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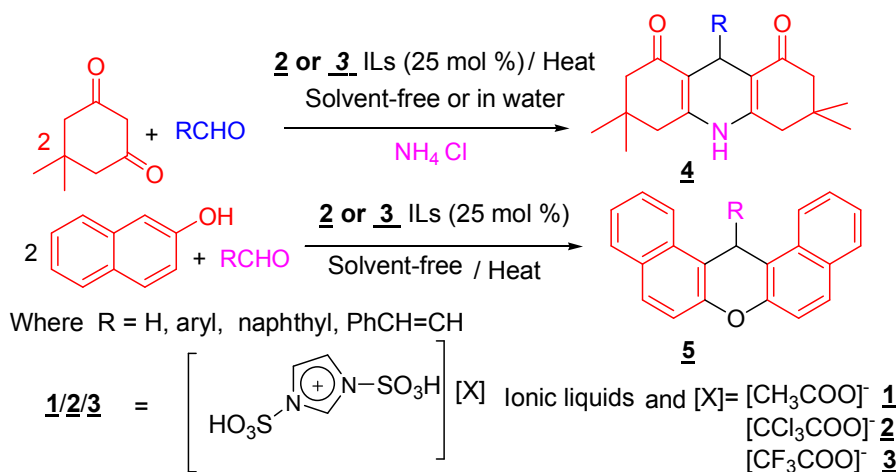
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Synthesis of Dibenzoxanthene and Acridine Derivatives Catalyzed by 1,3-Disulfonic Acid Imidazolium Carboxylate Ionic Liquids

Arup Kumar Dutta^a, Pinky Gogoi^a and Ruli Borah^a



1,3-Disulfonic acid imidazolium carboxylate [DSIM][X] (where X = [CH₃COO]⁻, [CCl₃COO]⁻, [CF₃COO]⁻) ILs were synthesized and examined their catalytic activity for the preparation of 14*H*-dibenzo[*a,j*]xanthene and 1,8-dioxo-decahydroacridine derivatives under solvent-free condition and in water at 80-100 °C.

ARTICLE

Synthesis of Dibenzoxanthene and Acridine Derivatives Catalyzed by 1, 3-Disulfonic Acid Imidazolium Carboxylate Ionic Liquids

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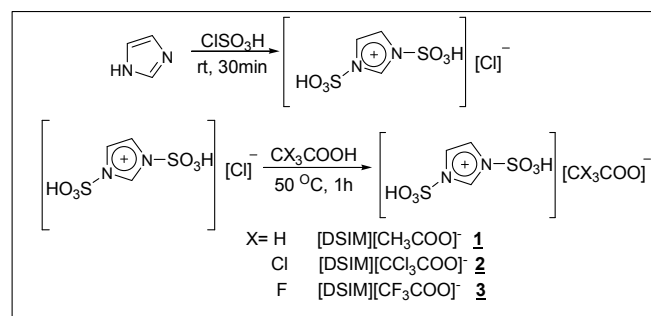
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New members of 1,3-disulfonic acid imidazolium carboxylate ionic liquids [DSIM][X] (where X = [CH₃COO]⁻, [CCl₃COO]⁻, [CF₃COO]⁻) were prepared and characterized by ¹H NMR, ¹³C NMR, FT-IR, TGA, UV-vis and elemental analysis. The more acidic ILs [DISM][CCl₃COO] and [DSIM][CF₃COO] were efficiently utilized as recyclable catalyst for the preparation of 14*H*-dibenzo[*a,j*]xanthene and 1,8-dioxo-decahydroacridine derivatives in short time under solvent-free condition at 80-100 °C with excellent yields. The above two ILs could be effectively utilized as catalysts for the synthesis of 1, 8-dioxo-decahydroacridine in water at the same temperature.

Introduction

The great capability of ionic liquids to replace volatile organic solvents, offer a new and environmentally benign approach toward modern chemical processes.¹ The implementation of task-specific ionic liquids with special functions further enhances the versatility of ionic liquids for those chemical reactions where both reagents and medium are coupled.² By virtue of the incorporated functional groups, these unique salts can act not only as solvents but also as catalysts in an array of synthetic, separations and electrochemical applications.³ Their solubility can also be tuned readily depending on the nature of cations and anions so that they can phase separate from organic as well as aqueous media.⁴ The preparation and application of such dual nature of acidic ionic liquids in organic reactions overcomes the common problems of traditional molecular solvents and acid catalysts. These ILs are non-flammable, thermally stable and exhibits negligible vapour pressure and recyclable. The higher potential of 1, 3-disulfonic acid imidazolium ILs in organic synthesis had encouraged us to develop new members of [DSIM][X] series where X = [CH₃COO]⁻, [CCl₃COO]⁻, [CF₃COO]⁻.⁵⁻⁶ In addition we also tried to observe the efficiency of [DSIM][X] ionic liquids for the one pot synthesis of 1,8-dioxo-decahydroacridine **4** and dibenzoxanthene **5** derivatives in environmentally benign condition. These heterocyclic compounds have wide applications in medicinal chemistry and material sciences.⁷⁻⁸ The traditional Brønsted or Lewis acid catalyzed synthesis of 1, 8-dioxo-decahydroacridine derivatives was performed in organic solvents such as HOAc^{9a}, CH₃CN^{9b} and DMF.^{9c} These conventional acids are often noxious, corrosive and non-recyclable, despite their higher catalytic activity.^{9d} The use of ionic liquids simplified the reaction conditions in terms of product selectivity, reaction time, recycling of catalyst and isolation of product.¹⁰ For the synthesis of dibenzoxanthene derivatives also few

reports utilized ionic liquids as catalyst or medium.¹¹ This work concerns on the synthesis, characterization and applications of 1,3-disulfonic acid imidazolium carboxylate ionic liquids and their applications in the synthesis of both classes of heterocyclic compounds.



Scheme 1. Synthesis of 1,3-disulfonic acid imidazolium carboxylate ILs

Results and Discussion

In continuation of our work¹² on the development of environmentally benign routes for organic synthesis, we synthesised three new members of 1,3-disulfonic acid imidazolium carboxylate ionic liquids ([DSIM][X] where X = [CH₃COO]⁻, [CCl₃COO]⁻, [CF₃COO]⁻) from the reactions of 1,3-disulfonic acid imidazolium chloride ([DSIM][Cl]) and three different carboxylic acids (AcOH, TCA and TFA) at room temperature stirring^{5a} (Scheme-1). The FT-IR spectra (Fig-1) of these ILs showed three strong absorption bands

at 1178-1192, 1048-1054 and 586-587 cm^{-1} corresponding to S-O asymmetric and symmetric stretching and bending vibration of $-\text{SO}_3\text{H}$ groups. The peaks around 875-878 cm^{-1} expressed the N-S

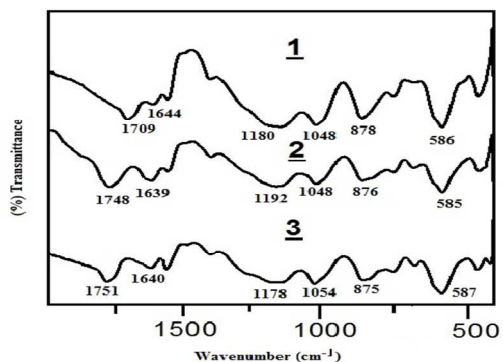


Figure 1. FT-IR spectra of **1**, **2** and **3** ionic liquids

stretching vibration. The carboxylate anions gave strong asymmetric bands around 1709-1751 cm^{-1} in the same region of imidazolium ions. The $-\text{OH}$ groups of the ionic liquids indicated broad and strong peaks at 3000-3660 cm^{-1} . In ^1H NMR spectra the characteristic acidic protons of two $-\text{SO}_3\text{H}$ groups of the ILs appeared in 14.2 and 11.3-13.4 ppm respectively. The presence of carbonyl signals at 158.6-172 ppm in ^{13}C NMR spectra confirmed the existence of carboxylate anions in these ionic liquids. The structures were further supported by the elemental analysis data.

The acidity of the ILs was determined on an UV-Visible spectrophotometer using 4-nitroaniline as basic indicator by following the Hammett equation that already reported in literature.¹³ The absorbance of the basic indicator [I] were determined in ionic liquid solutions which decreases with increasing the acidity of ionic liquids. The protonated form $[\text{IH}]^+$ of the indicator never appeared because of low molar absorptivity. The Hammett function H^0 for each ionic liquids was calculated using equation-1 by measuring the absorption differences $[\text{I}]/[\text{IH}]^+$.

$$H^0 = pK(\text{I})_{\text{aq}} + \log \frac{[\text{I}]}{[\text{IH}]^+} \quad (1)$$

Where $pK(\text{I})_{\text{aq}}$ is the pK_a value of the basic indicator in aqueous solution. The relative acidity of the ILs can be obtained by

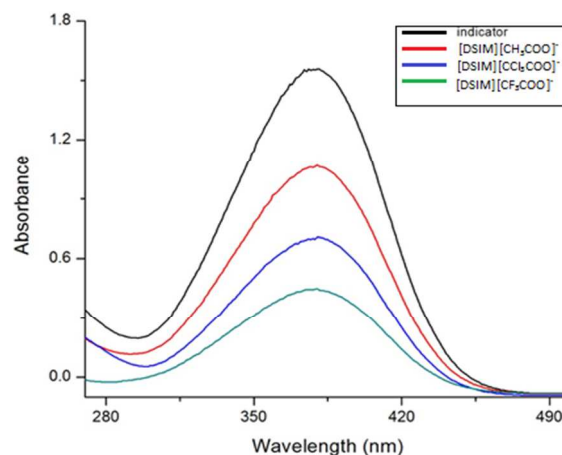


Figure 2. Hammett plot of ionic liquids using basic indicator 4-nitroaniline

determining the values of H^0 (table 1). The typical procedure involved the mixing of indicator 4-nitroaniline (5 mg/L, $pK_a=0.99$) and ionic liquid (5 mmol/L) in ethanol with equal concentration. The maximum absorbance observed at 382 nm in ethanol. The order of acidities of these ionic liquids were found from the Hammett plot (Fig-2) in the decreasing order as follows: $[\text{DSIM}][\text{CF}_3\text{COO}]^- > [\text{DSIM}][\text{CCl}_3\text{COO}]^- > [\text{DSIM}][\text{CH}_3\text{COO}]^-$ which was supported by the corresponding pK_a values¹⁴ included in table 1.

Table 1: Values of Hammett function (H^0) and pK_a for ionic liquids

Entry	IL	$A_{\text{max}}^{[a]}$	[I]%	[IH]%	H_0	pK_a
1	Blank	1.558	100.0	0	-	--
2	1	1.07	73.9	26.1	1.44	2.2
3	2	0.71	45.6	54.4	0.91	1.6
4	3	0.448	28.8	71.2	0.60	1.2

^[a] Indicator: 4-nitroaniline

The thermo gravimetric analysis of the three ionic liquids showed (Fig-3) slight weight loss below 100 $^{\circ}\text{C}$ due to absorbed moisture. All these ionic liquids were thermally stable in the temperature range of 260-281 $^{\circ}\text{C}$

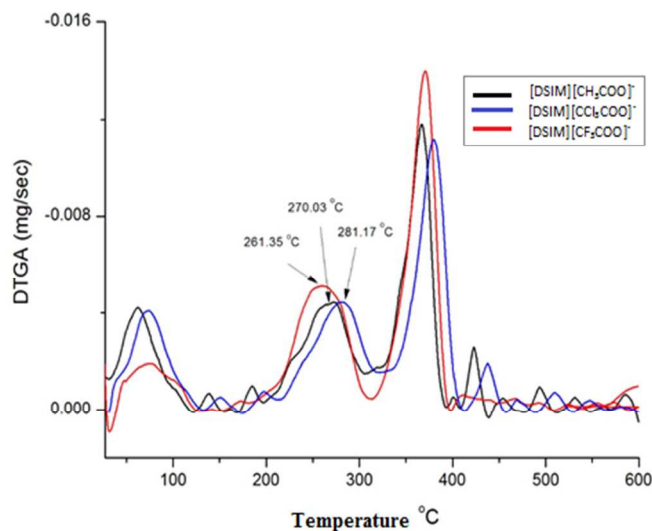
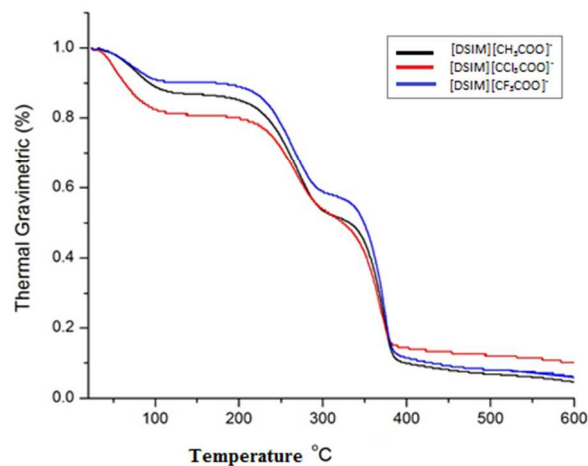
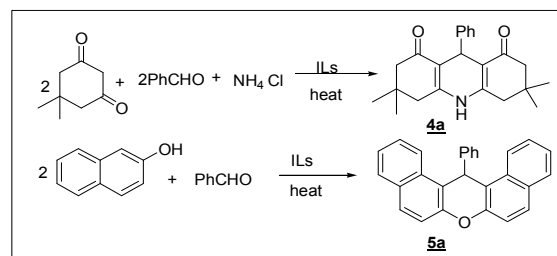


Figure 3. The TG/DTG diagrams of ILs

After calculating the acidity of ILs, we optimized the catalytic activity for the model synthesis of 1, 8-dioxo-decahydroacridine **4a** and dibenzoxanthene **5a** in various conditions (table 2). As a reaction medium the three ionic liquids didn't yield any product at ambient temperature for 4 hours. But at 100 °C the above reactions produced excellent yields within 10-15 minutes in [DSIM][CF₃COO] and [DSIM][CCl₃COO] ILs as reaction medium (table 2, entries 2,4,10,12) while [DSIM][CH₃COO] ionic liquid showed less amount of product (table 2, entries 1, 9). The best catalytic activity observed in solvent-free medium with 25 mol % of **3** at 90 °C for the synthesis of dibenzoxanthene (table 2, entry 14) whereas it was 80 °C for the 1, 8-dioxo-decahydroacridine (table 2, entry 6). Using 25 mol % of ionic liquid **2**, the product formation was decreased at 80 °C as compared to 100 °C under solvent-free condition (table 2, entries 3,7). Both ILs were effective for the formation of **4a** in water at 90 °C with the optimized amount in short time (table -2, entry 8).

Table 2: Ionic liquids catalysed optimization of the reaction conditions for the synthesis of **4a** and **5a**



Entry	IL (mol%)	T (°C)	Time(min)	% of Yield ^a
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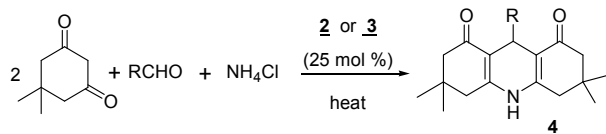
For acridine derivatives **4a**

1	1 (100)	100	25	65
2	2 (100/50)	100	10/10	98/98
3	2 (25/10)	100	10/45	96/65
4	3 (100/50)	100	10/10	100/100
5	3 (25/10)	100	10/45	100/82
6	3 (25)	90/80/70	10/10/25	100/100/70
7	2 (25)	80/90	20	70
8	2 (25)/ 3 (25)	90	25	93 ^b /98 ^b

For dibenzoxanthene derivatives **5a**

9	1 (100)	100	30	70
10	2 (100/50)	100	15/30	95/93
11	2 (25/10)	100	20/45	92/44
12	3 (100/50)	100	15/15	96/98
13	3 (25/10)	100	15/45	97/70
14	3 (25)	90/80/70	15/25/40	97/80/65
15	2 (25)	80/90	45	55
16	2 (25)/ 3 (25)	100	1 h	48 ^b /60 ^b

^a Isolated yields; ^b Using 1 ml of water

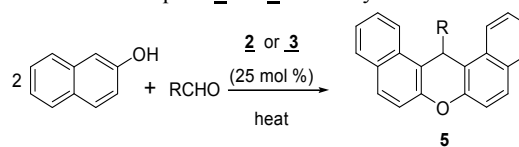
Table 3: Synthesis of 1,8-dioxo-decahydroacridines using ionic liquids **2** and **3** as catalysts

Where R = Ph (**a**), 4-NO₂Ph (**b**), 4-MePh (**c**), 4-ClPh (**d**), 4-MeOPh (**e**), H (**f**), 2-naphthyl (**g**), PhCH=CH- (**h**)

Entry	R	Time (mins)		% of yield ^{a,b}		Mp (°C)
		Solvent	In	Solvent	In	
		free	water	Free(A/B)	Water(C/D)	
1	a	10	25	96/100 4a	93/98 4a	287 ¹⁵
2	b	10	30	96/100 4b	92/96 4b	286 ¹⁶
3	c	15	35	94/98 4c	90/94 4c	279 ¹⁵
4	d	15	35	96/97 4d	93/94 4d	300 ¹⁵
5	e	15	35	95/99 4e	90/96 4e	280 ¹⁵
6	f	20	40	83/85 4f	80/83 4f	156
7	g	15	40	95/97 4g	92/93 4g	204
8	h	15	35	94/97 4h	90/94 4h	215

^a Method A: using 25 mol % of ionic liquid **2** at 100 °C; B: Using 25 mol % of ionic liquid **3** at 80 °C; C: Using 25 mol % of ionic liquids **2** at 90 °C; D: Using 25 mol % of ionic liquid **3** at 90 °C; ^bAll the products were characterized by ¹NMR, ¹³CNMR, FT-IR and CHN analysis.

Interestingly, the synthesis of **4a** was possible only up to 48-60 % yields in water at 100 °C along with unreacted 2-naphthol and aldehydes (table 2, entry 16). It may be due to heterogeneous phases of 2-naphthol in water medium. The standardized conditions were extended with various types of aldehydes using ILs **2** and **3** as catalysts for the synthesis of other derivatives of these heterocyclic compounds. All the results were included in table 3 and table 4. The results were observed to be satisfactory irrespective of the nature of aldehydes except with cinnamaldehyde and furaldehyde in both cases.

Table 4: Synthesis of dibenzoxanthene derivatives **5** using ionic liquids **2** and **3** as catalysts

Where R = Ph (**a**), 4-NO₂Ph (**b**), 4-MePh (**c**), 4-ClPh (**d**), 4-MeOPh (**e**), H (**f**), 2-naphthyl (**g**), PhCH=CH- (**h**)

Entry	R	Time(min)	% of yield ^b	Mp (°C)
		(A/B) ^a		
1	a	20/15	92/97 5a	186 ¹⁷
2	b	25/20	88/94 5b	312 ¹⁷
3	c	20/15	90/93 5c	228 ¹⁷
4	d	25/20	85/94 5d	288 ¹⁷
5	e	20/15	92/97 5e	204 ¹⁷
6	f	30/35	78/85 5f	135
7	g	20/15	90/96 5g	196-197
8	h	25/20	91/97 5h	181 ¹⁸

^a Method A: using 25 mol % of ionic liquid **2** at 100 °C; B: Using 25 mol % of ionic liquid **3** at 90 °C; ^b All the products were characterized by ¹NMR, ¹³CNMR, FT-IR and CHN analysis.

The reusability of the ionic liquids **2** and **3** were expressed by using bar diagram (Fig. 4) for three consecutive runs for the synthesis of 1, 8-dioxo-decahydroacridine **4a** and dibenzoxanthene **5a** under the optimized reaction conditions in solvent-free medium. Recyclability study indicated the high catalytic activity of the two ILs even after the third run with slight increasing reaction time.

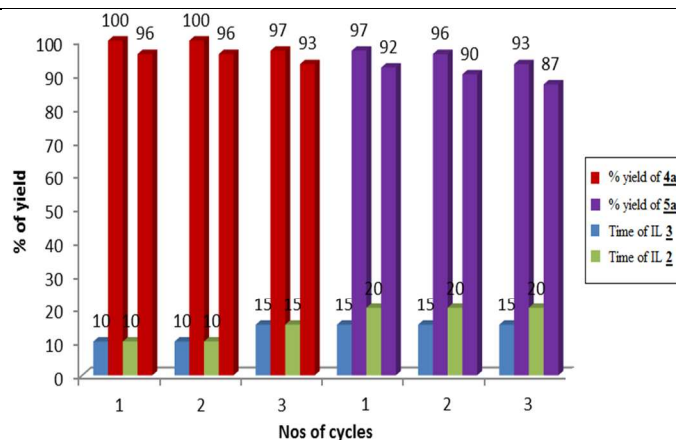


Figure 4: Recycling of the ILs during the synthesis of **4a** and **5a**

Conclusions

In summary, a new group of 1, 3 disulfonic acid imidazolium carboxylate ionic liquids was prepared and characterized by different analytical techniques. The acidity order of these ionic liquids in Hammett plot was identical with their catalytic activity for the one pot synthesis of 1,8-dioxodecahydroacridine and dibenzoxanthene derivatives in solvent-free medium in good to excellent yields.

Experimental Section

Experimental procedure

(a) Preparation of 1, 3 di-sulfonic acid imidazolium carboxylate ionic liquid [DSIM][X] where X= [CH₃COO]⁻, [CCl₃COO]⁻, [CF₃COO]⁻:

All these ionic liquids were prepared by mixing of equal amount of (5 mmol) [DSIM]Cl ionic liquids and carboxylic acids (CH₃COOH, CCl₃COOH, CF₃COOH) in dry dichloromethane (12 ml) in a 50 ml two necked round bottom flask at room temperature within 30 mins. The HCl gas outlet was connected to a vacuum system through water and an alkali trap. The mixture was continued to stirring for one hour to complete the elimination of produced HCl gas and then diluted with 10 ml of dry CH₂Cl₂. The CH₂Cl₂ layer was decanted and washed the residue by more of it (3 x 10 mL). The ionic liquid residue was dried under vacuum to get [DSIM][X] as reddish colored viscous liquids with 98-100 % yields.

(b) General procedure for the synthesis of 1,8-dioxodecahydroacridine derivatives **4**:

A mixture of dimedone (2 mmol), aldehydes (1mmol), ammonium chloride (1 mmol) and ionic liquid (25mol %) was heated in an oil bath for the specified temperature in absence of any solvent (or in 1 mL of water). After completion of the reaction as monitored by TLC, the product was extracted from the ionic liquid phases using dry dichloromethane (3 x 3 mL) as solvent. The product was isolated through distillation of dichloromethane solution under reduced pressure and the ionic liquid medium was again used for next cycle

of reaction. The solid product was purified by recrystallization in aqueous ethanol (15%) to get analytically pure product.

(C) General procedure for the synthesis of dibenzoxanthene derivatives **5**:

A mixture of 2-naphthol (2 mmol), aldehydes (1mmol) and ionic liquid (25 mol %) was heated in an oil bath for the specified temperature in absence of any solvent. After completion of the reaction as monitored by TLC, the product was extracted from the ionic liquid phases using dry dichloromethane (3 x 3 mL) as solvent. The product was isolated through distillation of dichloromethane solution under reduced pressure and the ionic liquid medium was again used for next cycle of reaction. The crude product, thus isolated was subjected to further purification by recrystallization in aqueous ethanol (15%) to get analytically pure product.

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Notes and references

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Electronic Supplementary Information (ESI) available:

This documents contains the ¹H NMR and ¹³C NMR spectral data of ILs (**1**, **2** and **3**), 1,8-dioxo-decahydroacridine (**4**) and dibenzoxanthene (**5**) with the soft copies of spectra. See DOI: 10.1039/b000000x/

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