivatives against Gram-positive and gram-negative bacteria as determined by disc diffusion and MIC method.

	4a	**4b**	**4c**	**4d**	**4e**	**4f**	**4g**	**4h**	**4i**	**4j**	**4k**	Tetracycline
S. aureus	0.2	-	0.09	0.14	0.23	0.22	0.05	-	0.17	-	0.08	0.01
E. coli	0.3	-	0.01	0.2	0.31	0.3	0.05	-	0.17	-	0.1	0.01



Scheme 3. Synthesis of *N-methyl-N′-propyl trimethoxysilylimidazolium chloride*



Scheme 4. Preparation of ionic liquid functionalized SBA-15 (SBA-IL).

By comparing the FT-IR spectrum of the SBA-15 and SBA-IL (Fig. 1), modification of SBA-15 surface with imidazolium groups is completely obvious. The absorption bands at 1100, 960 and 800 cm^{-1} correspond to the Si–O–Si asymmetric stretching, S–OH symmetric stretching and Si–O symmetric stretching vibrations, respectively. A broad band at 3403 cm^{-1} is due to the vibration of quaternary amine salt. The vibration band at 3107 cm^{-1} is for -HC=CH- aromatic ring of imidazolium groups. The band at 2948 cm^{-1} is for asymmetric vibration of CH_2 and CH_3 of organic groups onto the SBA-IL. Additionally, the band at 1569 cm^{-1} is for out of plane stretching of N-Me in imidazolium moiety.

The N_2 adsorption-desorption isotherms (Fig. 2) of SBA-15 and SBA-IL exhibited a typical irreversible type IV isotherm with an H1 hysteresis loop. Furthermore, the isotherm of SBA-IL shows a slight decreasing trend in overall nitrogen due to the grafting the organic groups onto the pores of SBA-15. The pore diameters of both SBA-15 and SBA-IL were obtained by BJH technique while surface area (S_{BET}) and the total pore volumes (V_{total}) were calculated from BET. The results are shown in Table 5 and confirmed that the internal surface of SBA-15 was functionalized with imidazolium groups since these properties were dropped, significantly.

The TGA curve of SBA-IL (Fig. 3) proved that the propyl imidazolium groups were grafted onto the pores of SBA-15. The weight reduction in TGA at the temperature below 200 °C is owing to the loss of physically adsorbed water molecules through hydrogen bonds. The weight loss at the temperature range between 200-800 °C shows that the amount of propyl imidazolium groups is about 2.0 mmol/g. DTA plot displays a weak exothermic peak between 120 and 330 °C, which is for the loss of water. An intense exothermic peak in the range of 330-580 °C is due to the decomposition of the organic groups.

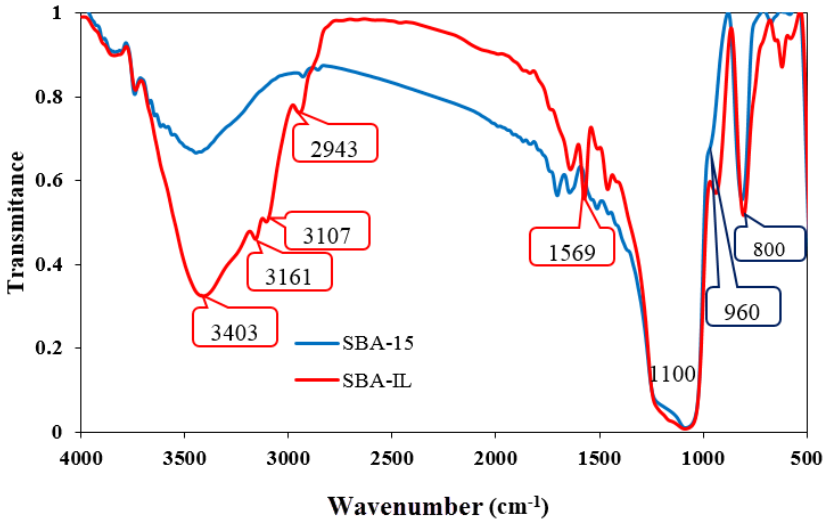


Fig. 1. FT-IR spectra of SBA-15 and SBA-IL.

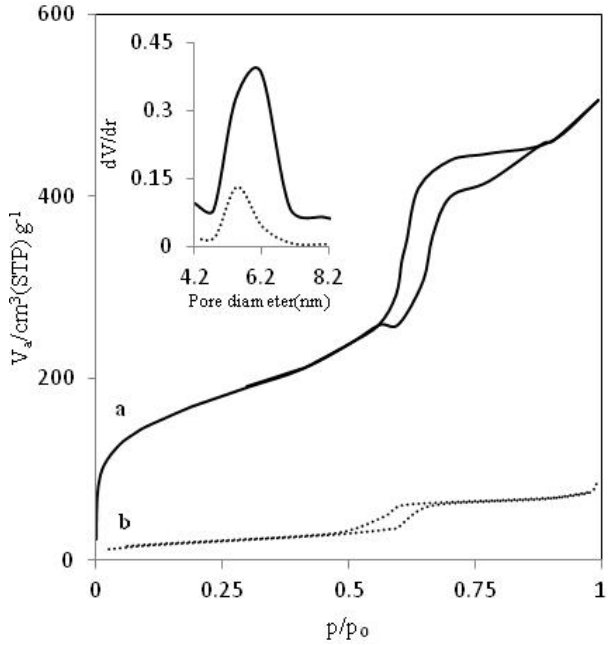


Fig. 2. N_2 adsorption-desorption isotherms of (a) SBA-15, (b) SBA-IL; (inset) BJH pore size distribution curves.

SEM image of SBA-IL (Fig. 4) displays uniform particles with dimensions of 480-857 nm. The same morphology was previously observed for SBA-15. Therefore, it proves that the surface morphology of SBA-IL was preserved without any changes during the modification procedure.

4. Conclusions

In conclusion, we could introduce a synthetic procedure for modifying SBA-15 with imidazolium groups. Then, SBA-IL was characterized and used as an efficient nanocatalyst for the synthesis of benzochromene derivatives *via* three-component reactions under solvent-free conditions. This procedure was a promising method and provided some advantages such as easy workup, high yield of products, and short reaction time. Additionally, the obtained benzochromenes exhibited antibacterial activity as well as tetracycline. The proposed mechanism for antibacterial action was same as that of for tetracycline due to similarity of benzochromenes with tetracycline.

Table 5. The pore diameters (D_{BJH}), BET surface area (S_{BET}) and the total pore volumes (V_{total}) from nitrogen adsorption-desorption for the SBA-15 and SBA-IL.

Molecular sieves	D_{BJH} (nm)	S_{BET} (m^2/g)	V_{total} (cm^3/g)
SBA-15	6.2	587	0.780
SBA-IL	5.5	71	0.128

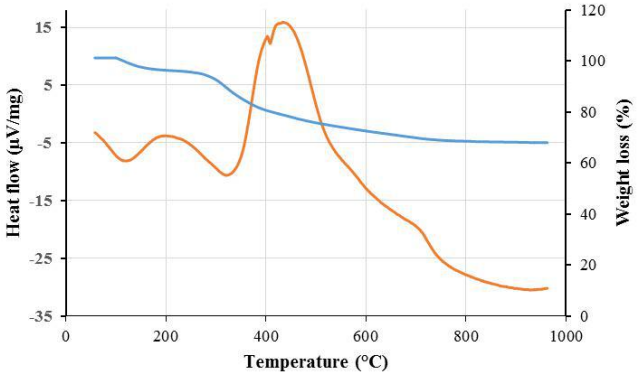


Fig. 3. TGA and DTA curves of SBA-IL.

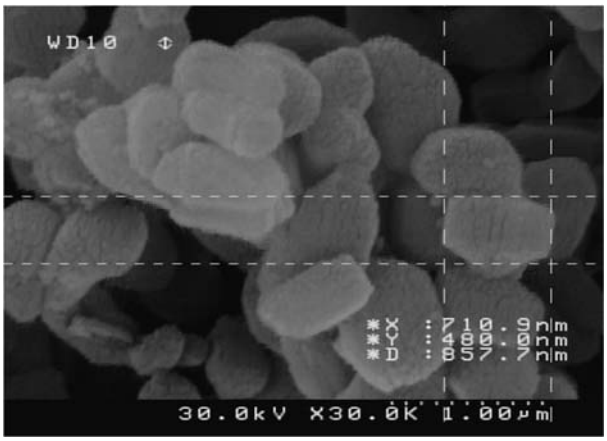


Fig. 4. SEM image of SBA-IL.

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