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Sulfonic acid functionalized pyridinium chloride [pyridine - SO_3H]Cl: novel homogeneous catalyst for solvent-free synthesis of dihydropyrimidinone derivatives

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Sulfonic acid functionalized pyridinium chloride [pyridine-SO_3H]Cl is used as a novel Brønsted acidic ionic liquid and effective catalyst under thermal solvent-free conditions. Using [pyridine-SO_3H]Cl as an efficient and recyclable catalyst leads to synthesis of 3,4-dihydropyrimidin-2(1*H*)-ones by the multicomponent Biginelli condensation of coupling between aromatic aldehydes, *β*-ketoesters and urea/thiourea under different conditions. Analytically, pure products are formed within 15–25 min in excellent yields. A simple and green chemical procedure for the synthesis of dihydropyrimidinones by two techniques, conventional heating and microwave irradiation, in the presence of a catalytic amount is investigated. Utilization of easy reaction condition, recyclable catalyst, and simple work-up make this methodology as an interesting option for the eco-friendly synthesis of Biginelli-like compounds.

Keywords: Sulfonic acid functionalized pyridinium chloride [Pyridine-SO_3H]Cl, Brønsted acidic ionic liquid, 3,4-dihydropyrimidin-2(1*H*)-ones, One-pot three component reaction, Solvent-free synthesis

INTRODUCTION

The development of simple and eco-friendly synthetic routes constitutes an important goal in green chemical technology. According to recent research, with the development of greener and eco-friendly processes, one-pot multi components reactions under solvent-free conditions with acidic ionic liquid or solid catalysts are considered as an important technique [1, 2]. As a promising approach to be environmentally consciousness, Solvent-free reactions are the subject of constant development because of its ease set-up, mild conditions, increased yields of products, efficiency and cleaner product formation compared to their solution counterparts [3, 4]. An ionic liquid (ILs) is a substance that is composed entirely of ions, and is a liquid at room temperature. Frequently the ionic liquid consists of organic cations and inorganic anions, although it is not limited to these combinations. Ionic liquids have low viscosity, essentially no vapor pressure, good heat transfer characteristics. Therefore, they are used as green solvents, catalysts, and reagents in a variety of organic transformations [5, 6]. Sulfonic acid functionalized pyridinium chloride [pyridine-SO_3H]Cl has been synthesized and used as an efficient catalyst for nitration of heterocyclic compounds [7]. Nitrogen heterocyclic compounds have been noticable to design biologically active molecules, because their structural subunits exist in many natural products as well as pharmaceuticals. Therefore, 3,4-dihydropyrimidin-2(1*H*)-ones is now recognized as a powerful heterocyclic compounds with a wide range of pharmaceutical and biological activities such as calcium channel blockers, antiviral, antihypertensive agents, antitumour, antibacterial and anti-inflammatory actions [8- 10]. Several techniques for the synthesis of 3,4-dihydropyrimidin-2(1*H*)-ones and its derivatives have been developed to improve and modify this reaction promoted by Brønsted acidic ionic liquid as well catalysts supported by non-metallic and non-toxic materials [11- 13].

Conventional heating and microwave irradiations have been established as important techniques in organic syntheses, but in all reactions it was found that the use of solvent free leads to a higher yield [14, 15]. In continuation to our interest in synthesis of heterocyclic compounds, we are reporting 1-Sulfopyridinium chloride as a recyclable homogeneous catalyst for greener synthesis of

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3,4-dihydro-pyrimidin-2-(1*H*)-ones in the one-pot multi-component condensation of aromatic aldehydes, *β*-ketoesters and urea/thiourea under classical reflux, solvent-free conditions, and microwave conditions [16- 18].

EXPERIMENTAL

General

The chemicals used in this work purchased from Fluka (Buchs, Switzerland) and were used without further purification. All known compounds were identified by comparison of their melting points and spectral data with those reported in the literature. Melting points were determined with an electrothermal 9100 apparatus and are uncorrected. IR spectra were determined as KBr discs on a Shimadzu IR-460 spectrophotometer. NMR spectra were obtained on a Bruker Avance DRX-300 MHz spectrometer (^{1}H NMR at 400 Hz, ^{13}C NMR at 100 Hz) in DMSO-d_6 using TMS as internal standard. Progress of the reactions was monitored by TLC using silica gel SIL G/UV 254 plates. Microwave reactions were carried out in a microwave oven (2500 W power; Micro-Synth, Milestone).

GENERAL PROCEDURE FOR THE SYNTHESIS OF FUSED 3,4-DIHYDROPYRIMIDINE-2(1H)-ONES (4A–N) AND THIONES(5A–C)

Classical heating method

To a magnetically stirred a mixture of an aromatic aldehyde (1 mmol), *β*-ketoesters (1 mmol), urea/thiourea (1.5 mmol) and then ionic liquid [pyridine-SO_3H]Cl (0-15% mol, Table 1) was finely mixed together in a test tube at 80°C under solvent-free conditions for an appropriate time. During the reaction process, a solid product spontaneously formed. the completion of the reaction was monitored by TLC. The reaction mixture was cooled to room temperature and diluted with ethyl acetate (10 mL) to give the pure recycled catalyst. The recovered catalyst was washed with ethyl acetate, dried under vacuum at 90 °C for about 3 h and reused for the next run of the reaction. The decanted solutions washed with water (20 mL), then stirred for 10-15 min to remove the excess of urea/thiourea and filtered. The results are summarized in (Table 2). The crude product was dissolved in hot ethanol, filtered off for removing

Microwave irradiation method

A 10 ml Pyrex tube was charged with a mixture of aromatic aldehyde (1 mmol), *β*-ketoesters (1 mmol), urea/thiourea (1.5 mmol), and [pyridine-SO_3H]Cl (0-15% mol). The reaction mixture was subjected to microwave irradiation for a suitable time till the reaction was completed (monitoring by TLC). The solid material washed with water (20 mL), then stirred for 10-15 min to remove the excess of urea/thiourea and filtered. The precipitate washed and recrystallized from EtOH to give pure crystals of pyrimidinones.

Representative spectral data

Ethyl-6-methyl-2-oxo-4-phenyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate (Table 2, 4a): White powder, FT-IR (KBr, cm^{-1}): 3337, 3226, 3106, 2949, 1700, 1670, 1652, 1420, 1339, 1239, 1092; ^{1}H NMR (400 MHz, DMSO-d_6): δ =2.26 (s, 3H, CH_3), 3.53 (s,3H, CH_3O), 5.16 (d, 1H, J=3.2, CH), 7.32-7.23 (m, 5H, Ar-H), 7.76 (s, 1H, NH), 9.22 (s, 1H, NH); ^{13}CNMR (100 MHz, DMSO-d_6): 17.80, 50.74, 53.77, 98.99, 126.13, 127.25, 128.41, 144.64, 148.62, 152.15, 165.80 ppm. Calcd for $C_{14}H_{16}N_2O_3$: C, 64.60; H, 6.20; N, 10.76; O, 18.44 %. Found: C, 64.52; H, 5.95; N, 11.10 %.

Ethyl-4-(4-chlorophenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (Table 2, 4b): White powder , FT-IR (KBr, cm^{-1}): 3443, 3244, 3121, 2982, 1727, 1700, 1649, 1492, 1333, 1286, 1089; ^{1}H NMR (400 MHz, DMSO-d_6): δ = 1.09 (t, 3H, J= 7.0, CH_3CH_2O), 2.26 (s, 3H, CH_3), 3.99 (q, 2H, J=6.9 , CH_3CH_2O), 5.16 (d, 1H, J=3.2, CH), 7.78-7.25 (m, 4H, Ar-H), 7.78 (d, 1H, J=2.6, NH), 9.26 (d, 1H, J=0.9, NH) ppm; ^{13}CNMR (100 MHz, DMSO-d_6): 14.02, 17.77, 53.41, 59.22, 98.83, 128.34, 131.77, 143.75, 148.67, 151.95, 157.68, 165.17 ppm. Calcd for $C_{14}H_{15}ClN_2O_3$: C, 57.05; H, 5.13; N, 9.50; O, 16.29 %. Found: C, 57.00; H, 5.06; N, 10.18 %.

Ethyl-4-(2-hydroxy-3-methoxyphenyl)-6-methyl-2-oxo-1,2,3,4-tetrahydropyrimidine-5-carboxylate (Table 2, 4c): White powder, FT-IR (KBr,

cm^{-1}): 3241, 3103, 2940, 1742, 1697, 1652, 1489, 1269, 1080. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 1.23 (t, J=7.0, 3H), 2.27 (s, 3H), 3.78 (s, 3H), 3.93 (q, J =3.2 Hz, 2H), 5.43 (s, 1H), 5.52 (d, J =3.0 Hz, 1H), 6.83-6.88 (m, 3H), 7.59 (s, 1H, NH), 9.10 (d, 1H, NH) ppm; ^{13}CNMR (100 MHz, DMSO-d_6): δ = 14.01, 17.66, 47.65, 55.82, 60.47, 98.02, 110.75, 111.54, 118.52, 118.95, 120.30, 143.45, 148.42, 152.18, 168.40 ppm. Calcd for $C_{15}H_{18}N_2O_5$: C, 58.82; H, 5.92; N, 9.15; O, 26.12 %. Found: C, 57.19; H, 5.76; N, 9.12 %.

Ethyl-4-(4-methoxyphenyl)-6-methyl-2-oxo-1, 2,3,4-tetrahydropyrimidine-5-carboxylate (Table 2, 4e): White powder, FT-IR (KBr, cm^{-1}): 3247, 3115, 2955, 2832, 1724, 1703, 1649, 1507, 1456, 1275, 1221, 1089; ^{1}H NMR (400 MHz, DMSO-d6): 1.16 (t, 3H, J= 7.0, CH_3CH_2O), 2.30 (s, 3H, CH_3), 3.78 (s, 3H, OCH_3), 4.04 (q, 2H, J=7.0 , CH_3CH_2O), 5.15 (d, 1H, J=3.1, CH), 6.93 (d, 2H, J=8.6, Ar-H), 7.21 (d, 2H, J=8.6, Ar-H), 7.72 (s, 1H, NH), 9.20 (s, 1H, NH) ppm; ^{13}C NMR (100 MHz, DMSO-d6): 14.07, 17.71, 53.30, 55.02, 59.10, 99.54, 113.67, 127.35, 137.02, 147.96, 152.11, 158.41, 165.34 ppm. Calcd for $C_{15}H_{18}N_2O_4$: C, 62.06; H, 6.25; N, 9.65; O, 22.04 %. Found: C, 60.16; H, 5.61; N, 10.35 %.

Ethyl-4-(4-Fluorophenyl)-6-methyl-2-oxo-1, 2, 3, 4-tetrahydropyrimidine- 5-carboxylate (Table 2, 4g): White powder, FT-IR (KBr, cm^{-1}): 3250, 3124, 2982, 1730, 1715, 1700, 1655, 1510, 1459, 1218, 1098; ^{1}H NMR (400 MHz, DMSO-d_6): δ =1.14 (t, 3H, J= 7.0, CH_3CH_2O), 2.30 (s, 3H, CH_3), 4.03 (q, 2H, J=7.0 , CH_3CH_2O), 5.20 (d, 1H, J=3.08, CH), 7.31-7.20 (m, 4H, Ar-H), 7.79 (s, 1H, NH), 9.27 (s, 1H, NH) ppm; ^{13}CNMR (100 MHz, DMSO-d_6): 14.03, 17.74, 53.30, 59.17, 114.97, 115.18, 128.16, 128.25, 148.48, 160.07, 162.48, 165.21 ppm. Calcd for $C_{14}H_{15}FN_2O_3$: C, 60.42; H, 5.43; N, 6.83; O, 10.07 %. Found: C, 60.11; H, 5.66; N, 7.57 %.

Ethyl-6-methyl-2-oxo-4-(1-phenylethyl)-1, 2, 3, 4-tetrahydropyrimidine-5-carboxylate (Table 2, 4h): White powder, FT-IR (KBr, cm^{-1}): 3379, 3244, 3096, 2979, 1715, 1664, 1522, 1456, 1257, 1170; ^{1}H NMR (400 MHz, DMSO-d_6): δ =1.06 (s, 3H, CH_3), 1.31 (d, 3H, J=7.2, CH_3), 2.09 (t, 3H, J= 7.0, CH_3CH_2O), 3.01-2.95 (m, 1H, CH), 3.90 (q, 2H, J=7.0, CH_3CH_2O), 4.62 (d, 1H, J=1.28, CH), 7.35-7.29 (m, 5H, Ar-H), 7.76 (d, 1H, J=15, NH), 8.90 (s, 1H, NH) ppm; ^{13}CNMR (100 MHz, DMSO-d_6): 15.52, 19.38, 45.46, 52.90, 65.28, 100.21, 126.62, 127.90, 128.54, 140.21, 152.40, 152.53, 159.70, 165.1 ppm. Calcd for $C_{16}H_{20}N_2O_3$: C, 66.65; H, 6.99; N, 9.72; O, 16.65 %. Found: C, 64.98; H, 5.63; N, 10.50 %.

Ethyl-6-methyl-4-phenyl-2-thioxo-1,2,3,4-tetrahydro-5-pyrimidinecarboxylate (Table 2, 5a): White powder, FT-IR (KBr, cm^{-1}): 3331, 3172, 2925, 1670, 1575, 1465, 1284, 1122, ^{1}H NMR (400 MHz, DMSO-d_6): δ = 1.10 (t, J=7.1, 3H), 2.30 (s, 3H), 4.01 (q, J = 7.0 Hz, 2H), 5.18 (d, J=3.6, 1H), 7.29 (m, 5H), 9.65 (d, 1H, NH), 10.33 (s, 1H, NH), ppm; ^{13}CNMR (100 MHz, DMSO-d_6): δ = 13.97, 17.12, 54.01, 59.56, 100.68, 126.35, 127.64, 128.52, 143.46, 144.99, 165.09, 174.20 ppm. Calcd for $C_{14}H_{16}N_2O_2S$: C, 60.85; H, 5.84; N, 10.14; O, 11.58 %; S; 11.60%. Found: C, 59.98; H, 5.21; N, 12.23 %.

Methyl 6-methyl-2-oxo-4-phenyl-1,2,3,4-tetrahydropyrimidine-5-carboxylate (Table 2, 4j): White powder, FT-IR (KBr, cm^{-1}): 3244, 3121, 2982, 1724, 1703, 1649, 1456, 1221, 1092; ^{1}H NMR (400 MHz, DMSO-d_6): δ =1.09 (t, 3H, J= 7.0, CH_3CH_2O), 2.26 (s, 3H, CH_3), 3.99 (q, 2H, J=7.0 , CH_3CH_2O), 5.15 (d, 1H, J=3.2, CH), 7.32-7.24 (m, 5H, Ar-H), 7.73 (s, 1H, NH), 9.19 (s, 1H, NH) ppm; ^{13}CNMR (100 MHz, DMSO-d_6): 14.04, 17.74, 53.94, 59.14, 99.24, 126.21, 127.22, 128.34, 144.83, 148.31, 152.11, 165.30 ppm. Calcd for $C_{13}H_{14}N_2O_3$: C, 63.40; H, 5.73; N, 11.38; O, 19.49 %. Found: C, 60.45; H, 5.83; N, 11.79 %.

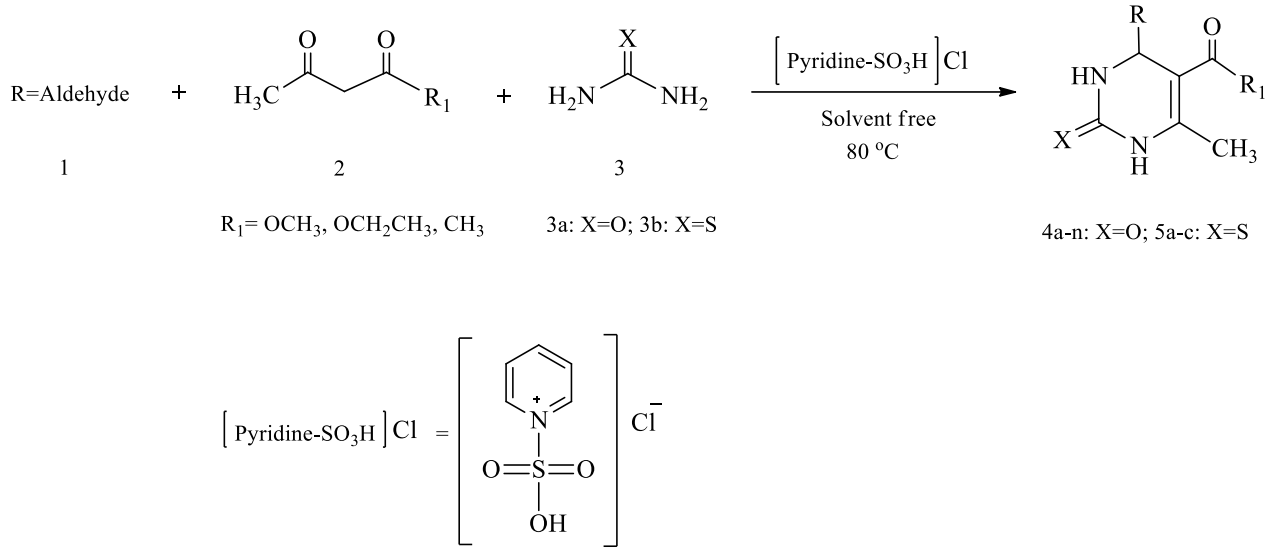
Methyl4-(4-methoxyphenyl)-6-methyl-2-oxo-1 ,2,3,4-tetrahydropyrimidine-5-carboxylate (Table 2, 4k): yellow powder, FT-IR (KBr, cm^{-1}): 3256, 3118, 2952, 1712, 1682, 1649, 1513, 1435, 1236. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 2.24 (s, 3H), 3.52 (s, 3H), 3.71 (s, 3H), 5.08 (d, J =3.2 Hz, 1H), 6.87 (d, J =8.8 Hz, 2H), 7.13 (d, J =8.8 Hz, 2H), 7.68 (s, 1H, NH), 9.18 (s, 1H, NH) ppm;

^{13}CNMR (100 MHz, DMSO-d_6): δ = 17.77, 50.72, 53.16, 55.00, 99.26, 113.72, 127.30, 136.81, 148.30, 152.17, 158.43, 165.83 ppm. Calcd for $C_{14}H_{16}N_2O_4$: C, 60.86; H, 5.84, N, 10.14; O, 23.16 %. Found: C, 60.15; H, 5.33; N, 10.86 %.

1-(6-Methyl-4-phenyl-2-thioxo-1,2,3,4-tetrahydro-5-pyrimidinyl) ethanone (Table 2, 5c): Orange powder, FT-IR (KBr, cm^{-1}): 3289, 3181, 2997, 1619, 1576, 1462, 1366, 1185. ^{1}H NMR (400 MHz, DMSO-d_6): δ = 2.24 (s, 3H), 2.41 (s, 3H), 5.37 (d, J = 3.7 Hz, 1H), 7.37 (m, 5H), 9.83 (d, 1H, NH), 10.36 (s, 1H, NH) ppm; ^{13}CNMR (100 MHz, DMSO-d_6): δ = 18.22, 30.40, 53.73, 110.43, 126.52, 127.67, 128.61, 142.88, 144.54, 174.04, 194.75 ppm. Calcd for $C_{13}H_{14}N_2OS$: C, 63.39; H, 5.73; N, 11.37; O, 6.50; S, 13.02 %. Found: C, 60.15; H, 5.23; N, 11.39 %.

RESULTS AND DISCUSSION

In our research, syntheses of dihydropyrimidinones were done in three different conditions, using ionic liquid as catalyst [19]. To outline the role The schematic representation is shown in (Scheme 1). The ionic liquid, [pyridine-SO_3H]Cl has been prepared according to the literature procedure [20]. After completion of the reaction, the catalyst was recovered from the reaction mixture by washing with warm ethyl acetate, dried under vacuum at 90 °C and reused for subsequent reactions [21-29]. The catalyst concentration was varied over a range of 0–15 mol% of catalyst on the basis of the total volume of the reaction mixture. To show the merit of the present work, we compared the yield and time of the reaction in the presence of different concentrations of the catalyst under solvent free conditions in (Table 1). The results showed that 10 mol % of catalyst were also efficient in this reaction at the expense of reaction time: 15 min, 94%.



Scheme 1. Synthesis of 3,4-dihydropyrimidin-2(1*H*)-ones and thiones.

Table 1. Investigation of catalyst effects in the synthesis of dihydropyrimidinones under solvent-free conditions

Mol % [pyridine-SO_3H]Cl	0	5	10	15
Time(min)	180	120	15	15
Yield(%)	10	45	94	94

To study the generality of this process, a variety of substituted aromatic aldehydes were examined. At these optimistic conditions (10 mol% of catalyst, solvent-free conditions, 80 °C), a series of 3,4-dihydropyrimidin-2(1*H*)-ones (4a–n) and thiones (5a–c) were obtained by varying the aromatic aldehyde and urea/thiourea with excellent yields in shorter reaction time. The results are listed in Table 2, which clearly indicate the generality of the reaction.

Table 2. Synthesis of different dihydropyrimidinones in presence of 10% mol of [pyridine-SO_3H]Cl as a acidic ionic liquid catalyst under solvent free conditions

	Aldehyde	R_1	X	[a]Product	Solvent free	M.p (°C)
					Time(min)/ Yield(%)	Found Reported[Ref]
1	Benzaldehy	OEt	O	4a	30/92	200-202 201-203[22]
2	4-Chloro benzaldehyde	OEt	O	4b	25/90	216-218 215-217[23]
3	2-Hydroxy-3-methoxy benzaldehyde	OEt	O	4c	25/90	211-215 new
4	Thiophene-2-carbaldehyde	OEt	O	4d	25/88	210-212 213-215[1]
5	4-Methoxy benzaldehyde	OEt	O	4e	15/96	201-203 207-208[5]
6	2-Chloro benzaldehyde	OEt	O	4f	25/90	216-218 216-218[1]
7	4-Fluoro benzaldehyde	OEt	O	4g	30/94	194-196 190-192[24]
8	2-Phenylpropanal	OEt	O	4h	30/88	210-212 new
9	3-Nitro benzaldehyde	OEt	O	4i	15/90	226-227 228-230[26]
10	benzaldehyde	OEt	S	5a	15/98	196-198 208-209[5]
11	benzaldehyde	OMe	O	4j	25/90	212-214 209-212[26]
12	4-Methoxy benzaldehyde	OMe	O	4k	30/90	187-189 192-194[12]
13	3-Nitro benzaldehyde	OMe	O	4l	15/92	280-281 284-286[26]
14	benzaldehyde	OMe	S	5b	15/92	224-226 228-229[26]
15	Benzaldehy	CH_3	O	4m	15/92	229-230 232-233[5]
16	4-Methoxy benzaldehyde	CH_3	O	4n	15/90	230-232 232-233[12]
17	Benzaldehyde	CH_3	S	5c	15/92	229-230 234-236[5]

[a]All reactions were carried out with a molar ratio of aldehyde, *β*-ketoester , urea/thiourea in the presence of [pyridine-SO_3H]Cl under solvent-free conditions. [b]Yields refer to isolated pure products.

The results showed that acidic ionic liquid catalyst such as [Bmim]$FeCl_4$, [Hmim]Tfa, [Cbmim]Cl, [Hmim]HSO4 and [pyridine-SO_3H]Cl exhibited a good catalytic activity for synthesis of

dihydropyrimidinones. As seen from Table 3, the ionic liquid [pyridine-SO_3H]Cl is an effective catalyst with high yield and shorter reaction time.

Table 3. Comparision of [pyridine-SO_3H]Cl with reported acidic ionic liquid catalysts for the synthesis of dihydropyrimidinones

Catalyst(mol%)	Temp(°C)	Time(min)	Yield(%)[Ref]
[Bmim]$FeCl_4$	90, s.f.[a]	120	90[27]
[Hmim]Tfa	50, s.f.[a]	45	90[28]
[Cbmim] Cl	80, s.f.[a]	30	90[5]
[Hmim]HSO4	80, s.f.[a]	90	96[29]
[pyridine-SO_3H]Cl	80, s.f.[a]	15	94

[a] s.f.: solvent free condition.

Table 4. Optimizing the reaction conditions

Entry	Mol % of [pyridine-SO_3H]Cl	Conditions Method/Solvent	Time (min)	Yield (%)[a]
1	-	r.t./Solvent-free	180	-
2	-	80 °C/Solvent-free	180	10
3	5	r.t./Solvent-free	120	-
4	5	80 °C/Solvent-free	120	45
5	5	Reflux/ethanol	120	30
6	10	r.t./Solvent-free	60	48
7	10	80 °C/Solvent-free	15	[b]94, 94, 92, 90
8	10	Reflux/water	30	76
9	10	Reflux/ethanol	30	59
10	10	Mv/Solvent free	6	68
11	10	Mv/Water	6	54
12	15	80 °C/Solvent-free	15	94
13	15	Reflux/water	30	78
14	15	Mv/Solvent free	6	68

[a]Isolated yield: [b]1-Sulfopyridinium chloride was run for four consecutive cycles

To evaluate the feasibility of 1-Sulfopyridinium chloride, a modal reaction involving benzaldehyde (1), *β*-ketoesters (2) and urea/thiourea(3) was carried out at different temperatures (r.t. and 80 °C) in the absence as well as in the presence of different amount of the catalyst (5– 15 mol%) under different conditions. At room temperature, without catalyst product formation was not observed. In the absence of the catalyst, as the temperature increases the yield of the product slightly increases. In the presence of the catalyst, maximum yield (94%) was observed at 80 °C with 10 mol% of the catalyst. The same reaction was also carried out with 10 mol% of catalyst under reflux(water and ethanol) and microwave conditions, but the yield of the obtained product was lower compared to the reaction under solver-free conditions (Table 4). In addition, the simple recovery for the synthesis of 3,4-dihydropyrimidin-2(1*H*)-ones, no significant loss was observed in the product yield when catalyst was used after four times recycling.

CONCLUSIONS

In conclusion, We have described a simple, one-pot, multi-component reaction between benzaldehyde, *β*-ketoesters and urea/thiourea in the presence of ionic liquid [pyridine-SO_3H]Cl under solvent-free conditions and all products of 3,4-dihydropyrimidin-2(1*H*)-ones gave good to

excellent yields of nitrogen heterocycles. The reactions carry the advantages that the starting materials are simply available and may be used without any purification or modification under neutral conditions.

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