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ORIGINAL PAPER

A green and efficient procedure for one-pot synthesis of xanthenes and acridines using silica boron–sulfuric acid nanoparticles (SBSANs) as a solid Lewis-protic acid

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Abstract Silica boron–sulfuric acid nanoparticles (SBSANs) as a solid Lewis-protic acid have been found to be an efficient heterogeneous catalyst in the synthesis of xanthene and acridine derivatives. The SBSAN-catalyzed reaction between carbonyl compound (aldehyde/ketone/ethyl orthoformate) and 5,5-dimethyl-1,3-cyclohexanedione (dimedone) for synthesis of xanthene derivatives is performed under mild conditions with the excellent isolated yield. Also, we can apply a broad scope of carbonyl compounds and amines for efficient synthesis of various acridine derivatives in the presence of SBSAN catalyst. In these multicomponent approaches the SBSAN catalyst can be reused for several times without any treatment in its catalytic activity.

Keywords Silica boron–sulfuric acid nanoparticles (SBSANs) · Solid acid · Lewis-protic acid · Xanthenes · Acridines

Introduction

Heterogeneous catalysts, especially solid acids have attracted increasing interest in organic synthesis, because they can provide a green and efficient media for organic transformations [1–7]. Solid acid catalysts help us to design a new procedure for excellent performing of acid-catalyzed reactions, with maximum product efficiency and minimum environmental impacts, as two important principles of green chemistry [8–10].

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The use of silica as substrate for preparation of solid acids is the most popular strategy in industrials and academic researches, due to ease of its availability and functionalization [11–13]. Solid acids with sulfonic/sulfuric acid groups based on silica have been used as heterogeneous catalysts in a large number of organic processes [14–17]. It is noteworthy that, when sulfonic/sulfuric groups as organic moieties have been grafted onto silica, their activity and selectivity are decreased due to a decreasing in the amount of reactant diffusion to the surface of catalyst [18, 19]. Thus, one of the goals in the preparation of solid acids is control of its reactivity with the ultimate goal of preparation a heterogeneous catalyst with the versatility of a homogenous catalyst in activity [20, 21]. One way that this aim can be achieved is that, the silica support is selected in nanometer scale, so the heterogeneous catalyst will even be dispersible in solution as pseudo-homogenous catalyst to form an emulsion to increase the velocity of reaction with increasing the diffusion rate [22, 23]. The other way is induction of Lewis acid sites' influence on the protic sites of catalysts which is known as Brønsted/Lewis acid synergy (BLAS) [24]. According to the BLAS concept, the coordination of Lewis acid sites to the nearest connected donor atom of protic sites (such as oxygen atom) has resulted in an enhancement in the acidity of Brønsted sites [25]. As a result, by use of these two most important strategies we can improve the reactivity of solid acids.

Previously, we have introduced the silica boron–sulfuric acid nanoparticles (SBSANs) as a new solid-acid catalyst based on nanometer scale silica support with two Lewis and protic acidic sites [26]. SBSAN catalyst has a nanoparticle nature and so its solution dispersibility has improved to some extent. It also has two acidic sites, one the protons of sulfuric acid group and other the boron atoms as Lewis acid. It seems that, in SBSAN catalyst there

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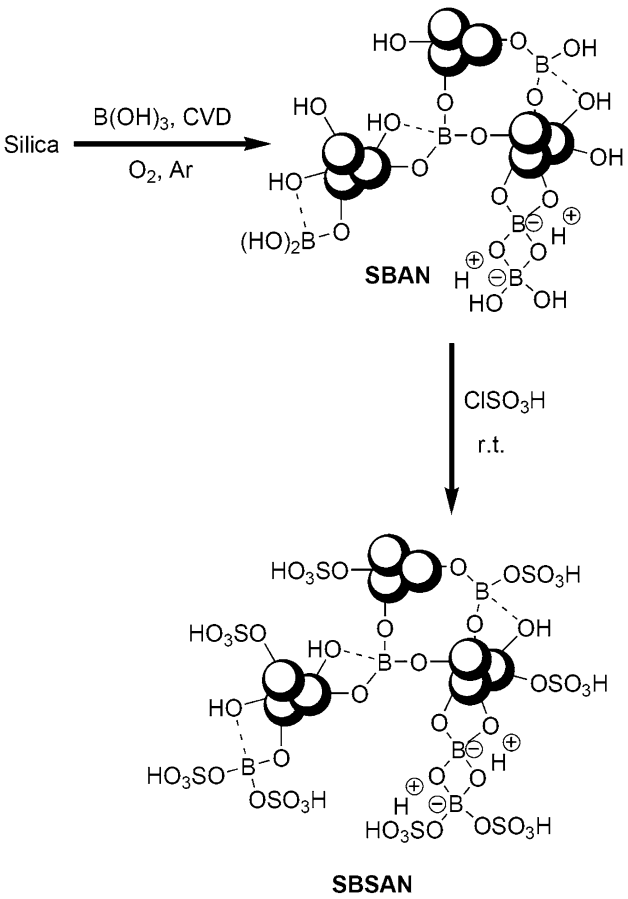
is a BLAS effect between Lewis and protic acidic sites, because its catalytic activity is decreased in donor solvents and on the other hand it shows a good reactivity in solvent-free conditions. The high reactivity of SBSAN catalyst became interesting for our research group because it effectively catalyzed the Ritter reaction as acid catalyzed at room temperature and under solvent free condition with the excellent results [26]. These interesting results prompted us to check other acid-catalyzed reactions. In the current study, we would like to present another important application of SBSAN as an efficient heterogeneous catalyst for synthesis of 1,8-dioxo-octahydroxanthenes and 1,8-dioxo-decahydroacridines under mild condition.

Two categories of materials which can be obtained using multicomponent reactions (MCRs) [27, 28] over an acid catalyst are 1,8-dioxo-octahydroxanthenes and 1,8-dioxo-decahydroacridines. Acridines, as a poly functionalized 1,4-dihydropyridine derivatives have attracted great interest for their widespread applications including the treatment of cardiovascular diseases, synthesizing of conjugated labels, for DNA-binding, synthesizing of dyes, in construction of fluorescent material for visualization of biomolecules, and in laser technologies [29–34]. Xanthenes have received significant attention due to their biological and pharmaceutical properties, such as antiviral effects, antibacterial properties, antinociceptive, and anti-inflammatory activities [35–38]. For synthesis of these classes of compounds there are several reports in the literature. Some of the catalysts which have been used for promotion of these reactions are as follows: $InCl_3{\cdot}4H_2O$ [39], diammonium hydrogen phosphate [40], *p*-dodecylbenzenesulfonic acid [41], Fe^{3+}-montmorillonite [42], $NaHSO_4$–SiO_2 or silica chloride [43], amberlyst-15 [44], silica sulfuric acid (SSA) [45], tetrabutylammonium hydrogen sulfate [46], 1-butyl-3-methylimidazolium hydrogen sulfate [47], montmorillonite K-10-supported [48], silica-bonded s-sulfonic acid (SBSSA) [49], trichloroisocyanuric acid [50], DABCO-bromine [51], silica-supported polyphosphoric acid (PPA–SiO_2) [52], and L-proline [53]. Although each of these methods has its benefits some of them often suffer from one or more disadvantages. Using of only reactive and conventional substrates, low yield, prolonged reaction time, tedious work-up processes, expensive reagents, and hazardous reaction conditions are most important deficiencies associated with these methods [54–59].

Herein, a convenient and efficient method for synthesis of 1,8-dioxo-octahydroxanthenes and 1,8-dioxo-decahydroacridines using SBSAN catalyst under solvent-free condition is described. These conditions are appropriate for applying a broad scope of carbonyl compounds and amines for synthesis of new xanthene and acridine derivatives. In this work, for the first time, ketones and ethyl orthoformate were used as carbonyl group source for the synthesis of new categories of xanthenes and acridines.



Scheme 1 Synthetic rout for preparation of SBSAN catalyst

Results and discussion

This study was initiated with synthesis of SBSAN catalyst based on our previous procedure [26]. The chemical processes for the preparation of SBSAN catalyst is depicted in Scheme 1.

According to the Scheme 1, SBSAN catalyst is prepared in a two-step method. The main part of the synthetic route for the preparation of SBSAN catalyst is the synthesis of silica boric acid nanoparticles (SBAN). This material was regularly produced during the modification of silica by boric acid over the chemical vapor deposition (CVD) process. The treatment of SBANs with chlorosulfonic acid ($ClSO_3H$) resulted in the formation of SBSAN catalyst. The SBSAN catalyst has been fully characterized using some different microscopic and spectroscopic techniques [26].

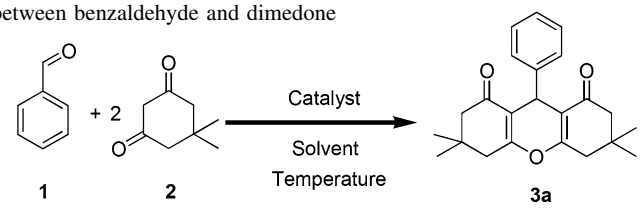
Due to the drastic reduction in catalytic activity of SBSAN catalyst in coordinating solvents, it seems that the boron atoms have a key role in the reactivity of this catalyst. In accordance with the proposed structure for SBSAN catalyst, Lewis acidity of the boron atoms is only resulted from the tri-coordinated borons which are

expected to be in the structure of this catalyst. In the structure of SBSAN catalyst there are also protic acidic sites which are related to the sulfonated boric acid and also hydroxy groups of silica. One of the possible structures for SBSAN catalyst, considering to the observation of B–O–B bond in the structure of this catalyst [26], is the complex nature of boron atoms. In the proposed structure for SBSAN, the counter ion of boronate complex is proton ions, which can be the protic sites of SBSAN catalyst. It is noticeable that the Lewis acidity of boron atoms in the structure of SBSAN catalyst is increased due to the presence of sulfonic group as electron withdrawing group on the boron atoms. In SBSAN catalyst between protic and Lewis sites there is a BLAS relationship, and perhaps this is the reason why the SBSAN catalyst effectively catalyzes the reactions under mild and solvent-less conditions.

To evaluate the catalytic activity of SBSAN catalyst in these multicomponent reactions, the reaction between benzaldehyde (**1**) and dimedone (**2**) was chosen as a model to found optimization conditions for the preparation of 1, 8-dioxo-octahydroxanthenes. The results of optimization study are summarized in Table 1.

According to the Table 1, the reaction between benzaldehyde and dimedone was performed in the presence of SBSAN catalyst at room temperature under solvent-free condition with 87 % isolated yield after 12 h (Table 1, entry 1). This reaction was also accomplished at 50 °C, and 95 % isolated yield of product was obtained after 2 h (Table 1, entry 2). The increasing of temperature to 80 °C did not have any effect on the reaction progress (Table 1, entry 3), so we decided to select 50 °C as optimum temperature for this reaction. The reaction was performed in refluxing ethanol, but the yield of product was decreased to some extent (Table 1, entry 4). In refluxing dichloromethane, the yield of desired product was enhanced to 85 % (Table 1, entry 5). In water as a green solvent, the yield of product is decreased significantly, and only 43 % yield of product was obtained (Table 1, entry 6). These comparisons simply reveal the effect of donor solvents on the catalytic activity of SBSAN. It can be seen that the yield of **3a** was decreased to 54 % when SBAN was used

Table 1 Optimization of reaction between benzaldehyde and dimedone



Entry	Catalyst	Solvent[a]	*T*/°C	Time/h	Yield of **3a** (%)[b]
1	SBSAN	None	r.t.	5	82 (87)[c]
2	SBSAN	None	50	2	95 (96)[c]
3	SBSAN	None	80	2	96 (97)[c]
4	SBSAN	EtOH	Reflux	12	82
5	SBSAN	CH_2Cl_2	Reflux	12	85
6	SBSAN	H_2O	Reflux	24	43
7	SBAN	None	80	24	54
8	Silica	None	80	12	Trace
9	$B(OH)_3$	None	80	24	Trace
10	Silica/$B(OH)_3$	None	80	24	Trace[d]
11	SSA	None	50	5	63
12	SBSAN	None	50	12	97[e]
13	SBSAN	None	50	24	68[f]

Amount of materials in all reactions: dimedone (2.2 mmol), benzaldehyde (1 mmol) and catalyst: SBSAN (0.05 g, 4.5 mol %), SBAN (0.1 g), silica (0.1 g) and $B(OH)_3$ (0.1 g)

[a] 5 mL of solvent was used

[b] Isolated yield

[c] The yields in practice are for time of 12 h

[d] A mixture of silica (0.05 g) and $B(OH)_3$ (0.05 g)

[e] Amount of catalyst used: 0.075 g (6.75 mol%)

[f] Amount of catalyst used: 0.025 g (2.25 mol%)

as catalyst at 80 °C in solvent free condition (Table 1, entry 7). The reaction did not carry out in the presence of silica and boric acid (Table 1, entries 8 and 9). The same result obtained when we used a mixture of silica and boric acid as catalyst (Table 1, entry 10). These results have also shown that the catalytic sites of SBAN are different from silica and boric acid in normal form. Also, the yield of desired product was obtained 63 %, when silica sulfuric acid (SSA) was used as catalyst under the same condition (Table 1, entry 11). This observation has also confirmed that the reactivity of SBSAN has not resulted only from the sulfuric acid groups.

The above results also confirmed that the boron atoms have effect on catalytic activity of SBSAN and so the existence of a BLAS between its protic and Lewis sites is unavoidable. However, the nano-particle nature of silica support should also be considered. It was seen that similar results were obtained by increasing the amount of catalyst (6.75 mol%) (Table 1, entry 12). As a result of decreasing the amount of catalyst (2.25 mol%), both reaction time and yield of product were changed (Table 1, entry 13). As clearly shown in Table 1, the best results for this protocol were obtained with catalyst loading of 4.5 mol% of SBSAN at 50 °C in solvent free condition. With this efficient protocol in our hand we set out to extend the scope of the substrates and so some new 1,8-dioxo-octahydroxanthenes were synthesized under optimized conditions. The results are depicted in Table 2.

SBSAN catalyst proved to be highly efficient and we realized a good general applicability of the protocol due to its significant functional group compatibility. One of the most important applications of this catalyst system is in the preparation of 1,8-dioxo-octahydroxanthenes whose corresponding aldehydes are aliphatic and have low boiling point (Table 2, entries 2–4). An interesting application of this protocol is the use of ethyl orthoformate as a carbonyl group source and 9-ethoxy-3,4,6,7-tetrahydro-3,3,6,6-tetramethyl-2H-xanthene 1,8(5H,9H)-dione as new ether derivative of 1,8-dioxo-octahydroxanthenes was obtained in the excellent yield (Table 2, entry 5). SBSAN catalyst provides efficient conditions for utilization of ketones as carbonyl component to produce xanthene derivatives with high structure diversity. For example, by the use of acetone as a simple ketone, 3,4,6,7-tetrahydro-3,3,6,6,9,9-hexamethyl-2H-xanthene-1,8(5H,9H)-dione compound was produced (Table 2, entry 6). Cyclic ketones were also used to generate the spiro-xanthenes as a new class of xanthene derivatives (Table 2, entry 7). These compounds may improve the biological activity of the 1,8-dioxo-octahydroxanthenes and/or create a new biological property for application in medicinal chemistry in future. Under optimized conditions by use of acetophenone any product was observed (Table 2, entry 8). Although the progress of this reaction depended on the aldehyde, all aldehydes, especially aromatic types, were rapidly converted to the corresponding products with excellent isolated yields in short reaction time (Table 2, entries 9–17). It seems that electron-withdrawing groups on the aryl ring increased the reaction rate, so that 4-nitrobenzaldehyde as well as 3,4-dimethoxybenzaldehyde reacted with dimedone in the presence of SBSAN catalyst (Table 2, entries 7 and 8). Interestingly, the reaction of dimedone and terephthaldehyde has resulted in the production of compound **3q** with 73 % isolated yield. This product has an aldehyde functional group which can be used for synthesizing of dual-polycyclic compounds or other compounds with a xanthene structural motif (Table 2, entry 17).

Condensation reaction between dimedone, benzaldehyde, and aniline (**4**) was checked as a simple model substrate for preparation of 1,8-dioxo-decahydroacridine derivatives using SBSAN catalyst, and after only 1 h the corresponding product was obtained with 96 % isolated yield at 50 °C under solvent-free condition (Scheme 2).

The above results encourage us to apply the SBSAN catalyst in synthesis of new 1, 8-dioxo-decahydroacridine derivatives (Table 3) using different carbonyl compounds and amines.

In first selection, we used adenine and adenosine as amine component to increase the structural diversity of this process and these multicomponent reactions lead to the formation of new nucleobase derivatives (Table 3, entries 1 and 2). The functional group compatibility of this reaction was highlighted using attached compounds or functionalized groups by spacer with primary amines (Table 3, entries 3 and 4). Another important feature of this method is the use of ammonia (ammonium carbonate) as an amine component to generate another class of dihydropyridine derivative (Table 3, entry 5). To evaluate the applicability of this protocol for aliphatic aldehydes we also synthesized the compounds **5g** and **5h** using 1,3,5-trioxane and acetaldehyde, respectively (Table 3). Although the SBSAN catalyst provides highly acidic environment, we can synthesize the compound **5i** with good yield using thiophene-2-carbaldehyde as an acid-sensitive substrate (Table 3). One of the most important applications of this protocol is the use of ketones instead of aldehydes to produce new derivatives of acridines which may be biologically active (Table 3, entry 9). If a cyclic ketone be used, the formed product is a spiro-acridine (Table 3, entry 10).

The recycling possibility of SBSAN catalyst was examined by the reaction of benzaldehyde and dimedone under optimizing conditions. When the reaction was completed, the reaction mixture was washed with hot ethanol and then catalyst separated by simple filtration.

Table 2 One-pot Synthesis of 1, 8-dioxo-octahydroxanthenes using SBSAN catalyst under mild conditions

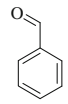
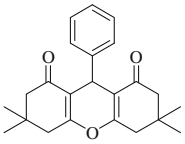
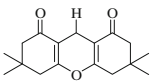
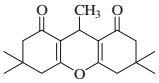
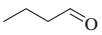
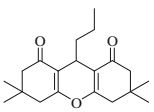
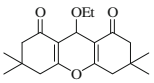
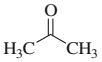
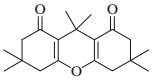
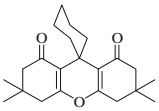
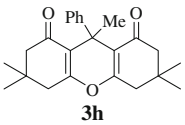
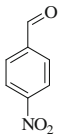
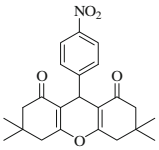
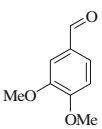
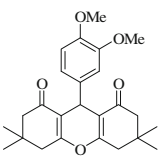
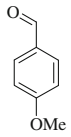
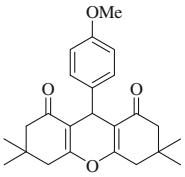
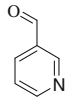
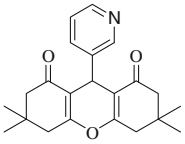
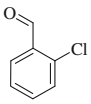
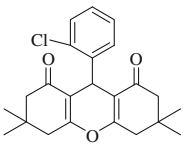
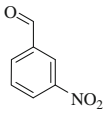
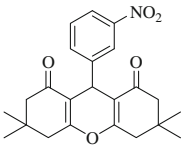
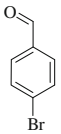
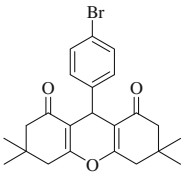
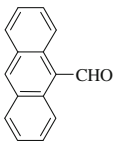
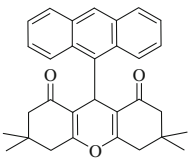
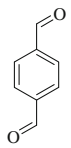
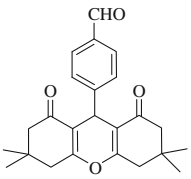
Entry	Substrate	Product	Time (h)	Yield (%)[a]	Mp (°C)
1		 **3a**	2	95	202–203 Lit: 201–203 [58]
2		 **3b**	3	91	Oil
3		 **3c**	3	92	Oil
4		 **3d**	2	90	65–68
5		 **3e**	3	91	Oil
6		 **3f**	4	90	Oil
7		 **3g**	5	91	Oil
8		 **3h**	12	0	–
9		 **3i**	1	95	221–222 Lit: 222 [41]
10		 **3j**	1	93	185–186 Lit: 184–186 [59]

Table 2 continued

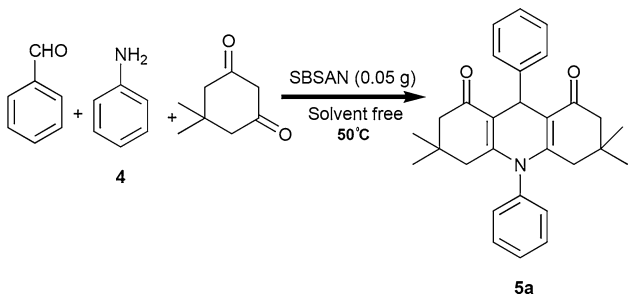
Entry	Substrate	Product	Time (h)	Yield (%)[a]	Mp (°C)
11		 **3k**	1	95	243–244 Lit: 242–244 [49]
12		 **3l**	1	94	184–185 Lit: 184–186 [49]
13		 **3m**	2	93	226–227 Lit: 226–228 [59]
14		 **3n**	1.5	93	169–170 Lit: 170–172 [49]
15		 **3o**	1	92	230–231 Lit: 230–232 [50]
16		 **3p**	2	90	203–204
17		 **3q**	1	73	185–187

Reaction condition: carbonyl compound (1 mmol), dimedone (2.2 mmol), and SBSAN (0.05 g, 4.5 mol%)

[a] Isolated yield

The recycled catalyst was heated in oven at 100 °C for 2 h and was saved for next run. The recycled catalyst could be reused seven times without any treatment (Fig. 1).

After eight times of reusability we checked the sulfur content of SBSAN catalyst using elemental analysis instrument, and the results have shown that only 0.6 % of



Scheme 2 Model substrates for synthesis of acridine using SBSAN catalyst. Reaction conditions: benzaldehyde (1 mmol), dimedone (2.2 mmol), and aniline (1 mmol)

sulfur of SBSAN catalyst was lost during this reaction. These results are in good agreement with catalyst activity of SBSAN catalyst after each recovery and no appreciable loss in the catalytic activity of SBSAN catalyst could be seen.

To compare the efficiency of our catalyst with the reported catalysts for the synthesis of xanthenes, we have tabulated the results of these catalysts for the synthesis of compounds **3a**, in Table 4.

As shown in Table 4, our catalyst is superior to some of the previously reported catalysts in terms of reaction condition, reaction time, and yield. Furthermore, in spite of most of the reported works, in our presented work, the preparation of both xanthenes and acridine derivatives using different carbonyl compounds have been achieved.

Conclusions

In summary, SBSAN with two protic and Lewis acidic sites was found to be an effective catalyst for the preparation of 1,8-dioxo-octahydroxanthene and 1,8-dioxo-9-aryl-10-aryl-decahydroacridine derivatives under green condition. By use of this heterogeneous catalyst in solvent-free condition, we could apply a broad scope of carbonyl compounds and amines to prepare a variety of xanthenes and acridines with high structural diversity. This new approach proceeded smoothly resulting in good to excellent yields. The SBSAN catalyst was separated easily from the reaction mixture by simple filtration. The use of SBSAN catalyst in these reactions provides a better and practical alternative to the existing procedures. Also, this procedure provides great promise toward further useful applications in other new multicomponent approach in future.

Experimental

Chemicals were purchased from Fluka and Aldrich Chemical Companies and used without further purification. ^{1}H and ^{13}C NMR spectra were recorded on a Bruker Avance 250 MHz spectrometer in DMSO or $CDCl_3$ solution with TMS as an internal standard. FTIR spectroscopy (Shimadzu FT-IR 8300 spectrophotometer) was employed for the characterization of the compounds. Melting points were determined in open capillary tubes in a Barnstead Electrothermal 9100 BZ circulating oil melting point apparatus. The reaction monitoring was accomplished by TLC on silica gel PolyGram SILG/UV254 plates.

Chemical vapor deposition (CVD) process was used for the synthesis of silica boric acid nanoparticles (SBANs). In this way, a brief mixture of argon and oxygen gases and the aerosols of the aqueous solution of boric acid ($\sim$2.0 g mL^{-1}) were injected into the solid silica supports positioned inside a quartz tube located in a tubing furnace at temperatures of about 600 °C. Silica boron–sulfuric acid nanoparticles (SBSANs) are prepared from the reaction of SBANs and chlorosulfonic acid ($ClSO_3H$) based on previous procedure [26].

General procedure for synthesis of 1,8-dioxo-octahydroxanthenes using SBSAN catalyst

A mixture of 5,5-dimethyl-cyclohexane-dione (0.308 g, 2.2 mmol) and aldehyde (1 mmol) in 0.05 g of SBSAN at 50 °C was stirred in a round-bottomed flask for the specified time for each compound. After completion of the reaction was confirmed by TLC, the reaction mixture was washed with hot ethanol (2 × 10 mL), and SBSAN catalyst was separated from the reaction mixture by filtration. The products were purified by recrystallization in ethanol.

General procedure for synthesis of 1,8-dioxo-9-aryl-10-aryl-decahydroacridine in the presence of SBSAN catalyst

In a round-bottomed flask a mixture of aldehyde (1 mmol), dimedone (0.308 g, 2.2 mmol), amine (1 mmol), and SBSAN (0.05 g) was stirred at room temperature for the specified time for each compound. When the reaction was completed (confirmed by TLC), the reaction mixture was washed with hot ethanol (2 × 10 mL), and SBSAN was separated from the reaction condition using a simple filtration. The separated solid after filtration, was recrystallized in ethanol to obtain pure product.

*3,3,6,6-Tetramethyl-3,4,5,6,7,9-hexahydro-1H-xanthene-1,8(2H)-dione (**3b**)*

Yield: 93 %; yellow oil. IR (liquid): ν = 2,861, 2,837, 1,640, 1,595 cm^{-1}. ^{1}H NMR (250 MHz, $CDCl_3$/TMS): δ = 0.95 (s, 6H), 1.05 (s, 6H), 2.25–2.41 (m, 8H), 4.30 (s, 2H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): δ = 27.3, 32.4, 35.0, 39.1, 46.1, 113.3, 157.6, 185. Anal. Calcd for

Table 3 The synthesized acridine derivatives using SBSAN catalyst

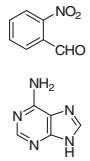
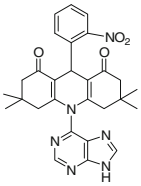
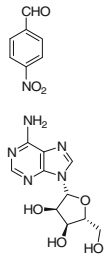
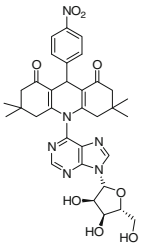
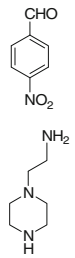
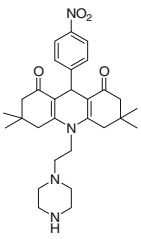
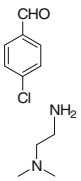
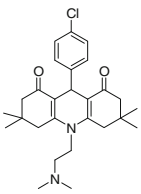
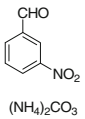
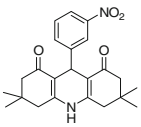
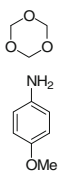
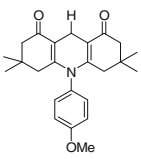
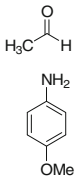
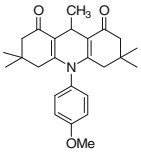
Entry	Substrates	Product	Time/h	Yield (%)[a]
1		 **5b**	3	89
2		 **5c**	3	87
3		 **5d**	2	91
4		 **5e**	2	90
5		 **5f**	2	93[a]
6		 **5g**	4	89
7		 **5h**	2	92

Table 3 continued

Entry	Substrates	Product	Time/h	Yield (%)[a]
8		**5i**	2	89
9		**5j**	5	90
10		**5k**	5	88[b]

Reaction conditions: aldehyde (1 mmol), dimedone (2.2 mmol), amine (1 mmol), and SBSAN (0.05 g)

[a] Isolated yield

[b] 2 mmol of $(NH_4)_2CO_3$ was used

Fig. 1 Catalytic recovery times of SBSAN catalyst for eight runes. Reaction condition: benzaldehyde (2 mmol), dimedone (4.4 mmol), SBSAN (0.1 g), solvent free and 50 °C. Reaction time = 2 h

$C_{17}H_{22}O_3$ (274.35): C, 74.42; H, 8.08; found: C, 74.35; H, 8.01.

*3,3,6,6,9-Pentamethyl-3,4,5,6,7,9-hexahydro-1H-xanthene-1,8(2H)-dione (**3c**)*

Yield: 89 %; pale yellow oil. IR (liquid): v = 2,860, 2,831, 1,645, 1,596 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.98$ (t, $J = 1.3$ Hz, 3H), 1.06 (s, 12H), 2.09–2.10 (m, 4H), 2.24–2.25 (m, 4H), 5.01 (q, $J = 1.2, 2.1$, 1H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 21.1$, 24.3, 29.1, 32.3, 35.0, 50.3, 99.3, 155.3, 162.4, 195. Anal. Calcd for $C_{18}H_{24}O_3$ (288.38): C, 74.97; H, 8.39; found: C, 74.91; H, 8.33.

*3,3,6,6-Tetramethyl-9-propyl-3,4,5,6,7,9-hexahydro-1H-xanthene-1,8(2H)-dione (**3d**).*

Yield: 91 %; yellow solid; mp 65–68 °C. IR (KBr): v = 2,863, 2,829, 1,640, 1,599 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.62$ (t, $J = 1.0$ Hz, 3H), 0.87 (s, 6H), 1.01 (s, 6H), 1.52–1.60 (m, 4H), 2.75–2.82 (m, 8H), 4.26 (t, $J = 1.8$ Hz, 1H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 14.1$, 20.1, 28.1, 32.5, 37.1, 44.8, 51.0, 115.3, 152.6, 192.2. Anal. Calcd for $C_{20}H_{28}O_3$ (316.43): C, 75.91; H, 8.92; found: C, 75.87; H, 8.89.

*9-Ethoxy-3,3,6,6-tetramethyl-3,4,5,6,7,9-hexahydro-1H-xanthene-1,8(2H)-dione (**3e**)*

Yield: 90 %; colorless oil. IR (liquid): v = 2,850, 2,828, 1,645, 1,597, 1,121 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.88$ (s, 6H), 0.89 (s, 6H), 1.07 (t, $J = 2.3$ Hz, 3H), 2.30–2.50 (m, 8H), 3.60–3.66 (m, 2H), 5.41 (s, 1H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 17.4$, 27.5, 31.1, 44.0, 52.5, 69.1, 111.3, 152.4, 196.5. Anal. Calcd for $C_{19}H_{26}O_4$ (318.41): C, 71.67; H, 8.23; found: C, 71.61; H, 8.21.

Table 4 Comparison of the results of the synthesis of compound **3a**, using SBSAN catalyst with those obtained by the reported catalysts

Entry	Catalyst and conditions	**3a**		References
		Time (h)	Yield (%)	
1	SBSAN, Solvent-free, 50 °C	2	95	This work
2	[bmim][BF_4]/$InCl_3$, 80 °C	5	95	[39]
3	Silica chloride, CH_3CN, reflux	6	90	[43]
4	Silica sulfuric acid, solvent free, 100 °C	1	97	[45]
5	Montmorillonite K10, solvent free, 100 °C	2	82	[48]
6	SBSSA, EtOH, reflux	10	98	[49]
7	Trichloroisocyanuric acid, EtOH, reflux	2	95	[50]
8	DABCO-bromine, H_2O, reflux	1	90	[51]
9	PPA–SiO_2, solvent-free, 140 °C	0.5	93	[52]
10	L-proline, DCE, 60 °C	6	83	[53]

*3,3,6,6,9,9-Hexamethyl-3,4,5,6,7,9-hexahydro-1H-xanthene-1,8(2H)-dione (**3f**)*

Yield: 88 %; yellow oil. IR (liquid): $v = 3{,}327$, 2,856, 2,832, 1,643, 1,593 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.98$ (s, 6H), 1.00 (s, 6H), 1.18 (s, 3H), 1.22 (s, 3H), 2.05–2.35 (m, 6H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 14.1$, 20.2, 28.7, 32.4, 42.7, 50.3, 60.6, 64.1, 101.1, 176.4, 199.5. Anal. Calcd for $C_{22}H_{30}O_3$ (342.47): C, 77.16; H, 8.83; found: C, 77.00; H, 8.78.

*3′,3′,6′,6′-Tetramethyl-3′,4′,6′,7′-tetrahydrospiro[cyclohexane-1,9′-xanthene]-1′,8′(2′H,5′H)-dione (**3g**)*

Yield: 91 %; yellow oil. IR (liquid): $v = 3{,}312$, 2,993, 2,891, 1,637, 1,547 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.98$–1.02 (m, 12H), 1.16–1.32 (m, 6H), 2.08 (s, 4H), 2.13–2.22 (m, 8H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 13.6$, 22.2, 27.4, 31.94, 42.3, 55.6, 100.1, 176.4, 199.5. Anal. Calcd for $C_{19}H_{26}O_3$ (302.41): C, 75.46; H, 8.67; found: C, 75.36; H, 8.59.

*9-(Anthracen-9-yl)-3,3,6,6-tetramethyl-3,4,5,6,7,9-hexahydro-1H-xanthene-1,8(2H)-dione (**3p**)*

Yield: 92 %; yellow solid; mp 203–204 °C. IR (KBr): $v = 2{,}967$, 1,670, 1,265, 1,057, 853 cm^{-1}. ^1H NMR (250 MHz, DMSO-d_6/TMS): $\delta = 0.97$ (s, 6H), 1.03 (s, 6H), 2.46–2.83 (m, 8H), 4.68 (s, 1H), 7.33–7.49 (m, 4H), 7.80–7.84 (m, 4H), 8.12 (s, 1H). ^{13}C NMR (62.5 MHz, DMSO-d_6/TMS): $\delta = 15.9$, 22.5, 27.8, 51.7, 52.7, 108.1, 124.9, 125.4, 126.2, 127.4, 128.7, 129.9, 130.5, 138.1, 160.3, 196.5. Anal. Calcd for $C_{31}H_{30}O_3$ (450.57): C, 82.64; H, 6.71; found: C, 82.57; H, 6.65.

*4-(3,3,6,6-Tetramethyl-1,8-dioxo-2,3,4,5,6,7,8,9-octahydro-1H-xanthen-9-yl)benzaldehyde (**3q**)*

Yield: 73 %; white solid; mp 185-187 °C. IR (KBr): $v = 2{,}859$, 2,831, 1,645, 1,561 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.96$ (s, 6H), 1.06 (s, 6H), 2.17–2.51 (m, 8H), 4.70 (s, 1H), 7.07 (t, $J = 0.9$ Hz, 2H), 7.26 (q, $J = 1.2$, 1.0 Hz, 2H), 11.84 (s, 1H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 27.7$, 30.0, 40.0, 45.0, 50.1, 113.1, 129.5, 129.7, 130.8, 144.7, 156.5, 191.4, 197.8. Anal. Calcd for $C_{24}H_{26}O_4$ (378.46): C, 76.17; H, 6.92; found: C, 76.12; H, 6.88.

*10-(1H-Benzo[d]imidazol-7-yl)-3,3,6,6-tetramethyl-9-(2-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (**5b**)*

Yield: 91 %; pale yellow solid; mp 245–247 °C. IR (KBr): $v = 3{,}243$, 3,085, 1,615, 1,591, 1,550, 1,461, 1,330 cm^{-1}. ^1H NMR (250 MHz, DMSO-d_6/TMS): $\delta = 1.05$ (s, 6H), 1.12 (s, 6H), 2.41–2.51 (m, 8H), 5.4 (s, 1H), 7.14 (d, $J = 3.2$ Hz, 1H), 7.26 (q, $J = 2.0$, 1.5 Hz, 1H), 7.33 (q, $J = 2.7$, 1.3 Hz, 1H), 7.80 (d, $J = 1.5$ Hz, 1H), 8.29 (d, $J = 3.2$ Hz, 2H), 9.80 (brs, 1H). ^{13}C NMR (62.5 MHz, DMSO-d_6/TMS): $\delta = 27.3$, 33.1, 55.4, 56.0, 109.9, 119.1, 126.5, 127.5, 129.8, 130.4, 134.5, 137.6, 148.1, 149.0, 150.3, 151.4, 164.2, 192.1. Anal. Calcd for $C_{30}H_{30}N_4O_4$ (510.58): C, 70.57; H, 5.92; N, 10.97; found: C, 70.49; H, 5.87; N, 10.95.

*10-(1-((2R,3R,4S,5R)-3,4-Dihydroxy-5-(hydroxymethyl)tetrahydrofuran-2-yl)-1H-benzo[d]imidazol-4-yl)-3,3,6,6-tetramethyl-9-(4-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (**5c**)*

Yield: 88 %; pale yellow solid; mp >300 °C. IR (KBr): $v = 3{,}125$, 2,968, 2,881, 1,635, 1,591, 1,564, 1,473, 1,447,

1,365, 1,312, 1,225, 1,148, 991, 560 cm^{-1}. ^1H NMR (250 MHz, DMSO-d_6/TMS): $\delta = 1.03$ (s, 6H), 1.10 (s, 6H), 2.30–2.42 (m, 8H), 3.65 (d, $J = 4.2$ Hz, 1H), 3.76–3.82 (m, 2H), 4.23 (d, $J = 7.6$ Hz, 1H), 4.63 (d, $J = 8.7$ Hz, 1H), 4.90 (s, 1H), 5.14 (brs, 3H), 5.82 (d, $J = 1.3$ Hz, 1H), 7.42 (d, $J = 4.2$ Hz, 2H), 8.16 (d, $J = 5.2$ Hz, 2H), 8.27 (s, 1H), 8.57 (s, 1H). ^{13}C NMR (62.5 MHz, DMSO-d_6/TMS): $\delta = 28.7$, 51.4, 61.0, 74.0, 77.5, 84.1, 90.0, 112.2, 115.1, 118.3, 119.2, 129.1, 138.8, 147.1, 148.6, 149.3, 150.0, 151.1, 151.6. Anal. Calcd for $C_{35}H_{38}N_4O_8$ (642.70): C, 65.41; H, 5.96; N, 8.72; found: C, 65.38; H, 5.91; N, 8.70.

*3,3,6,6-Tetramethyl-9-(4-nitrophenyl)-10-(2-(piperaz-in-1-yl)ethyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H, 5H)-dione (**5d**)*

Yield: 93 %; yellow solid; mp 233–235 °C. IR (KBr): $\nu = 3{,}131$, 2,967, 2,875, 1,643, 1,590, 1,566, 1,489, 1,451 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.83$ (s, 6H), 86 (s, 6H), 2.24–2.42 (m, 20H), 5.45 (s, 1H), 7.34 (d, $J = 8.3$ Hz, 2H), 7.64 (d, $J = 8.8$ Hz, 2H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 27.3$, 28.3, 32.8, 33.4, 43.1, 45.4, 50.3, 55.4, 65.4, 97.5, 114.0, 126.6, 140.0, 157.5, 162.4, 197.6. Anal. Calcd for $C_{29}H_{38}N_4O_4$ (506.64): C, 68.75; H, 7.56; N, 11.06; found: C, 68.71; H, 7.53; N, 11.01.

*9-(4-Chlorophenyl)-10-(2-(dimethylamino)ethyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (**5e**)*

Yield: 91 %; yellow solid; mp 217–219 °C. IR (KBr): $\nu = 2{,}868$, 2,537, 1,644, 1,635, 1,601, 1,524, 1,326 cm^{-1}. ^1H NMR (250 MHz, DMSO-d_6/TMS): $\delta = 1.00$ (s, 6H), 1.11 (s, 6H), 2.17 (d, $J = 1.8$ Hz, 2H), 2.25–2.41 (m, 8H), 2.45 (d, $J = 1.8$ Hz, 3H), 2.61 (t, $J = 2.6$ Hz, 3H), 3.26 (t, $J = 2.3$ Hz, 2H), 5.17 (s, 1H), 7.09–7.18 (m, 2H), 7.34–7.43 (m, 2H). ^{13}C NMR (62.5 MHz, DMSO-d_6/TMS): $\delta = 27.3$, 32.2, 41.0, 47.0, 51.5, 56.2, 66.5, 130.0, 131.1, 132.4, 136.4, 141.7, 161.2, 193.2. Anal. Calcd for $C_{27}H_{35}ClN_2O_2$ (455.03): C, 71.27; H, 7.75; N, 6.16; found: C, 71.21; H, 7.72; N, 6.13.

*3,3,6,6-Tetramethyl-9-(3-nitrophenyl)-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (**5f**)*

Yield: 95 %; white solid; mp 211–213 °C. IR (KBr): $\nu = 2{,}971$, 2,929, 1,641, 1,592, 1,514, 1,453, 1,331 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.92$ (s, 6H), 0.94 (s, 6H), 2.35–2.44 (m, 8H), 5.48 (s, 1H), 7.42 (q, $J = 5.2$, 2.3 Hz, 2H), 7.42 (q, $J = 5.2$, 2.3 Hz, 2H), 7.81 (d, $J = 3.6$ Hz, 1H), 8.31 (d, $J = 9.4$ Hz, 1H), 9.27 (s, 1H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 27.3$, 31.2, 38.3, 42.5, 50.8, 110.6, 118.2, 124.9, 128.1, 134.6, 142.6, 148.9, 192.4. Anal. Calcd for $C_{23}H_{26}N_2O_4$ (394.46): C, 70.03; H, 6.64; N, 7.10; found: C, 69.97; H, 6.61; N, 7.04.

*10-(4-Methoxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (**5g**)*

Yield: 90 %; pale yellow solid; mp 156–158 °C. IR (KBr): $\nu = 3{,}347$, 2,877, 2,824, 1,648, 1,583, 1,554, 1,354 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.93$ (s, 6H), 0.94 (s, 6H), 2.04–2.21 (m, 8H), 3.60 (s, 3H), 5.21 (s, 2H), 6.77 (d, $J = 3.2$, 2H), 7.00 (d, $J = 3.3$, 2H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 35.8$, 44.4, 50.6, 56.0, 97.5, 114.4, 126.6, 131.0, 157.5, 162.4, 197.6. Anal. Calcd for $C_{24}H_{29}NO_3$ (379.49): C, 75.96; H, 7.70; N, 3.69; found: C, 75.91; H, 7.64; N, 3.65.

*10-(4-Methoxyphenyl)-3,3,6,6,9-pentamethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione (**5h**)*

Yield: 92 %; pale yellow solid; mp 159–161 °C. IR (KBr): $\nu = 2{,}938$, 2,829, 1,639, 1,589, 1,443, 1,335, 1,261, 1,045 cm^{-1}. ^1H NMR (250 MHz, $CDCl_3$/TMS): $\delta = 0.91$ (s, 3H), 0.93 (s, 6H), 0.94 (s, 6H), 2.09–2.25 (m, 8H), 3.68 (s, 3H), 5.25 (s, 1H), 6.72 (d, $J = 3.6$ Hz, 2H), 7.08 (d, $J = 7.9$ Hz, 2H). ^{13}C NMR (62.5 MHz, $CDCl_3$/TMS): $\delta = 28.3$, 32.8, 43.1, 50.3, 55.4, 97.3, 114.4, 126.1, 131.0, 157.5, 162.4, 197.6. Anal. Calcd for $C_{25}H_{31}NO_3$ (393.52): C, 76.30; H, 7.94; N, 3.56; found: C, 76.23; H, 7.89; N, 3.51.

*9,9′-(1,4-Phenylene)bis(10-(4-methoxyphenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione) (**5j**)*

Yield: 90 %; yellow solid; mp 295–297 °C. IR (KBr): $\nu = 3{,}321$, 2,865, 2,834, 1,675, 1,648, 1,586, 1,567, 1,432, 1,419, 1,324, 1,311, 1254, 1,044, 832, 768 cm^{-1}. ^1H NMR (250 MHz, DMSO-d_6/TMS): $\delta = 1.02$ (s, 12H), 1.03 (s, 12H), 2.21–2.48 (m, 16H), 3.45 (s, 6H), 5.2 (s, 2H), 6.91–6.99 (m, 4H), 7.28 (t, $J = 5.7$ Hz, 4H), 7.34–7.39 (m, 4H). ^{13}C NMR (62.5 MHz, DMSO-d_6/TMS): $\delta = 28.0$, 31.1, 50.5, 111.5, 115.0, 116.6, 129.4, 134.4, 138.0, 151.1, 154.0, 191.1. Anal. Calcd for $C_{54}H_{60}N_2O_6$ (833.06): C, 77.85; H, 7.26; N, 3.36; found: C, 77.81; H, 7.22; N, 3.34.

*10,10′-(1,4-Phenylene)bis(9-(4-bromophenyl)-3,3,6,6-tetramethyl-3,4,6,7,9,10-hexahydroacridine-1,8(2H,5H)-dione) (**5k**)*

Yield: 87 %; yellow solid; mp 291–293 °C. IR (KBr): $\nu = 3{,}334$, 3,312, 2,895, 2,879, 1,668, 1,653, 1,581, 1,554, 1,430, 1,412, 1,319, 1,308, 1,232, 1,040, 830, 776 cm^{-1}. ^1H NMR (250 MHz, DMSO-d_6/TMS): $\delta = 0.98$ (s, 12H), 1.08 (s, 12H), 2.37–2.41 (m, 16H), 5.19 (s, 2H), 7.01

(t, $J = 3.2$ Hz, 4H), 7.24 (d, $J = 4.1$ Hz, 4H), 7.36 (t, $J = 3.2$ Hz, 4H). ^{13}C NMR (62.5 MHz, DMSO-d_6/TMS): $\delta = 27.4$, 32.3, 51.3, 112.0, 117.1, 120.3, 130.4, 141.1, 153.3, 179.5. Anal. Calcd for $C_{52}H_{54}Br_2N_2O_4$ (930.80): C, 67.10; H, 5.85; N, 3.01; found: C, 67.03; H, 5.81; N, 2.97.

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References

1. H.I. Ryoo, L.Y. Hong, S.H. Jung, D.P. Kim, J Mater Chem **20**, 6419 (2010)
2. B.K. Gorityala, J. Ma, K.K. Pasunooti, S. Cai, X.W. Liu, Green Chem **13**, 573 (2011)
3. C. Tagusagawa, A. Takagaki, A. Iguchi, K. Takanabe, J.N. Kondo, K. Ebitani, T. Tatsumi, K. Domen, Chem Mater **22**, 3072 (2010)
4. T. Wang, R.D. Ma, L. Liu, Z.P. Zhan, Green Chem **12**, 1576 (2010)
5. Y. Xu, W. Gu, D.L. Gin, J Am Chem Soc **126**, 1616 (2004)
6. D.J. Lipomi, J.S. Panek, Org Lett **7**, 4701 (2005)
7. S.V. Vyver, L. Peng, J. Geboers, H. Schepers, F.D. Clippel, C.J. Gommes, B. Goderis, P.A. Jacobs, B.F. Sels, Green Chem **12**, 1560 (2010)
8. M. Hino, K. Arata, Green Chem **3**, 170 (2001)
9. K. Arata, Green Chem **11**, 1719 (2009)
10. T. Okuhara, Chem Rev **102**, 3641 (2002)
11. S. Wang, J.A. Guin, Chem. Commun. 2499 (2000)
12. M.H. Lim, C.F. Blanford, A. Stein, Chem Mater **10**, 467 (1998)
13. S. Minakata, M. Komatsu, Chem Rev **109**, 711 (2009)
14. B. Karimi, M. Khalkhali, J Mol Catal A: Chem **232**, 113 (2005)
15. M.B. Gawande, V. Polshettiwar, R.S. Varma, R.V. Jayaram, Tetrahedron Lett **48**, 8170 (2007)
16. G.B. Brieva, M.P.D.F. Escrig, J.M.C. Martin, J.L.G. Fierro, Green Chem **12**, 1163 (2010)
17. M.P. Kapoor, W. Fujii, Y. Kasama, M. Yanagi, H. Nanbu, L.R. Juneja, J Mater Chem **18**, 4683 (2008)
18. P.D. Stevens, J.D. Fan, H.M.R. Gardinmalla, M. Yen, Org Lett **7**, 2085 (2005)
19. T.J. Yoon, W. Lee, Y.S. Oh, New J Chem **27**, 227 (2003)
20. Z.L. Lu, E. Lindner, H.A. Mayer, Chem Rev **102**, 3543 (2002)
21. A.P. Wight, M.E. Davis, Chem Rev **102**, 3589 (2002)
22. S. Banerjee, J. Das, R.P. Alvarez, S. Santra, New J Chem **34**, 302 (2010)
23. S.D. Jackson, J.S.J. Hargreaves, D. Lennon, K.M.K. Yu, S.C. Tsang, Catal. Appl. 161 (2003)
24. S. Li, A. Zheng, Y. Su, H. Zhang, L. Chen, J. Yang, C. Ye, F. Deng, J Am Chem Soc **129**, 11161 (2007)
25. A. Corma, V. Fornes, F. Rey, Appl. Catal. **59**, 267 (1990)
26. A. Khalafi-Nezhad, H.O. Foroughi, M.M. Doroodmand, F. Panahi, J Mater Chem **21**, 12842 (2011)
27. K.K. Pasunooti, C.N. Jensen, H. Chai, M.L. Leow, D.W. Zhang, X.W. Liu, J Comb Chem **12**, 577 (2010)
28. A. Khalafi-Nezhad, F. Panahi, Synthesis. 984 (2011)
29. E. Antman, J. Muller, S. Goldberg, R. Macalpin, M. Rubenfire, B. Tabatznik, C. Liang, F. Heupler, S. Achuff, N. Reichek, E. Geltman, N.Z. Kerin, R.K. Neff, E.N. Raunwald, Engl. J. Med. **302**, 1269 (1980)
30. M. Guazzi, M. Olivari, A. Polese, C. Fiorentini, F. Margrini, P. Moruzzi, Clin Pharmacol Ther **22**, 528 (1977)
31. R.S. Hornung, B.A. Gould, R.I. Jones, T.N. Sonecha, E.B. Raferty, Am J Cardiol **51**, 1323 (1983)
32. E. Delfourne, C. Roubin, J. Bastide, J Org Chem **65**, 5476 (2000)
33. J. Antonini, P. Polucci, A. Magnano, S. Martelli, J Med Chem **44**, 3329 (2001)
34. M.G. Ferlin, C. Mar-zano, G. Chiarelotto, F. Baccichetti, F. Bordin, Eur J Med Chem **35**, 827 (2000)
35. S.M. Menchen, S.C. Benson, J.Y.L. Lam, W. Zhen, D. Sun, B.B. Rosenblum, S.H. Khan, M. Taing, U. S. Patent US6583168 2003, Chem. Abstr. **139**, 54287f (2005)
36. R.C. Hunter, T. Beveridge, J. Appl. Environ. Microbiol. **71**, 2501 (2005)
37. E.F. Llama, C.D. Campo, M. Capo, M. Anadon, Eur J Med Chem **24**, 391 (1989)
38. J.P. Poupelin, G. Saint-Rut, O. Foussard-Blanpin, G. Narcisse, G. Uchida-Ernouf, R. Lacroix, Eur J Med Chem **13**, 67 (1978)
39. X. Fan, X. Hu, X. Zhang, J. Wang, Can J Chem **83**, 16 (2005)
40. F. Darviche, S. Balalaie, F. Chadegani, P. Salehi, Synth Commun **37**, 1059 (2007)
41. T.S. Jin, J.S. Zhang, J.C. Xiao, A.Q. Wang, T.S. Li, Synlett. 866 (2004)
42. G. Song, B. Wang, H. Luo, L. Yang, Catal Commun **8**, 673 (2007)
43. B. Das, P. Thirupathi, I. Mahender, K.R. Reddy, B. Ravikanth, L. Nagarapu, Catal Commun **8**, 535 (2007)
44. B. Das, P. Thirupathi, I. Mahender, V.S. Reddy, Y.K. Rao, J Mol Catal A: Chem **2347**, 233 (2006)
45. M. Seyyedhamzeh, P. Mirzaei, A. Bazgir, Dyes Pigm **76**, 836 (2008)
46. H.N. Karade, M. Sathe, M.P. Kaushik, ARKIVOC **13**, 252 (2007)
47. K. Niknam, M. Damya, J Chin Chem Soc **56**, 659 (2009)
48. A. Sharifi, M.S. Abaee, A. Tavakkoli, M. Mirzaei, A. Zolfaghari, Synth Commun **38**, 2958 (2008)
49. K. Niknam, F. Panahi, D. Saberi, M. Mohagheghnejad, J. Heterocyclic Chem. **47**, 292 (2010)
50. M.A. Bigdeli, F. Nemati, G.H. Mahdavinia, H. Doostmohammadi, Chin Chem Lett **20**, 1275 (2009)
51. M. Bigdeli, Chin Chem Lett **21**, 1180 (2010)
52. S. Kantevari, S.V.N. Vuppalapati, D.O. Biradar, L. Nagarapu, J Mol Catal A: Chem **269**, 53 (2007)
53. B. Das, J. Kashanna, R.A. Kumar, P. Jangili, Synth Commun **42**, 2876–2884 (2012)
54. A.C. Cole, J.L. Jensen, I. Ntai, K.L.T. Tran, K.J. Weaver, D.C. Forbes, J.J.H. Davis, J Am Chem Soc **124**, 5962 (2002)
55. Z.Q. Tang, Y. Chen, C.N. Liu, K.Y. Cai, S.J. Tu, J. Heterocyclic Chem. **47**, 363–367 (2010)
56. T.S. Jin, J.S. Zhang, A.Q. Wang, T.S. Li, Synth Commun **35**, 2339 (2005)
57. S. Rostamizadeh, A. Amirahmadi, N. Shadjou, A.M. Amani, J. Heterocyclic Chem. **49**, 111 (2011)
58. A. John, P.J.P. Yadav, S. Palaniappan, J Mol Catal A: Chem **248**, 121 (2006)
59. M.T. Maghsoodlou, S.M. Habibi-Khorassani, Z. Shahkarami, N. Maleki, M. Rostamizadeh, Chin Chem Lett **21**, 686 (2010)