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Acidic ionic liquid immobilized on cellulose: an efficient and recyclable heterogeneous catalyst for the solvent-free synthesis of hydroxylated trisubstituted pyridines†

Cite this: *RSC Advances*, 2013, **3**, 3184

Received 24th November 2012,
Accepted 8th January 2013

DOI: 10.1039/c3ra23052j

www.rsc.org/advances

Shailesh P. Satasia, Piyush N. Kalaria and Dipak K. Raval*

The synthesis of a novel cellulose supported acidic ionic liquid (Cell-IL) for the solvent-free synthesis of hydroxylated trisubstituted pyridines is reported. Cell-IL was prepared by immobilization of an acidic ionic liquid [1-butyl-3-(3-trimethoxypropyl)-1*H*-imidazol-3-ium hydrogen sulfate] on cellulose. The viability of this concept has been confirmed by FT-IR, TGA-DTG, ^{1}H NMR and elemental analysis. The Cell-IL showed good thermal stability and exhibited a high catalytic activity in the synthesis of a series of hydroxylated trisubstituted pyridines. It was recovered and reused for the model reaction three times without an appreciable change in its activity.

1. Introduction

In recent years, green chemistry has focused mainly on environmentally friendly reactions, sustainable resources and reusable catalysts. To design a catalyst with high selectivity and efficiency, which is kind to the environment and can be easily recovered, is an attractive and rapidly developing area in chemistry. Ionic liquids (ILs) have attracted extensive research interest as environmentally benign solvents due to their excellent properties such as negligible vapour pressure, solvating ability, non-flammability, high thermal stability and reusability.[1–3] Compared to pure acidic ILs, IL-based heterogeneous catalysts offer more advantages. They are required in lower amounts, are easy to separate and have a competent catalyst recovery.[4–6] The concept of ''immobilized'' or ''heterogenised'' liquids is well known for supported liquid phase (SLP) catalysts. The immobilization process aims to transfer the desired catalytic properties of the liquids to a solid catalyst.[7]

It is likely that renewable bio-polymeric supports will play a key role in the future for the development of clean processes. In this regard, biopolymers are attractive candidates to explore their application in supported catalysis.[8,9] Several interesting biopolymers such as alginate,[10] gelatin,[11,12] starch[13] and chitosan[14,15] derivatives have been utilized as the support for catalytic applications. Cellulose, one of the most abundant natural biopolymers, has unique properties such as being biodegradable, inexpensive, extremely inert, non-toxic and environmentally benign. These properties make it an attractive alternative to conventional organic or inorganic supports for catalytic applications,[16] offering a relatively low contamination risk to the environment.

Substituted pyridines are an important class of compounds that feature profoundly in a number of pharmaceutical targets and are a common motif in many natural products.[17,18] They have attracted continuous interest to explore newer methods for their construction.[19–23] These compounds have also been synthesized through the reaction of *N*-phenacylpyridinium salts with α,β-unsaturated ketones in the presence of ammonium acetate (Kröhnke's synthesis).[24,25] The pyridinium salts and the unsaturated ketones have to be synthesized first, which makes the method relatively expensive. Kröhnke pyridines bearing a hydroxyl group are usually prepared by SPOT-synthesis,[26] or multi-step protection/deprotection strategies.[27,28] Many improved methods for the preparation of 2,4,6-trisubstituted pyridines have also been reported.[29–34] Recently, Yin *et al.* reported the synthesis of hydroxylated trisubstituted pyridines under microwave irradiation and have investigated their photophysical properties.[35]

As part of a program directed toward the synthesis of various heterocyclic compounds,[36–41] we envisage the use of a catalytic system based on cellulose functionalized with an acidic IL for the synthesis of hydroxylated trisubstituted pyridines. The present protocol, operating without the need for protection of the hydroxyl group, is a novelty. An acidic ionic liquid [1-butyl-3-(3-trimethoxypropyl)-1*H*-imidazol-3-ium hydrogen sulfate] was immobilized on cellulose to prepare Cell-IL. This is the first ever report describing the synthesis of hydroxylated trisubstituted pyridines using Cell-IL as the catalyst under solvent free conditions.

2. Experimental section

2.1 Materials and instrumentation

All the reactions were performed with commercially available reagents. They were used without further purification. Organic

Department of Chemistry, Sardar Patel University, Vallabh Vidyanagar-388 120, Gujarat, India. E-mail: dipanalka@yahoo.com; Fax: +91-02692-236475; Tel: +91-02692-226856 – Ext. - 211

† Electronic supplementary information (ESI) available. See DOI: 10.1039/c3ra23052j

solvents were purified by standard methods.[42] Reactions were carried out on an *Equitron* round oil bath (model no. 8481ISDWO). All the reactions were monitored by thin-layer chromatography carried out on fluorescent coated plates (aluminium plates coated with silica gel 60 F254, 0.25 mm thickness, Merck) and detection of the components was made by exposure to UV light. Melting points were measured in a μThermoCal$_{10}$ (Analab Scientific pvt Ltd., India) and the reported values are uncorrected. The synthesized compounds were identified by ^{1}H and ^{13}C NMR spectra recorded in MeOD on a Bruker Avance 400 MHz spectrometer (Bruker Scientific Corporation Ltd., Switzerland) using the residual solvent signal as an internal standard at 400 MHz and 100 MHz respectively. IR spectra were recorded on a Bruker alpha E- FTIR spectrophotometer in the range 4000–400 cm^{-1} and frequencies of only characteristic peaks are expressed. Elemental analyses were performed on a PerkinElmer 2400 series II elemental analyzer (PerkinElmer, USA) and all results are found within ±0.4% of the theoretical compositions. TGA data were obtained with a heating rate of 10 °C min^{-1} on a TGA-DTG (TA instruments model 5000/2960 thermo gravimetric analyzer, USA).

2.2 Synthesis of 1-butyl-3-(3-trimethoxysilylpropyl)-1*H*-imidazol-3-ium hydrogen sulfate (IL-HSO$_4$)

IL1, 1-butyl-3-(3-trimethoxysilylpropyl)-1*H*-imidazol-3-ium chloride was prepared according to the known process.[43] Then it was modified, concerning the anion of the IL. Conc. H_2SO_4 (1.35 mL, 25 mmol) was added drop wise into a solution of the above in ethanol (35 mL) over 30 min. The final mixture was stirred at 50 °C for another 8 h and evaporated under reduced pressure to give a viscous orange liquid in 96% yield (**IL2**).

^{1}H NMR (400 MHz, $CDCl_3$): δ = 0.942–0.980 (m, 2 H), 1.172 (t, J = 7.4 Hz, 3 H), 1.341 (m, 2 H), 1.907–1.938 (m, 4H), 3.501 (s, 9 H), 4.290 (m, 4 H), 7.469–0.477 (m, 2H), 9.139 (s, 1 H), 14.524 (s, 1 H), IR: ν = 3140, 2932, 2870, 1566 cm^{-1}; anal. calcd for $C_{13}H_{27}ClN_2O_3Si$: C 48.35, H 8.43, N 8.68; found: C 48.68, H 8.58, N 8.81.

2.3 Immobilization of IL-HSO$_4$ on cellulose

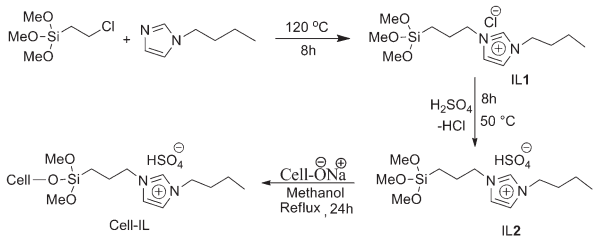
In the preparation of Cell-IL, cellulose (5.0 gm) was first activated by treatment with 25 mL 0.1 M aqueous sodium hydroxide solution. The mixture of IL2 (2.0 g) and the sodium salt of cellulose was stirred in 30 mL methanol at reflux for 24 h. The ionic liquid was immobilized on cellulose upon etherification (Scheme 1). The obtained Cell-IL was cooled to room temperature, filtered and washed thrice with water, dried under vacuum at (144 mm of Hg) 60 °C for 2–3 h to give the supported acidic ionic liquids in powder form.

IR: 3139, 2949, 2872, 1644, 1564, 1454 cm^{-1}

Elemental analysis found: C 32.56, H 4.89, N 4.26.

2.4 Synthesis of hydroxylated 2,4,6-trisubstituted pyridines

A mixture of 4-hydroxybenzaldehyde **1** (25 mmol), 4-hydroxyacetophenone **2a** (50 mmol) and NH_4OAc (150 mmol) was heated at 110 °C for 30–35 min in the presence of Cell-IL (60 mg) under solvent-free conditions. The progress of the reaction was monitored by TLC. After completion of the reaction, the solid product gradually formed upon cooling was poured into cold



Scheme 1 Synthesis of the cellulose supported IL catalyst.

water. The solid was separated by filtration through a sintered funnel under suction. The residue was treated with methanol to recover the insoluble catalyst from the product. The solvent was then evaporated under vacuum and the desired product was obtained as a yellow solid. The structures of the products were confirmed by ^{1}H NMR, ^{13}C NMR, IR spectroscopy and also supported by elemental analysis. The filtered catalyst was dried at 60 °C under vacuum (144 mm Hg) for 4 h and was recycled thrice for the model reaction to check its catalytic efficiency.

2.5 Spectral data of synthesized compounds

4,4′,4′′-(Pyridine-2,4,6-triyl)triphenol (3a). Yield: 88%, mp: 281–282 °C, ^{1}H NMR (400 MHz, MeOD): δ (ppm) 8.032 (d, J = 8.8, 4H), 7.797 (s, 2H, Py–H), 7.727 (d, J = 8.8, 2H), 6.973–6.921 (m, 6H); ^{13}C NMR (100 MHz, MeOD): δ (ppm) 158.5, 158.11, 156.6, 150.09, 131.10, 129.78, 128.26, 128.12, 115.55, 115.06, 114.49. IR (cm^{-1}): 3312, 1713, 1598, 1521, 1389, 1242, 1186, 826. Anal. calcd $C_{23}H_{17}N$ (355.12): C, 77.73; H, 4.82; N, 3.94. Found: C, 77.56; H, 4.96; N, 3.76.

3-(2,6-Bis(4-methoxyphenyl)pyridin-4-yl)phenol (3i). Yield: 81%, mp: 138–139 °C, ^{1}H NMR (400 MHz, MeOD): δ (ppm) 8.133 (d, J = 8.8 Hz, 4H), 7.824 (s, 2H, Py–H), 7.224–7.383 (m, 3H), 7.069 (d, J = 8.8 Hz, 4H), 6.904–6.933 (m, 1H), 3.882 (s, 6H, OCH_3); ^{13}C NMR (100 MHz, MeOD): δ (ppm) 160.83, 157.87, 157.14, 150.44, 140.29, 132.06, 129.85, 128.24, 128.03, 118.03, 115.44, 115.23, 113.77, 113.64, 54.47, 54.25. IR (cm^{-1}): 3412, 1614, 1504, 1409, 1248, 1163, 1026, 879, 761. Anal. calcd $C_{25}H_{21}NO_3$ (383.44): C, 78.31; H, 5.52; N, 3.65. Found: C, 78.55; H, 5.74; N, 3.42.

3-(2,6-Di([1,1′-biphenyl]-4-yl)pyridin-4-yl)phenol (3l). Yield: 84%, mp: 233–234 °C, ^{1}H NMR (400 MHz, MeOD): δ (ppm) 8.305 (s, 2H, Py–H), 8.071 (d, J = 8.4 Hz, 4H), 7.928 (d, J = 8.4 Hz, 4H), 7.711 (d, J = 7.2 Hz, 4H), 7.096–7.516 (m, 10H) : ^{13}C NMR (100 MHz, MeOD): δ (ppm) 159.04, 158.64, 158.37, 153.84, 145.09, 139.16, 136.19, 130.73, 129.11, 128.85, 127.79, 126.96, 121.37, 118.97, 114.44. IR (cm^{-1}): 3421, 1607, 1523, 1401, 1239, 1180, 1123, 842, 754. Anal. calcd $C_{35}H_{25}NO$ (475.19): C, 88.39; H, 5.30; N, 2.95. Found: C, 88.63; H, 5.14; N, 3.02.

3. Results and discussion

3.1 Characterization of catalysts

The resulting biopolymer supported acidic IL catalyst was characterized by FT-IR spectroscopy and TGA analysis. Fig. 1 shows the FT-IR spectra of Cell-IL and Cell-ONa catalysts. The FT-IR spectrum of Cell-IL (curve 1) exhibited some characteristic stretching vibration bands due to the imidazole ring at 1644 and

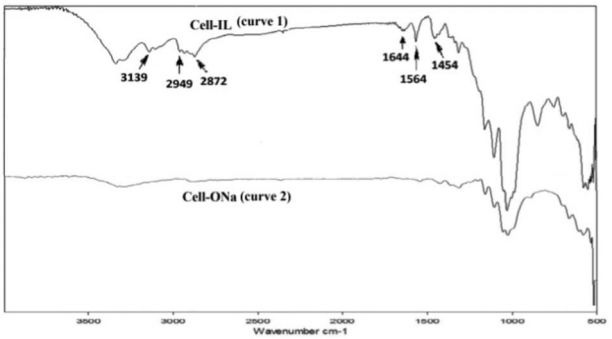


Fig. 1 FT-IR spectra comparison of Cell-ONa and immobilized IL.

1564 cm^{-1}, which may be attributed to the C=N and C=C vibration peaks.[44] Additional bands at 3139, 2949 and 1454 cm^{-1} are due to C–H stretching and deformation vibrations of the imidazole moiety and alkyl chain. These are the additional characteristic peaks of the functionalized IL which are absent in the IR spectrum of Cell-ONa (curve 2).

The thermal stability of the prepared catalyst was checked by TGA. TG and DTG curves are shown in Fig. 2. The TG curve indicates an initial weight loss of 1.9% up to 100 °C due to the adsorbed water in Cell-IL. A major weight loss in the IL supported onto the cellulose was recorded in the temperature range 220 °C to 400 °C. The weight loss was about 66.25%. The DTG curve shows that decomposition of the organic structure mainly occurred in one step from 220 °C to 360 °C, which is related to the main weight loss of 62.05%.

3.2 Applications of Cell-IL as the catalyst for synthesis of hydroxylated trisubstituted pyridines derivatives

Initial screening for the catalytic efficiency of Cell-IL in the model three-component condensation reaction was carried out in the presence of 40 mg immobilized IL (Cell-IL) under solvent free conditions at 100 °C. The efficiency of the reaction was affected by the amount of the catalyst. From the performed sets of reactions, it was found that 60 mg of Cell-IL was the optimum amount of catalyst required to carry out the reactions with maximum yield (Table 1, entry 3).

The model reaction was also optimized for the reaction temperature. At room temperature, the reaction did not proceed. The reaction progressed with 88% yield within 36 min at 110 °C

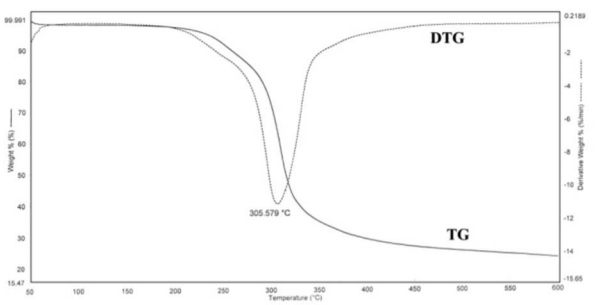


Fig. 2 TG-DTG analyses for the Cell-IL catalyst.

Table 1 Optimization of amounts of Cell-IL in the formation of **3a**[a]

Entry	Immobilized IL (mg)	Time (min)[b]	Yield[c] (%)
1	40	80	66
2	50	60	77
3	60	52	81
4	70	56	80
5	80	54	80

[a] 4-Hydroxy benzaldehyde (25 mmol), 4-hydroxy acetophenone (50 mmol), ammonium acetate (150 mmol). [b] All reactions were run until completion as indicated by TLC. [c] Isolated yield.

Table 2 Effect of temperature on the synthesis of **3a**[a]

Entry	Temperature (°C)	Time (min)[b]	Yield[c] (%)
1	30	180	—
2	50	120	23
3	80	95	51
4	100	52	81
5	110	36	88
6	120	50	86

[a] 4-Hydroxy benzaldehyde (25 mmol), 4-hydroxy acetophenone (50 mmol), ammonium acetate (150 mmol), Cell-IL (60 mg). [b] All reactions were run until completion as indicated by TLC. [c] Isolated yield.

(Table 2, entry 5). This temperature was chosen as the optimum reaction temperature for a series of compounds (**3b–3p**) synthesized to study the effect of various substituted building blocks on the reaction.

A number of various catalysts were tried for the synthesis of **3a**. The model reaction was attempted without any catalyst (Table 3, entry 1), which afforded compound **3a** in poor yield. Acetic acid, *p*-TSA, Amberlyst-15, montmorillonite K-10 and Dowex50WX4 also promoted the reaction with low yields (Table 3, entries 2–6). The results indicated that Cell-IL was found to be the most effective catalyst (Table 3, entry 7), leading to 88% yield of **3a** without any added solvent.

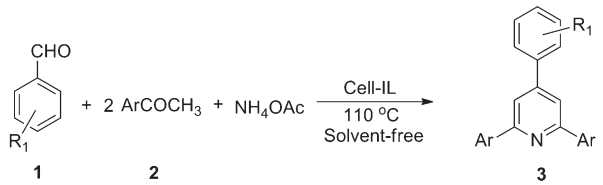
With these optimized conditions in hand, different aromatic aldehydes and ketones were attempted to prepare a series of trisubstituted hydroxylated pyridines to explore the scope and generality of the reaction (Table 4). In **3a–3l**, the corresponding

Table 3 Influence of different catalysts for the synthesis of **3a**[a]

Entry	Catalyst (amount)[b]	Time[c]	Yield[d] (%)
1	Nil	120	39
2	Acetic acid	52	51
3	*p*-TSA	68	53
4	Amberlyst-15	84	42
5	Montmorolonite K-10	43	62
6	Dowex50WX4	73	53
7	**Cell-IL**	**36**	**88**

[a] 4-Hydroxy benzaldehyde (25 mmol), 4-hydroxy acetophenone (50 mmol), ammonium acetate (150 mmol). [b] Amount of catalyst was 60 mg. [c] All reactions were run until completion as indicated by TLC. [d] Isolated yield.

Table 4 Cell-IL catalyzed synthesis of **3**[a] under solvent free conditions



1 2 3

Entry	Compounds	Aldehydes (R_1)	Ketones (Ar)	Time (min)[b]	Yield[c] (%)	m.p. (°C) Found	m.p. (°C) Reported[31]
1	**3a**	4-OH	4-OH-C_6H_5	36	88	281–282	281–282
2	**3b**	4-OH	-C_6H_5	35	86	179–180	209–210
3	**3c**	4-OH	4-Me-C_6H_5	32	89	275–276	274–275
4	**3d**	4-OH	4-OMe-C_6H_5	30	88	243–244	243–244
5	**3e**	4-OH	4-F-C_6H_5	33	81	275–276	—
6	**3f**	4-OH	2-furyl	26	91	262–263	263–264
7	**3g**	4-OH	2-thienyl	30	88	138–139	136–137
8	**3h**	4-OH	4-Ph-C_6H_5	32	85	224–225	223–225
9	**3i**	3-OH	4-OMe-C_6H_5	40	81	138–139	—
10	**3j**	3-OH	2-furyl	33	82	162–164	163–164
11	**3k**	3-OH	2-thienyl	38	80	124–125	124–125
12	**3l**	3-OH	4-Ph-C_6H_5	31	84	233–234	—
13	**3m**	2-OH	4-Ph-C_6H_5	43	67	224–225	226–227
14	**3n**	4-N$(CH_3)_2$	4-OH-C_6H_5	34	82	244–246	246–247
15	**3o**	2-thienyl	4-OH-C_6H_5	39	78	232–233	231–232
16	**3p**	H	4-OH-C_6H_5	34	88	197–198	229–230

[a] Aldehydes (25 mmol), acetophenones (50 mmol), ammonium acetate (150 mmol), Cell-IL (60 mg). [b] All reactions were run until completion as indicated by TLC. [c] Isolated yield.

trisubstituted hydroxylated pyridines were obtained in good to excellent yield. However, the *ortho*-substituted aromatic aldehydes under the above optimized conditions gave lower yields (Table 4, entry 13).

The reaction was feasible without protecting the hydroxyl group present in the aromatic aldehydes and ketones. All the prepared compounds were obtained in pure form after recrystallization from alcohol. The present method is better in terms of yield as well as mildness of the reaction conditions compared to the methods reported earlier.[30,33,45]

3.3 Recyclability of the catalyst

To investigate the catalytic efficiency of the recycled catalyst, three successive cycles of the model reaction were run under the optimal reaction conditions using recycled Cell-IL from the previous run (Table 5). The activity of the catalyst did not show any significant decrease in the yields after three successive runs for the model reaction. It was revealed that the catalyst displayed very good reusability.

Table 5 Reusability study[a]

No. of runs	Fresh	1	2	3
% Yield (**3a**)[a]	88	85	84	82

[a] Loss of catalyst (<5%) during handling.

4. Conclusions

A simple and efficient supported ionic liquid catalyst for the solvent-free synthesis of hydroxylated trisubstituted pyridines is developed. The work provides an example for the applications of cellulose supported ionic liquid catalysts as environmentally benign efficient alternatives in organic synthesis. The attractive features of this approach are a simple reaction procedure, short reaction time, easy work up and reusability of the Cell-IL. Cell-IL has proven to be a highly efficient heterogeneous catalyst exhibiting a high catalytic activity for a series of targeted pyridine derivatives with easy recovery and reusability for three successive turns.

Acknowledgements

SPS and PNK acknowledge UGC, New Delhi for providing research fellowships. The authors also thank the Head, Department of Chemistry, Sardar Patel University for providing research facilities and SICART, Vallabh Vidyanagar for elemental analysis and TGA at concessional rates.

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