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ORIGINAL PAPER

An efficient, one-pot synthesis of pyrido[2,3-*d*:6,5-*d*′]dipyrimidines using SBA-15-supported sulfonic acid nanocatalyst under solvent-free conditions

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Abstract An acidic nanocatalyst SBA-15-SO_3H with a high surface area and high acid content was readily synthesized, characterized, and effectively used in the synthesis of some new and known pyrido[2,3-*d*:6,5-*d*′]dipyrimidine derivatives by condensation reaction of 6-aminouracil and different aldehydes under solvent-free conditions. The catalyst was reused for 6 runs without significant loss of its activity. Short reaction times, high yield of the products, solvent-free conditions, and use of a robust and reusable catalyst are worthwhile advantages of the present method.

Keywords Nanocatalyst · SBA-15-SO_3H · Pyrido[2,3-*d*:6,5-*d*′]dipyrimidine · Solvent-free

Introduction

Dipyrimidines are useful heterocyclic compounds with a broad range of biological, medicinal, and pharmacological properties, such as antitumor [1], antibacterial [2], antifungal [3], antiviral [4], and anti-oxidant [5]. In spite of the wide variety applications of dipyrimidines, only few methods have been reported for the preparation of these compounds. Dipyrimidines are generally synthesized by the reaction of active methylene carbons with 5-dimethylaminomethylene-6-imino-1,3-dimethyluracil (the Vilsmeier intermediate) [6–12]. Reaction of barbituric acid with aldehydes and aminouracil derivatives also gave fused dipyrimidines [13–15]. All of the aforementioned methods for the preparation of dipyrimidines suffer from one or more drawbacks, such as long reaction times, use of toxic solvent, tedious work-up, low yield of products, and use of microwave irradiation.

Due to the importance of dipyrimidines and to overcome the drawbacks of the reported methods, there is a still need to develop a new route to synthesize these compounds. Designing nanocatalysts is an interesting area in organic synthesis due to their high surface area, stability, durability, activity as well as their reusability. SBA-15 (Santa Barbara Amorphous-15) mesoporous nanocatalyst, with high surface area, well ordered hexagonal arrays of cylindrical channels, fine pore size distributions, high thermal stability, large pore size, thick silica walls, and appreciable number of silanol groups at the surface of its channels, is an interesting candidate for the support in catalytic reactions [16–18]. This efficient support can be modified to extend its physical and chemical properties. Acidifying the SBA-15 can boost its acid character in order to use it as an efficient, heterogeneous, and reusable catalyst in acid-catalyzed reactions.

In continuation of our interest toward the synthesis and application of new catalytic systems for the synthesis of heterocyclic compounds [19–25], we wish to report a simple and green method for the synthesis of pyrido[2,3-*d*:6,5-*d*′]dipyrimidine derivatives by the use of sulfonic acid-modified SBA-15 (SBA-15-SO_3H) as a new catalyst (Scheme 1).

Experimental

All chemicals were purchased from Sigma-Aldrich and Merck companies. X-ray powder diffraction (XRD) was implemented on a Philips X'Pert diffractometer using CuKα radiation. The prepared catalyst pore structure

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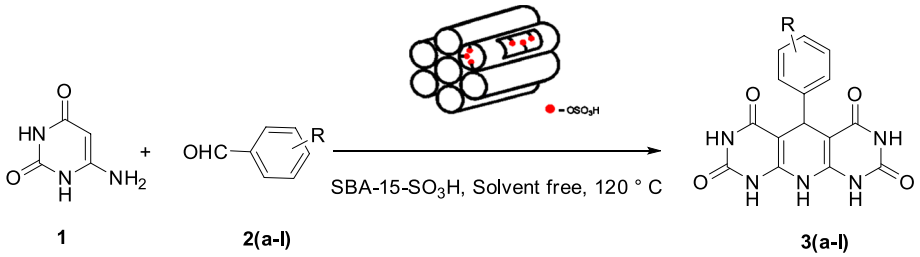
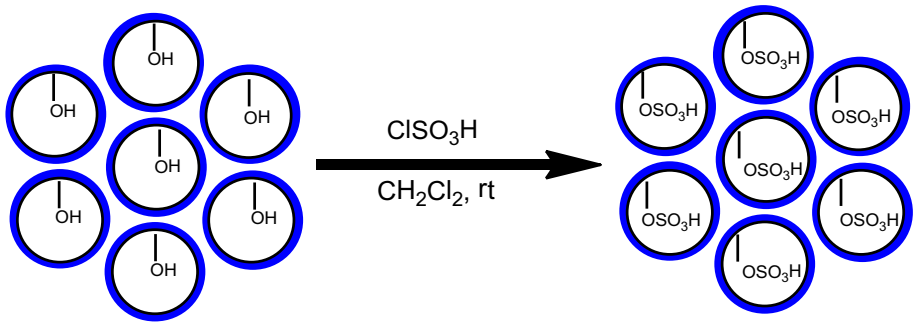
Scheme 1 SBA-15-SO_3H as a novel acid catalyst for the preparation of pyrido[2,3-*d*:6,5-*d′*]dipyrimidine derivatives



Scheme 2 Modification of SBA-15 with chlorosulfonic acid



was verified by the nitrogen sorption isotherm ([5.0.0.3] Belsorp, BEL Japan, Inc.) and also Micrometrics Tristar II 3020. Transmission electron micrographs (TEM) were recorded on a Philips EM-208 instrument on an accelerating voltage of 100 kV. The morphology of the catalyst was determined by scanning the electron microscope model VEGA\\ TESCAN-XMU instrument with an accelerating voltage of 20 kV. Melting points were determined using melting point IA 8103 apparatus; IR spectra were recorded on an ABB Bomem model FTLA200-100 instrument. ^{1}H and ^{13}C NMR spectra were measured on a Bruker DRX-300 spectrometer at 300 and 75 MHz, respectively. Mass spectra were measured on a Shimadzu QP 1100 EX mass spectrometer with 70-eV ionization potential. The number of acid sites on SBA-15-SO_3H was determined by acid–base titration. The thermal analysis (C, H, N) was carried out using Carlo ERBA Model EA 1108 analyzer.

Preparation of SBA-15-SO_3H

SBA-15 was prepared according to the common method reported in the literature [26].

Modification of SBA-15

For modification of the Support (SBA-15), chlorosulfonic acid ($ClSO_3H$) was used as an acidic source which is innovative for modifying SBA-15; 9 mmol of $ClSO_3H$ was added dropwise to 1 g of SBA-15 in 20 mL dichloromethane and stirred for 60 min at room temperature. Then the reaction mixture was filtered and washed with dichloromethane (20 mL) and dried in a vacuum oven to obtain SBA-15-SO_3H (Scheme 2).

General procedure for the synthesis of pyrido[2,3-*d*:6,5-*d′*]dipyrimidine derivatives in the presence of SBA-15-SO_3H under solvent-free conditions

In a reaction vessel, a mixture of 6-aminouracil (2 mmol), aldehyde (1 mmol), and SBA-15-SO_3H (0.05 g) was heated at 120 °C under solvent-free conditions for a specified time (completion of the reaction was monitored by TLC). After completion of the reaction, hot DMF (5 mL) was added and the catalyst was separated by simple filtration. Then water (15 mL) was added to the filtrate to give the solid product.

Spectral data of some compounds

5-(4-Nitrophenyl)-9,10-dihydropyrido[2,3-d:6,5-d′] dipyrimidine-2,4,6,8(1H,3H,5H,7H)-tetraone (Table 3, compound 3b)

IR (KBr, cm^{-1}): 3336, 3150, 1719, 1668. ^{1}HNMR (300 MHz, DMSO) δ (ppm): 10.78 (s, NH), 10.36 (s, NH), 8.97 (s, NH), 8.10 (d, $J = 9$ Hz, 2H-Ar) 7.94 (s, 2NH), 7.43 (d, $J = 9$ Hz, 2H-Ar), 4.83 (s, 1H). Anal.Calcd for $C_{15}H_{10}N_6O_6$: C, 37.66; H, 2.09; N, 13.39. Found: C, 37.84; H, 2.25, N, 13.48.

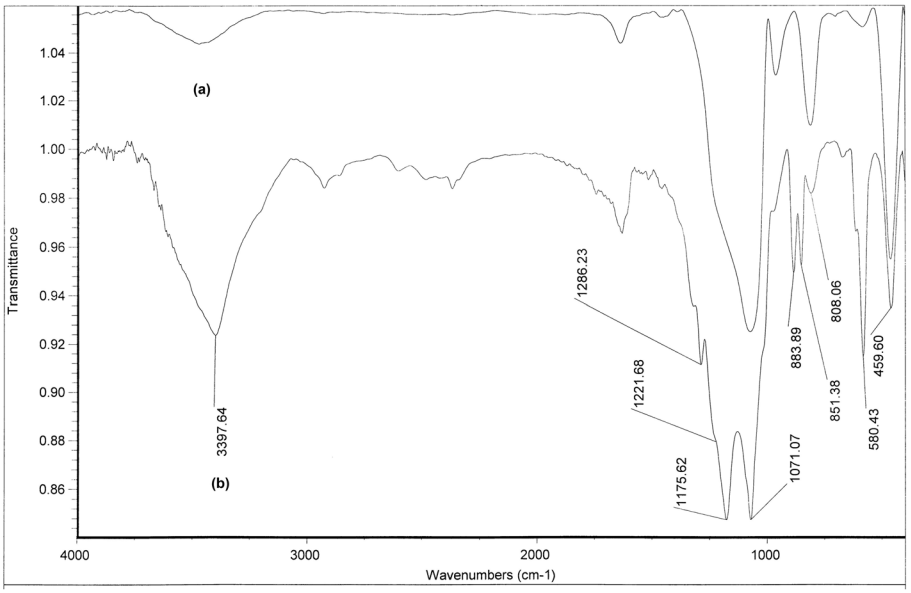


Fig. 1 The IR spectra of **a** SBA-15; **b** SBA-15-SO_3H

5-(4-Chlorophenyl)-9,10-dihydropyrido[2,3-d:6,5-d′] dipyrimidine-2,4,6,8(1H,3H,5H,7H)-tetraone (Table 3, compound 3c)

IR (KBr, cm^{-1}): 3325, 3148, 1720, 1667. ^{1}HNMR (300 MHz, DMSO) δ (ppm): 10.51 (s, 2NH), 10.32 (s, 2NH), 7.15 (d, J = 6 Hz, 2H-Ar), 7.06 (d, J = 6 Hz, 2H-Ar), 6.70 (s, NH), 5.27 (s, 1H). ^{13}CNMR (75 MHz, DMSO) δ (ppm): 165.32, 154. 29, 149.75, 138.68, 129.49, 128.56, 127.53, 85.72, 32.14. MS (*m/z*): 360 (M^++1). Anal.Calcd for $C_{15}H_{10}N_5O_4$ Cl: C, 50.07; H, 2.78; N, 19.47. Found: C, 50.12; H, 2.85, N, 19.53.

5-(2,4-Dichlorophenyl)-9,10-dihydropyrido[2,3-d:6,5-d′] dipyrimidine-2,4,6,8 (1H,3H,5H,7H)- tetraone (Table 3, compound 3d)

IR (KBr, cm^{-1}): 3465, 3327, 3168, 1716, 1634.^{1}HNMR (300 MHz, DMSO) δ (ppm): 10.26 (s, 2NH), 10.19 (s, 2NH), 7.36 (s, H–Ar), 7.29 (d, J = 9 Hz, H–Ar), 7.25 (d, J = 9 Hz, H–Ar), 6.45 (s, NH), 5.23 (s, 1H). ^{13}CNMR (75 MHz, DMSO) δ (ppm): 162.36, 153.96, 149.84, 149.70, 137.87, 133.23, 130.64, 130.45, 128.70, 85.00, 35.82. Anal.Calcd for $C_{15}H_9N_5O_4$ Cl_2: C, 45.68; H, 2.28; N, 17.77. Found: C, 45.70; H, 2.30, N, 17.79.

5-(2-Bromophenyl)-9,10-dihydropyrido[2,3-d:6,5-d′] dipyrimidine-2,4,6,8(1H,3H,5H,7H)-tetraone (Table 3, compound 3e)

IR (KBr, cm^{-1}): 3336, 3168, 1712, 1635. ^{1}HNMR (300 MHz, DMSO) δ (ppm): 10.45 (s, 2NH), 10.21 (s, 2NH), 7.07 (m, 4H-Ar), 6.70 (s, NH), 5.18 (s, 1H). ^{13}CNMR (75 MHz, DMSO) δ (ppm): 169.70, 165.76, 153.60, 149.70, 135.65, 133.22, 127.93, 127.61, 124.00, 84.83, 35.50. Anal.Calcd for $C_{15}H_{10}N_5O_4$ Br: C, 44.56; H, 2.47; N, 17.33. Found: C, 45.02; H, 2.53, N, 17.42.

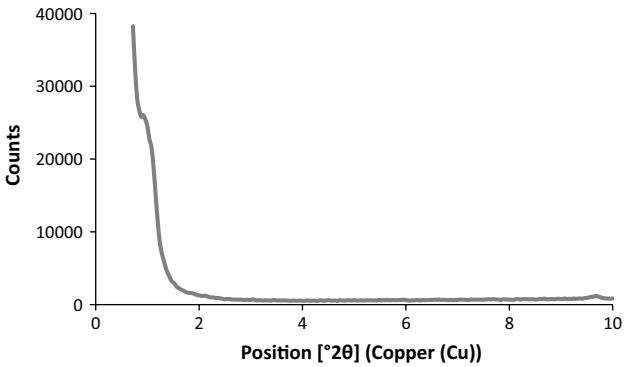


Fig. 2 The XRD pattern of SBA-15-SO_3H in low angle region of 1.0° (2*θ*) to 10.0° (2*θ*)

5-(3-Nitrophenyl)-9,10-dihydropyrido[2,3-d:6,5-d′] dipyrimidine-2,4,6,8(1H,3H,5H,7H)-tetraone (Table 3, compound 3f)

IR (KBr, cm^{-1}): 3354, 3153, 1711, 1632. ^{1}HNMR (300 MHz, DMSO) δ (ppm): 10.62 (s, 2NH), 10.44 (s, 2NH), 7.74 (m, 4H-Ar), 6.73 (s, NH), 5.38 (s, 1H).^{13}CNMR (75 MHz, DMSO) δ (ppm): 165.46, 154.50, 149.81, 147.82, 142.44, 133.89, 129.23, 121.30, 120.39, 85.19, 32.63. Anal. Calcd for $C_{15}H_{10}N_6O_6$: C, 37.66; H, 2.09; N, 13.39. Found: C, 37.70; H, 2.11, N, 13.48.

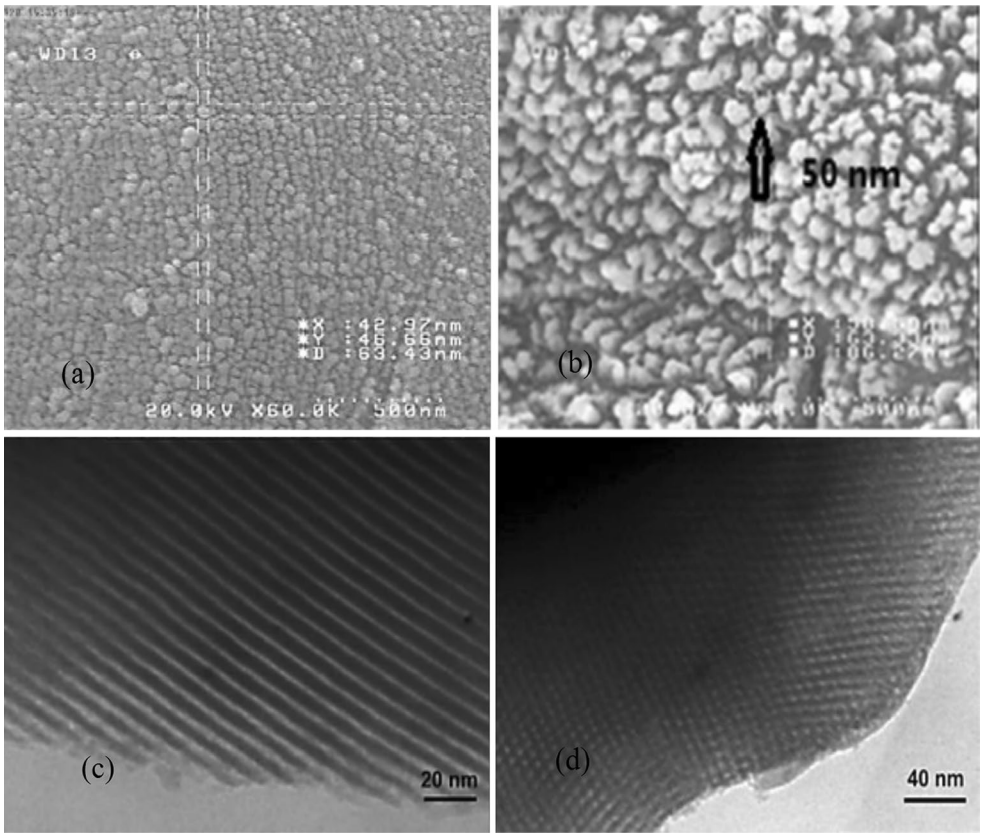


Fig. 3 The SEM images of SBA-15 and SBA-15-SO_3H (**a, b**), TEM images of SBA-15-SO_3H (**c, d**)

Table 1 Surface area, pore volume, and average pore size of SBA-15-SO_3H

Adsorbent	BET surface area (m^2/g)	BJH desorption cumulative volume of pores (cm^3/g)	BJH desorption average pore width (nm)
SBA-15	798	0.73	4.47
SBA-15-SO_3H	318	0.41	5.18

5-(4-Methylphenyl)-9,10-dihydropyrido[2,3-d:6,5-d′] dipyrimidine-2,4,6,8(1H,3H,5H,7H)-tetraone (Table 3, compound 3g)

IR (KBr, cm^{-1}): 3389, 3255, 1732, 1514. ^{1}HNMR (300 MHz, DMSO) δ (ppm): 10.49 (s, 2NH), 10.29 (s, 2NH), 6.98 (d, J = 9 Hz, 2H-Ar), 6.93 (d, J = 9 Hz, 2H-Ar), 6.69 (s, NH), 5.25 (s, 1H), 2.21 (s, 3H, CH_3).^{13} CNMR (75 MHz, DMSO) δ (ppm): 165.42, 162.37, 154.25, 149.84, 136.31, 128.32, 126.51, 86.22, 32.12, 20.54. Anal.Calcd for $C_{16}H_{13}N_5O_4$: C, 56.64; H, 39.23; N, 20.65. Found: C, 56.70; H, 39.28, N, 20.67.

5-(4-Acetamidophenyl)-9,10-dihydropyrido[2,3-d:6,5-d′] dipyrimidine-2,4,6,8(1H,3H,5H,7H)-tetraone (Table 3, compound 3h)

IR (KBr, cm^{-1}): 3339, 3169, 1737, 1638. ^{1}HNMR (300 MHz, DMSO) δ (ppm): 10.48 (s, 2NH), 10.28 (s, 2NH), 7.93 (s, NH), 7.36 (d, J = 9 Hz, 2H-Ar), 6.95 (d, J = 9 Hz, 2H-Ar,) 6.69 (s, NH), 5.23 (s, 1H), 1.73 (s, 3H, CH_3). ^{13} CNMR (75 MHz, DMSO) δ (ppm): 23.90, 35.80, 84.85, 118.63, 126.72, 133.95, 136.42, 149.79, 162.33, 167.91, 169.72.; MS (*m/z*): 382(M^+). Anal.Calcd for $C_{17}H_{14}N_6O_5$: C, 53.40; H, 3.66; N, 21.99. Found: C, 53.50; H, 3.80, N, 22.01.

5-(5-Bromo-2-hydroxyphenyl)-9,10-dihydropyrido[2,3-d:6,5-d′]dipyrimidine-2,4,6,8(1H,3H,5H,7H)-tetraone (Table 3, compound 3j)

IR (KBr, cm^{-1}): 3400, 3289, 3177, 1730. ^{1}HNMR (300 MHz, DMSO) δ (ppm): 11.73 (s, NH), 10.89 (s, NH), 10.03 (s, NH), 10.01 (s, NH), 7.13 (m, 3H-Ar), 6.47 (s, NH), 4.78 (s, 1H).^{13} CNMR (75 MHz, DMSO) δ (ppm):

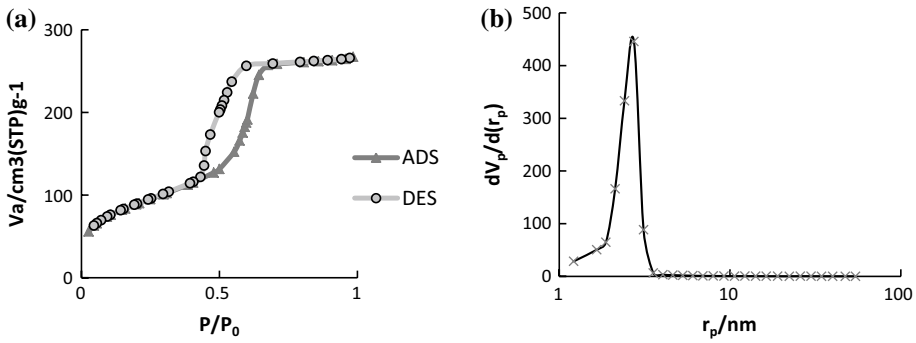


Fig. 4 N_2 adsorption–desorption isotherm and BJH of SBA-15-SO_3H (**a, b**)

Table 2 Optimization of the reaction conditions

Entry	Catalyst (g)	Solvent	Temperature (°C)	Time (min)	Yield (%)
1	–	Solvent free	120	60	Trace
2	0.02	Solvent free	120	60	85
3	0.05	Solvent free	120	10	96
4	0.1	Solvent free	120	10	95
5	0.05	EtOH	Reflux	120	70
6	0.05	CH_3CN	Reflux	120	65
7	0.05	DMF	Reflux	120	75
8	0.05	H_2O	Reflux	120	50
9	0.05	Solvent free	100	60	65
10	0.05	Solvent free	110	60	90
11	0.05	Solvent free	120	10	96
12	0.05	Solvent free	130	10	96

163.31, 154.43, 151.57, 150.18, 149. 62, 148.86, 130.88, 130.16, 127.53, 89.33, 26.70. MS (*m/z*):421 (M^++2). Anal. Calcd for $C_{15}H_{10}N_5O_4Br$: C, 42.96; H, 2.39; N, 16.71. Found: C, 43.15; H, 2.58, N, 16.89.

5-(4-Hydroxyphenyl)-9,10-dihydropyrido[2,3-d:6,5-d′] dipyrimidine-2,4,6,8(1H,3H,5H,7H)-tetraone (Table 3, compound 3i)

IR (KBr, cm^{-1}): 3171, 1712, 1628. ^{1}HNMR (300 MHz, DMSO) δ (ppm) : 10.45 (s, 2NH), 10.23 (s, 2NH), 8.98 (s, OH), 6.81 (d, J = 9 Hz, 2H-Ar), 6.68 (s, NH), 6.57 (d, J = 9 Hz, 2H-Ar), 5.19 (s, 1H).^{13}CNMR (75 MHz, DMSO) δ (ppm): 165.78, 154.71, 153.64, 149.79, 129.17, 127.42, 114.49, 84.85, 35.81. Anal.Calcd for $C_{15}H_{11}N_5O_6$: C, 50.42; H, 3.08; N 19.60. Found: C, 50.55, H, 3.15; N, 19.70.

Catalyst characterization

In the IR spectra (Fig. 1a, b), the bands appearing at 3427 and 3397 cm^{-1} are due to the stretching of the OH groups. In comparison with the IR spectra of SBA-15, increasing the intensity of the OH band in SBA-15-SO_3H confirms that the modification of SBA-15 has been occurred. The existence of strong peaks at 1175 and 1286 cm^{-1} is related to the symmetrical and unsymmetrical stretching S=O bonds in SBA-15-SO_3H. A peak at 580 cm^{-1} can be assigned to S–O bond in the synthesized catalyst. In both spectra (Fig. 1a, b), a peak appeared at 1071 cm^{-1} which is due to the Si–O stretching vibration [27]. The concentration of acidic groups in the SBA-15 determined by acid–base titration showed that the amount of H^+ in the catalyst is 3.45 mmol g^{-1}.

The XRD pattern of SBA-15-SO_3H (Fig. 2) shows a peak at 1.05° (2θ) and one small peak at 9.80° (2θ) which completely confirms the mesoporous structure of the catalyst. The SEM images of SBA-15 (Fig. 3a, b) show spherical morphology before and after the modification of the catalyst with different sizes of spheres. They indicate that the surface of SBA-15 is covered with sulfonic groups after modification (Fig. 3b). It can be concluded that its spherical morphology remains unchanged during the surface modification. Figure 3b shows uniformity in particles. This figure also shows nanoparticles with the size about 50 nm. The TEM images (Fig. 3c, d) show the hexagonal arrangement of the functionalized catalyst.

Table 3 Synthesis of pyrido[2,3-*d*:6,5-*d*′]dipyrimidine derivatives (3) from reaction of 6-aminouracil (1) and aldehydes (2) in the presence of SBA-15-SO_3H as catalyst

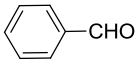
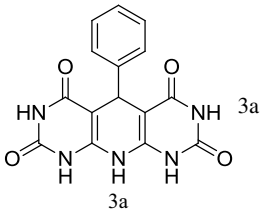
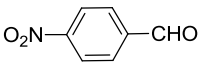
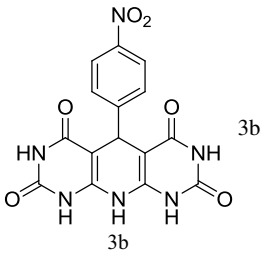
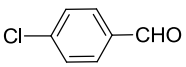
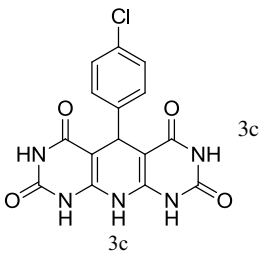
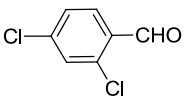
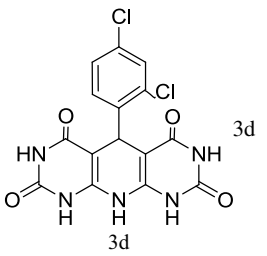
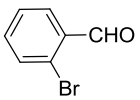
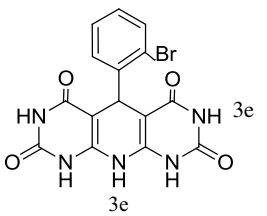
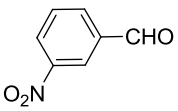
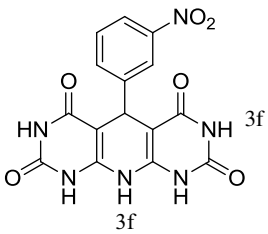
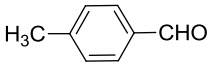
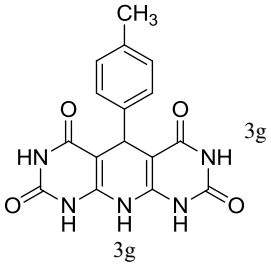
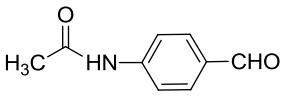
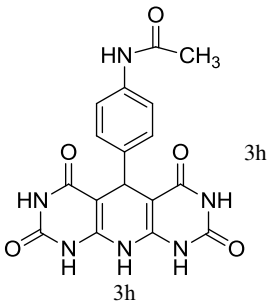
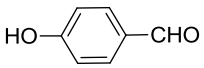
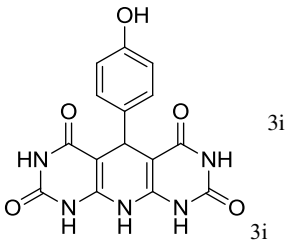
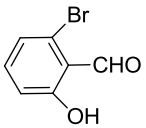
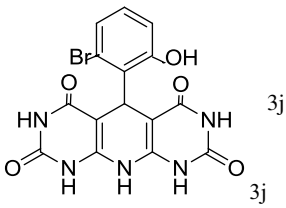
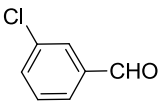
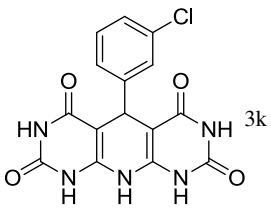
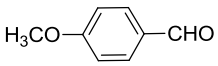
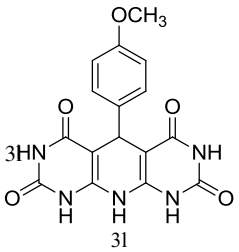
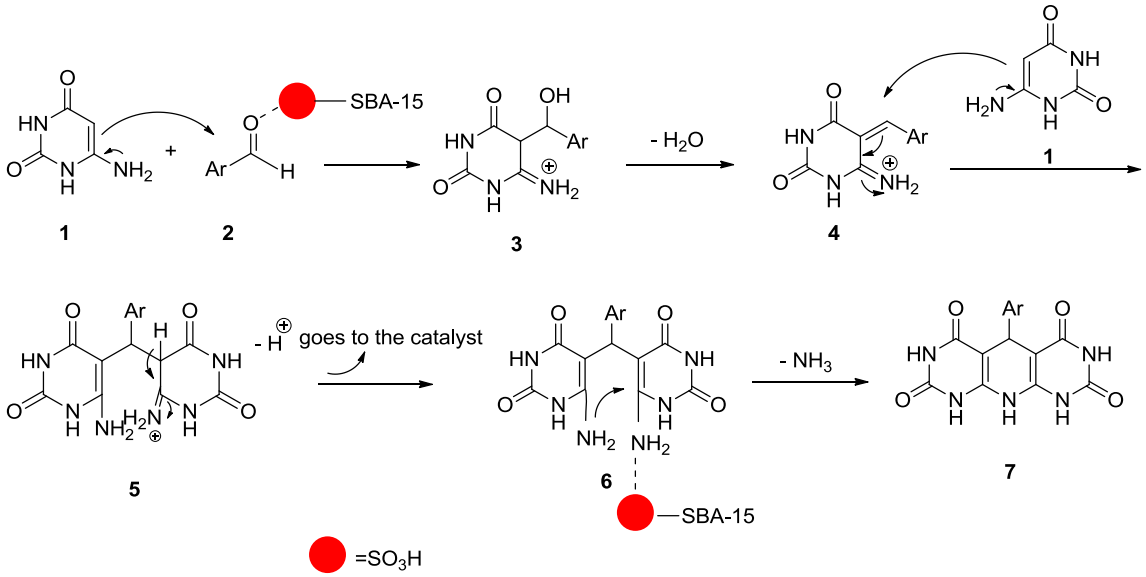
Entry	Aldehyde 2(a–l)	Product[a] 3(a–l)	Time (min)	Yield[b] (%)	Mp (°C)/Ref.
1		 3a	30	96	295/[14]
2		 3b	15	95	230/[14]
3		 3c	10	96	300/[14]
4		 3d	15	85	>300/[14]
5		 3e	60	73	>300
6		 3f	30	98	250/[14]

Table 3 continued

Entry	Aldehyde 2(a–l)	Product[a] 3(a–l)	Time (min)	Yield[b] (%)	Mp (°C)/Ref.
7		 3g	45	88	>300/[14]
8		 3h	60	77	>300
9		 3i	30	90	>300
10		 3j	15	92	>300
11		 3k	30	85	>300/[14]
12		 3l	30	90	228/[28]

[a] Reaction conditions: T = 120° C, 6-aminouracil (2 mmol), aldehyde (1 mmol), and SBA-15-SO_3H (0.05 g)

[b] Isolated yields; all the synthesized compounds were known except 3e, 3h, 3i, 3j and their physical and spectral data agree with the reported values



Scheme 3 Plausible mechanism for the synthesis of pyrido[2,3-*d*:6,5-*d*′]dipyrimidine derivatives

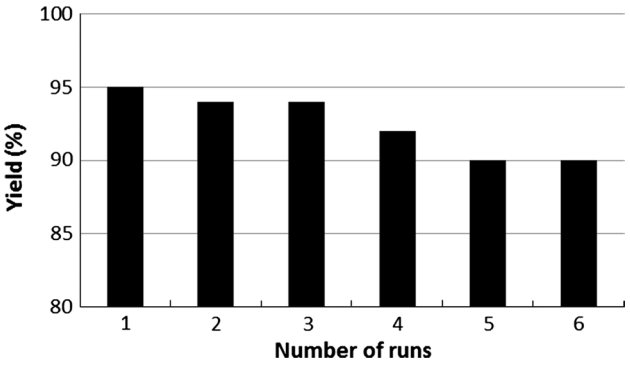


Fig. 5 Reusability of the catalyst

BET studies of the catalyst show that the surface area and pore volume decreased after modification of SBA-15 with $ClSO_3H$ (Table 1). BJH analysis shows pore volume and pore area distribution in the mesoporous range (Fig. 4b).

Catalytic activity of SBA-15-SO_3H

The catalytic activity of SBA-15-SO_3H was investigated during the synthesis of pyrido[2,3-*d*:6,5-*d*′]dipyrimidine derivatives. The reaction between 6-aminouracil (1) and 4-chlorobenzaldehyde (2) (Scheme 1) was chosen as a model reaction to find the best condition for the preparation of pyrido[2,3-*d*:6,5-*d*′]dipyrimidine derivatives. The results of optimization studies are summarized in Table 2.

According to Table 2, the best result was obtained when 0.05 g of SBA-15-SO_3H was used (Table 2, entry 3). Increasing the amount of the catalyst did not have any effect on the product yield and the reaction time (Table 2, entry 4). It should be mentioned that when the reaction proceeded in the absence of the catalyst, only a trace amount of the product was obtained (Table 2, entry 1). The performance of the SBA-15-SO_3H in various solvents (EtOH, CH_3CN, DMF and H_2O) was also investigated (Table 2, entries 5–8). The results demonstrate that solvent-free condition was the best choice (Table 2, entry 3). In order to determine the optimum temperature for this reaction, the condensation reaction was carried out under solvent-free conditions at 100 to 130 °C using SBA-15-SO_3H as catalyst. At 100 °C, the yield of the reaction was 65 % and increased to 96 % at 120 °C (Table 2, entries 9 and 11). There was no significant change in the yield and reaction time at higher temperature (Table 2, entry 12).

After optimization of the reaction conditions, a series of pyrido[2,3-*d*:6,5-*d*′]dipyrimidine derivatives were synthesized by the condensation of 6-aminouracil (1) and different aldehydes (2a–l) under optimized reaction conditions (Table 3). All the reactions were completed within 10–60 min. As Table 3 shows, the aldehydes with electron withdrawing substituents afford the desired product in shorter reaction times and higher yields (Table 3, entries 2, 3 and 6). In the case of 2-bromobenzaldehyde, the steric hindrance resulted in the low yield of the product (Table 3, entry 5). A plausible mechanism for the formation of the pyrido[2,3-*d*:6,5-*d*′]dipyrimidine derivatives is shown in Scheme 3. First, acidic sites on the surface of the catalyst activate the aldehyde (**2**). In the second step, nucleophilic

addition takes place between 6-aminouracil (**1**) and aldehyde (**2**), and consequently water elimination produces the intermediate (**4**). In the next step, nucleophilic attack of the other 6-aminouracil molecule (**1**) to intermediate (**4**) leads to the production of the intermediate (**5**). NH_2 activation in the intermediate (**6**) by SBA-15-SO_3H helps in the elimination of ammonia which is a driving force for obtaining the desired product (**7**).

The stability and reusability of the catalyst

The reusability of the catalyst was investigated using 6-aminouracil and 4-nitrobenzaldehyde as model substrates. After completion of the reaction, the decanted catalyst was washed with dichloromethane and dried in an oven at 60 °C for 2 h. Then it was subjected to another reaction with identical substrates. As can be seen in Fig. 5, the recovered catalyst can be used for at least 6 runs without significant loss of its activity.

Conclusion

In summary, a new highly acidic recyclable catalyst has been designed by direct grafting of sulfonic groups on the silica-based SBA-15 and used in the synthesis of some new and known pyrido[2,3-*d*:6,5-*d*′]dipyrimidine derivatives. Short reaction times, high yield of the products, and solvent-free conditions are worthwhile advantages of the present method.

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