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Yashvant V Khandare
Department of Chemistry, Amolakchand Mahavidyalaya, Yavatmal, Maharashtra, India

Prashant N Muskawar
Department of Chemistry, Amolakchand Mahavidyalaya, Yavatmal, Maharashtra, India

Correspondence
Prashant N Muskawar
Department of Chemistry, Amolakchand Mahavidyalaya, Yavatmal, Maharashtra, India

Mild synthesis of chromeno[4,3-B]chromene derivatives using novel Lewis-acid hexyl-benzimidazolium based ionic liquid surfactant combined catalyst

Yashvant V Khandare and Prashant N Muskawar

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Abstract
A new 1-hexyl, 3-butane sulfonate benzimidazolium ionic liquid combined with Lewis acid facilitates a novel catalyst for the synthesis of chromeno[4,3-b]chromene derivatives. Chromene derivatives were prepared through the multicomponent reaction among Aromatic aldehydes, 4-hydroxy coumarin and 1,3-dicarbonyl compounds in acetonitrile solvent at 70°C. The significant advantages of this new method include low reaction time, easy work-up, cost-effective, wide substrate scope, excellent yield, and complete atom economy of the final products. Moreover, the prepared catalyst could be frequently recovered many times with only a little decrease in the catalytic activity. The structures of the LAHBimILSC and synthesized chromeno[4,3-b]chromene derivatives investigated by using spectral data (UV-Vis, IR, NMR and mass spectra).

Keywords: 4-hydroxy coumarin and 1, 3-dicarbonyl compounds, chromeno[4,3-b]chromene derivatives, hexyl-benzimidazolium based ionic liquid

1. Introduction
Green chemistry principles exhibit designing of synthesis of new products and processes in such a way which reduces generation of waste and hazardous substances to health and the nature. It includes environmentally friendly methods of formation of new products. It reduces waste by using catalyst. Catalysts effectively work in low amount to carry out reactions in less time. Heterocyclic compounds containing coumarin has the most interested nuclei for many researchers all over the world. This class of compounds exhibits excellent biological activities such as antiviral, antifungal, antibacterial, anticancer and anti-inflammatory [1-3].
The coumarins were extracted from clove flowers and synthesized in 1868 [4]. Recently, near about 1400 species of coumarin explored in the literature which are found in many different families of the plant kingdom [5, 6]. Bicyclic heterocyclic compounds including oxygen as a hetero atom in the structure the fusion of the benzene ring with the 2H-pyron or 4H-pyron rings are designated 2H-Chromene (2H-1-benzopyran) and 4H-chromene (4H-1-benzopyron) [7]. Chromene derivatives occurred in the structures of the natural compounds. Chromenes appeared in alkaloids, tocopherols and flavonoids. Moreover, substituted chromenes played important role in the medicinal chemistry [8-10] Chromenes explored in various natural extracts like visnadine and Khellactone [11, 12].
Recently, several applications of chromene derivatives, large methods of preparations were explored which includes: copper-catalysed intramolecular coupling of aryl bromides with 1,3-dicarbonyls.[13] cycloaddition reaction between propargylic alcohols with 2-naphthols or phenols bearing electron-donating groups, via allenylidene intermediates, leading to the formation of the respective 1H-naphtho[2,1-b]pyrans and 4H-1-benzopyrans,[14] cyclization reaction between different substituted α, α-dicyanoolefins with β-naphthol in the presence of efficient bifunctional thiourea catalyst, [15] among others [16-19]. Chromeno[4,3-b]chromene derivatives synthesizes via multicomponent reaction between 4 hydroxycoumarin (3),1,3-cyclohexanedione (4) and different aryl aldehydes(5a-n) promoted by niobium pentachloride [20]. Lewis acid-surfactant-combined (LASC) catalysts were developed with aqueous-phase [21] which act both as lewis acid to catalyse the reaction and as a surfactant to solubilize the organic substrates in the aqueous medium.

A surfactant-type Bronsted acid [22] forms stable dispersion systems with organic substrates and thus acts both as a catalyst to activate the molecule and as a protector to create a hydrophobic environment within the micelle. Surfactant is well known compound having colloidal nature in aqueous medium and play phase transfer catalyst role in organic reaction. Benzimidazolium Ionic Liquid used as a surfactant combined with Lewis acid played key role in organic reactions [23].

In the literature, different types of Lewis acid-surfactant-combined catalysts (LASC) like Scandium trisdodecylsulfate (STDS) [24], Aluminum tris (dodecyl sulfate) trihydrate [25], Aluminium dodecyl sulfate trihydrate [26], Zirconium tetrakis(dodecyl sulfate) [27], nano-TiO2 on dodecyl-sulfated silica support (NTDSS [28] Iron(III) dodecyl sulphate [29], Zirconium tetrakis(dodecylsulfate) [Zr(DS)$_4$] [30], Fe(SD)$_3$ catalyst [31], SDS with CuSO4 in aqueous media under ultrasonic conditions [32] Were introduced for organic transformation. The synthesis of Bis(indolyl)methanes by the reaction between indole (1) and 4-nitrobenzaldehyde was carried out by using LASSC catalyst gives 99% yield of the product [33].

Ionic Liquids (ILs) have a great attention from chemists. ILs are used as a catalytic medium and to be developed as a green solvents for different chemical processes [34]. Benzimidazole derivatives was reported have a useful in medicinal chemistry and other medicinal agents [35]. Benzimidazolium based ionic liquids were developed as a green and eco-friendly catalysts for different organic reactions [36-38] and an effectively worked as catalysts in the organic reactions to improve synthetic productivity [39-41].

The environmentally benign procedure with reusable catalyst promoted us to develop a safe alternate method for the synthesis of chromeno[4,3-b]chromene derivatives. We have developed an efficient procedure for the synthesis of Lewis-acid benzimidazolium based Ionic Liquid surfactant combined Catalyst (LAHSBim) from 1-Hexyl-3-butane sulfonate benzimidazolium ionic liquid reaction with unhydrous Aluminium chloride ($AlCl_3$). LAHSBim is an efficient catalyst for the synthesis of chromeno[4,3-b]chromene derivatives via a one-pot three-component reaction. This acidic catalyst facilitates the interaction with the substrate molecules and consequently can serve as an efficient catalyst towards the synthesis of chromeno[4,3-b]chromene derivatives.

2. Experimental

2.1 Materials

All the chemicals and solvents are available commercially. Benzimidazole, sodium hydride (60%), Sodium sulphate (were purchased from LOBA Chemical, INDIA. Tetrahydrofuran dried in Lab. Using Toulene, chloroform. 4-hydroxy coumarin, dimedone, 4-nitrobenzaldehyde, 2-nitrobenzaldehyde, acetonitrile, Methanol, ethyl acetate and all other solvent as well as TLC plate Silica gel GF-254 were procured from Merck India used as received unless it is specified. Butane sultone purchased from Sigma-Aldrich India. All melting points have been determined by open capillary method.

2.2. Characterization

The Ionic liquid catalyst and All synthesized chromeno[4,3-b)chromene derivatives were characterized using analytical techniques as IR, ^{1}H NMR and ^{13}C NMR spectroscopy. Also the melting points were measured for all synthesized derivatives.

2.3 Method

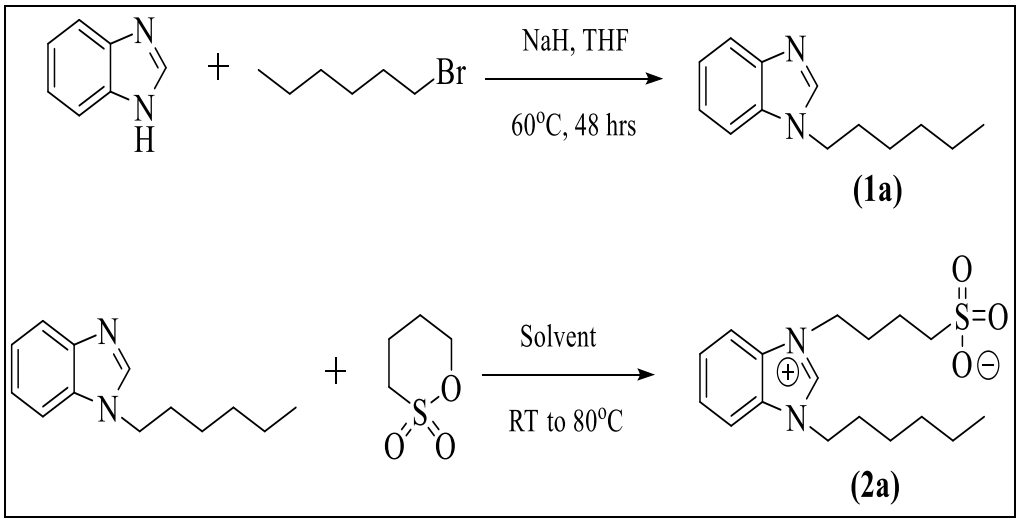
2.3.1 Scheme 1: Synthesis of surfactant 4-(1-hexyl-1H-benzo[d]imidazol-3-ium-3yl)-1-sulfonate ionic liquids

Step I: Preparation of Hexyl Benzimidazole (1a)

The 1-Hexyl benzimidazole was synthesized using known procedure from the literature with minor modifications and Analytical data encountered with literature [37, 42, 43].

Step-II: Synthesis of surfactant 4-(1-hexyl-1H-benzo[d]imidazol-3-ium-3yl)-1-sulfonate ionic liquids. (2a)

In a round bottom flask, A solution of toluene (100 ml) with Hexyl Benzimidazole (0.1 mol) and Butane Sultone (0.1 mol) was taken. Reaction mixture stirred for 48 hours on the magnetic stirrer at room temperature to 80 °C temperature. White precipitate was formed. After completion of reaction, toluene was removed using rotary evaporator and then compound washed with diethyl ether 3 times by adding 10ml. After washing with diethyl ether compound finally washed with ethyl acetate (20 ml). Decanted the solvent and dried over vacuum pump to remove remaining solvent and moisture from it. The dry product of 1-Hexyl Butane Sulfonate Benzimidazolium Ionic Liquid was obtained. Yield: 92%.

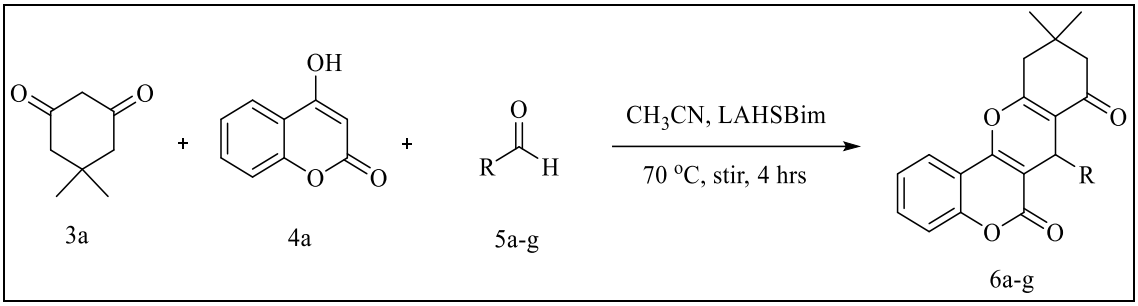


Scheme 1: Synthesis of surfactant 4-(1-hexyl-1H-benzo[d]imidazole-3-ium-3-yl)butane-1-sulfonate (HSBim).

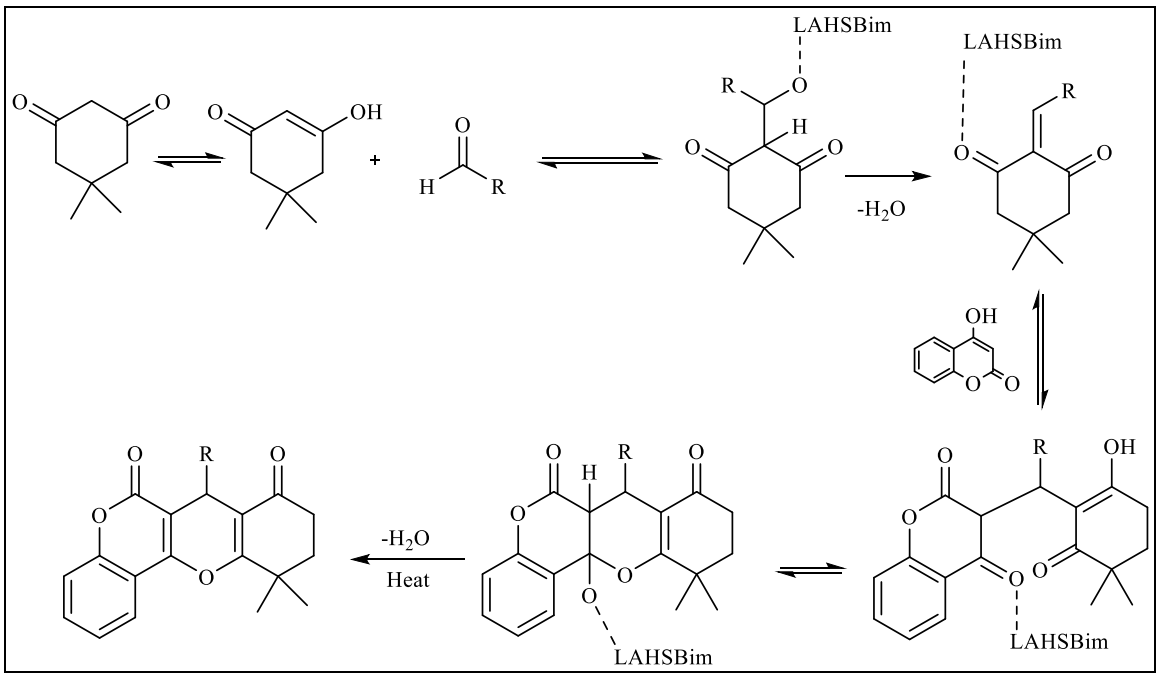
White solid (93%); ^{1}H NMR (400 MHz, DMSO-d_6) δ= 9.90 (s, 1H), 8.08-8.14 (m, *J*=3.40 Hz, 2H), 7.66-7.69 (m, *J*=2.34 Hz, 2H), 4.47-4.55 (m, *J*=7.13 Hz, 4H), 2.53-2.57 (t, *J*=7.53 Hz, 2H), 2.01-2.05 (t, *J*=7.19 Hz, 2H), 1.89-1.92 (t, *J*=6.03 Hz, 2H), 1.64-1.68 (t, *J*=7.38 Hz, 2H). 1.27-1.28 (d, *J*=3.85 Hz, 6H). 0.81-0.85 (t, *J*=7.06 Hz, 3H). ^{13}C NMR (100 MHz, DMSO-d_6) δ= 143.0, 131.52, 126.70, 114.45, 55.20, 51.0, 46.80, 40.0, 31.50, 39.0, 38.0, 35.50, 32.50, 14.60. FT-IR (KBr, ν /cm^{-1}): 3480, 3150, 2950, 2875, 1650, 1500, 1250, 1155, 850, 650, 551. TOF MS (US+) Calculated for $C_{17}H_{26}N_2O_3S^+$; 338.17 [M]; Found 339.17 [M$^+$].

2.3.2. Scheme 2: Synthesis of Chromeno[4,3-b]Chromene Derivatives (6a-g)

In a 50 mL RB flask equipped with a distillation condenser, the catalyst Lewis-acid benzimidazolium based Ionic Liquid surfactant combined catalyst (LAHSBim) (10 mmol%) was added to a mixture of 4-hydroxycoumarin (1 mmol), aromatic aldehyde (1 mmol) and dimedone (1 mmol) in Acetonitrile (5 mL). The reaction mixture was stirred and reflux at 70°C temperature and monitored by TLC. After completion of the reaction (4 hrs), the reaction mixture was filtered in hot condition to remove LAHSBim catalyst. Acetonitrile was removed from the filtrate under rotary evaporator and crude solid compound was recrystallized from ethanol to give the corresponding chromeno[4,3-b]chromene derivative.



Scheme 2: MCR of 4-hydroxycoumarin (4a), 1,3-Dicarbonyl Compounds (3a) and arylaldehydes (5a-g) promoted by in the presence of LAHSBim catalyst at reflux condition.



Scheme 2: Mechanistic proposal for multicomponent reaction promoted by LAHSBim for the Synthesis of Chromin[4,3,b]Chromene Derivatives

3. Results and Discussion

Firstly, the multicomponent reaction between 4-hydroxy coumarin (1 equiv.), 1,3-Dicarbonyl Compounds (1 equiv.) and benzaldehyde (1 equiv.) in the presence of different concentrations (0.0, 10, and 25 mol%) of LAHBim catatlysts and A solvent acetonitrile was used as a model in order to develop a protocol for the optimization of the reaction conditions. The results are summarized in Table 1. As shown in Table1, we were unable to obtain the expected product by using 0.0mol% of LAHSBim catatlyst. The use of 10 mol% of LAHSBim catatlysts produced the best results for the tested solvents, in which presented the best yield (96% in 4h) (entry 3, Table 1). Based on these results, it was established that Entry 3,4,5 (Table 1) presents the best reaction conditions to be applied in the reactions with the other studied aldehydes. We investigated the reaction profile of this reaction by using other aldehydes (5a-g) containing electron-donating and electron-withdrawing groups obtaining the chromeno[4,3-b]chromene derivatives in analogous yields. The results are summarized in Table 2. The products were recrystallized in two steps. Firstly, it was done from ethanol and the second time from ethyl acetate.

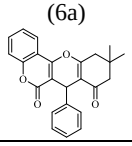
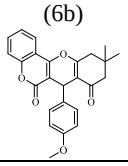
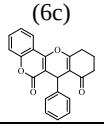
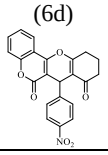
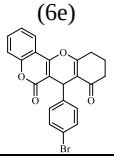
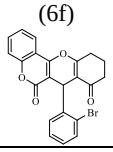
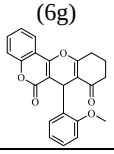
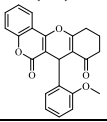
The products were characterized by spectroscopic methods. The results in Table 2 show that by using 10 Mol% equivalent of LAHSBim catatlysts and a reaction time of 4h, it was possible to obtain chromeno[4,3-b]chromene derivatives (6a-g) with good yields (88-96%). No large yields differences were occurred by using different aldehydes.

Table 1: Optimization ofmulticomponent reaction between 4-hydroxycoumarin (4a), 1,3 1,3-Dicarbonyl Compounds (3a-h) and benzaldehyde (5a-h) promoted by LAHSBim.

Entry	LAHBim Catalyst (Mol%)	Solvent	Time (Hrs)	Yield%
1	0.0	Acetonitrile	24	0.0
2	5	Acetonitrile	24	30
3	10	Acetonitrile	4	96
4	25	Acetonitrile	4	96
5	50	Acetonitrile	4	96

[a]Reaction conditions: 4-hydroxycoumarin (4a) (1.0mmol), 1,3-Dicarbonyl Compounds (3a-b) (1.0mmol), benzaldehyde (5a-h) (1.0mmol) and LAHSBim catatlysts (0-50mol%) in Acetonitrile (10 mL) with LAHSBim 0.0, 5, 10, 25, 50 mol% at reflux. [b]Isolatedyields.

Table2: Results for the synthesis of chromeno[4,3-b]chromene derivatives (6a-h)

Sr. No.	Aryl Aldehyde	1,3-Dicarbonyl Compounds	Product	Yield%
1	Benzaldehyde (5a)	5,5-dimethylcyclohexane-1,3-dione (3a)	(6a) 	96
2	4-methoxybenzaldehyde (5b)	5,5-dimethylcyclohexane-1,3-dione (3a)	(6b) 	96
3	Benzaldehyde (5c)	Cyclohexane-1,3-dione (3b)	(6c) 	95
4	4-nitrobenzaldehyde (5d)	Cyclohexane-1,3-dione (3b)	(6d) 	95
5	4-bromobenzaldehyde (5e)	Cyclohexane-1,3-dione (3b)	(6e) 	89
6	2-bromobenzaldehyde (5f)	Cyclohexane-1,3-dione (3b)	(6f) 	90
7	4-(dimethyl)aminobenzaldehyde (5g)	Cyclohexane-1,3-dione (3b)	(6g) 	88
8	2-methoxybenzaldehyde (5h)	Cyclohexane-1,3-dione (3b)	(6h) 	94

Besides, these methodology was applied successfully on aromatic aldehydes and corresponding chromeno[4,3-b]chromene derivatives were obtained in good yields. For comparison, our results for the multicomponent reaction between 4-hydroxycoumarin (3) (1mmol), 1,3-Dicarbonyl Compounds dione (1 mmol) and 4-nitrobenzaldehyde (1 mmol) were compared with other studies described in the literature (Table 3) [29]. When compared with other Lewis acids, [29] LAHSBim is more effective, requiring shorter reaction times and providing better yields.

Based on this work and our experimental results we proposed a plausible mechanism that begins with the

Knoevenagel condensation reaction between the enol form of 1,3-Dicarbonyl Compounds and aldehydes activated by LAHSBim followed by the elimination of H_2O, producing, as intermediate, the b-dicarbonyl enone (I), which can act as Michael acceptors. The b-dicarbonyl enone (I) forms a complex with LAHSBim, and can be attacked by 4-hydroxycoumarin via Michael addition, giving rise to a novel intermediate II that can be readily converted into the product followed by loss of water in the presence of LAHSBim catalyst, leading to 6a products (Scheme 2). Finally, we can conclude that the use of LAHSBim catalyst as a promoter agent in the multicomponent reaction between 4-hydroxycoumarin, 1,3-Dicarbonyl Compounds and aryl aldehydes, produces chromeno[4,3-b]chromene derivatives with good-yielding in short reaction times.

10,10-dimethyl-7-phenyl-10,11-dihydrochromeno [4,3-b]chromene-6,8 (7H,9H)-dione, (6a)
White solid. NMR ^{1}H (400 MHz, $CDCl_3$): δ (ppm)= 7.65 (dd, 1H), 7.41 (dd, 1H), 7.85-7.83 (d, 1H), 7.39 (d, 1H), 7.19 (d, 2H), 7.17 (dd, 2H), 7.09 (dd, 1H), 4.66 (s, 1H), 2.46 (s, 2H), 2.05-2.05 (s, 2H), 1.05 (s, 6H).
NMR ^{13}C (100 MHz, $CDCl_3$): δ= 196.52, 165.22, 163.37, 153.0, 152.77, 144.72, 128.51, 128.32, 127.15, 124.44, 123.8, 116.27, 115.38, 115.05, 114.87, 106.27, 50.48, 40.0, 38.87, 32.31, 26.88.

7-(4-methoxyphenyl)-10,10-dimethoxy,10,11-dihydrochromeno[4,3-b]chromene-6,8(7H,9H)-dione. Yield: 69%, M.P. 184-186 °C, (6b)
White solid. NMR ^{1}H (400 MHz, $CDCl_3$): δ (ppm)= 7.94-7.93 (d, 1H), 7.69 (s, 1H), 7.45-7.43 (t, 2H), 7.18-7.16 (d, 2H). 6.86-6.79 (d, 2H), 4.64 (s, 1H), 3.68 (s, 3H), 2.76 (s, 2H), 2.35-2.31 (d, 2H), 1.09 (s, 3H), 0.99 (s, 3H). NMR ^{13}C (100 MHz, $CDCl_3$): δ= 196.44, 162.90, 160.43, 158.49, 153.90, 152.37, 135.44, 129.88, 129.46, 125.24, 123.07, 117.03, 115.06, 114.42, 113.97, 106.51, 55.45, 50.51, 40.19, 39.35, 32.32, 27.19.

7-phenyl-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene-6,8-dione, (6c)
White solid. NMR ^{1}H (400 MHz, $CDCl_3$): δ= 7.83 (dd, 1.5 Hz, 1H), 7.45-7.41 (m, 1H), 7.43-7.39 (m, 2H), 7.33-7.27 (m, 2H), 7.28-7.23 (m, 1H), 7.21-7.15 (m, 2H), 5.20 (s, 1H), 3.01-2.94 (m, 1H), 2.87-2.75 (m, 1H), 2.67-2.54 (m, 2H), 2.45-2.36 (m, 2H) ppm. NMR ^{13}C (100 MHz, $CDCl_3$): δ= 196.8, 163.4, 160.8, 154.6, 152.8, 144.1, 132.20, 128.3, 127.5, 126.9, 126.20, 124.1, 115.9, 115.1, 113.1, 105.5, 36.8, 33.2, 27.01, 21.1 ppm.

7-(4-nitrophenyl)-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene-6,8-dione, (6d)
White solid. NMR ^{1}H (400 MHz, $CDCl_3$): δ= 8.38-8.29 (m, 1H), 8.15-8.13 (m, 1H), 7.95 (dd, 1.5 Hz, 1H), 7.70-7.62 (m, 2H), 7.57-7.53 (m, 2H), 7.39-7.35 (m, 1H), 5.20 (s, 1H), 2.92-2.78 (m, 2H), 3.21-3.10 (m, 2H), 2.26-2.14 (m, 2H) ppm. NMR ^{13}C (100 MHz, $CDCl_3$): δ= 197.9, 163.2, 161.5, 153.5, 152.4, 145.9, 142.4, 128.7, 127.8, 125.4, 123.7, 123.5, 122.8, 116.1, 116.8, 115.7, 113.7, 105.6, 35.08, 33.6, 27.35, 21.2 ppm.

7-(4-bromophenyl)-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene-6,8-dione, (6e)
White solid. NMR ^{1}H (400 MHz, $CDCl_3$): δ= 7.83 (dd, 1.3 Hz, 1H), 7.65-7.55 (m, 1H), 7.45-7.33 (m, 4H), 7.31-7.25 (m, 2H), 5.01 (s, 1H), 2.83-2.75 (m, 2H), 2.48-2.43 (m, 2H), 2.17-2.13 (m, 2H) ppm. NMR ^{13}C (100 MHz, $CDCl_3$): δ= 196.9, 163.6, 161.5, 154.20, 152.8, 142.5, 133.5, 132.4, 131.4, 125.3, 123.5, 121.2, 117.1, 116.1, 113.7, 105.5, 37.1, 32.9, 27.2, 21.3 ppm.

7-(2-bromophenyl)-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene-6,8-dione, (6f)
White solid. NMR ^{1}H (400 MHz, $CDCl_3$): δ= 7.89 (dd, 1.5 Hz, 1H), 7.68-7.60 (m, 1H), 7.54-7.43 (m, 2H), 7.42-7.34 (m, 2H), 7.52 (d, Hz,1H), 7.29-7.23 (m, 1H), 5.35 (s, 1H), 2.79-2.73 (m, 2H), 2.44-2.34 (m, 2H), 2.15-2.05 (m, 2H) ppm. NMR ^{13}C (100 MHz, $CDCl_3$): δ= 196.7, 163.5, 160.4, 154.7, 152.7, 138.7, 132.9, 132.5, 128.4, 128.2, 127.3, 127.1, 123.8, 123.0, 117.0, 115.1, 113.7, 104.9, 37.2, 36.1, 27.2, 21.3 ppm.

7-(4-(dimethylamino)phenyl)-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene-6,8-dione, (6g)
Yellow solid. NMR ^{1}H (400 MHz, $CDCl_3$): δ= 7.88 (dd, 1.5 Hz, 1H), 7.67-7.61 (m, 1H), 7.55-7.49 (m, 1H), 7.25-7.21 (m, 2H), 7.12-7.06 (m, 1H), 6.67-6.61 (m, 2H), 5.12 (s, 1H), 2.91 (s, 3H), 2.89 (s, 3H), 2.78-2.66 (m, 2H), 2.51-2.36 (m, 2H), 2.19-2.07 (m, 2H) ppm. NMR ^{13}C (100 MHz, $CDCl_3$): δ= 196.7, 162.9, 161.8, 153.4, 152.7, 150.4, 131.5, 129.3, 127.4, 124.9, 124.3, 124.1, 122.9, 116.3, 116.2, 116.1, 114.2, 107.5, 41.2, 37.3, 36.1, 32.3, 27.3, 21.0 ppm.

7-(2-methoxyphenyl)-7,9,10,11-tetrahydro-6H,8H-chromeno[4,3-b]chromene-6,8-dione, (6h)
White solid. NMR ^{1}H (400 MHz, $CDCl_3$): δ= 7.88 (dd, 1H), 7.61-7.53 (m, 2H), 7.39-7.27 (m, 2H), 7.15 (td, 1.8 Hz, 1H), 6.88 (td, 1H), 6.79 (d, 1H), 5.10 (s, 1H), 3.77 (s, 3H), 2.89-2.72 (m, 2H), 2.47-2.35 (m, 2H), 2.12-1.94 (m, 2H) ppm. NMR ^{13}C (100 MHz, $CDCl_3$): δ= 196.4, 164.2, 161.3, 157.9, 155.1, 153.3, 131.9, 131.8, 128.9, 128.7, 124.1, 122.1, 120.7, 117.1, 114.7, 114.2, 112.3, 105.7, 56.2, 37.5, 31.9, 27.6, 21.2 ppm.

4. Conclusion
From green chemistry point of view, a catalyst is more interesting when it can be easily recovered and re-used. We have developed a convenient method for the selective synthesis of chromeno[4,3-b]chromene derivatives via the use of heterogeneous and recyclable Lewis-acid Benzimidazolium based ionic liquid surfactant combined catalyst. The reaction were carried out in acetonitrile at reflux condition but at low reaction time is reported by us for the first time. The notable advantages of this work are simple workup, avoidance of toxic solvents, high reaction rates and good yields, no side reactions, ease of preparation and handling of the catalyst, effective recovery and reusability of the catalyst. This method is bestowed with several unique merits, such as high conversions, simplicity in operation, and cost efficiency. It contributes to the practice of green chemistry.

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