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An Efficient Green Protocol for Synthesis of 2,3-Dihydroquinazolin-4(1*H*)-ones Using SBA-16/GPTMS-TSC-Cu^{I} under Solvent-Free Conditions

Mohammad Anwar Erfan, Batool Akhlaghinia,* and Sara S. E. Ghodsinia[a]

Herein a rapid, efficient, facile and environmentally benign synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones using SBA-16/GPTMS-TSC-Cu^{I} (Cu^{I} anchored onto mesoporous SBA-16 functionalized by aminated 3-glycidyloxypropyltrimethoxysilane with thiosemicarbazid) is reported. The aforesaid mesostructured catalyst with a unique "super-cage" structure and narrow particle size distribution (3-7 nm) exhibited excellent catalytic activity in high yielding preparation of 2,3 dihydroquinazolin-4(1*H*)-ones from the condensation of 2-aminobenzamide and an aldehyde in solvent-free conditions. Furthermore, SBA-16/GPTMS-TSC-Cu^{I} as a heterogeneous catalyst was stable under reaction conditions and can be recycled at least five times without any loss of its catalytic efficiency.

Introduction

Heterocyclic chemistry as half of all organic chemistry research is an important branch of organic chemistry. In particular, nitrogen-containing heterocycles due to their applications in many biological processes have attracted considerable interest.[1,2] Amongst them, 2,3-dihydroquinazolin-4(1*H*)-one derivatives as key and supplementary blocks in agro-chemistry, medicinal chemistry and organic chemistry[3] are found widely in nature, especially in biologically and pharmaceutically active molecules. Also, they can act as "core structures" in drug discovery owing to their important pharmacological properties.[4–6] Recent literature studies clearly show that they display an extensive important biological and pharmacological activities as well as medicinal properties including anticancer,[7] antitumor,[8] antibiotic,[9] and antihypertension action.[10] There are a variety of methods in the literature for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one derivatives with their own merits and demerits: (a) condensation of anthranilamide with an aldehyde or ketone;[11] (b) desulfurization of 2-thioxo-4(3*H*)-quinazolinones;[12] (c) reaction of isatoic anhydride with Schiff bases;[13] (d) one-step conversion of 2-nitrobenzamides to 2,3-dihydro-4(1*H*)-quinazolinones;[14] (e) condensation of anthranilamide with benzyl;[15] (f) two-step synthesis starting from isatoic anhydride and amines, then was annulated with ketones;[16] (g) a one-pot three-component condensation of isatoic anhydride, aldehydes and amines.[17,18] Of these, condensation of anthranilamide with an aldehyde or ketone is one of the simplest and direct methods of 2,3-dihydroquinazolin-4(1*H*)-onse preparation using a variety of homogeneous or heterogeneous catalysts such as molecular iodine (I_2),[19] cyanuric chloride,[20] morpholinoethanesulfonic acid[21] NH_4Cl,[22] tetrabutylammonium bromide (TBAB),[23] p-sulfonic acid calix[4] arene,[24] [Al$(H_2PO_4)_3$],[25] [bmim]HSO_4,[26] Sc$(OTf)_3$,[27] $ZrCl_4$,[28] Y$(OTf)_3$,[29] $BiBr_3$,[30] $NaHSO_4$,[31] lactic acid,[32] heteropoly acids,[33,34] β-cyclodextrin-SO_3H,[35] SiO_2-PPA,[36] [PYC_4SO_3H][HSO_4]/A300SiO_2,[37] Fe_3O_4 nanoparticles,[38] cellulose-SO_3H,[39] β-cyclodextrin,[40] Cu$(NO_3)_2$/F_3O_4-DETA,[41] amberlyst-15,[42] Fe_3O_4-chiff base of Cu(II),[43] Sc$(OTf)_3$ fluorous bis(oxazolines),[44] chiral SPINOL-phosphoric acids, 3 A°MS,[45] SiO_2-$H_3PW_{12}O_{40}$,[46] TBAHS[47] or by electrochemical reactions.[48] Although, the reported synthetic protocols produce good results in many instances, some of them associated with at least one of the following imperfections: harsh reaction conditions, prolonged-time period, unsatisfactory yields, and use of environmentally harmful solvent, expensive moisture-sensitive catalysts, hazardous acid catalysts, high catalyst loading, expensive reagent, and tedious workup conditions. Despite the advances in synthetic methodologies, as a consequence of pharmacological properties of 2,3-dihydroquinazolin-4(1*H*)-ones, development of a new, simple, environmentally benign, high-yielding, and clean catalytic route towards this direction is an attractive target in synthetic chemistry.

During the past decade, the development of environmentally compatible processes has become one of the aims of research in efforts designed to produce chemical compounds.[49] Alongside, implementing safer practices such as waste prevention has been started by industries using new catalysts. In this regard, in the twenty-first century, new environmentally benign, economical, practical and efficient processes using recoverable heterogeneous catalysts were designed by chemists as a central focus area in green chemistry research. Particularly, ease of recovery and repeated use of the catalyst can be provided by supported metal catalysts. The overall performance, catalytic activity, and reusability of heterogeneous catalysts depend on the choice of the catalyst

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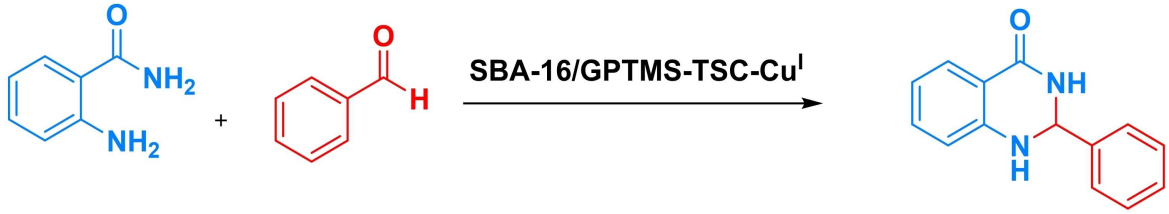
support. In recent years, SBA-16 as a novel mesoporous silica material has been identified as a choice of support material for several metal catalysts owing to its regularly arranged pore structure,[50] narrow pore size distribution,[51] high pore volume, large specific surface area, and excellent hydrothermal stability which have shown considerable superiority among other solid supports.

Recently, the successfully synthesized Cu^{I} anchored onto mesoporous SBA-16 functionalized by aminated 3-glycidyloxypropyltrimethoxysilane with thiosemicarbazide (SBA-16/GPTMS-TSC-Cu^{I}) (**IV**) (Scheme 1) has been used as a novel, efficient, and heterogeneous mesostructured catalyst for synthesis of symmetrical diaryl sulfides from the cascade reaction of aryl halides with S_8/ thiourea under solvent-free conditions.[52] Owing to increasingly stringent environmental standards and economic pressures and also as a part of our continuing research work in green chemistry to explore the catalytic activity of heterogeneous catalysts[53,54] for various organic transformations, we are interested in developing an efficient simple, mild and expeditious synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones in high yields, using Cu^{I} anchored onto mesoporous SBA-16 functionalized by aminated 3-glycidyloxypropyltrimethoxysilane with thiosemicarbazide (SBA-16/GPTMS-TSC-Cu^{I}) (Scheme 2).

Results and Discussion

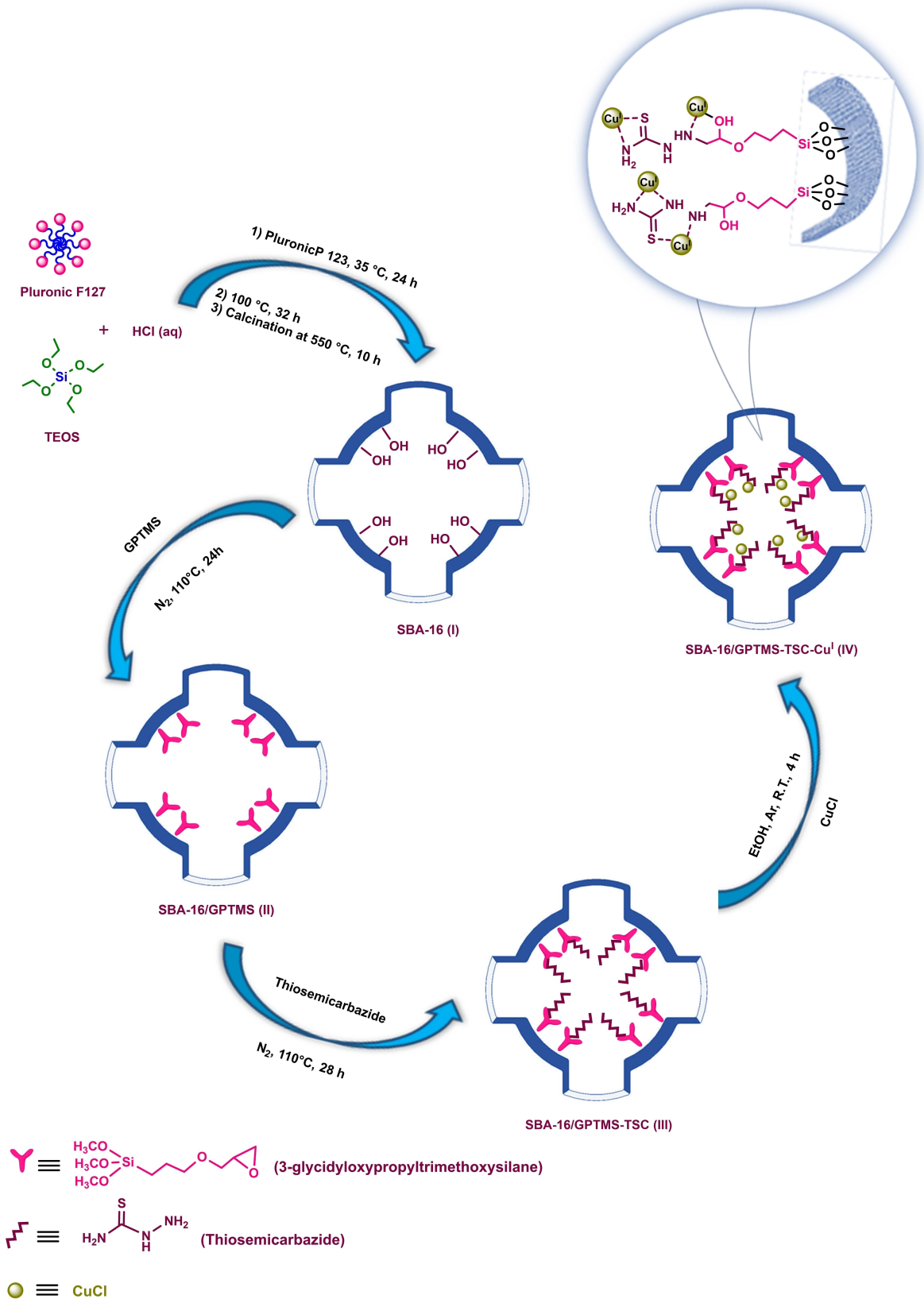
In order to study the catalytic activity of SBA-16/GPTMS-TSC-Cu^{I} in preparation reaction of 2,3-dihydroquinazolin-4(1*H*)–ones, the condensation reaction of 2-aminobenzamide with benzaldehyde was selected as a model reaction, and the effect of various parameters such as solvent, temperature and catalyst loading on this reaction was studied. The screening of various conditions is listed in Table 1. At the outset, in order to elucidate the role of the catalyst, the reaction between 2-aminobenzamide (1.05 mmol) and benzaldehyde (1 mmol) was conducted in refluxing H_2O in the absence of any catalyst. Table 1 shows that in the absence of the catalyst, desired product has been obtained as a trace within a reaction time of 10 h (Table 1, entry 1). It was observed that at the same reaction conditions, the model reaction was progressed smoothly in the presence of SBA-16/GPTMS-TSC-Cu^{I} (Table 1, entry 2). Then by considering the crucial role of the SBA-16/GPTMS-TSC-Cu^{I}, the present study was concentrated on the improvement of the reaction yield by screening different solvents and solvent-free conditions (Table 1, entries 2–8). It can be seen that H_2O and solvent-free conditions clearly stand out as the choice with their fast reaction rates and high isolated yields. As the reaction temperature has an important role in

Table 1. Synthesis of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one from the reaction of 2-aminobenzamide with benzaldehyde catalyzed by SBA-16/GPTMS-TSC-Cu^{I} under different reaction conditions.



Isolated Yield (%)	Time (min)	Temperature (°C)	Solvent	Catalyst (mol %)	Entry
Trace	10 (h)	Reflux	H_2O	-	1
98	20	Reflux	H_2O	9.3	2
25	20	Reflux	EtOH	9.3	3
64	20	Reflux	CH_3CN	9.3	4
55	20	Reflux	Toluene	9.3	5
23	20	Reflux	1,4-Dioxane	9.3	6
0	20	Reflux	CH_2Cl_2	9.3	7
98	20	100	-	9.3	8
10	20	r.t	H_2O	9.3	9
10	20	r.t	-	9.3	10
45	20	60	H_2O	9.3	11
98	20	60	-	9.3	12
98	20	70	-	9.3	13
78	20	50	-	9.3	14
60	20	40	-	9.3	15
98	20	60	-	11.1	16
98	**20**	**60**	**-**	**7.4**	**17**
24	20	60	-	5.6	18
45	20	60	-	0.04(g)	19[a]
45	20	60	-	0.04(g)	20[b]
45	20	60	-	0.04(g)	21[c]
24	20	60	-	7.4	22[d]

[a] The reaction was performed in the presence of SBA-16. [b] The reaction was performed in the presence of SBA16/GPTMS. [c] The reaction was performed in the presence of SBA-16/GPTMS/TSC. [d] The reaction was performed in the presence of CuCl.



Scheme 1. Overall flowchart for the fabrication of SBA-16/GPTMS-TSC-Cu^{I}.

R= Ph, 4-FPh, 4-ClPh, 2-ClPh, 3-BrPh, 4-O_2NPh, 4-CH_3Ph, 3-CH_3Ph, 4-CH_3OPh, 4-HOPh, 2-HOPh, 3,4-$(HO)_2$Ph, 3-HOPh, 4-$(CH_3)_2$NPh, 4-C_5H_4N, 2-C_4H_3S, 2-C_4H_3O, PhCH=CH, C_2H_5, $(CH_3)_2CH$, $CH_3(CH_2)_7CH_2$

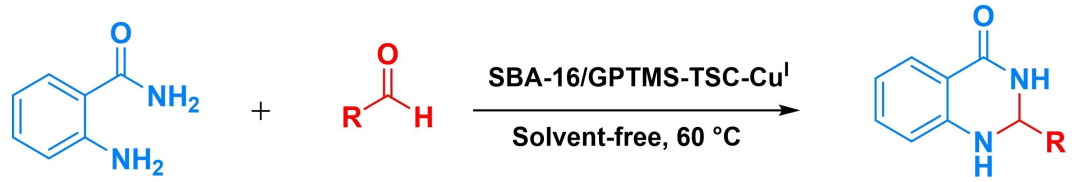
Scheme 2. Synthesis of 2,3- dihydroquinazolin-4(1*H*)-ones from the condensation reaction of 2-aminobenzamide with different aldehydes in the presence of SBA-16/ GPTMS-TSC-Cu^{I} under solvent-free conditions.

this reaction, we tried to optimize the reaction rates and high isolated yields. As the reaction temperature has an important role in this reaction, we tried to optimize the reaction temperature in the model reaction. Thermal evaluation of the reaction (in H_2O and solvent-free conditions) revealed that the best result was obtained at 60 °C in solvent-free conditions (Table 1, entries 9–14). Different amount of the catalyst loading was also optimized and it was found that 7.4 mol% of the mesostructured SBA-16/GPTMS-TSC-Cu^{I} gave the maximum yield of the product (Table 1, entries 15–17). To emphasize the efficiency of SBA-16/GPTMS-TSC-Cu^{I} in the preparation reaction of 2,3-dihydroquinazolin-4(1*H*)–ones, by applying the optimized reaction conditions, in a further set of experiments, the model reaction was also performed using SBA-16, SBA-16/GPTMS, SBA-16/GPTMS/TSC and CuCl (Table 1, entries 18–21). It was found that no satisfactory product yield was obtained in all cases. Accordingly, a comprehensive screening of the process parameters reveals that the highest yield and shortest reaction time for preparation of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one occur in the presence of 7.4 mol% of (SBA-16/GPTMS-TSC-Cu^{I}) catalyst in solvent-free conditions at 60 °C (Table 1, entry 17).

These identified optimized conditions turned our attention to examine the scope of catalytic condensation of 2-aminobenzamide with a variety of electronically divergent aldehydes towards the corresponding 2,3-dihydroquinazolin-4(1*H*)-ones. The results are compiled in Table 2. The condensation reaction of a variety of electronically divergent aromatic aldehydes was first investigated. In most cases, aromatic aldehydes possessing different functionalities (electron-withdrawing groups such as F, Cl, Br, NO_2 and electron-donating groups such as CH_3, OCH_3, OH, $N(CH_3)_2$) underwent the condensation reaction with 2-aminobenzamide with ease and in excellent yields of the desired 2,3-dihydroquinazolin-4(1*H*)-ones (Table 2, entries 1–14). The above observations reveal that the efficiency of the catalyst was so good that only slight differences in yield and reaction time were seen on functional group changes at various positions. We further expanded the scope of this reaction to heteroaromatic and α,β-unsaturated aldehydes. They were observed to be well tolerated under optimized conditions furnishing the corresponding products with satisfactory yields (Table 2, entries 15–18). These promising results prompted us to investigate the catalytic efficiency of SBA-16/GPTMS-TSC-Cu^{I} for condensation of 2-aminobenzamide with aliphatic aldehydes. Aliphatic aldehydes were found to be less reactive compared to aromatic, heteroaromatic and α,β-unsaturated ones (Table 2, entries 19–21). Further, we also examined the catalytic condensation reaction of 2-aminobenzamide with ketones. No product was obtained when ethyl methyl ketone, acetophenone and cyclohexanone were used in this reaction under the same conditions even after a prolonged reaction time (data are not given in Table 2). Due to the lower reactivity of ketones as compared to the aldehydes, the progress of this transformation was not convincing with ketone.

All the synthesized 2,3-dihydroquinazolin-4(1*H*)-ones were known and isolated, purified and initially identified by comparison of their melting points with those reported previously or with authentic samples prepared by the conventional methods. The mass spectra of these compounds displayed molecular ion peaks at the appropriate *m/z* values. For more clarification, the structures of some selected compounds (3a, 3c, 3 f, 3 g, 3i. 3j, 3k, 3 l, 3 m, 3n, 3p, 3q 3r, 3u) were characterized in details using FT-IR, ^{1}H NMR and ^{13}C NMR spectroscopy. The spectral data of all products are given in the experimental section. (*see Supporting Information file*) FT-IR spectra exhibited a characteristic absorption band at 1670–1648 cm^{-1} due to the carbonyl group of dihydroquinazolin ring. The stretching vibrations of two N–H bonds appeared around 3337–3182 cm^{-1}. In the ^{1}H and ^{13}C NMR spectra, all of the signals are in good accord with the structure, wherein two broad singlets at δ 8.91-8.24 and δ 7.30-5.98 ppm correspond to the N–H documented the successful synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones through the ring-closing reaction. The singlet resonating around 6.71-5.71 ppm established the presence of C–H of the dihydroquinazolin ring. The signals related to the protons of the phenyl rings appear at around 7.66-6.35 ppm (as multiplets). Besides, in the ^{13}C NMR spectra, the resonating signal at δ

Table 2. Scope and functional group tolerance of SBA-16/GPTMS-TSC-Cu^{I} catalyzed preparation of 2,3-dihydroquinazolin-4(1*H*)-ones under solvent-free conditions.



1 2 (3a-3u)

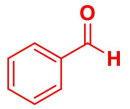
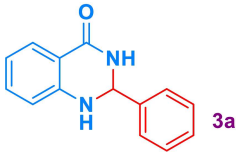
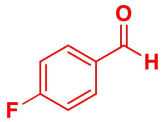
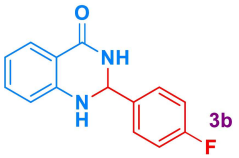
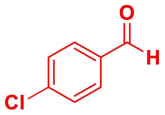
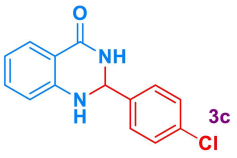
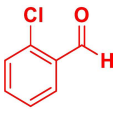
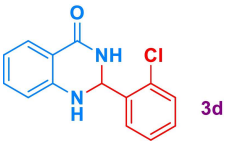
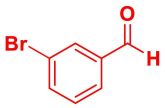
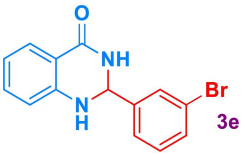
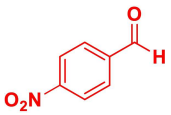
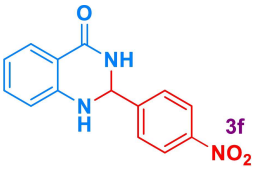
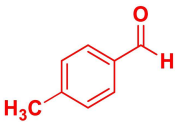
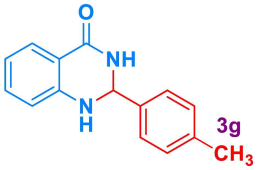
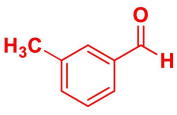
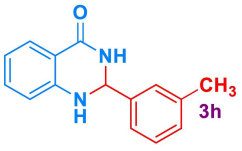
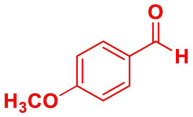
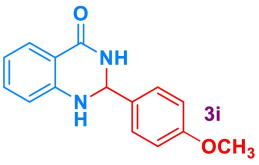
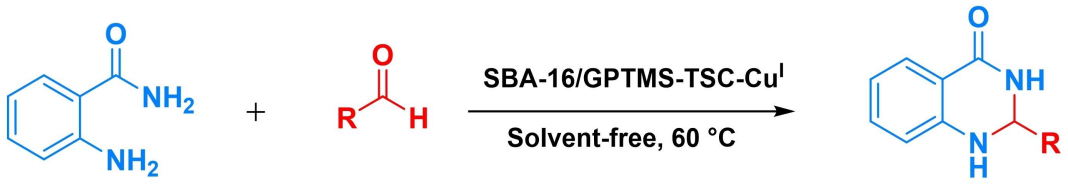
Entry	aldehyde	Product	Time (min)	Isolated Yield (%)
1		 3a	20	98
2		 3b	30	90
3		 3c	35	95
4		 3d	50	85
5		 3e	30	93
6		 3f	35	90
7		 3g	25	93
8		 3h	40	87
9		 3i	25	92

Table 2. continued



1 2 (3a-3u)

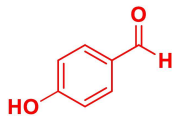
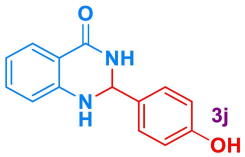
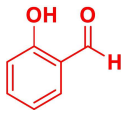
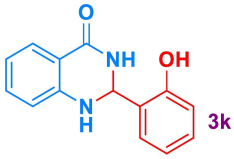
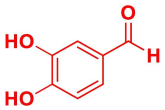
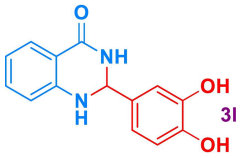
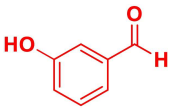
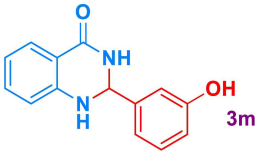
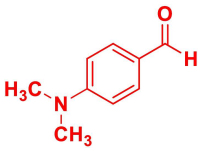
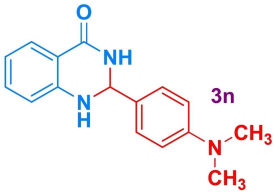
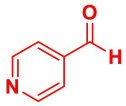
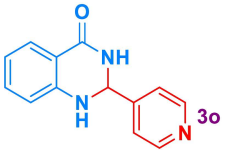
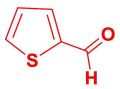
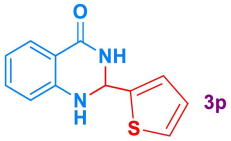
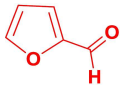
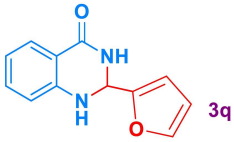
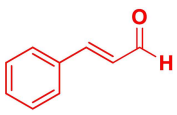
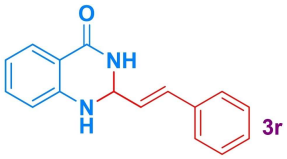
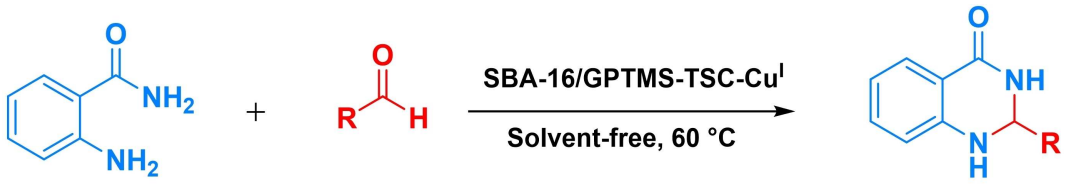
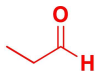
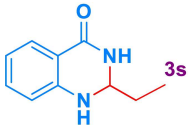
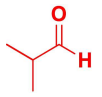
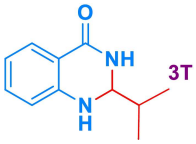
Entry	aldehyde	Product	Time (min)	Isolated Yield (%)
10			25	92
11			60	82
12			35	85
13			30	88
14			35	80
15			40	85
16			35	88
17			30	85
18			50	95

Table 2. continued



1 2 (3a-3u)

Entry	aldehyde	Product	Time (min)	Isolated Yield (%)
19			12 h	65
20			12 h	80
21			12 h	75

166.6-149.2 ppm corresponds to C=O and aromatic carbons resonate at δ 159–113 ppm which approve the formation of desired products.

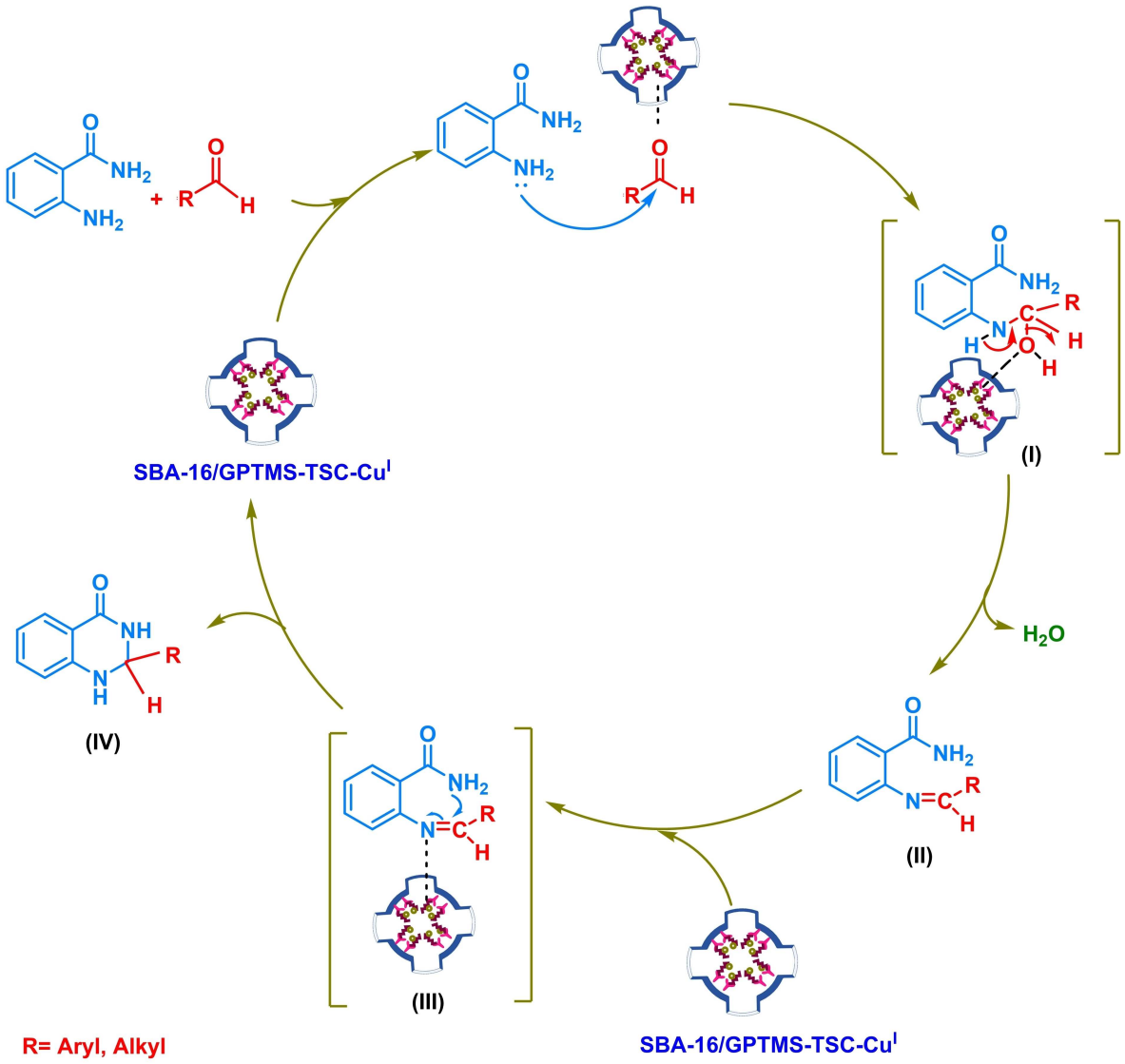
Consistent with previous suggestions in the literature,[55] we would like to propose a plausible mechanism for the formation of 2,3-dihydroquinazolin-4(1*H*)-ones from the condensation reaction of 2-aminobenzamide with aromatic and aliphatic aldehydes as shown in Scheme 3. It is supposed that the acidic property of SBA-16/GPTMS-TSC-Cu^{I} plays an essential role in accelerating the formation of 2,3-dihydroquinazolin-4(1*H*)-ones as is evident from the illustrated mechanism. Initially, the electrophilicity of the carbonyl group in aldehyde is enhanced with the help of supported metal (Cu^{I}) towards nucleophilic attack of 2-aminobenzamide (-NH_2). Subsequent dehydration of intermediate **(I)** affords the imine intermediate **(II)**. Thereafter, the imine part of intermediate **(II)**, which is activated by supported metal as well, produced intermediate **(III)**. In the following, the intramolecular nucleophilic attack of amide nitrogen on activated imine group produces 2,3-dihydroquinazolin-4(1*H*)-one **(IV)**. In the entire mechanism, SBA-16/GPTMS-TSC-Cu^{I} is regenerated and reused for the consecutive runs.

From an environmental point of view, economic and commercial consideration the reusability of catalysts is one of their most important advantages. In order to investigate this issue, the recycling of the catalyst was investigated using the model reaction (condensation of 2-aminobenzamide with benzaldehyde) under the optimized reaction conditions (Table 1, entry 17). Upon completion of the reaction, the reaction mixture was cooled to room temperature and then diluted by EtOAc (5 mL). Thereafter, the catalyst was easily separated from the reaction medium by simple filtration, washed with distilled water (2×5 mL) and ethanol (2×5 mL) to remove residual product. Afterward, the catalyst was dried under vacuum at 60 °C and fresh substrates were added to the recycled catalyst without any activation and employed for the next reaction run. As shown in Table 3, the catalyst can be recycled for up to five successive runs without obvious loss of its activity. The average isolated yield for five successive runs is 94.8%, which clearly demonstrates the practical recyclability of SBA-16/GPTMS-TSC-Cu^{I}.

Based on ICP-OES analysis, the exact amount of Cu in the recycled catalyst after five successive runs is found to be 1.80 mmol g^{-1}, while the freshly-prepared catalyst was shown to contain 1.86 mmol of Cu per 1.000 g of SBA-16/GPTMS-TSC-Cu^{I}. Therefore, the obtained results clearly certified the strong coordination of Cu^{I} ions with organic moieties on the surface of the mesostructured catalyst. It means that the mesostructured catalyst has good stability and reusability without any significant leaching of Cu in preparation reaction of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one under the optimized reaction conditions.

Table 3. Synthesis of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one in the presence of recycled SBA-16/GPTMS-TSC-Cu^{I}.

Run	Time (min)	Conversion (%)	Isolated yield (%)
1	20	98	96
2	20	98	96
3	20	97	95
4	20	97	95
5	20	96	92



Scheme 3. Recommended mechanism pathway for the preparation of 2,3 dihydroquinazolin-4(1*H*)-ones catalyzed by SBA-16/GPTMS-TSC-Cu^{I}.

Also, Fourier transform infrared (FT-IR) spectroscopy of the 5th recycled catalyst demonstrated no significant changes in the intensities, frequencies and shapes of absorption bands in comparison with the fresh catalyst. (Figure 1)

The hot filtration test was performed for the synthesis of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one under the optimized reaction conditions to investigate the heterogeneous nature of the catalytic species. In this line, in half time of the reaction (10 min), the reaction was stopped by separation of the mesostructured catalyst from the reaction mixture. Then the reaction mixture was allowed to react for another 10 min under similar conditions. The progress of the reaction was followed by thin-layer chromatography. We found no further substantial improvement in the yield of condensation reaction in the second half of the reaction time, which confirmed that the leaching of Cu in the reaction mixture is negligible. The obtained results from ICP-OES analysis of the reaction mixture showed that only 0.01 ppm of Cu species were leached out from the surface of SBA-16/GPTMS-TSC-Cu^{I} established the heterogeneous nature of the mesostructured catalyst.

The merit of the present synthetic methodology in comparison with protocols previously reported in the literature was shown in Table 4. It is obvious that the condensation reaction of 2-aminobenzamide with benzaldehyde in the presence of SBA-16/GPTMS-TSC-Cu^{I} is superior in terms of reaction media, (Table 4, entries 2, 4, 7, and 11) reaction time, (Table 4, entries 4, 7, 9, 11) and reusability of catalyst, (Table 4, entries 2, 4–8, and 10–11) while each of these catalytic systems has their own advantages. Also, this new mesostructured catalyst is comparable in terms of price, non-toxicity, stability and ease of separation.

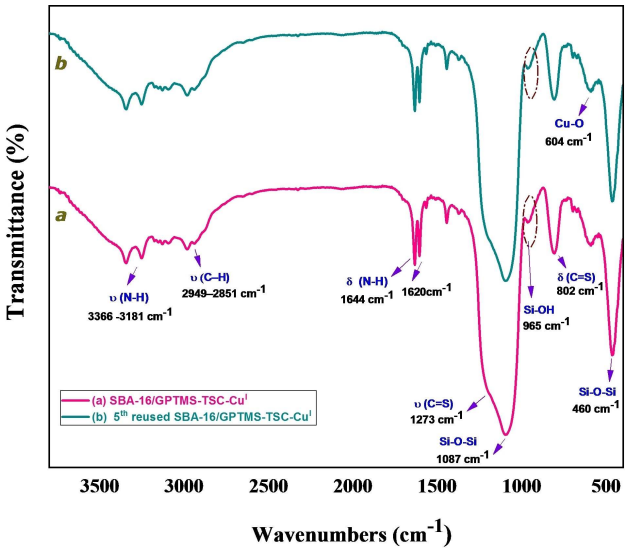


Figure 1. FT-IR spectra of (a) fresh SBA-16/GPTMS-TSC-Cu^{I} (**IV**) (b) and 5th reused SBA-16/GPTMS-TSC-Cu^{I}.

Conclusion

In the present study, SBA-16/GPTMS-TSC-Cu^{I} (with a unique "super-cage" structure and narrow particle size distribution (3-7 nm)) was introduced as an efficient, eco-friendly, non-hazardous, chemically stable and heterogeneous mesostructured catalyst in a condensation reaction of 2-aminobenzamide with substituted aromatic and aliphatic aldehydes for construction of 2,3 dihydroquinazolin-4(1*H*)-ones under solvent-free conditions. The method consistently has the advantages of environmental acceptability, high yields, short reaction times, excellent functional group compatibility and easy experimental and work-up procedures. Moreover, results clearly indicate the excellent recycling capability of catalyst (up to five runs with little loss of activity), which is the advantage of the present method over existing ones for the synthesis of titled compounds.

Experimental

General

All the chemical reagents and solvents were purchased from Merck chemical company and were used as received without any further purification. The purity determinations of the products and reaction progresses were obtained by TLC on silica gel polygram STL G/UV 254 plates. The melting points of the products were determined with an Electrothermal Type 9100 melting point apparatus. The FT-IR spectra were recorded on an AVATAR 370 FT-IR spectrometer (Therma Nicolet spectrometer, USA) using KBr plates at room temperature in the range between 4000 and 400 cm^{-1} with a resolution of 4 cm^{-1}. Mass spectra were recorded with a CH7 A Varianmat Bremen instrument at 70 eV electron impact ionization, in *m/z* (rel.%). The NMR spectra were recorded on a Bruker Avance 300 MHz instrument in DMSO-d_6 and $CDCl_3$ as the solvents. ICP-OES was carried out on a 76004555 SPECTRO ARCOS ICP-OES analyzer. Cu^{I} anchored onto mesoporous SBA-16 functionalized by aminated 3-glycidyloxypropyltrimethoxysilane with thiosemicarbazide (SBA-16/GPTMS-TSC-Cu^{I}) was prepared by the method reported in the literature.[51] All the yields refer to the isolated products after purification by recrystallization from ethanol or thin-layer chromatography.

Typical procedure for preparation of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one in the presence of SBA-16/GPTMS-TSC-Cu^{I}

A mixture of 2-aminobenzamide (1.05 mmol, 0.143 g), benzaldehyde (1 mmol, 0.106 g) and SBA-16/GPTMS-TSC-Cu^{I} (7.4 mol%, 0.039 g) was stirred at 60 °C. Reaction progress was monitored by TLC (ethylacetate:*n*-hexane, 1:3). After the required time (20 min), the reaction mixture was allowed to cool to room temperature. Thereafter, EtOAc (5 ml) was added to the reaction mixture and the mesostructured catalyst was separated by simple filtration, washed with distilled water (2 × 5 mL), ethanol (2 × 5 mL), and dried under vacuum at 60 °C for the use in the next run. The crude product was then extracted with ethyl acetate (any inorganic impurities was removed by

Table 4. Comparison of the catalytic activity of SBA-16/GPTMS-TSC-Cu^{I} with some literature precedents for preparation reaction of 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one

Ref	Reusability	IsolatedYield (%)	Time(min)	Temperature (°C)	Solvent	Catalyst	Entry
41	8	97	50	Reflux	H_2O	$Cu(NO_3)_2/F_3O_4$-DETA	1
42	-	85	60	80	CH_3CN	Amberlyst-15	2
54	6	99	60	Reflux	EtOH	Fe_3O_4-chiff base of Cu (II)	3
53	-	84	48 (h)	Reflux	CH_2Cl_2	$Sc(OTf)_3$, Fluorous bis(oxazolines)	4
31	-	95	40	r.t	EtOH	$NaHSO_4$	5
29	-	93	1.5 (h)	r.t	EtOH	$Y(OTf)_3$	6
45	1	99	24 (h)	r.t	$CHCl_3$	Chiral SPINOL-phosphoric acids, 3 Å MS	7
32	-	92	20	70	-	Lactic acid	8
46	3	96	24	Reflux	H_2O	SiO_2-$H_3PW_{12}O_{40}$	9
30	-	95	30	r.t	CH_3CN	$BiBr_3$	10
47	-	90	2 (h)	Reflux	MeOH	TBAHS[a]	11
Present study	5	98	20	60	-	SBA-16/GPTMS-TSC-Cu^{I}	12

[a]TBAHS = Tetrabutylammonium hydrogen sulfate

adding water to the filtrate) and the organic layer was dried over anhydrous Na_2SO_4. After the evaporation of the solvent, the residue was purified by recrystallization from EtOH to yield the desired pure 2-phenyl-2,3-dihydroquinazolin-4(1*H*)-one (0.219 g, 98%).

Supporting Information Summary

Experimental section, FT-IR, ^{1}HNMR, ^{13}CNMR, Mass spectrum and CHNS analysis of products were described.

Acknowledgements

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Keywords: 2,3-dihydroquinazolin-4(1*H*)-ones · SBA-16/GPTMS-TSC-Cu^{I} · aldehyde · 2-aminobenzamide · solvent-free conditions

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