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Short Communication

Basic-functionalized recyclable ionic liquid catalyst: A solvent-free approach for Michael addition of 1,3-dicarbonyl compounds to nitroalkenes under ultrasound irradiation

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ABSTRACT

A task-specific ionic liquid (TSIL) has been introduced as a recyclable catalyst in Michael addition. A series of nitroalkenes and various C-based nucleophiles were reacted in the presence of 30 mol% of recyclable basic-functionalized ionic liquid. Good to excellent yields were obtained in 30 min under ultrasound irradiation.

1. Introduction

Ionic liquids (IL's) are emerging solvents of interest as greener alternatives to conventional organic solvents aimed at facilitating sustainable chemistry. As a consequence of their unusual physical properties, reusability, and eco-friendly nature, ionic liquids have attracted the attention of chemists [1].

It is becoming evident from the increasing number of reports on the use of ionic liquids as solvents, catalysts, and reagents in organic synthesis that they are not totally inert under many reaction conditions [2]. While in some cases, their unexpected reactivity has proven fortuitous and in others, it is imperative that when selecting an ionic liquid for a particular synthetic application, attention must be paid to its compatibility with the reaction conditions.

The Michael addition is a powerful reaction for the formation of carbon–carbon bonds [3]. Moreover, the addition products are important synthetic intermediate, which can be further manipulated into a range of different classes of biologically active compounds [4]. This type of reaction is traditionally promoted by quantitative amount of strong bases that often lead to undesirable side reactions [5]. On the other hand, a range of Lewis acids are found to catalyze this reaction, and these procedures are also not free from disadvantages [6]. Thus, a number of milder reagents such as Al_2O_3, K_2CO_3, rhodium and ruthenium complex, clay-supported nickel bromide, quaternary ammonium salt, and *N*-phenyl-tris(dimethylamino)imino-phosphorane immobilized on polystyrene resin have been developed over the past few years [7]. Moreover, room temperature ionic liquids, particularly BMIM-BF_4, have been used as alternative green solvents to carry out the Michael addition using Ni(acac)$_2$ and Cu(II) triflate as catalysts [8].

Recent development of IL's turned on designing suitable ionic liquids with specific application repetition of ILs many times that can be used both as catalysts/promoters and solvents [9]. Several innovative synthetic procedures on this task lead to successful results and emerge as a new field in task specific ionic liquids (TSIL's). In this way, TSIL's were utilized in many chemical transformations and especially the Michael addition reaction have much paid attention due to the formation of C–C bond [10]. For example, Ranu et al. explored the influence of a new tailor-made, task-specific ionic liquid BMIM-OH on Michael addition reactions. The BMIM-OH was used in quantitative amount which would function as solvent and catalyst [11].

A pressing challenge for organic chemists is to develop new catalytic processes that are not only efficient, byproduct free, and high yielding but also eco-compatible [12]. On account of these factors, here in we report the solvent free Michael addition of 1,3-dicarbonyl compounds to nitroalkenes under ultrasound irradiation using catalytic amount of basic-functionalized ionic liquid as a recyclable catalyst (Fig. 1).

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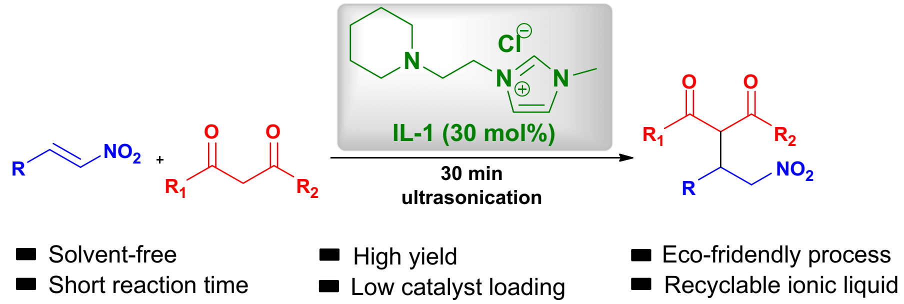


Fig. 1. Michael addition reactions of 1,3-dicarbonyl compounds to nitroalkenes.

2. Results and discussion

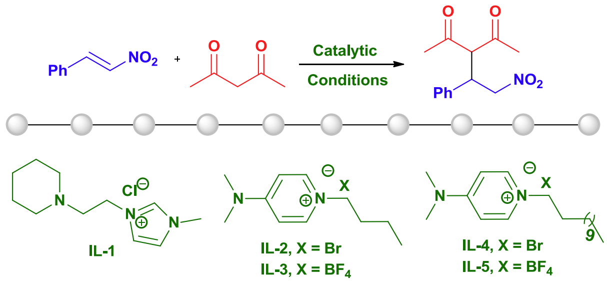
The efficacy of this protocol was initially evaluated by the reaction of 2,4-pentanedione (2 equivalents) to trans-β-nitrostyrene (1 equivalent) in the presence of 30 mol% of five different ionic liquids under sonication for 30 min. In the initial experiment, a range of ionic liquids were screened for the Michael addition (Table 1, entries 1–5). The result for IL-1 significantly better as compared with other ionic liquids (entry 1, Table 1) [13]. After selecting the appropriate ionic liquid, the amount of ionic liquid required to promote the completion of the reaction was also evaluated. Reactions with 20 and 10 mol% of IL-1 showed significant decrease in yields (Table 1, entries 1 *vs* 6 and 7). A decrease in yield was observed when the reaction time was reduced from 30 to 15 min (Table 1, entry 1 *vs* entry 8). In the absence of ultrasonication, the conjugate adduct was obtained in only 73% of yield in 120 min reaction time at room temperature under magnetic stirring (entry 9). No product was formed in the absence of IL-1 (entry 10).

Therefore, an optimum combination for the conjugate addition of nitroalkenes with 1,3-dicarbonyl compounds is using 30 mol% of IL-1 catalyst under 30 min ultrasonication under solvent free conditions.

The results of the optimization process to decode the scope and applicability of reactions are summarized in Tables 2 and 3. The Michael addition of 2,4-pentanedione to a variety of nitroalkenes was examined considering the usefulness and versatility of adducts in organic synthesis [14]. It is apparent from results that all reactions of nitroalkenes proceeded smoothly affording the desired products with good to excellent yields. Electronic effects had small influence on the reaction course, with electron-withdrawing groups attached to the aromatic ring of β-nitrostyrenes afforded the products with good to excellent yield (Table 2, entries 2–5 and 8–9). By using 30 mol% IL-1, the reactions of nitrostyrenes, bearing *ortho* or *para* electron donating substituent, with 2,4-pentanedione proceeded smoothly, and afforded the corresponding Michael adducts in 99% yield (Table 2, entries 6 and 7). We also employed other nitrostyrene in this reaction, e.g. heterocyclic. For instance, 2-(2-nitrovinyl) thiophene reacted with 2,4-pentanedione, allowing the conjugate adduct in 76% yield (Table 2, entry 10).

Next, the scope of the reaction was investigated with different nucleophilic species. As shown in Table 3, various ketoesters effectively reacted with trans-β-nitrostyrene in the presence of 30 mol% IL-1 (Table 3, entries 1–3). An important feature of our methodology is the use of different nucleophiles, such as cyclic-β-ketoester, giving the corresponding products in good yields (Table 3, entries 4–6). This result demonstrates the potential wide ranging utility of this methodology by the preparation of various conjugate adducts.

Table 1
Screening of reaction: ionic liquid, catalyst loading, and reaction time.



Entry	TSIL's	IL (% mol)	Time (min)	Yield (%)[a,b]
1	IL-1	30	30	98
2	IL-2	30	30	42
3	IL-3	30	30	10
4	IL-4	30	30	23
5	IL-5	30	30	16
6	IL-1	20	30	81
7	IL-1	10	30	66
8	IL-1	30	15	88
9[c]	IL-1	30	120	73
10[d]	–	–	30	–

[a] Unless otherwise specified, the reactions were performed using trans-β-nitrostyrene (0.25 mmol), 2,4-pentanedione (0.5 mmol), and ionic liquid (30 mol%) under ultrasonication for 30 min.

[b] Isolated yield.

[c] Reaction was performed without ultrasonication under room temperature for 2 h.

[d] Reaction was performed without ionic liquid.

Although steps toward sustainability can be made by reusing solvents, recycling is rarely accomplished with complete efficiency. Hence, we attempted to reuse the ionic liquid/catalyst, which was one of the prime objectives in our quest. In this regard, we performed a set of experiments to explore whether the ionic liquid can be reused for further reactions (Fig. 2). After completion of the reaction, task-specific ionic liquid catalyst was recovered and subjected to another run, affording the product in 98% yield. This process was repeated four more times, affording the desired product in excellent yields. The simple experimental and product isolation procedures combined with the ease of recovery and reuse of the reaction medium is expected to contribute to the development of a green strategy for the Michael addition reactions.

3. Experimental section

For visualization, TLC plates were either placed under ultraviolet light, or stained with iodine vapor, or acidic vanillin. All solvents were used as purchased unless otherwise noted. Purification of products was carried out by flash chromatography on silica gel.

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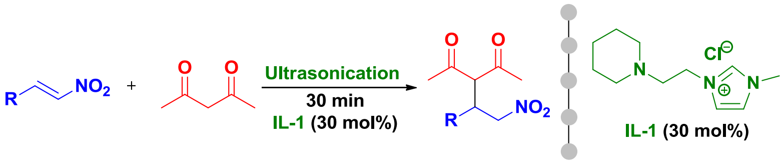
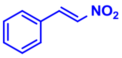
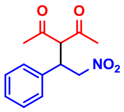
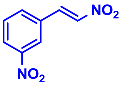
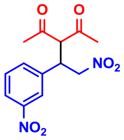
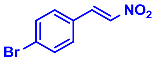
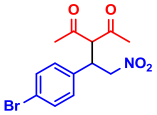
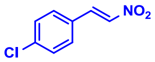
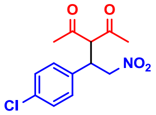
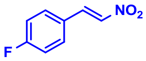
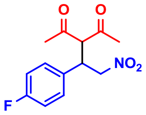
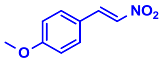
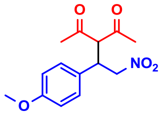
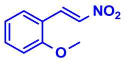
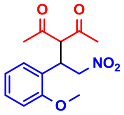
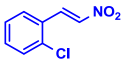
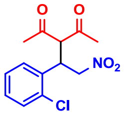
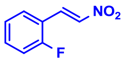
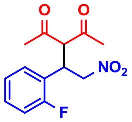
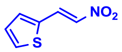
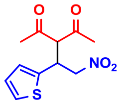
Table 2
Michael addition reactions of 2,4-pentanedione to nitroalkenes.



Entry	Substrate	Product	Yield [a,b]
1			98
2			52
3			71
4			74
5			99
6			99
7			99
8			>99
9			81
10			61(76)[c]

[a] Unless otherwise specified, the reactions were performed using nitrostyrene (0.25 mmol), 2,4-pentanedione (0.5 mmol) and ionic liquid (30 mol%) under ultrasonication for 30 min at room temperature.

[b] Isolated yield.

[c] Yield in parenthesis refer to 1 h reaction time under ultrasonication.

Chemical yields refer to pure isolated substances. ^1H and ^{13}C NMR spectra were obtained using deuterated solvents ($CDCl_3$ and DMSO-d_6) in a Bruker Avance III spectrometer. Chemical shifts are reported in ppm from tetramethylsilane with the solvent as the internal standard. All ionic liquids were prepared according literature procedure [13]. ^1H and ^{13}C NMR spectral data of the compounds are identical to those reported.

3.1. 1General synthetic procedure of ionic liquid

3.1.1. Synthesis of-1-methyl-3-(2-(piperidin-1-yl)ethyl)-1H-imidazol-3-ium-chloride (IL-1)

In a two neck 100 mL round-bottomed flask equipped with reflux condenser and magnetic stirrer, N-methyl imidazole (1.025 g, 12.5 mmol), 1-(2-chloroethyl)piperidine hydrochloride (1.84 g,

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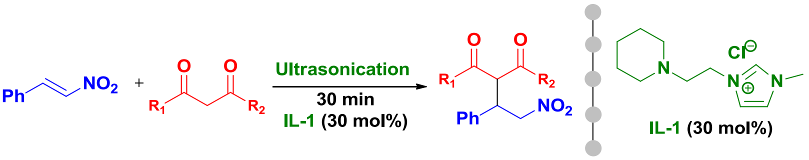
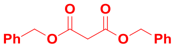
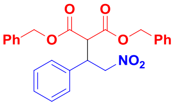
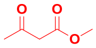
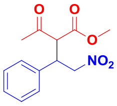
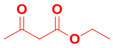
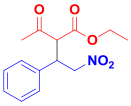
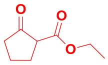
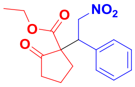
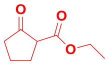
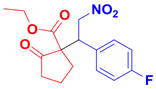
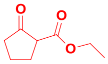
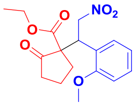
Table 3
Variation of nucleophiles in reaction with trans-β-nitrostyrene catalyzed byIL-1.



Entry[a]	Nucleophile	Product	Yield[b,c]
1			92
2			83(1:1)
3			80(1:1)
4			62(8:2)
5			>99(7:3)
6			89(7:3)

[a] Unless otherwise specified, the reactions were performed using trans-β-nitrostyrene (0.25 mmol), nucleophile (0.5 mmol), and ionic liquid (30 mol%) under ultrasonication.

[b] Isolated yield.

[c] d.r. (in parenthesis) was determined by NMR analysis.

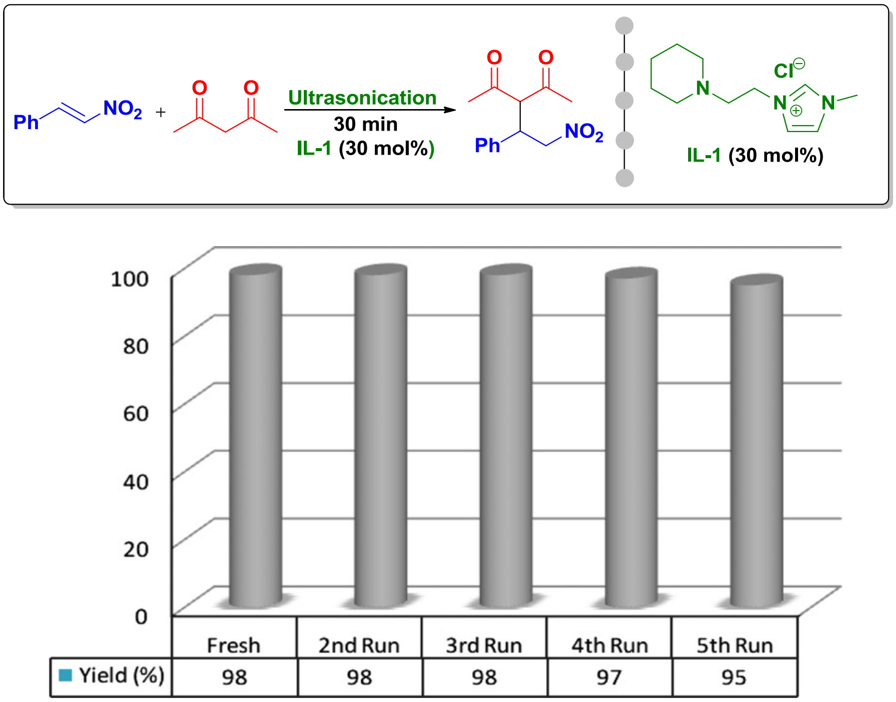


Fig. 2. Recyclability of ionic liquid for the conjugate addition.

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10 mmol) and absolute ethanol (10 mL) were added. The mixture was refluxed for 24 h. After the reaction, the solvent was removed under vacuum, the residue was washed with dichloromethane and dried at 70 °C under vacuum. The white solid was dissolved in the mixture of ethanol (5 mL) and water (5 mL), and neutralized by NaOH (0.4 g, 10 mmol). After removal of solvents, the product was extracted with dichloromethane, dried at 70 °C under vacuum for 10 h. Pale yellow oily liquid was obtained in 90% yield. ^{1}H NMR (400 MHz, DMSO-d_6) δ: 1.52–1.37 (m, 6 H), 2.53–2.59 (m, 4 H), 2.90–2.94 (m, 2 H), 3.86 (s, 3 H), 4.42–4.47 (m, 2 H), 7.76 (s, 1 H), 7.86 (s, 1H), 9.41 (s, 1H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6) δ: 23.1, 24.4, 35.7, 45.0, 53.2, 56.5, 122.5, 123.2, 137.0 ppm.

3.1.2. Synthesis of 1-butyl-4-(dimethylamino)pyridinium bromide (IL-2)

A mixture of 4-dimethylaminopyridine (5 mmol) and butyl bromide (6 mmol) and MeCN (10 mL) was allowed to stir 24 h at 70 °C. The resulting mixture was then evaporated affording the yellow crystals. The resulting crystalline mass was washed twice with ether (20 mL) and, after vacuum drying, a pale yellow crystals was obtained in 95% yield. ^{1}H NMR (400 MHz, DMSO-d_6) δ: 0.88 (t, J = 7.3 Hz, 3 H), 1.27–1.16 (m, 2 H), 1.76–1.68 (m, 2 H), 3.17 (s, 6 H), 4.17 (t, J = 7.3 Hz, 2 H), 7.03 (d, J = 7.8 Hz, 2 H), 8.34 (d, J = 7.7 Hz, 2 H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6) δ: 13.3, 18.7, 32.3, 39.7, 56.3 (2C), 107.7, 142.0, 155.8 ppm.

3.1.3. Synthesis of 1-butyl-4-(dimethylamino)pyridinium tetrafluoroborate (IL-3)

A mixture of 1-butyl-4-(dimethylamino)pyridinium bromide (5 mmol), sodium tetrafluoroborate (6 mmol) and distilled water (1 mL) was vigorously stirred for 60 min. The lower aqueous phase was separated and discarded and, to the remaining liquid, dichloromethane (20 mL) was added. The organic phase was separated and solvent evaporation afforded the desired 1-butyl-3-methylimidazolium tetrafluoroborate in quantitative yield. ^{1}H NMR (400 MHz, DMSO-d_6) δ: 0.88 (t, J = 7.3 Hz, 3 H), 1.27–1.16 (m, 2 H), 1.76–1.68 (m, 2 H), 3.17 (s, 6 H), 4.17 (t, J = 7.3 Hz, 2 H), 7.03 (d, J = 7.8 Hz, 2 H), 8.34 (d, J = 7.7 Hz, 2 H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6) δ: 13.3, 18.7, 32.3, 39.7, 56.3 (2C), 107.7, 142.0, 155.8 ppm.

3.1.4. Synthesis of 4-(dimethylamino)-1-dodecylpyridinium bromide (IL-4)

A mixture of 4-dimethylaminopyridine (5 mmol) and dodecyl bromide (6 mmol) and MeCN (10 mL) was allowed to stir 24 h at 70 °C. The resulting mixture was then evaporated affording the yellow crystals. The resulting crystalline mass was washed twice with ether (20 mL) and, after vacuum drying, pale yellow crystals was obtained in 99% yield. ^{1}H NMR (400 MHz, DMSO-d_6) δ: 0.83 (t, J = 7.25 Hz, 3 H), 1.30–1.13 (m, 18 H), 1.78–1.69 (m, 2 H), 3.17 (s, 6 H), 4.16 (t, J = 7,2 Hz, 2H), 7.03 (d, J = 7,6 Hz, 2H), 8.33 (d, J = 7.8 Hz, 2H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6) δ: 13.9, 25.4, 28.4, 28.7, 28.8, 28.9, 29.0 (2C), 31.3, 39.7, 56.6, 107.6, 142.0, 155.8 ppm.

3.1.5. Synthesis of 4-(dimethylamino)-1-dodecylpyridinium tetrafluoroborate (IL-5)

A mixture of 4-(dimethylamino)-1-dodecylpyridinium bromide (5 mmol), sodium tetrafluoroborate (6 mmol) and distilled water (1 mL) was vigorously stirred for 60 min. The lower aqueous phase was separated and discarded and, to the remaining liquid, dichloromethane (20 mL) was added. The organic phase was separated and solvent evaporation afforded the desired 4-(dimethylamino)-1-dodecylpyridinium tetrafluoroborate in 100% yield. ^{1}H NMR (400 MHz, DMSO-d_6) δ: 0.83 (t, J = 7.25 Hz, 3 H), 1.30–1.13 (m, 18 H), 1.78–1.69 (m, 2 H), 3.17 (s, 6 H), 4.16 (t, J = 7,2 Hz, 2H), 7.03 (d, J = 7,6 Hz, 2H), 8.33 (d, J = 7.8 Hz, 2H) ppm. ^{13}C NMR (100 MHz, DMSO-d_6) δ: 13.9, 25.4, 28.4, 28.7, 28.8, 28.9, 29.0 (2C), 31.3, 39.7, 56.6, 107.6, 142.0, 155.8 ppm.

3.2. General procedure for the synthesis of Michael adduct

In a vial, ionic liquid (30 mol%), trans-β-nitrostyrene (0.25 mmol) and 1,3-dicarbonyl compound (0.5 mmol) was added and the reaction mixture was allowed under ultrasonication for 30 min. After completion of the reaction (monitored by TLC), the reaction mixture was washed with Et_2O (3 × 5 mL) and dried over Na_2SO_4. The crude was purified by silica column chromatography affording the corresponding pure Michael adducts. The NMR data's of products were in accordance with literature data [15].

3.3. General procedure for ionic liquid recycle experiments

Following extraction with diethyl ether, the ionic liquid solution was subjected to vacuum to remove traces of diethyl ether, flushed with inert gas and charged with further portions of *trans*-β-nitrostyrene (1 eq) and 2,4-pentanedione (2 eq) at room temperature.

4. Conclusions

In conclusion, we have developed an efficient solvent free conjugate Michael addition of 1,3-dicarbonyl compounds to various nitroalkenes in the presence of catalytic amount of base-behavior ionic liquid under ultrasonication providing the desired conjugate adducts in good to excellent yields. The versatility, economic and high yield of this method, in addition to the shorter reaction time and low loading of catalyst/ionic liquid, highlights the potential for the use of this developed method in large scale library synthesis involving carbon–carbon bond formation. Most importantly, the recovery and recycling of the ionic liquid in further reactions was successfully achieved and can be reused for at least four successive runs without observing significant decrease in yield.

We believe that the ionic liquids which enable the easy recycling in organic reactions will have great synthetic value and potentially find wide applications in organic synthesis. Further investigations to clarify the mechanism and explore applications in asymmetric transformations are currently underway in our laboratory.

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