

RESEARCH PAPER

## A Green Multicomponent One-pot Synthesis of 9, 10-Diaryl-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one Derivatives using Nanoporous Base Silica (SBA-Pr-NH<sub>2</sub>) as Catalyst

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### ABSTRACT

Organic-inorganic hybrid mesoporous materials such as amino functionalized silica (SBA-Pr-NH<sub>2</sub>) have received considerable attention due to their basic catalytic applications and adsorption functions. In this work, we investigated the efficient role of SBA-Pr-NH<sub>2</sub> as a heterogeneous nano catalyst in the synthesis of 9,10-diaryl-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one derivatives. Application of SBA-Pr-NH<sub>2</sub> as an eco-friendly and reusable catalyst resulted in reducing reaction time and temperature in this method. The preparation method was developed using a green one-pot three-component reaction of acenaphthoquinone, benzils and ammonium acetate under solvent-free condition. Besides, the synthesis of benzil derivatives as starting material was carried out. The new compounds were characterized by IR, mass, and NMR spectroscopy. The significant merits of this protocol are its simplicity, short reaction times, low reaction temperature, catalyst reusability, and non-chromatographic purification of products. Also, imidazole and benzimidazole moieties represent significant substructures of a wide variety of bioactive compounds and also significant structural motifs in biological systems, natural products, and drugs.

### How to cite this article

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### INTRODUCTION

The SBA-15 (SBA = Santa Barbara Amorphous) mesoporous silica with high specific surface area, great pore wall thickness, large pore size, and high thermal stability is a unique inorganic solid support [1]. Surface modification of mesoporous materials has a significant role in the employment of these materials in many interesting applications [2, 3]. Recently, organic-inorganic hybrid mesoporous materials such as amino functionalized nanoporous silica (SBA-Pr-NH<sub>2</sub>) have received considerable attention with regard to applications as excellent adsorbents and heterogeneous catalysts [4-6]. Moreover, amino-functionalized mesoporous silica SBA-15 materials

were found to be beneficial as support for drug delivery systems [7] and enzyme immobilization [8], absorbent for removal of heavy metals [9] and finally spacer for further chemical modifications [10].

Imidazole and benzimidazole derivatives are an important class of N-containing heterocyclic organic compounds that found to possess various biological and pharmacological properties [11] such as antibacterial [12], anti-inflammatory [13], antimalarial [14], and cyclin-dependent kinase inhibitors [15]. They are also common structural motifs in marine natural products that represent key moieties responsible for observed biological activities [16, 17]. Despite their importance from

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pharmacological point of view, with a broad range of applications in many different fields, imidazoles have drawn a great deal of interest. They are used in photography as photosensitive compounds [18] and as fluorescent emitting materials for organic light-emitting diodes (OLEDs) [19, 20].

As part of our program for applying heterogeneous solid catalysts to multicomponent reactions [21-26], in this work, we investigate the effective role of SBA-Pr-NH<sub>2</sub> as a nano base catalyst in the synthesis of fused imidazoles, 9,10-diaryl-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one derivatives from the three-component reaction of acenaphthoquinone, benzils, and ammonium acetate (Fig. 1). To the best of our knowledge, there is only one report for the synthesis of these compounds through the heating a mixture of acenaphthoquinone, a benzil, and ammonium acetate under solvent-free conditions which resulted in the formation of 7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-ones in 3 h and at 200 °C and the products were purified by column chromatography [27]. Herein, application of SBA-Pr-NH<sub>2</sub> as an eco-friendly and reusable catalyst resulted in reducing reaction time and temperature in this method.

## MATERIALS AND METHODS

All chemicals were obtained commercially and used without further purification. IR spectra were established from KBr disk using a FT-IR Bruker Tensor 27 instrument. Melting points were measured using the capillary tube method with an Electrothermal 9200 apparatus. The <sup>1</sup>H NMR and <sup>13</sup>C NMR (300 MHz and 100 MHz) were run on a Bruker DPX using TMS as an internal standard (CDCl<sub>3</sub> solution). Gas chromatography-mass spectrometry (GC-MS) analysis was performed on an Agilent 6890-5973 GC/MS detector.

Nitrogen adsorption-desorption isotherms were recorded at -196 °C using a BELSORP-mini II. All samples were degassed at 100 °C for 3 h under vacuum and argon gas flow before analysis. The specific surface area ( $S_{BET}$ ) was evaluated using the Brunauer-Emmett-Teller (BET) equation, and the pore size distribution ( $D_{BJH}$ ) was measured from the desorption branches by means of the Barret-Joyner-Halenda (BJH) model, with the pore volume being taken at  $P/P_0 = 0.995$ . Transmission electron microscopy (TEM) analysis was obtained on a Tecnai G<sup>2</sup> F30 at 300 kV.

### SBA-15 nanoporous silica: Synthesis and functionalization

The nanoporous compound SBA-15 was synthesized and functionalized according to our previous report [28].

### General procedure for the preparation of benzil compounds (2c-2e) [29]

In the first step, benzaldehyde (47.5 mL, 0.47 mol) and a solution of 5 g sodium cyanide (96%) were refluxed in water (50 mL) for 4 h. The reaction mixture was cooled in iced water, and then the crude benzoin was obtained after filtering and washing with cold water. In the second step, 20 g benzoin was heated with concentrated nitric acid (100 mL) on the water-bath until the evolution of nitrogen oxides from the stirring mixture was ceased. After pouring the reaction mixture into the cold water benzil crystals (**2c-e**) were obtained and recrystallized in ethanol.

### General procedure for the preparation of products (4a-h)

The SBA-Pr-NH<sub>2</sub> (0.02 g) was activated in vacuum at 100 °C to remove any surface humidity and then

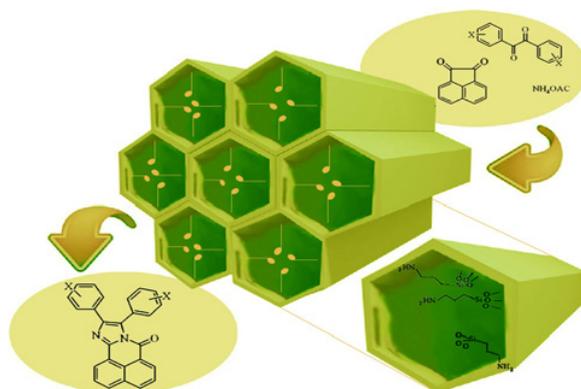


Fig. 1. Synthesis of 9,10-diaryl-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-ones in the presence of SBA-Pr-NH<sub>2</sub> as a nano-reactor.

was cooled to room temperature. A mixture of acenaphthoquinone (1 mmol), benzils (1 mmol), and ammonium acetate (2 mmol) was added to the catalyst under solvent-free condition, and the resulting mixture was stirred at 100 °C for 15-40 min. Upon completion of the reaction (monitored by TLC), the resulting solid product was dissolved in dimethyl formamide (DMF), and was filtered for removing the unsolvable catalyst and then the filtrate was cooled and crystallized. In addition, the recovered catalyst could be washed sequentially with diluted aqueous Et<sub>3</sub>N solution, water, and acetone to reuse without any noticeable loss in reactivity.

#### Selected spectral data

##### 9,10-Bis(4-methoxyphenyl)-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one (4c)

Yellow crystals (yield 66%); mp: 253-254 °C. IR (KBr,  $\nu_{\max}$  (cm<sup>-1</sup>)): 3061, 2925, 2855, 1719 (C=O), 1595, 1305, 1013, 830, 776. <sup>1</sup>H NMR (350 MHz, CDCl<sub>3</sub>):  $\delta$  3.83 (s, 3H, OMe), 3.84 (s, 3H, OMe), 7.81-7.83 (m, 5H, Ar-H), 8.03 (d, J = 7.0, 3H, Ar-H), 8.24-8.27 (4H, Ar-H), 8.54 (d, J = 7.3, 2H, Ar-H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>):  $\delta$  59.8 (OMe), 60.6 (OMe), 120.0, 123.8, 124.1, 125.3, 125.8, 126.1, 128.3, 129.0, 129.4, 129.6, 130.9, 131.1, 132.5, 132.9, 142.8, 143.3, 143.5, 151.9, 152.6, 160.8 (C=O). EI-MS: 432 (M<sup>+</sup>), 391, 282, 182, 154, 126, 74, 63, 50.

##### 9,10-Bis(3-methoxyphenyl)-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one (4e)

Yellow crystals (yield 68%); mp: 248-250 °C. IR (KBr,  $\nu_{\max}$  (cm<sup>-1</sup>)): 3051, 2923, 1693 (C=O), 1535, 1470, 1277, 819, 764. <sup>1</sup>H NMR (350 MHz, CDCl<sub>3</sub>):  $\delta$  3.07 (s, 3H, OMe), 3.08 (s, 3H, OMe), 7.80-7.82 (m, 5H, Ar-H), 8.05 (d, J = 7.0, 4H, Ar-H), 8.26 (d, J = 8.3, 4H, Ar-H), 8.40 (d, J = 8.0, 1H, Ar-H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>):  $\delta$  56.4 (OMe), 56.9 (OMe), 122.3, 123.6, 124.0, 125.5, 125.8, 126.2, 128.3, 129.2,

129.4, 129.6, 130.8, 131.1, 132.4, 132.9, 142.6, 143.3, 143.5, 151.9, 152.6, 160.9 (C=O). EI-MS: 432 (M<sup>+</sup>), 344, 315, 288, 172, 164, 152, 138, 75, 50.

##### 9,10-Bis(2,3-dichlorophenyl)-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one (4f)

Yellow crystals (yield 76%); mp >300 °C. IR (KBr,  $\nu_{\max}$  (cm<sup>-1</sup>)): 3053, 2924, 2854, 1723 (C=O), 1586, 1305, 1124, 1012, 776. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.83-7.91 (m, 4H, Ar-H), 8.14 (d, J = 7.0, 2H, Ar-H), 8.30-8.36 (m, 4H, Ar-H), 8.65 (d, J = 7.3, 2H, Ar-H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>):  $\delta$  120.2, 120.5, 123.5, 124.3, 126.1, 126.4, 126.9, 128.6, 129.2, 129.4, 129.8, 130.8, 131.1, 132.0, 132.3, 142.1, 142.6, 142.9, 150.9, 151.5, 160.3 (C=O). EI-MS: 508 (M<sup>+</sup>), 344, 198, 182, 154, 126, 98, 74, 50.

##### 9,10-Bis(2,6-dichlorophenyl)-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one (4g)

Yellow crystals (yield 81%); mp: 255-256 °C. IR (KBr,  $\nu_{\max}$  (cm<sup>-1</sup>)): 3051, 2923, 1718 (C=O), 1598, 1304, 1013, 830, 756. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.82-7.90 (m, 4H, Ar-H), 8.12 (d, J = 7.0, 3H, Ar-H), 7.29-8.36 (m, 4H, Ar-H), 8.36 (d, J = 7.3, 1H, Ar-H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>):  $\delta$  122.8, 123.6, 124.5, 126.6, 126.8, 126.9, 128.3, 129.2, 129.5, 129.9, 130.6, 131.0, 132.2, 132.5, 142.2, 142.4, 142.8, 150.8, 151.0, 160.6 (C=O). EI-MS: 508 (M<sup>+</sup>), 475, 391, 369, 344, 293, 216, 154, 126, 105, 77, 57.

##### 9,10-Bis(3-nitrophenyl)-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one (4h)

Yellow crystals (yield 79%); mp >300 °C. IR (KBr,  $\nu_{\max}$  (cm<sup>-1</sup>)): 3061, 2924, 1694 (C=O), 1535, 1469, 1335, 1280, 908, 819, 766. <sup>1</sup>H NMR (300 MHz, CDCl<sub>3</sub>):  $\delta$  7.62-7.71 (m, 3H, Ar-H), 7.78-7.89 (m, 4H, Ar-H), 8.08-8.11 (m, 3H, Ar-H), 8.32 (d, J = 8.2, 1H, Ar-H), 8.42 (d, J = 7.0, 1H, Ar-H), 8.77 (d, J = 7.4, 1H, Ar-H), 8.88 (d, J = 7.3, 1H, Ar-H). <sup>13</sup>C NMR (100 MHz, CDCl<sub>3</sub>):  $\delta$  121.4, 121.8, 123.1, 124.6, 125.2,

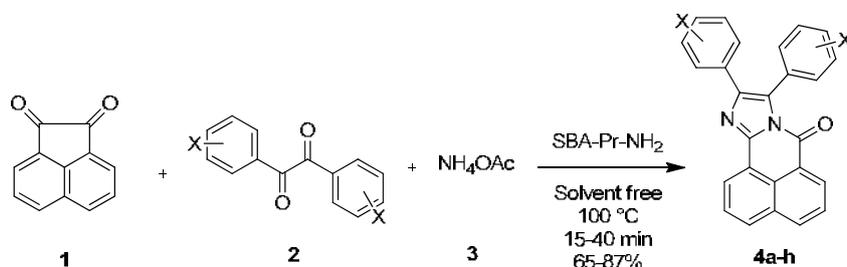


Fig. 2. Synthesis of 9,10-diaryl-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one derivatives 4a-h in the presence of SBA-Pr-NH<sub>2</sub>.

125.5, 126.1, 128.0, 129.2, 129.3, 129.5, 131.0, 131.4, 131.9, 132.4, 132.6, 141.3, 141.6, 146.3, 146.9, 150.4, 160.5 (C=O). EI-MS: 462 (M<sup>+</sup>), 340, 293, 218, 172, 154, 126, 105, 77, 57.

## RESULTS AND DISCUSSION

A green one-pot synthesis of 9,10-diaryl-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one derivatives **4** was performed by the reaction of acenaphthoquinone **1**, various benzils **2**, and ammonium acetate **3** in the presence of SBA-Pr-NH<sub>2</sub> as a heterogeneous base catalyst (Fig. 2). The goals of this study were: (1) using a suitable heterogeneous base catalyst in a multicomponent reaction, (2) illustration of this heterogeneous catalyst efficiency, and (3) applying the present

catalytic system in the synthesis of a variety of fused imidazoles.

In order to optimize the reaction condition, various solvents such as CH<sub>3</sub>CN, H<sub>2</sub>O, EtOH, DMF, and also solvent-free system were studied for the synthesis of 9,10-diaryl-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-ones (Table 1). It was found that solvent-free condition is the most effective system for the reaction of acenaphthoquinone **1**, benzil **2** and ammonium acetate **3** which affords the desired product **4a** in higher yield (87%) within 15 min at 100 °C in the presence of SBA-Pr-NH<sub>2</sub> (Table 1, entry 7).

After completion of the reaction (monitored by TLC), the crude product was dissolved in hot DMF, the heterogeneous solid catalyst was simply

Table 1. The optimization of reaction condition.

Entry	Solvent	Temperature (°C)	Catalyst	Time (min)	Yield (%)
1	CH <sub>3</sub> CN	Reflux	SBA-Pr-NH <sub>2</sub>	15	65
2	H <sub>2</sub> O	Reflux	SBA-Pr-NH <sub>2</sub>	2 h	trace
3	EtOH	Reflux	SBA-Pr-NH <sub>2</sub>	20	70
4	DMF	rt.	-	30	67
5	DMF	Reflux	SBA-Pr-NH <sub>2</sub>	20	75
6	EtOH	rt.	SBA-Pr-NH <sub>2</sub>	2 h	trace
7	Solvent free	100	SBA-Pr-NH <sub>2</sub>	15	87
8	Solvent free	200	-	3 h	92 [27]

Table 2. Synthesis of 9,10-diaryl-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one derivatives in the presence of SBA-Pr-NH<sub>2</sub>.

Entry	X	Product	Time (min)	Yield (%)	mp (°C)	mp [Lit]
1	H	4a	15	87	>300	>300 [27]
2	4-Cl	4b	25	85	>300	>300 [27]
3	4-OMe	4c	30	66	253-254	Not reported
4	4-Me	4d	25	65	>300	>300 [27]
5	3-OMe	4e	25	68	248-250	Not reported
6	2,3-Cl <sub>2</sub>	4f	35	76	>300	-
7	2,6-Cl <sub>2</sub>	4g	35	81	255-256	-
8	3-NO <sub>2</sub>	4h	40	79	>300	-

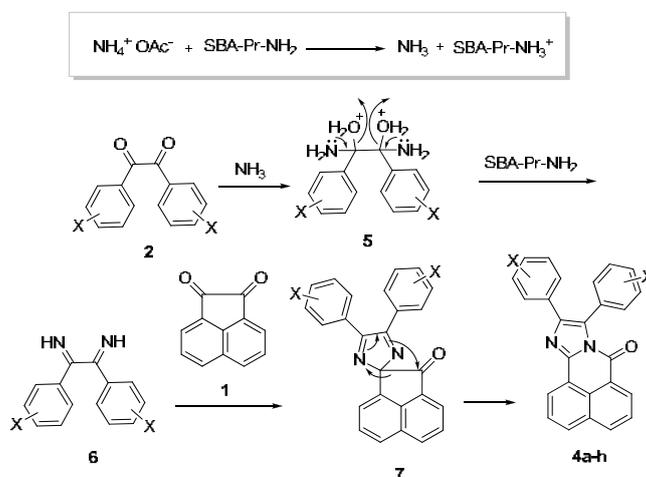


Fig. 3. The proposed mechanism.

removed by filtration, and after cooling of the filtrate during the time, the pure yellow crystals were obtained. The catalyst can be reactivated, and then reused without considerable loss of reactivity. The new compounds were characterized by IR, mass, and NMR spectroscopy. The desired products were illustrated in Table 2.

The proposed mechanism for the synthesis of the fused imidazoles **4a-h** starts by crucial role of SBA-Pr-NH<sub>2</sub> in activating ammonium acetate as the source of ammonia. The condensation of benzil **2** and ammonium acetate **3** is occurred by releasing two water molecules to form imine intermediate **6**, which subsequently is condensed with carbonyl group of acenaphthoquinone **1** to form the intermediate **7**. Finally, a ring opening-closure occurs to form target compounds **4a-h**, as illustrated in Fig. 3.

Synthesis of benzil derivatives was performed at the first step by condensation of aromatic aldehydes treated with an alkali sodium cyanide in aqueous solution to prepare  $\alpha$ -hydroxyketone or benzoin **9**. In the second step, oxidation of

benzoin was performed in the presence of nitric acid to afford benzil derivatives **2c-2e** (Fig. 4). The results are summarized in Table 3.

The characterization of the nanocatalyst by different techniques such as N<sub>2</sub> adsorption-desorption isotherm, XRD, FT-IR spectra, and TEM image was discussed in our previous reports [28, 32]. For example, three textural parameters of SBA-15 and SBA-Pr-NH<sub>2</sub> including specific surface area (BET method), total pore volume, and pore diameter (BJH method) are provided in Table 4. A decrease in pore size and BET surface area, as well as pore volume are apparently indicative of the successful attachment of organic moieties within the mesopores of SBA-15.

Fig. 5 displays the low-angle XRD patterns of SBA-15 and SBA-Pr-NH<sub>2</sub>. There were three well-resolved reflections for SBA-Pr-NH<sub>2</sub> similar to that for SBA-15 which demonstrates that functionalized samples possess mesoordered two-dimensional hexagonal structures.

The TGA analysis of SBA-Pr-NH<sub>2</sub> (Fig. 6) confirmed the amount of grafted organic groups

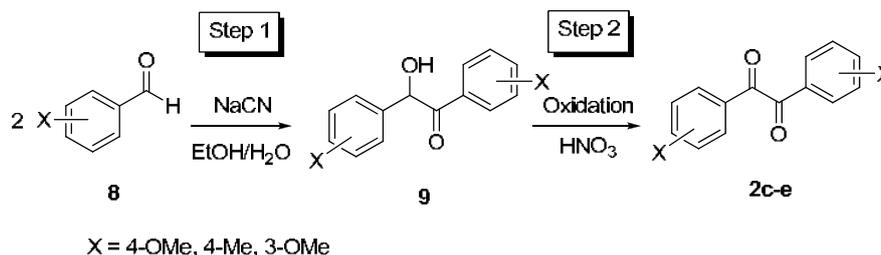


Fig. 4. Synthesis of benzil derivatives.

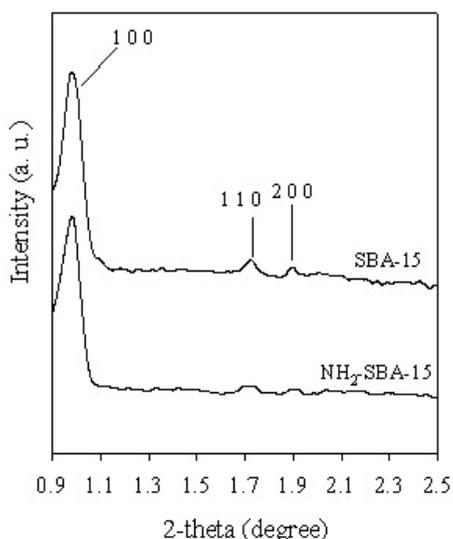


Fig. 5. Low-angle XRD patterns of SBA-15 and SBA-Pr-NH<sub>2</sub>.

Table 3. Preparation of various synthetic benzil compounds (2c-e).

Entry	X	Product	Yield (%)	mp (°C)
1	4-OMe	2c	81	131-133 [30]
2	4-Me	2d	80	100-102 [30]
3	3-OMe	2e	85	81-82 [31]

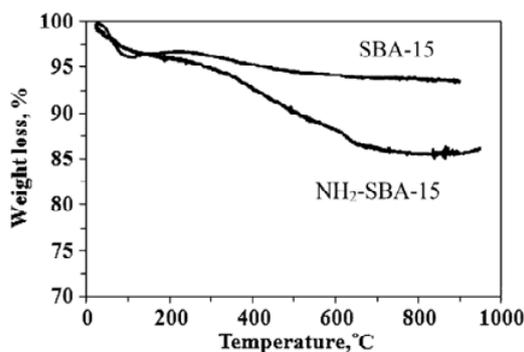
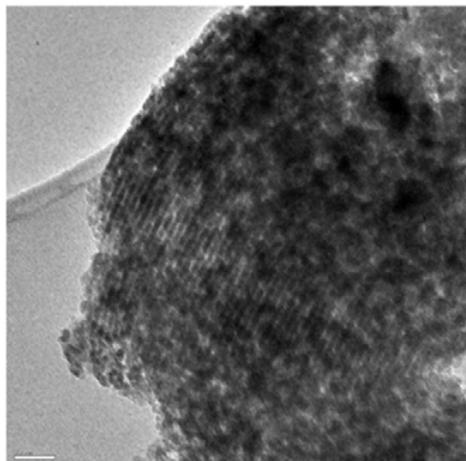


Fig. 6. Thermogravimetric analysis (TGA) of SBA-Pr-NH<sub>2</sub>.

Table 4. Characteristics of the synthesized materials derived from nitrogen adsorption-desorption.

Sample	Textural properties		
	$S_{BET}$ (m <sup>2</sup> /g)	$V_{total}$ (cm <sup>3</sup> /g)	$D_{BJH}$ (nm)
SBA-15	481	1.3	5.9
SBA-Pr-NH <sub>2</sub>	356	1.0	3.6

Fig. 7. TEM image of SBA-Pr-NH<sub>2</sub>.

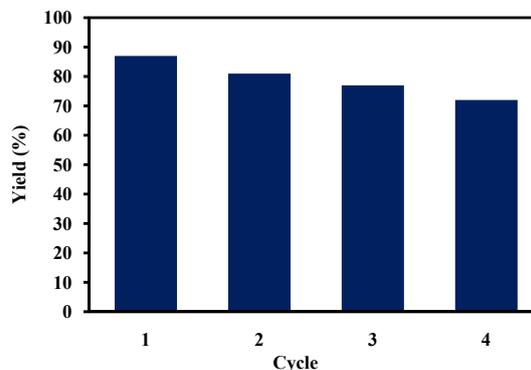
on the surface of SBA-15. The weight reduction of SBA-Pr-NH<sub>2</sub> in the temperature range between 200 and 600 °C indicated the amount of organic functional groups.

The TEM image was employed to directly visualize the pore geometry of SBA-Pr-NH<sub>2</sub> (Fig. 7). The TEM image reveals parallel channels resembling the configuration of the pores of SBA-15. This indicates that the pores of SBA-Pr-NH<sub>2</sub> have not been collapsed during the functionalization reaction.

Finally, the recovered catalyst could be washed sequentially with diluted aqueous Et<sub>3</sub>N solution, water, and acetone to reuse without any noticeable loss in reactivity. Reusability of the catalyst was evaluated under optimized conditions for the preparation of the model compound **4a**. As illustrated in Fig. 8, the process of recycling was completed four times with no significant decrease in the activity of the catalyst. The yields for the four runs were established to be 87, 81, 77, and 72%, respectively.

## CONCLUSIONS

In summary, we have successfully synthesized 9,10-diaryl-7H-benzo[d,e]imidazo[2,1-a]isoquinolin-7-one derivatives in the presence of amino functionalized nanoporous silica (SBA-Pr-NH<sub>2</sub>) as an efficient catalyst under solvent-free conditions. The attractive features of this protocol are simple method, short reaction times,

Fig. 8. Reusability of SBA-Pr-NH<sub>2</sub> in the synthesis of the model compound **4a**.

low reaction temperature, simple workup, and non-chromatographic purification of products. Catalyst reusability and ease of recovery makes this method a benign, economic and waste-free chemical procedure.

## ACKNOWLEDGEMENTS

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## CONFLICT OF INTERESTS

The authors declare that there is no conflict of interests regarding the publication of this paper.

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