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One-pot synthesis of tetrahydropyrimido[4,5-*b*]quinoline derivatives using sulfonic acid functionalized SBA-15 and their antimicrobial activities

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ABSTRACT

A Simple and efficient method for synthesis of tetrahydropyrimido[4,5-*b*]quinoline derivatives via three component reaction of aromatic aldehydes, dimedone and 6-amino uracil derivatives using a catalytic amount of sulfonic acid functionalized nanoporous silica (SBA-15-Pr-SO_3H) is described. The advantages of this method are easy and clean work-up, high yield, mild reaction condition, reusable catalyst and environmentally benign solvents. Antibacterial and antifungal activities of synthesized compounds were measured against gram positive, gram negative bacteria and fungus. Only compounds **4c** and **4g** showed activity against *Staphylococcus aureus*.

***Keywords**: 4(6)-Amino uracil, aldehydes, Dimedone, SBA-15-Pr-SO_3H, Multi-component reaction.*

1. Introduction

Multi-component reactions (MCRs) have created strong strategies. MCRs have been used widely to form carbon-carbon and carbon-heteroatom bonds in synthetic chemistry [1,2]. They have become a powerful tool in organic, synthetic, and medicinal chemistry because of their simplicity and simultaneously multiplex bond forming [3-5]. They are also environmentally and economically useful because of reducing dangerous, toxic and expensive solvents after each synthesis steps.

The importance of uracil and its derivatives are well known by synthetic [5], and biological [6] chemists. Because of anticancer and antiviral activities of uracil compounds [7,8], they have received significant attention in organic synthesis [9,10]. They are versatile building blocks for the synthesis of a wide range of heterocyclic compounds, such as pyridopyrimidines [11], pyrimidoquinolines [12], benzopyrano pyrimidines [13] and pyrazolopyrimidines [14]. Annulated uracils containing pyrido[2,3-*d*]pyrimidine core display a wide range of biological activities like antitumour [15], cardiotonic [16], antimalarial [17], antimicrobial [18], antioxidant [19], and antifungal [18] activities.

Santa Barbara Amorphous (SBA-15) is a nanoporous silica with hexagonal structure. Because of its high surface area, large pore size, high chemical and thermal stability, high selectivity, and easy isolation from products, it has the potential to use as a catalyst support [20-21]. Sulfonic-acid-functionalized SBA-15 is one of the most important functionalized nanoporous material used as green and environmentally catalyst in organic reactions [22-23]. In continuation of our previous works on the application of heterogeneous catalysts in organic synthesis [24-30], herein, we would like to report an efficient method in the synthesis of tetrahydropyrimido[4,5-*b*]quinoline derivatives using sulfonic acid functionalized SBA-15.

2. Experimental

Melting points were measured by using the capillary tube method with an Electro thermal 9200 apparatus. Mass spectrometry analysis was performed on a model 5973 mass-selective detector (Agilent). Infrared (IR) were recorded from KBr disc using a FT-IR Bruker Tensor 27 instrument. The ^{1}HNMR was run on a

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Bruker, 300 or 500 MHz. The ^{13}CNMR was run on a Bruker, 75 or 125 MHz.

2.1. General procedure for the synthesis of tetrahydro pyrimido[4,5-b]quinoline derivatives 4 (a-l)

In a 50 mL two-neck round-bottom flask equipped to a condenser, a mixture of 6-amino uracil derivative **1** (1 mmol), aromatic aldehyde **2** (1 mmol), dimedone **3** (1 mmol), SBA-Pr-SO_3H (0.02 g) and H_2O:EtOH (1:1) (3 mL) were added and heated in oil bath at 90 °C. After completion of the reaction (monitored by TLC), the crude product was dissolved in hot ethanol and water, the heterogeneous solid catalyst was removed easily by simple filtration, and after cooling of the filtrate, the pure crystals of products **4** were obtained.

Spectral data for new compounds

*8,8-dimethyl-5-(3-nitrophenyl)-7,8,9,10-tetrahydropyrimido[4,5-b]quinoline-2,4,6(1H,3H,5H)-trione (**4b**):*

Yellow solid, m.p.= 298-300 °C. IR (KBr): $\bar{\nu}$ = 3572, 3411, 3176, 3080, 2965, 1710, 1651, 1616, 1530, 1474, 1380, 1348, 1226, 821, 771, 734, 689, 561 cm^{-1}. ^{1}HNMR (300 MHz, DMDO-d_6): δ= 10.81 (s, 1H, NH), 10.41 (br s, 1H, NH), 8.99 (br s, 1H, NH), 7.99 (s, 1H, 1CH arom.), 7.96 (d, 1H, *J*=7.3 Hz, 1CH arom.), 7.65 (d, 1H, *J*=7.3 Hz, 1CH arom.), 7.50 (t, 1H, *J*=7.7 Hz, 1CH arom.), 4.84 (s, 1H, CH), 2.48 (m, 2H, 2CH), 2.21 (d, 1H, *J*=16.2 Hz, 1CH), 2.01 (d, 1H, *J*=16.2 Hz, 1CH), 1.01 (s, 3H, CH_3), 0.87 (s, 3H, CH_3) ppm. ^{13}CNMR (75 MHz, DMDO-d_6): δ= 194.47, 162.77, 150.10, 149.96, 148.57, 147.43, 134.52, 129.34, 122.10, 121.09, 110.26, 88.80, 49.95, 33.73, 32.20, 28.96, 26.36 ppm. EIMS (rel. int.): m/z= 382 [M$^+$] (8), 380 (100), 363 (30), 324 (40), 273 (80), 217 (50), 152 (35), 127 (90), 68 (45), 55 (70).

*5-(4-methoxyphenyl)-8,8-dimethyl-7,8,9,10-tetrahydropyrimido[4,5-b]quinoline-2,4,6(1H, 3H,5H)-trione (**4f**):*

Light yellow solid. m.p.>300 °C. IR (KBr): $\bar{\nu}$ = 3454, 3208, 2957, 2830, 2362, 2050, 1707, 1651, 1532, 1509, 1484, 1418, 1379, 1343, 1261, 1230, 1206, 1172, 1151, 1110, 1087, 1027, 977, 861, 788, 757, 720, 676, 609, 547 cm^{-1}. ^{1}HNMR (300 MHz, DMDO-d_6): δ= 10.71 (s, 1H, NH), 10.21 (s, 1H, NH), 8.73 (s, 1H, NH), 7.07 (d, 1H, *J*=8.2 Hz, 1CH arom.), 6.73 (d, 1H, *J*=8.2 Hz, 1CH arom.), 4.67 (s, 1H, CH), 3.65 (s, 3H, CH_3), 2.42 (m, 2H, 2CH), 2.17 (d, 1H, *J*=16.1 Hz, 1CH), 1.98 (d, 1H, *J*=16.1 Hz, 1CH), 0.99 (s, 3H, CH_3), 0.87 (s, 3H, CH_3) ppm. ^{13}CNMR (75 MHz, DMDO-d_6): δ= 194.43, 162.78, 157.43, 149.91, 148.92, 143.60, 138.83, 128.52, 113.13, 111.54, 89.97, 54.93, 50.20, 33.17, 29.03, 26.51 ppm. EIMS (rel. int.): m/z= 368 [M+1] (50), 367 [M$^+$] (100), 352 (70), 259 (30), 203 (65), 178 (15), 108 (15), 77 (10).

*5-(2,6-dichlorophenyl)-8,8-dimethyl-7,8,9,10-tetrahydropyrimido[4,5-b]quinoline-2,4,6(1H,3H,5H)-trione (**4g**):*

Light yellow solid. m.p.>300 °C, IR (KBr): $\bar{\nu}$ = 3569, 3180, 2958, 1716, 1652, 1534, 1434, 1377, 1334, 1270, 1229, 1207, 1170, 1151, 1079, 1008, 840, 789, 766, 673, 602, 562, 545, 523 cm^{-1}. ^{1}HNMR (500 MHz, DMDO-d_6): δ= 10.59 (s, 1H, NH), 10.20 (s, 1H, NH), 8.86 (s, 1H, NH), 7.29 (d, 1H, *J*=8.0 Hz, 1CH arom.), 7.21 (d, 1H, *J*=8.0 Hz, 1CH arom.), 7.08 (t, 1H, *J*=8.0 Hz, 1CH arom.), 5.55 (s, 1H, CH), 2.42 (d, 1H, *J*=17.0 Hz, 1CH), 2.28 (d, 1H, *J*=17.0 Hz, 1CH), 2.14 (d, 1H, *J*=16.0 Hz, 1CH), 1.92 (d, 1H, *J*=16.0 Hz, 1CH), 1.00 (s, 3H, CH_3), 0.92 (s, 3H, CH_3) ppm. ^{13}CNMR (125 MHz, DMDO-d_6): δ= 194.06, 162.15, 150.67, 144.83, 138.25, 137.80, 134.07, 129.35, 127.83, 127.68, 108.25, 86.79, 50.22, 32.37, 31.64, 28.85, 26.52 ppm. EIMS (rel. int.): m/z= 407 [M+2] (10), 405 [M$^+$] (20), 385 (40), 383(100), 370 (40), 347 (80), 323 (40), 273 (40), 260 (75), 217 (30), 161 (20), 55 (22).

*5-(2,3-dimethoxyphenyl)-8,8-dimethyl-7,8,9,10-tetrahydropyrimido[4,5-b]quinoline-2,4,6(1H,3H,5H)-trione (**4h**):*

Light yellow solid. m.p.>300 °C. IR (KBr): $\bar{\nu}$ = 3582, 3345, 3168, 3036, 2962, 2833, 1722, 1676, 1610, 1540, 1482, 1382, 1329, 1285, 1253, 1222, 1201, 1160, 1064, 1005, 916, 838, 801, 769, 749, 672, 609, 588, 549, 511 cm^{-1}. ^{1}HNMR (500 MHz, DMDO-d_6): δ= 10.59 (s, 1H, NH), 10.16 (s, 1H, NH), 8.70 (s, 1H, NH), 6.84-6.73 (m, 3H, 3CH arom.), 4.90 (s, 1H, CH), 3.72 (s, 3H, OCH_3), 3.71 (s, 3H, OCH3), 2.41 (d, 1H, *J*=17.0 Hz, 1CH), 2.35 (d, 1H, *J*=17.0 Hz, 1CH), 2.11 (d, 1H, *J*=16.1 Hz, 1CH), 1.95 (d, 1H, *J*=16.2 Hz, 1CH), 0.98 (s, 3H, CH_3), 0.87 (s, 3H, CH_3) ppm. ^{13}CNMR (125 MHz, DMDO-*d*6): δ= 193.96, 162.46, 152.16, 149.81, 148.99, 148.85, 146.91, 143.95, 139.05, 122.83, 122.21, 111.01, 110.50, 89.37, 59.53, 55.38, 50.37, 32.05, 30.1165, 28.62, 26.71 ppm. EIMS (rel. int.): m/z= 398 [M+1] (2), 397 [M$^+$] (5), 379 (7), 368 (15), 313 (12), 259 (10), 236 (12), 109 (20), 97 (35), 83 (52), 69 (80), 55 (100).

3. Results and Discussion

3.1. Synthesis and functionalization of SBA-15

The nanoporous compound SBA-15 was synthesized and functionalized according to our previous report as shown in Fig. 1 [31].

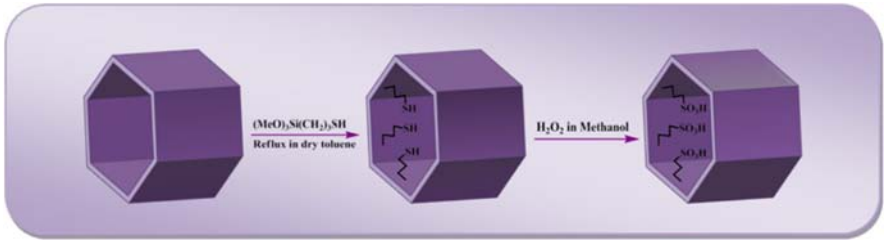


Fig. 1. Preparation of SBA-15-Pr-SO_3H.

The surface of the catalyst was analyzed by different methods such as TGA, BET and other methods which were demonstrated that the organic groups (propyl sulfonic acid) were immobilized into the pores. The Thermal gravimetric analysis (TGA) of SBA-15-Pr-SO_3H shows two major decomposition states: one below 100 °C, assigned to loss of water surface and one more mass loss between 200 °C and 600 °C, corresponded to the decomposition of organic groups grafted onto silica. Therefore the concentration of immobilized acid (from the weight loss) can be estimated as 1.2 mmol g^{-1} [32,33].

Fig. 2 illustrates the SEM and TEM images of SBA-Pr-SO_3H. SEM image (Fig. 2 left) shows uniform particles about 1µm. The same morphology was observed for SBA-15. It can be concluded that morphology of solid was saved without change during the surface modifications. On the other hand, the TEM image (Fig. 2 right) reveals the parallel channels, which resemble the pores configuration of SBA-15. This indicates that the pore of SBA-Pr-SO_3H was not collapsed during two steps reactions.

3.2. Synthesis of tetrahydropyrimido[4,5-b]quinoline derivatives

In the present study, condensation of 6-amino uracil derivatives **1**, aromatic aldehydes **2** and dimedone **3** in the presence of SBA-15-Pr-SO_3H was described (Scheme 1).

In order to determine the optimum conditions we first studied the reaction between 6-amino uracil, benzaldehyde and dimedone in different solvents using SBA-15-Pr-SO_3H (Table 1).

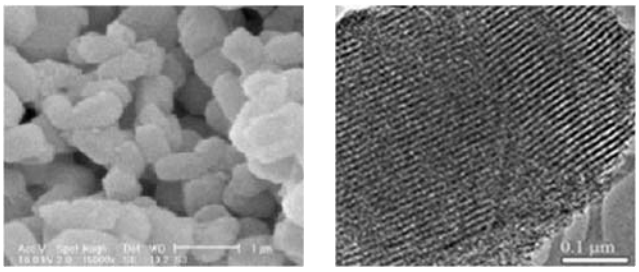
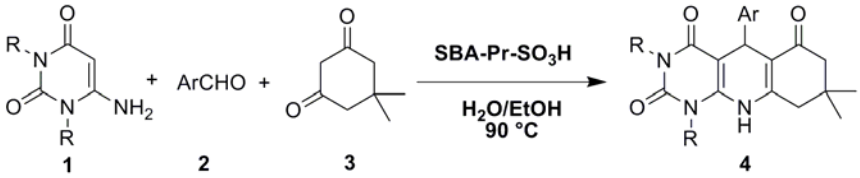


Fig. 2. SEM (Left) and TEM (Right) images of SBA-Pr-SO_3H.



Scheme 1. Synthesis of tetrahydropyrimido[4,5-*b*]quinoline derivatives in the presence of SBA-15-Pr-SO_3H.

Table 1. The optimization of reaction conditions for the synthesis of **4a**.

Entry	Solvent	Condition	Time (h)	Yield (%)[a]
1	H_2O	Reflux	6	47
2	EtOH	Reflux	5:30	65
3	H_2O:EtOH	90 °C	4	93
4	-	120 °C	5	Trace

[a]Isolated yield. Reaction conditions: 6-amino uracil (1 mmol), benzaldehyde (1 mmol), dimedone (1 mmol), SBA-15-Pr-SO_3H (0.02 g).

As shown results in Table 1, the shortest reaction time with high yield was obtained after 4 hours in EtOH:H_2O (1:1) and 90 °C.

After optimizing the conditions, we examined this condition for different aromatic aldehydes (Scheme 1). By this method, the reactions were carried out easily in the presence of SBA-15-Pr-SO_3H (0.02 g) in H_2O:EtOH at 90 °C to produce tetrahyropyrimido[4,5-*b*]quinoline derivatives in good to excellent yields (74-94%). This protocol offers advantages such as its simple procedure and work-up, use of the green and reusable catalyst, and environmentally benign solvent. The experimental procedure has the ability to tolerate a variety of functional groups under the reaction conditions. As shown in Table 2, entries 10-12, using 6-Amino-1,3-dimethyluracil instead of 6-amino uracil will increase the reaction rate and reduce the reaction time. After completion of the reaction which monitored by TLC, the reaction mixture was dissolved in hot mixture of ethanol and water, the heterogeneous catalyst was removed by filtration. After cooling of the filtrate, the pure crystals of products were obtained. The products were characterized by melting points, ^{1}HNMR, ^{13}CNMR, IR and Mass Spectrometry. Melting points are compared with reported values in literature as shown in Table 2.

The acid catalyst can be reactivated by simple washing subsequently with diluted acid solution, water and acetone, and then reused without noticeable loss of reactivity. The reusability of the catalyst was investigated under optimized conditions for the synthesis of the model compound **4a**. The recycling process was completed four times with no significant decrease on the catalyst activity. The yields for the four runs were found to be 93%, 87%, 84%, and 81% respectively.

A reasonable mechanism for the synthesis of tetrahydropyrimido[4,5-*b*]quinoline derivatives **4** is proposed in scheme 2. SBA-15-Pr-SO_3H acts as a source of H^+, which can protonate carbonyl groups of both aldehyde and dimedone to create more reactive species. The Knoevenagel condensation between aldehyde **2 (a-l)** and dimedone **3** after dehydration yields α,β-unsaturated ketone **5**. Subsequent Michael-type addition of 6-amino uracil derivative **1** to α,β-unsaturated ketone **5** gives the intermediate **6**. Cyclization of intermediate **6** followed by dehydration affords the corresponding products **4 (a-l)**. The high yields of reactions are due to the effect of nano pore of solid acid catalyst, which could act as nano-reactor (Fig. 3).

Table 2. SBA-15-Pr-SO_3H catalyzed the synthesis of tetrahyropyrimido[4,5-*b*]quinoline **4**(**a-l**).

Entry	Ar	R	Product	Time (h)	Yield (%)	m.p. (°C)		Ref.
						Found	Reported	
1	C_6H_5	H	**4a**	4	93	>300	>300	[34]
2	3-$NO_2C_6H_4$	H	**4b**	6	94	298-300	New	This work
3	4-ClC_6H_4	H	**4c**	3:30	78	>300	>300	[34]
4	4-OHC_6H_4	H	**4d**	3	84	>300	>300	[35]
5	4-$CH_3C_6H_4$	H	**4e**	5	74	>300	>300	[34]
6	4-$OCH_3C_6H_4$	H	**4f**	4	79	>300	New	This work
7	2,6-$Cl_2C_6H_3$	H	**4g**	4	74	>300	New	This work
8	2,3-$(OCH_3)_2C_6H_3$	H	**4h**	4	76	>300	New	This work
9	3,4-$(OCH_3)_2C_6H_3$	H	**4i**	3	83	>300	>300	[35]
10	C_6H_5	CH_3	**4j**	2:30	84	268-269	268	[34]
11	4-ClC_6H_4	CH_3	**4k**	1	85	284-285	291	[34]
12	4-$OCH_3C_6H_4$	CH_3	**4l**	2:30	86	>300	>300	[36]

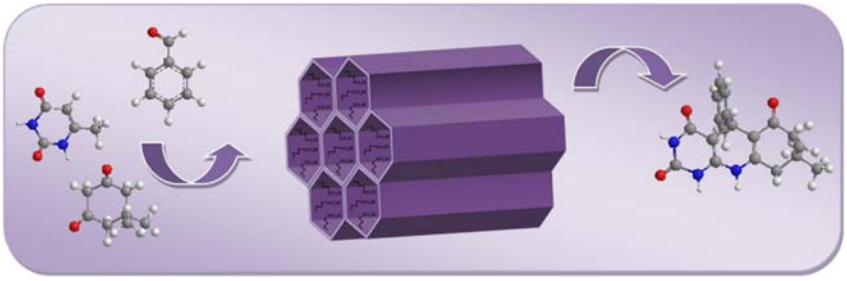


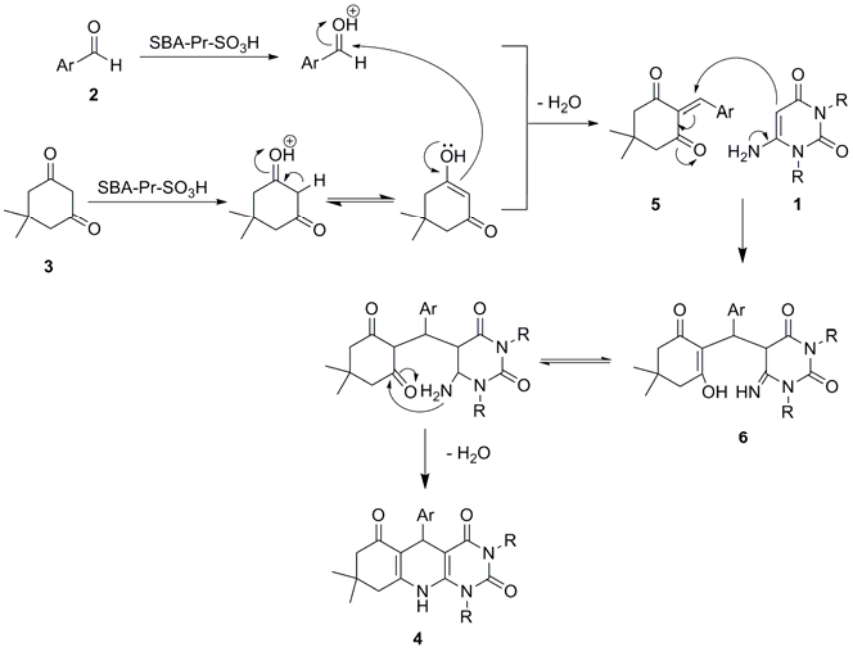
Fig. 3. SBA-15-Pr-SO_3H catalyst acts as nano-reactor.

The synthesis of tetrahydropyrimido[4,5-*b*]quinoline derivatives have been studied with several conditions in literature [26, 34-37], but they are few reports which used the same starting materials for synthesis of these compounds. Table 3 shows different conditions using the same starting material in the synthesis of tetrahydropyrimido[4,5-*b*]quinolines. In contrast with other existing methods, the present methodology offers several advantages such as heterogeneous and reusable catalyst, good yields, short reaction times and simple work-up. As shown in Table 3, in compare to present work, entry 1 have long reaction times. Entry 2 used *p*-TSA which is not a heterogeneous catalyst and also it's purification from reaction mixture is not simple; furthermore it is not a recyclable catalyst.

3.3. Antimicrobial activities of tetrahydropyrimido [4,5-b]quinoline derivatives

All synthesized compounds were screened for antimicrobial activity using disc diffusion method (Table 4). In this project, five microorganisms were used such as *Pseudomonas aeruginusa* ATCC 85327 and *Escherichia coli* ATCC 25922 (gram negative bacteria), *Staphylococcus aureus* ATCC 25923 and *Bacillus subtilis* ATCC 465 (gram positive bacteria) and *Candida albicans* ATCC 10231 (fungi). Tetrahydropyrimido[4,5-*b*]quinolines (100 µg) were dissolved in DMSO (1 mL) and 25 µL of them were loaded to 6 mm paper discs. On the other hand, 100 microliters of microorganisms suspensions (10^9 cell/mL) were spread on sterile Muller-Hilton Agar plates, then the discs were placed on the surface of culture plates. The inhibition zones of compounds around the disc were compared with three commercial antibiotics such as Chloramphenicol, Gentamicin and Nystatin as shown in Table 4. Among them only compounds **4c** and **4g** show activity against *S. aureus*.

The minimum inhibitory concentration (MIC) of synthesized compounds were also determined by microdillution method [38] in compare with three commercial antibiotics (Chloramphenicol, Gentamicin and Nystatin).



Scheme 2. Proposed mechanism for the synthesis of tetrahydropyrimido[4,5-*b*]quinolines.

Table 3. Comparison of different conditions in the synthesis of pyrimido[4,5-*b*]quinolones.

Entry	Catalyst	Solvent	Condition	Time (h)	Yield (%)	Year
1	TEBAC	H_2O	90ºC	8-22	75-98	2009 [35]
2	*p*-TSA	H_2O	90ºC	2.5-3.5	62-95	2012 [37]
3	SBA-Pr-SO_3H	H_2O/EtOH	90 ºC	1-6	74-94	This work

As shown in Table 5, the MIC value of compounds **4c** and **4g** for the *B. subtilis* and *S. aureus* were higher than the range of those reported for Chloramphenicol and Gentamicin to National Committee for Clinical Laboratory Standards (NCCLS 2000). It means in compare to commercial antibiotics, higher inhibitory concentrations of synthesized compounds are needed to show antibacterial activity against tested microorganisms.

4. Conclusion

In summary we have developed an easy method for preparation of tetrahydropyrimido[4,5-*b*]quinoline derivatives via one-pot three-component condensation of 6-amino uracil derivatives, aldehydes and dimedone in the presence of SBA-15-Pr-SO_3H. The advantages of this method are easy work-up, high yield, clean reaction, mild reaction condition and environmental friendly solvents. We have reported SBA-15-Pr-SO_3H as an effective heterogeneous solid acid catalyst that can be removed by simple filtration from reaction and can be reactivated by simple washing subsequently with diluted acid solution, water and acetone, and then reused without noticeable loss of reactivity.

Acknowledgments

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Table 4. Inhibition zone (mm) of synthesized compounds against fungi and gram positive and negative bacteria by disc diffusion method.

Compound	*B. subtilis*	*S. aureus*	*E. coli*	*P. aeruginosa*	*C. albicans*
4a	0	0	0	0	0
4b	0	0	0	0	0
4c	9	11	0	0	0
4d	0	0	0	0	0
4e	0	0	0	0	0
4f	0	0	0	0	0
4g	0	17	0	0	0
4h	0	0	0	0	0
4i	0	0	0	0	0
4j	0	0	0	0	0
4k	0	0	0	0	0
4l	0	0	0	0	0
Chloramphenicol	26	22	24	8	-
Gentamicin	28	20	20	18	-
Nystatin	-	-	-	-	18

Table 5. Minimum inhibitory concentration (μg/mL) of synthesized compounds against fungi and gram positive and negative bacteria.

Compound	*B. subtilis*	*S. aureus*	*E. coli*	*P. aeruginosa*	*C. albicans*
4a	-	-	-	-	-
4b	-	-	-	-	-
4c	512	256	-	-	-
4d	-	-	-	-	-
4e	-	-	-	-	-
4f	-	-	-	-	-
4g	-	64	-	-	-
4h	-	-	-	-	-
4i	-	-	-	-	-
4j	-	-	-	-	-
4k	-	-	-	-	-
4l	-	-	-	-	-
Chloramphenicol	4	8	4	256	-
Gentamicin	0.125	0.5	0.5	1	-
Nystatin	-	-	-	-	8

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