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Synthesis of benzo[4,5]imidazo[1,2-a]pyrimidines and 2,3-dihydroquinazolin-4(1H)-ones under metal-free and solvent-free conditions for minimizing waste generation†

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Brønsted acidic ionic liquid was found to be an efficient and recyclable catalyst for the synthesis of benzo[4,5]imidazo[1,2-a]pyrimidines and 2,3-dihydroquinazolin-4(1H)-ones. The reactions proceeded smoothly with a broad scope of substrates providing the expected products in good to excellent yields under an atom-economical pathway. The low-cost recyclable catalyst, metal- and solvent-free conditions, and the ease of product isolation are the highlighted advantages in solving the issue of trace metal contamination in synthesized pharmaceuticals.

Introduction

The invention of new catalysts for the synthesis of biologically active compounds is essential to the advancement of green chemistry.[1–3] Classical aqueous inorganic acids including H_2SO_4, HF, HCl, HNO_3, and H_3PO_4 have been used for various organic transformations. Nevertheless, these homogeneous Brønsted acids are not suitable for widespread application due to separation and environmental issues.[4] Ionic liquids have attracted considerable attention in many fields due to their structural diversity, low cost, and low toxicity.[5–7] The use of ionic liquids as green catalysts for chemical reactions has presented many advantages in a sustainable synthesis.[8–13] These homogeneous catalysts are thermally stable, highly active, and easily recyclable.[14] The most popular Brønsted acidic ionic liquids have been easily prepared by the reaction of 1,4-butane sultone or 1,3-propane sultone with N-methylimidazole.[15] Thus, these Brønsted acidic ionic liquids have been widely used as green catalysts for various organic transformations.[16–24] Herein, we report the synthesis of benzo[4,5]imidazo[1,2-a]pyrimidines and 2,3-dihydroquinazolin-4(1H)-ones catalyzed by a Brønsted acidic ionic liquid under solvent-free conditions. The presented method is considered an essential solution for the environmental problems in industrial processes. The current synthesis scheme contributes to the sustainable and low-cost pathway for the preparation of benzo[4,5]imidazo[1,2-a]pyrimidines and 2,3-dihydroquinazolin-4(1H)-ones.

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Results and discussion

Synthesis of benzo[4,5]imidazo[1,2-a]pyrimidines

Pyrimidine derivatives are backbones of various pharmaceutical drugs with various applications including anticancer,[25–29] anti-inflammatory,[30–32] antibacterial,[31,33–35] and antifungal activities.[35–37] Over the past decade, many approaches have been achieved for the preparation of pyrimidine derivatives.[38–42] However, multi-step or low-yield procedures and harmful conditions could limit the widespread use of these approaches. Recently, multicomponent reactions have provided new prospects for the advancement in organic synthesis to eliminate these drawbacks.[43,44] Herein, the Brønsted acidic ionic liquid was used for the first time to catalyze the synthesis of benzo[4,5]imidazo[1,2-a]pyrimidines via a one-pot multicomponent reaction. Initially, [(4-SO₃H)BMIM]HSO₄ ionic liquid was prepared as reported in the literature.[22] The structure of [(4-SO₃H)BMIM]HSO₄ was confirmed by NMR, FT-IR, TGA, and HR-MS (ESI, Section S2†). The as-synthesized [(4-SO₃H)BMIM]HSO₄ was obtained in high yield after a simple purification step. Thermogravimetric analysis shows that [(4-SO₃H)BMIM]HSO₄ is thermally stable (up to 250 °C). Next, we investigated the model reaction between 2-aminobenzimidazole, ethyl acetoacetate, and benzaldehyde in the presence of different catalysts (10 mol%) at 100 °C for 120 min (Table 1). When the model reaction was performed using metal chlorides such as $AlCl_3$, $FeCl_3$, $FeCl_2$, and $ZnCl_2$ as the catalysts at 100 °C for 120 min, the expected product was obtained in 32% to 60% yield (Table 1, entries 1–4). These results encouraged us to test a series of Lewis acidic deep eutectic solvents with zinc chloride. However, the yields of the desired product drastically decreased (Table 1, entries 5–7). Other deep eutectic solvents were investigated by

Table 1 Yields for reaction of 2-aminobenzimidazole, ethyl acetoacetate, and benzaldehyde with different catalysts[a]

Entry	Catalyst	Yield[b] (%)
1	$AlCl_3$	57
2	$FeCl_3$	60
3	$FeCl_2$	32
4	$ZnCl_2$	60
5	[CholineCl][ZnCl]	0
6	[CholineCl][ZnCl]$_2$	22
7	[CholineCl][ZnCl]$_3$	12
8	[CholineCl][ethylene glycol]$_2$	59
9	[CholineCl][glucose]	48
10	[CholineCl][oxalic acid]$_2$	52
11	[EMIM]Cl	67
12	[BMIM]BF$_4$	69
13	[(4-SO$_3$H)BMIM]HSO$_4$	89

[a] Reaction condition: 2-aminobenzimidazole (1.0 mmol), ethyl acetoacetate (1.0 mmol), benzaldehyde (1.0 mmol), and catalyst (10 mol%) at 100 °C for 120 min. [b] Isolated yield.

replacing $ZnCl_2$ with ethylene glycol, oxalic acid or glucose (Table 1, entries 8–10), but the yields of the product were not satisfactory. Thus, we focused on ionic liquids. Three types of ionic liquids, namely, [EMIM]Cl, [BMIM]BF$_4$, and [(4-SO$_3$H) BMIM]HSO$_4$, were tested for the model reaction. When the reactions were performed in ionic liquid media, the yields of the desired product drastically increased (Table 1, entries 11–13). Among these ionic liquids, [(4-SO$_3$H)BMIM]HSO$_4$ showed the best efficiency, allowing the reaction to afford 89% yield (Table 1, entry 13). As shown in Table 1, metal chlorides could be employed in the model reaction. However, these metal chlorides are moisture sensitive and easily decompose in the presence of a small amount of water. Furthermore, these metal chlorides could not be recovered and reused after the aqueous workup. The prominent feature of [(4-SO$_3$H)BMIM]HSO$_4$ is stability in water and in almost all cases, catalytic use, recovery, and reuse of [(4-SO$_3$H)BMIM]HSO$_4$ are possible. In our study, we tested different ratios of [(4-SO$_3$H)BMIM]HSO$_4$ with the aim of obtaining the desired product in high yield. The best yield was obtained in the presence of 10 mol% [(4-SO$_3$H)BMIM]HSO$_4$. When the amount of catalyst was increased to 15 mol%, the yield slightly improved. All attempts to reduce the amount of [(4-SO$_3$H)BMIM]HSO$_4$ loading led to diminished yields.

With the optimized conditions in hand, we focused on using a variety of aromatic aldehydes. The results are presented in Scheme 1. A series of *p*-substituted benzaldehyde smoothly reacted with 2-aminobenzimidazole and ethyl acetoacetate to provide benzo[4,5]imidazo[1,2-*a*]pyrimidines in excellent yields. Electron-donating group on benzaldehyde, such as methoxybenzaldehyde, gave the expected products in excellent yield (90%), but longer reaction time was required (150 min). Electron-withdrawing groups on benzaldehyde, such as 2-nitrobenzaldehyde and 4-nitrobenzaldehyde, were all effectively reactive and provided the corresponding products in excellent yields within shorter reaction times (75 min). Benzaldehyde bearing halogen substituents could also react smoothly to

attain the expected products in high yields (88–92%), and the substituent position did not significantly affect the reaction yield. In the interest of extending the scope of reaction, we examined acetonylacetone under the current method. To our delight, the reactions proceed smoothly to provide the desired products in high yields (88–93%) and shorter reaction times (60–90 min). Moreover, the method avoids the use of volatile organic solvents, harmful conditions, and additives making it greener than the previous reports. In addition, the pure products were obtained easily by recrystallization; thus, this method shows promise for application in industry.
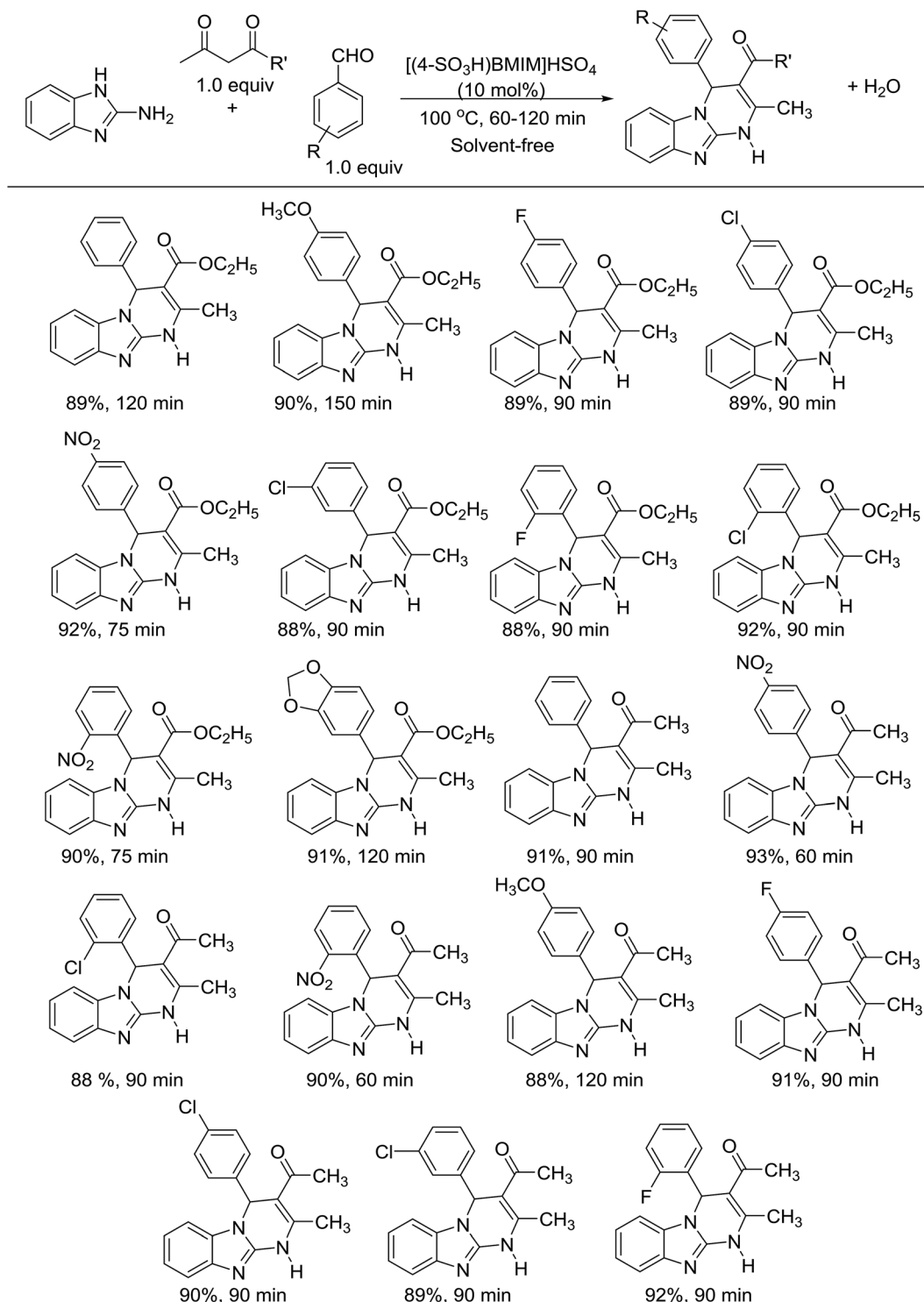
The comparison of the present method with previously reported literature is shown in Table 2. The multicomponent reaction of 2-aminobenzimidazole, ethyl acetoacetate, and benzaldehyde in the presence of [(4-SO$_3$H)BMIM]HSO$_4$ afforded the expected product in good yield within 2 h under metal-free and solvent-free conditions (Table 2, entry 6). The previous studies reported that the same reaction also achieved the target products in moderate to good yields, but some methods required longer reaction time and/or high temperature (Table 2, entries 1–5). More importantly, the [(4-SO$_3$H)BMIM]HSO$_4$ could be recovered and reused without any considerable loss in catalytic activity in a test of six cycles of reuse.

To get more information about the plausible mechanism, we performed three control experiments under the optimized conditions. 2-Benzylidene-3-oxobutanoate intermediate was detected as the major product (by GC-MS) when the model reaction was performed for 20 min. Thus, the reaction mainly followed Knoevenagel condensation between benzaldehyde and ethyl acetoacetate in the first step, where the [(4-SO$_3$H)BMIM] HSO$_4$ acted as a Brønsted acidic catalyst. The control experiments with only two components, *i.e.*, the reaction of benzaldehyde with ethyl acetoacetate, also provided the desired product (2-benzylidene-3-oxobutanoate) in 70% yield, which was higher than the reaction yield of 2-aminobenzimidazole and benzaldehyde. The intermediate could completely convert into the desired product when 2-aminobenzimidazole was added. Based on the above control experiments and previous literature, the plausible mechanism is proposed, as presented in Scheme 2. The reaction first followed Knoevenagel condensation between benzaldehyde and ethyl acetoacetate to produce 2-benzylidene-3-oxobutanoate as intermediate, which could further react with 2-aminobenzimidazole through Michael reaction. Finally, the desired product was obtained through cyclization and dehydration. The [(4-SO$_3$H)BMIM]HSO$_4$ played a vital role in four steps of the reaction.

Synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones

Quinazolinones are valuable bioactive compounds possessing antitumor,[45–47] antiviral,[48,49] antibacterial,[50,51] and anti-inflammatory activities.[52,53] Traditionally, both homogeneous and heterogeneous acidic catalysts can catalyze the synthesis of 2,3-dihydroquinazolin-4(1*H*)-ones.[54–56] Recently, various protocols have been reported for the preparation of 2,3-dihydroquinazolin-4(1*H*)-ones with the aid of improving the reaction efficiency.[57–61] Moreover, 2,3-dihydroquinazolin-4(1*H*)-
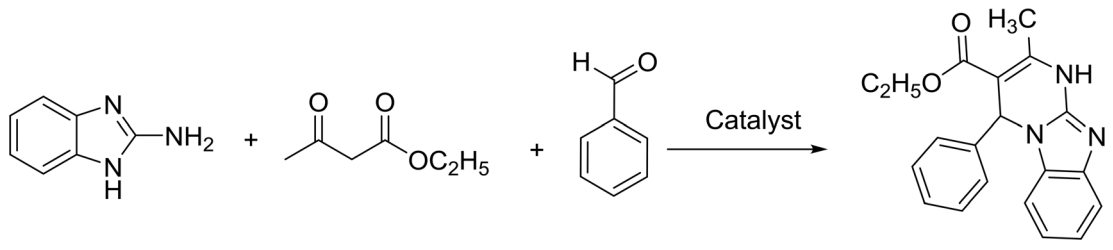
Scheme 1 [(4-SO$_3$H)BMIM]HSO$_4$-catalyzed synthesis of benzo[4,5]imidazo[1,2-a]pyrimidines.

ones have been successfully synthesized using ionic liquids including [BMIM]BF$_4$,[62] poly(4-vinylpyridine) supported acidic ionic liquids,[63] and imidazolium triflate.[64] However, methods that involved these ionic liquids presented some drawbacks, including high temperature, prolonged reaction time, and the use of volatile organic solvents. In this study, we report that [(4-SO$_3$H)BMIM]HSO$_4$ acidic ionic liquid could be used as an active catalyst for the preparation of 2,3-dihydroquinazolin-4(1H)-ones presumably due to strong effect of Brønsted acidic groups on the ionic liquid. Interestingly, various aromatic aldehydes reacted successfully under the optimized conditions, and the expected product was obtained in excellent yields in the
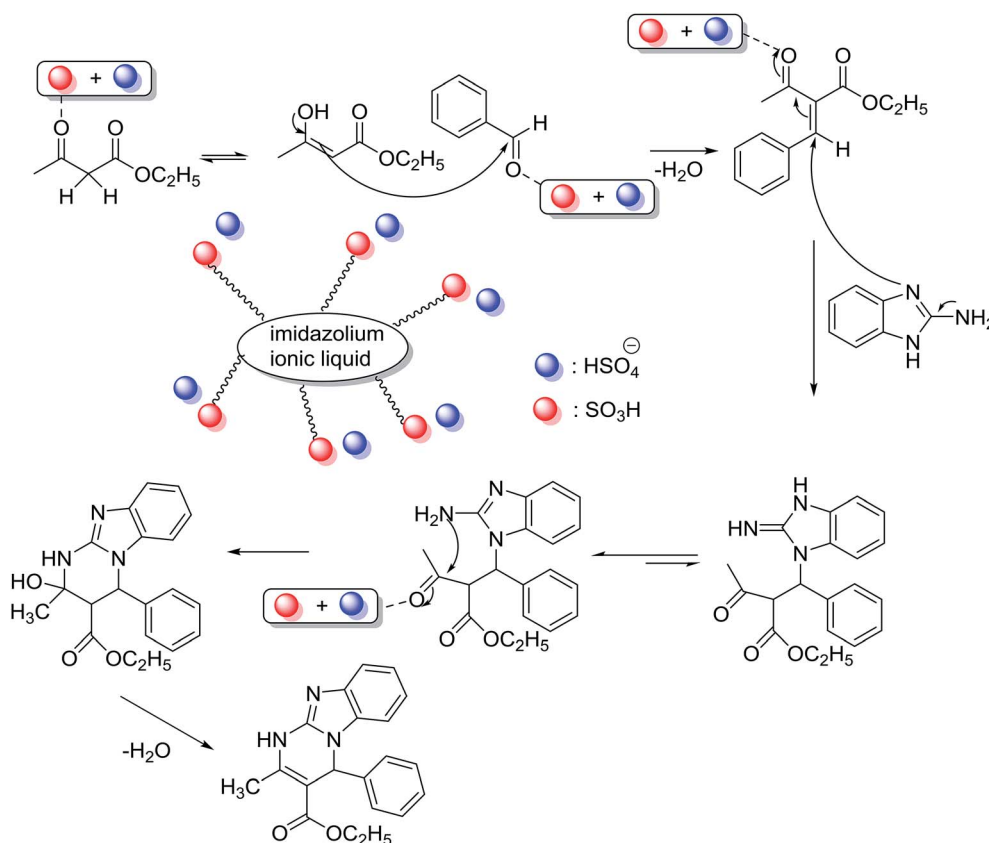
Table 2 Comparison of the present method with previous literature for the one-pot multicomponent synthesis of benzo[4,5]imidazo[1,2-a]pyrimidine
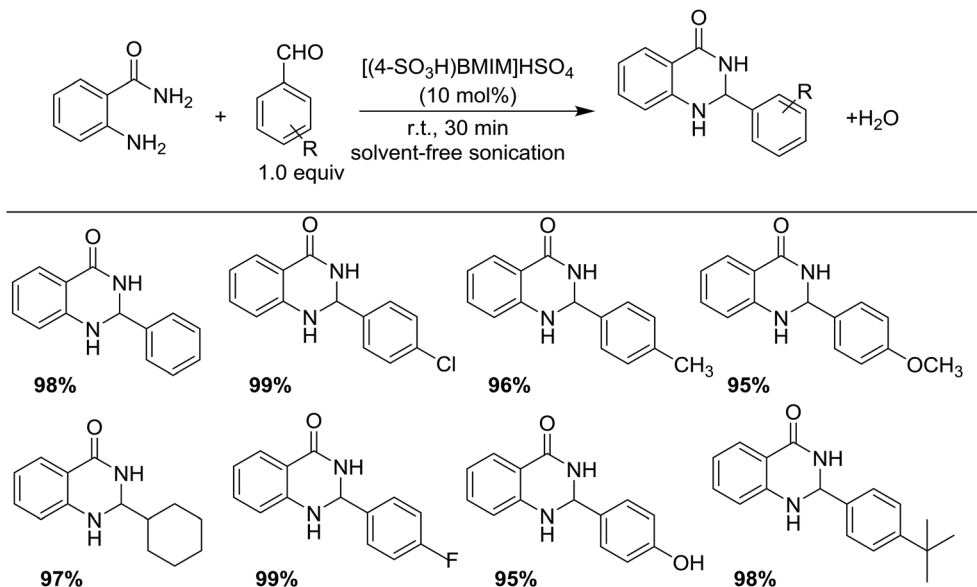

Entry	Catalytic system	Reaction conditions	Yield (%)
1	1,1,3,3-N,N,N′,N′-Tetramethylguanidinium trifluoroacetate (30 mol%)	100 °C, 5 h	73 (ref. 26)
2	[bmim][BF$_4$] (3 mL)	90 °C, 7 h	86 (ref. 36)
3	L-Proline (20 mol%), H$_2$O	Reflux, 3 h	91 (ref. 30)
4	Thiamine hydrochloride (5 mol%), H$_2$O	Reflux, 3 h	90 (ref. 41)
5	α-Zirconium sulfophenylphosphonate (12 mol%)	80 °C, 20 h	73 (ref. 42)
6	Current work: [(4-SO$_3$H)BMIM]HSO$_4$ (10 mol%), solvent-free	100 °C, 2 h	92

presence of [(4-SO$_3$H)BMIM]HSO$_4$ (Scheme 3). The substituent on the benzene did not affect the reaction rate. In addition, pure products were obtained easily by recrystallization. Remarkably, the condensation between anthranilamide and benzaldehyde was successfully performed on a 50 mmol scale, and the expected yield was obtained equally with 1 mmol scale. This method avoids the use of volatile organic solvents, harmful conditions, and metal catalysts, thus showing promise for application in industrial processes.

The comparison of the presented method with previously reported literature is shown in Table 3. The reaction of anthranilamide and benzaldehyde in the presence of [(4-SO$_3$H)

Scheme 2 Proposed mechanism for the preparation of benzo[4,5]imidazo[1,2-a]pyrimidines.

Scheme 3 Reaction scope with aldehydes.

BMIM]HSO$_4$ afforded the expected product in excellent yield at room temperature for 30 min under metal-free and solvent-free conditions (Table 3, entry 6). The previous studies reported that the same reaction also achieved the target products in good to excellent yields, but some methods required the use of a solvent, high temperature and/or longer reaction time (Table 3, entries, 1–5). Additionally, the [(4-SO$_3$H)BMIM]HSO$_4$ could be recovered and reused without any considerable loss of catalytic activity in a test of six cycles.

On the basis of our experiments and the literature, we proposed the reaction mechanism for the cyclocondensation between anthranilamide and aromatic aldehydes (Scheme 4).[56,69] The first step could be the formation of imine as a critical intermediate under the presence of [(4-SO$_3$H)BMIM]HSO$_4$. Next, the imine would be activated by the Brønsted

acidic ionic liquid and the desired product could be obtained from the intermediate by the intramolecular nucleophilic attack of the nitrogen (–NH$_2$) on the amide group to imine carbon.
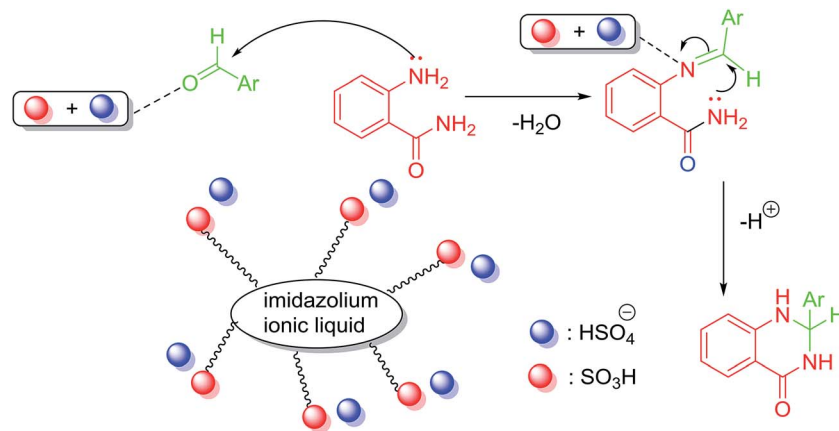
Recyclability of the catalyst is a necessary feature for the-application in industrial processes. The recyclability of [(4-SO$_3$H)BMIM]HSO$_4$ in this study was investigated in the model reactions with benzaldehyde under optimal conditions (Fig. 1). After completion of the reaction, the reaction mixture was diluted with ethanol, and the desired product was obtained by filtration. The catalyst was recovered, dried under vacuum for 6 h and reused for next cycles. The yields of the product slightly decreased after the sixth cycle. The FT-IR spectra confirmed that there was no change in functional groups of [(4-SO$_3$H)BMIM]HSO$_4$ (Fig. 2).

Table 3 Comparison of the presented method with previous literature for the synthesis of 2-phenyl-2,3-dihydroquinazolin-4(1H)-one

Entry	Catalytic system	Reaction conditions	Yield (%)
1	[BMIM]PF$_6$ (3.0 mL)	75 °C, 35 min	89 (ref. 62)
2	Cp$_2$TiCl$_2$ (1 mol%), ethanol (0.5 mL)	30 °C, 8 min	96 (ref. 65)
3	Fe$_3$O$_4$@SiO$_2$@GMSI-VB1 (10 mg), H$_2$O (1.0 mL)	80 °C, 80 min	95 (ref. 66)
4	MNPs/DETA-SA (15 mg), H$_2$O (2.0 mL)	90 °C, 40 min	94 (ref. 67)
5	p-Sulfonic acid calix[4]arene(1.0 mol%), H$_2$O (1.0 mL)	r.t., 18 min	94 (ref. 68)
6	Current work: [(4-SO$_3$H)BMIM]HSO$_4$ (10 mol%), solvent-free	r.t., 30 min	98

Scheme 4 A plausible mechanism for the synthesis of 2,3-dihydroquinazolin-4(1*H*)-one.
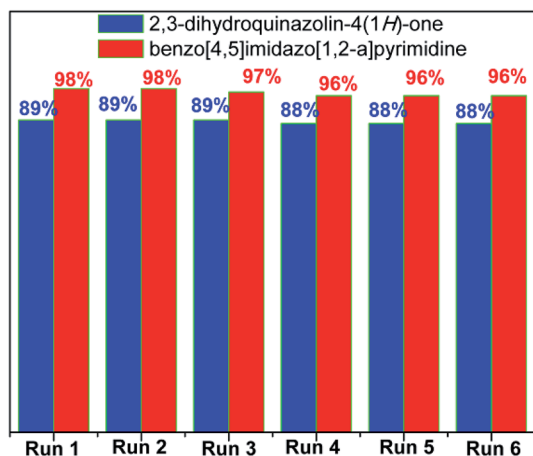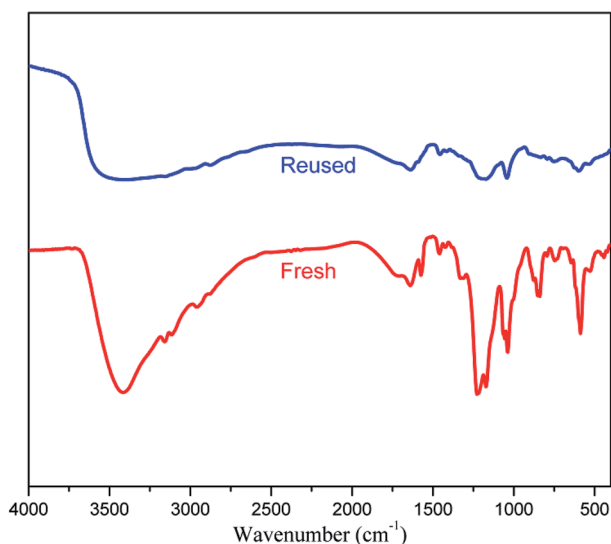

Fig. 1 Recycling test.

Fig. 2 FT-IR spectra of the fresh and the six-times reused catalyst.

Conclusions

We have developed a facile, efficient, and atom-economic method for preparing benzo[4,5]imidazo[1,2-*a*]pyrimidines and 2,3-dihydroquinazolin-4(1*H*)-ones under metal- and solvent-free conditions. The presented method provides a straightforward and green approach for the preparation of biologically nitrogen-heterocyclic compounds from the readily available starting materials in good to excellent yields within short reaction time. Remarkably, [(4-SO$_3$H)BMIM]HSO$_4$ Brønsted acidic ionic liquid showed to be particularly suitable for these transformations as highlighted by its environmental friendliness, low-cost, recyclability, and simplicity of operation.

Experimental

Materials and instrumentation

All the reagents were purchased from Merck, Acros, and Sigma-Aldrich and used without further purification. Ultrasonic irradiation was performed in Elma sonic S30H. FT-IR spectra were analyzed using a Bruker Vertex 70 (KBr pellets). Thermogravimetric analysis was performed on Analyzer TGA Q5000. ^{1}H and ^{13}C NMR spectra were performed on a Bruker Advance 500. High-resolution electrospray ionization mass spectrometry (HRMS-ESI) was performed on a Bruker micrOTOF-QII MS at 80 eV.

Synthesis of [(4-SO$_3$H)BMIM]HSO$_4$ under solvent-free sonication

[(4-SO$_3$H)BMIM]HSO$_4$ was synthesized *via* a one-pot two-step procedure under solvent-free sonication according to our previous literature report.[22]

General procedure for the synthesis of benzo[4,5]imidazo[1,2-*a*]pyrimidines

A mixture of benzaldehyde (106 mg, 1 mmol), ethyl acetoacetate (130 mg, 1 mmol), 2-aminobenzimidazole (133 mg, 1 mmol) and [(4-SO$_3$H)BMIM]HSO$_4$ (31.6 mg, 0.1 mmol) was heated 100 °C and the progress of the reaction was monitored by TLC.

After completion of the conversion, the reaction mixture was quenched with cold ethanol (10 mL). The crude product was filtered and washed with petroleum ether (10 mL), and then purified by recrystallization from ethanol to obtain the desired pure product.

General procedure for the synthesis 2,3-dihydroquinazolin-4(1H)-one

A mixture of anthranilamide (136 mg, 1 mmol), benzaldehyde (106 mg, 1 mmol), and [(4-SO$_3$H)BMIM]HSO$_4$ (31.6 mg, 0.1 mmol) was sonicated for 30 min at room temperature and the progress of the reaction was monitored by TLC or GC-MS. After completion of the conversion, the reaction mixture was quenched with ethanol (10 mL). The crude product was filtered and washed with petroleum ether (2 × 5 mL), and then purified by recrystallization from ethanol to afford the desired pure product.

Conflicts of interest

There are no conflicts to declare.

Acknowledgements

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