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A novel dicationic ionic liquid as a highly effectual and dual-functional catalyst for the synthesis of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones

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Abstract A novel dicationic ionic liquid, *N*,*N*,*N*′,*N*′-tetramethyl-*N*,*N*′-bis(sulfo)ethane-1,2-diaminium mesylate [TMBSED][OMs]$_2$), has been produced, and identified by analysis of its ^{1}H NMR, ^{13}C NMR, FT-IR, mass and thermal gravimetric data. Thereafter, it has been utilized as a highly effectual, homogeneous and dual-functional catalyst to promote the multi-component reaction of ethyl acetoacetate, hydroxylamine hydrochloride and arylaldehydes under solvent-free conditions for the synthesis of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones. Due to the dual-functionality of [TMBSED][OMs]$_2$ (possessing acidic and basic sites), and also having two sites of each, it was a highly effective and general catalyst for the synthesis. Moreover, a plausible and attractive mechanism based on the dual-functionality of the catalyst has been proposed.

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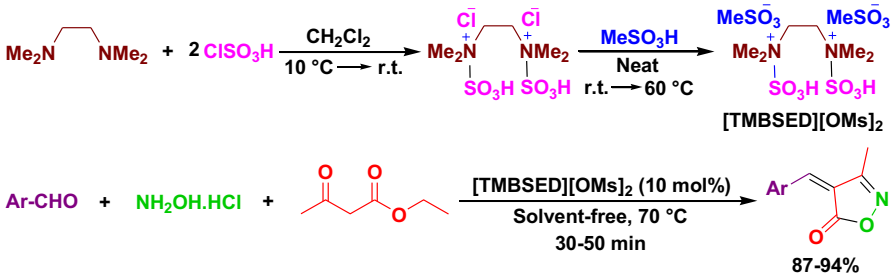
Graphical Abstract



Keywords *N*,*N*,*N*′,*N*′-Tetramethyl-*N*,*N*′-bis(sulfo)ethane-1,2-diaminium mesylate ([TMBSED][OMs]$_2$) · Dicationic ionic liquid · Dual-functional catalyst · 3-Methyl-4-arylmethylene-isoxazole-5(4*H*)-one · Multi-component reaction · Solvent-free

Introduction

Isoxazole derivatives are a significant group of 5-membered heterocycles which have a N–O bond in their ring. They have various utilizations in organic and medicinal chemistry. Many compounds bearing the isoxazole moiety represent medicinal and biological activities, such as antitumor [1], antifungal [2], antibacterial [2], analgesic [3], hypoglycemic [4], anti-oxidant [5], anti-HIV [6], and COX-2 inhibitor [7]. These compounds are also applied as merocyanine dyes [8], fluorescent chemosensors for fluoride anions [9], and androgen antagonists [10]. 3-Methyl-4-arylmethylene-isoxazole-5(4*H*)-ones, as an important member of isoxazole derivatives, can be prepared by the multi-component reaction of ethyl acetoacetate, hydroxylamine hydrochloride and arylaldehydes in the presence of a catalyst, e.g., sodium acetate/hν [11], nanoporous Na^+-montmorillonite perchloric acid [12], phthalimide-*N*-oxyl salts [13], DL-tartaric acid [14], 2-hydroxy-5-sulfobenzoic acid [15], *N*-bromosuccinimide [16], sulfated polyborate [17], Ag/SiO_2 [18], citric acid [19] and montmorillonite nanoclay/ultrasound [20].

In recent times, ionic liquids have been broadly exploited as catalysts to promote organic transformations. This extensive utilization is because of their unbeatable chemical and physical properties, including effectiveness, generality, capability to catalyze numerous types of organic reactions, ability to use in solvent-free or solution conditions, capacity to modify their physical and chemical properties by changing cation and anion structures, negligible vapor pressure, adjustable hydrophobics, broad liquid range, unique electrochemical properties, controlled miscibility, high thermal and chemical stability, good ionic conductivity and non-flammability [21–28].

Production of compounds by multi-component techniques has been broadly used in organic and pharmaceutical chemistry. In this technique, at least three reactants are condensed in one-pot to produce a complex product in which there are the main elements of all the reactants. Accordingly, it is associated with many benefits with respect to classical multi-step reactions, including enhancement of yield, decrement of reaction time, minimizing application of volatile organic solvents, decreasing generation of side-products/waste, saving energy and time, and adoption with green chemistry [29–31].

Another important practical technique in synthetic organic chemistry is carrying out reactions in solvent-free conditions. This technique has many advantages compared with solution conditions, which consist of compliance with green chemistry, higher yields, shorter reaction times, simplicity of reaction procedure, workup and purification, increment of selectivity, need for milder conditions, high effectiveness and minimization of by-product/waste synthesis [32–35].

Considering the above issues, it can be said that the production of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones by a multi-component reaction using a novel ionic liquid-catalyst under solvent-free conditions, is an ideal process. We report this ideal process in this research with the production of a novel ionic liquid, i.e., *N*,*N*,*N*′,*N*′-tetramethyl-*N*,*N*′-bis(sulfo)ethane-1,2-diaminium mesylate ([TMBSED][OMs]$_2$), and its characterization by analysis of ^{1}H NMR, ^{13}C NMR, FT-IR, mass and TGA data. Thereafter, we have introduced [TMBSED][OMs]$_2$ as a highly effective, homogeneous and dual-functional catalyst for the synthesis of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones via the multi-component reaction of ethyl acetoacetate, hydroxylamine hydrochloride and arylaldehydes under solvent-free conditions.

Experimental

Materials and methods

The reactants and solvents were purchased from Merck, Fluka or Acros. Identification of the known compounds was accomplished by comparing their melting points and spectroscopic data with the reported ones. Monitoring progress of the reactions was achieved by thin layer chromatography (TLC). Recording the melting points was performed using a Büchi B-545 device in open capillary tubes. Spectra were recorded on the following devices: ^{1}H NMR (400 or 500 MHz) and ^{13}C NMR (100 or 125 MHz) on Bruker Advance DPX, FT-NMR spectrometers; TG on Linseis STAPT 1000; IR on Shimadzu IR-60; and mass spectra on a spectrometer 5975C VL MSD model Tripe-Axis Detector.

Preparation of [TMBSED][OMs]$_2$

A solution of *N*,*N*,*N*′,*N*′- tetramethylethane-1,2-diamine (5 mmol, 0.581 g) in dry CH_2Cl_2 (20 mL) was added dropwise to a stirring solution of chlorosulfonic acid (10 mmol, 1.165 g) in dry CH_2Cl_2 (200 mL) over a period of 10 min at 10 °C. After

that, the reaction mixture was allowed to heat to room temperature (accompanied by stirring), and stirred for another 4 h. The solvent was evaporated under reduced pressure, and the liquid residue was triturated with dry petroleum ether (3 × 2 mL), and dried under powerful vacuum at 90 °C to give $[TMBSED][Cl]_2$ [24]. Then, methanesulfonic acid (10 mmol, 0.96 g) was added dropwise to $[TMBSED][Cl]_2$ (5 mmol, 1.74 g) over a period of 3 min at room temperature under pressure of nitrogen gas (to remove the HCl produced during the reaction). The resulting mixture was stirred for 12 h at room temperature, and 2 h at 60 °C under a continuous flow of nitrogen gas to give $[TMBSED][OMs]_2$ as a viscous light-brown liquid in quantity yield.

Spectroscopic data of $[TMBSED][OMs]_2$

IR (KBr) ν, cm^{-1}: 874, 1045, 1173, 2000–3600. ^1H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.47 (6H, s) 2.86 (12H, s), 3.51 (4H, s), 9.94 (2H, br.). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 39.9, 43.2, 50.9. Mass, *m/z*: 468 [M^+], 469 [$M^+ + 1$].

General procedure for the production of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones

A mixture of ethyl acetoacetate (1 mmol, 0.130 g), hydroxylamine hydrochloride (1.2 mmol, 0.083 g) and $[TMBSED][OMs]_2$ (0.1 mmol, 0.047 g) was stirred at 70 °C for 1 min. Then, aldehyde (1 mmol) was added to it, and the resulting mixture was stirred at the same temperature. After the reaction was completed (as monitored by TLC), the mixture was cooled to room temperature, and the resulting precipitate was washed by water (2 × 2 mL), dried under vacuum, and recrystallized from methanol to give the p ure product.

Selected spectroscopic data of the products

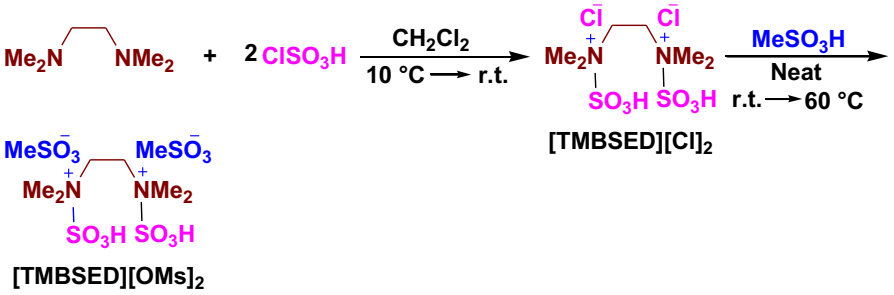
3-Methyl-4-phenylmethylene-isoxazole-5(4H)-one (***A***) ^1H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.32 (3H, s, CH_3), 7.61 (2H, t, J = 7.6 Hz, H_{Ar}), 7.68 (1H, t, J = 7.2 Hz, H_{Ar}), 8.00 (1H, s, = CH), 8.43 (2H, d, J = 7.2 Hz, H_{Ar}). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 11.8, 119.3, 129.4, 132.9, 134.0, 134.4, 152.2, 162.7, 168.3. Mass, *m/z*: 187 [M^+].

3-Methyl-4-(2,4-dimethylphenyl)methylene-isoxazole-5(4H)-one (***B***) ^1H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.30 (3H, s, CH_3-heterocycle), 2.35 (3H, s, CH_3-Ph), 2.46 (3H, s, CH_3-Ph), 7.15 (1H, d, J = 7.9 Hz, H_{Ar}), 7.20 (1H, s, H_{Ar}), 8.01 (1H, s, =CH), 8.33 (1H, d, J = 8.1 Hz, H_{Ar}). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 11.2, 19.5, 21.3, 126.3, 126.5, 127.9, 130.8, 131.1, 131.4, 141.1, 144.3, 149.3, 162.1. Mass, *m/z*: 215 [M^+].

3-Methyl-4-(4-methoxyphenyl)methylene-isoxazole-5(4H)-one (***C***) ^1H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.28 (3H, s, CH_3), 3.91 (3H, s, OCH_3), 7.18 (2H, d, J = 8.8 Hz, H_{Ar}), 7.89 (1H, s, =CH), 8.55 (2H, d, J = 8.8 Hz, H_{Ar}). ^{13}C NMR

(100 MHz, DMSO-d_6) δ (ppm): 11.8, 56.3, 115.2, 115.7, 126.3, 137.4, 151.8, 162.8, 164.7, 169.1. Mass, *m/z*: 217 [M^+].

3-Methyl-4-(3,4-dimethoxyphenyl)methylene-isoxazole-5(4H)-one (***D***) IR (KBr) ν, cm^{-1}: 776, 1247, 1276, 1376, 1459, 1508, 1558, 1589, 1608, 1730, 2939, 3096. ^{1}H NMR (500 MHz, DMSO-d_6) δ (ppm): 2.26 (3H, s, CH_3), 3.83 (3H, s, OCH_3), 3.90 (3H, s, OCH_3), 7.20 (1H, d, J = 8.6 Hz, H_{Ar}), 7.85 (1H, s, =CH), 8.01 (1H, d, J = 8.6 Hz, H_{Ar}), 8.49 (1H, d, J = 2.0 Hz, H_{Ar}). ^{13}C NMR (125 MHz, DMSO-d_6) δ (ppm): 11.7, 55.9, 56.4, 112.1, 115.5, 116.0, 126.5, 131.5, 148.8, 152.2, 154.8, 162.7, 169.3. Mass, *m/z*: 247 [M^+].



Scheme 1 Preparation of [TMBSED][OMs]$_2$

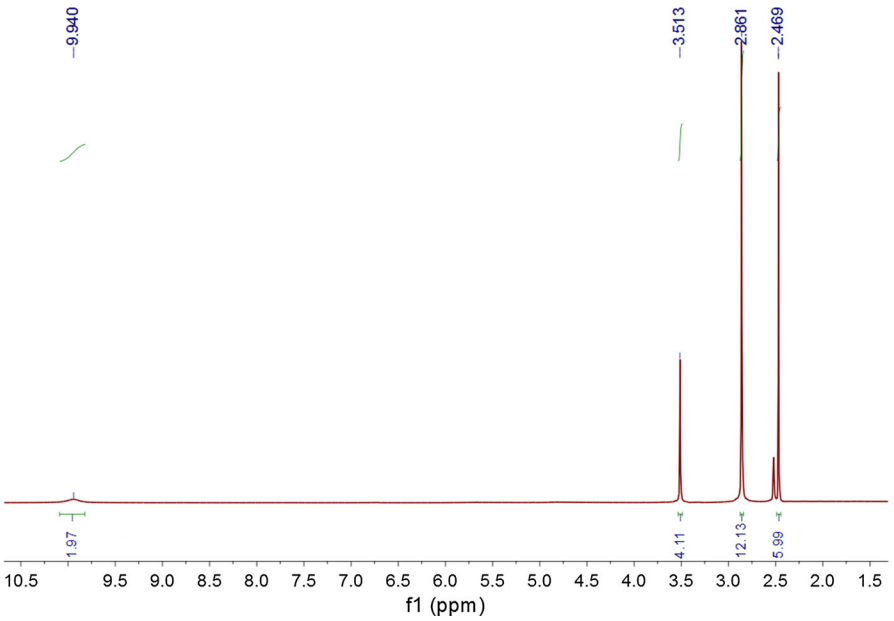
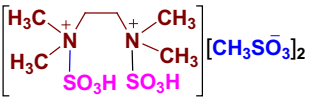


Fig. 1 ^{1}H NMR spectrum of [TMBSED][OMs]$_2$

Table 1 Analysis of the ^{1}H NMR data of $[TMBSED][OMs]_2$



Chemical shift (ppm)	2.47	2.86	3.51	9.94
Integral	5.99	12.13	4.11	1.97
Splitting pattern	Singlet	Singlet	Singlet	Broad
Hydrogen kind	CH_3–SO_3	CH_3–N	CH_2	SO_3H
Hydrogen number	6	12	4	2

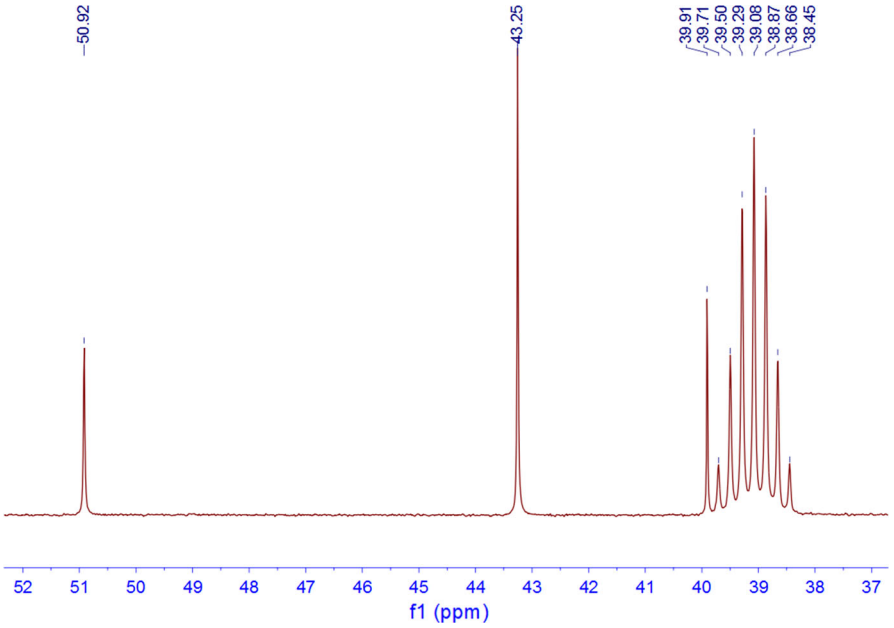
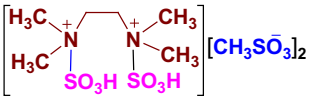


Fig. 2 ^{13}C NMR spectrum of the $[TMBSED][OMs]_2$

3-Methyl-4-(2-methoxyphenyl)methylene-isoxazole-5(4H)-one (***E***) ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.31 (3H, s, CH_3), 3.85 (3H, s, OCH_3), 7.27 (1H, d, J = 8.4 Hz, H_{Ar}), 7.52 (1H, t, J = 8.0 Hz, H_{Ar}), 7.94 (1H, d, J = 7.6 Hz, H_{Ar}), 7.97 (1H, s, =CH), 8.21 (1H, s, H_{Ar}). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 11.8, 55.8, 118.0, 119.5, 120.8, 127.1, 130.4, 134.2, 152.1, 159.6, 162.7, 168.3. Mass, *m/z*: 217 [M^+].

3-Methyl-4-(4-hydroxyphenyl)methylene-isoxazole-5(4H)-one (***F***) ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.27 (3H, s, CH_3), 6.97 (2H, d, J = 8.8 Hz, H_{Ar}), 7.83 (1H, s, = CH), 8.48 (2H, d, J = 8.8 Hz, H_{Ar}), 11.06 (1H, s, OH). ^{13}C NMR

Table 2 Analysis of the ^{13}C NMR data of [TMBSED][OMs]$_2$



Chemical shift (ppm)	38.4–39.71	39.9	43.2	50.9
Carbon kind	DMSO-d_6 (solvent)	CH_3–SO_3	CH_3–N	CH_2

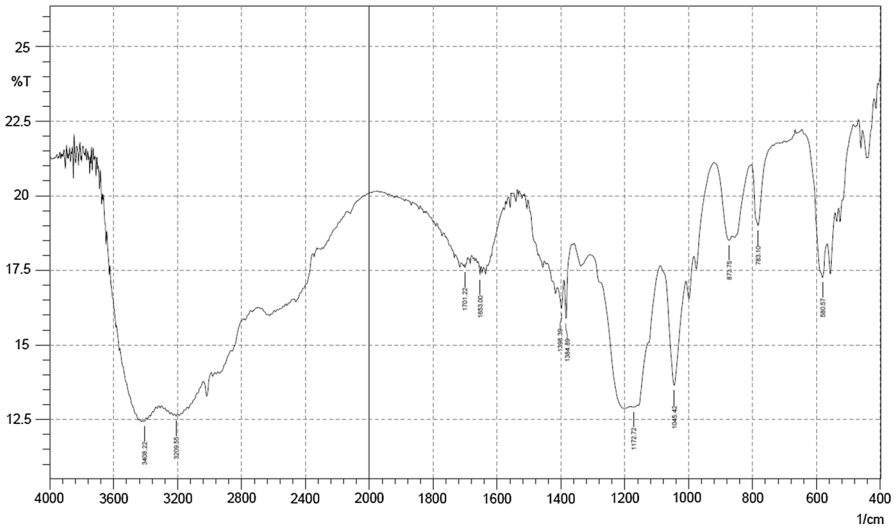


Fig. 3 IR spectrum of [TMBSED][OMs]$_2$

(100 MHz, DMSO-d_6) δ (ppm): 11.8, 114.4, 116.6, 125.1, 138.0, 152.1, 162.8, 164.3, 169.3. Mass, *m/z*: 203 [M^+].

3-Methyl-4-(2-hydroxyphenyl)methylene-isoxazole-5(4H)-one (***G***) ^1H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.25 (3H, s, CH_3), 6.93 (1H, t, J = 7.5 Hz, H_{Ar}), 7.0 (1H, d, J = 8.4 Hz, H_{Ar}), 7.49 (1H, t, J = 7.7 Hz, H_{Ar}), 8.08 (1H, s, =CH), 8.73 (1H, d, J = 8.1 Hz, H_{Ar}), 11.04 (1H, s, OH). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 11.2, 116.0, 116.5, 119.1, 119.5, 132.3, 136.7, 136.8, 145.1, 159.6, 162.2. Mass, *m/z*: 203 [M^+].

3-Methyl-4-(4-dimethylaminophenyl)methylene-isoxazole-5(4H)-one (***H***) ^1H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.23 (3H, s, CH_3), 3.16 (6H, s, $N(CH_3)_2$), 6.89 (2H, d, J = 8.8 Hz, H_{Ar}), 7.65 (1H, s, =CH), 8.49 (2H, d, J = 8.8 Hz, H_{Ar}). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 11.8, 40.4, 109.5, 112.1, 121.5, 138.1, 151.0, 154.9, 162.6, 170.3. Mass, *m/z*: 230 [M^+].

Table 3 FT-IR data of $[TMBSED][OMs]_2$

Peak (cm^{-1})	Related bond or functional group
874	Symmetric N–S stretching vibration
1045	Bending of S–OH
1173	Stretching of S=O in mesylate
2000–3600	OH groups of the two SO_3H

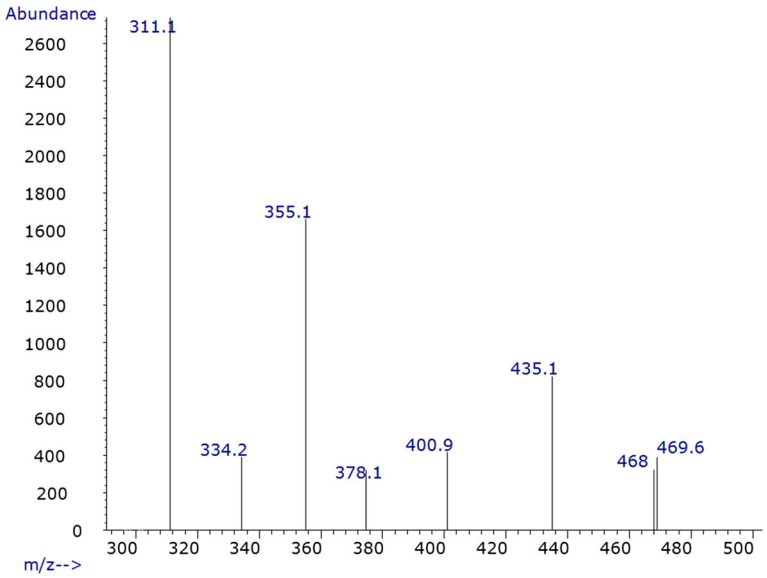


Fig. 4 Mass spectrum of the catalyst

3-Methyl-4-(5-bromo-2-hydroxyphenyl)methylene-isoxazole-5(4H)-one (***I***) IR (KBr) ν, cm^{-1}: 582, 825, 1105, 1259, 1368, 1477, 1578, 1602, 1735, 2918, 3076, 3000–3350. ^{1}H NMR (500 MHz, DMSO-d_6) δ (ppm): 2.26 (3H, s, CH_3), 6.97 (1H, d, J = 8.8 Hz, H_{Ar}), 7.63 (1H, d, J = 8.8 Hz, H_{Ar}), 7.96 (1H, s, =CH), 8.92 (1H, d, J = 2.5 Hz, H_{Ar}), 11.33 (1H, br., OH). ^{13}C NMR (125 MHz, DMSO-d_6) δ (ppm): 11.6, 110.5, 118.4, 118.8, 121.6, 134.3, 138.9, 143.7, 159.1, 162.5, 168.6. Mass, *m/z*: 282 [M^+].

3-Methyl-4-(4-chlorophenyl)methylene-isoxazole-5(4H)-one (***J***) ^{1}H NMR (400 MHz, DMSO-d_6) δ (ppm): 2.31 (3H, s, CH_3), 7.70 (2H, d, J = 8.4 Hz, H_{Ar}), 8.00 (1H, s, =CH), 8.45 (2H, d, J = 8.4 Hz, H_{Ar}). ^{13}C NMR (100 MHz, DMSO-d_6) δ (ppm): 11.8, 119.9, 129.6, 131.7, 135.5, 139.1, 150.5, 162.7, 168.3. Mass, *m/z*: 221 [M^+].

3-Methyl-4-(2-fluorophenyl)methylene-isoxazole-5(4H)-one (***K***) IR (KBr) ν, cm^{-1}: 769, 1118, 1236, 1381, 1482, 1568, 1606, 1618, 1735, 3112. ^{1}H NMR (500 MHz, DMSO-d_6) δ (ppm): 2.30 (3H, s, CH_3), 7.37–7.43 (2H, m, H_{Ar}), 7.70 (1H, m, H_{Ar}), 7.96 (1H, s, =CH), 8.55 (1H, t, J = 7.8 Hz, H_{Ar}). ^{13}C NMR

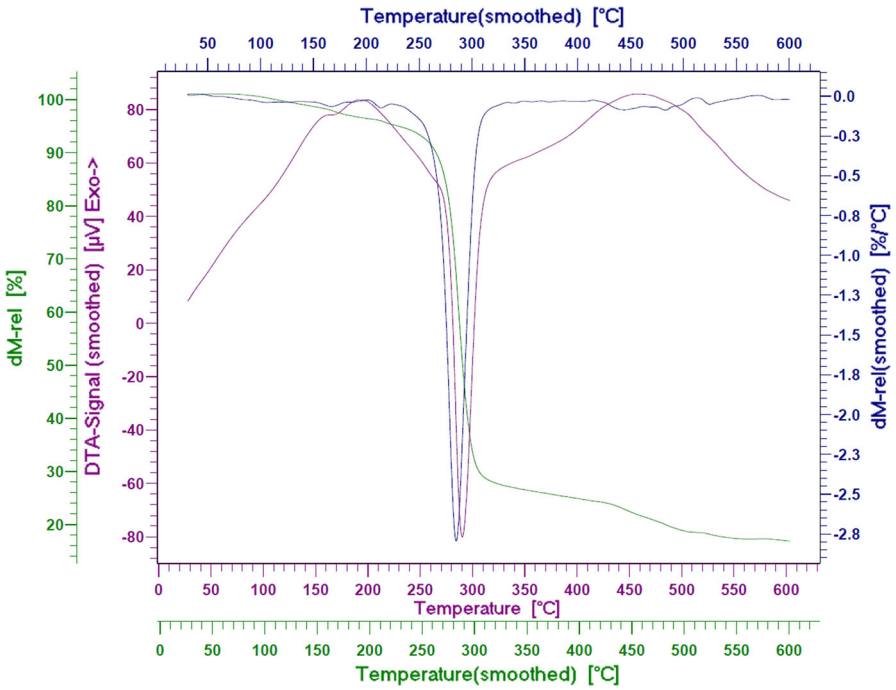


Fig. 5 TG, DTG and DTA diagrams of $[TMBSED][OMs]_2$

(125 MHz, DMSO-d_6) δ (ppm): 11.6, 116.3, 120.4, 121.4, 125.0, 132.8, 136.5, 142.2, 162.3, 162.8, 167.6. Mass, *m/z*: 205 $[M^+]$.

Results and discussion

Production and characterization of $[TMBSED][OMs]_2$

At the outset, the dicationic ionic liquid *N*,*N*,*N*′,*N*′-tetramethyl-*N*,*N*′-bis(-sulfo)ethane-1,2-diaminium mesylate ($[TMBSED][OMs]_2$) was produced by the reaction of *N*,*N*,*N*′,*N*′- tetramethylethane-1,2-diamine (1 eq.) with chlorosulfonic acid (2 eq.), and then with methanesulfonic acid (2 eq.) (Scheme 1).

$[TMBSED][OMs]_2$ was characterized by analyzing its ^1H NMR, ^{13}C NMR, FT-IR, mass and thermal gravimetric analysis (TGA) data.

The ^1H NMR spectrum of $[TMBSED][OMs]_2$ is shown in Fig. 1. There are four kinds of hydrogen in this compound; interpretation of the spectrum is summarized in Table 1.

In the ^{13}C NMR spectrum (Fig. 2), three peaks can be seen. The carbons related to each peak are displayed in Table 2.

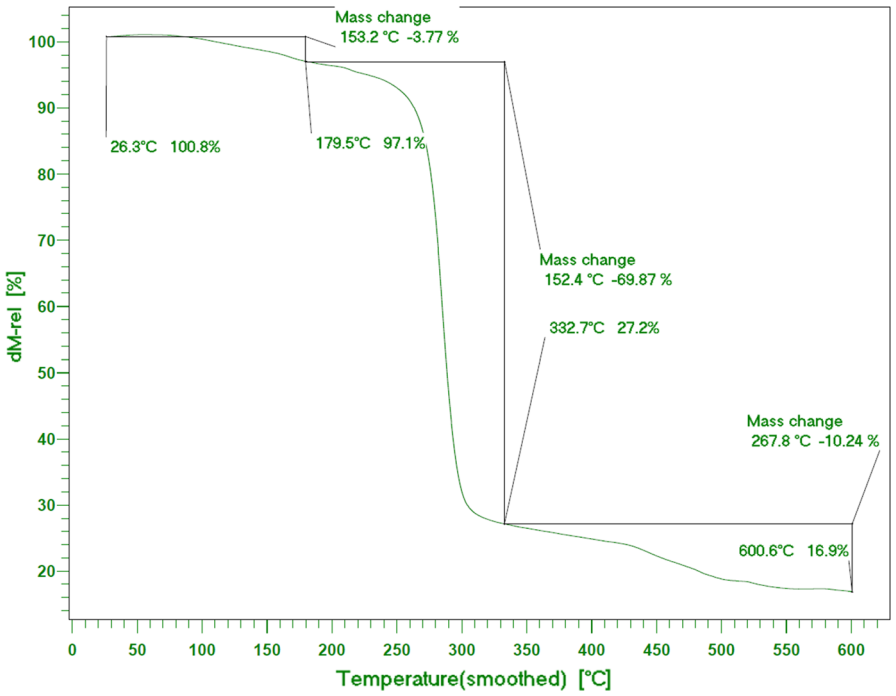
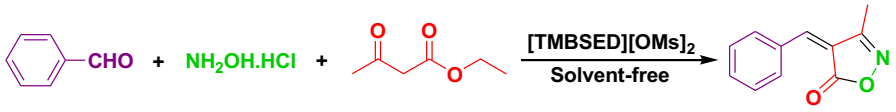


Fig. 6 TG diagram of the ionic liquid



Scheme 2 Reaction of ethyl acetoacetate with hydroxylamine hydrochloride and benzaldehyde

The FT-IR spectrum (Fig. 3) verified the presence of the expected functional groups and bonds in $[TMBSED][OMs]_2$. The main IR data are summarized in Table 3.

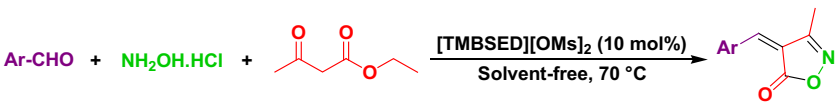
In the mass spectrum of $[TMBSED][OMs]_2$ (Fig. 4), the peaks observed at *m/z* 468 and 469 corresponding to the molecular mass (M^+) and ($M^+ + 1$).

Thermogravimetric analysis of $[TMBSED][OMs]_2$ was also studied. The corresponding diagrams are illustrated in Figs. 5 and 6. The thermogravimetry (TG), differential thermogravimetry (DTG) and differential thermal analysis (DTA) show the main weight loss in one step (about at 285 °C).

Catalytic performance of $[TMBSED][OMs]_2$ for the synthesis of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones

The catalytic performance of the ionic liquid was examined for the synthesis of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones. For the selection of the best

Table 4 Production of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones using $[TMBSED][OMs]_2$



Product	Ar	Time (min)	Yield[a] (%)	M.p. (°C) [Ref.]
A	C_6H_5	40	93	141–143 (141–143) [20]
B	$2,4\text{-}(Me)_2C_6H_3$	40	91	192–194 (193–195) [18]
C	$4\text{-}MeOC_6H_4$	30	90	178–179 (175–177) [16]
D	$3,4\text{-}(MeO)_2C_6H_3$	40	92	134–136 (135) [11]
E	$2\text{-}MeOC_6H_4$	50	89	157–159 (159–160) [19]
F	$4\text{-}HOC_6H_4$	50	92	213–215 (214–215) [20]
G	$2\text{-}HOC_6H_4$	30	91	199–201 (198–200) [16]
H	$4\text{-}(Me_2N)C_6H_4$	30	91	226–227) 224–226 ([20]
I	$5\text{-}Br\text{-}2\text{-}HOC_6H_3$	40	87	199–201
J	$4\text{-}ClC_6H_4$	30	93	128–130 (128–130) [14]
K	$2\text{-}FC_6H_4$	40	94	155–157

[a]Isolated yield

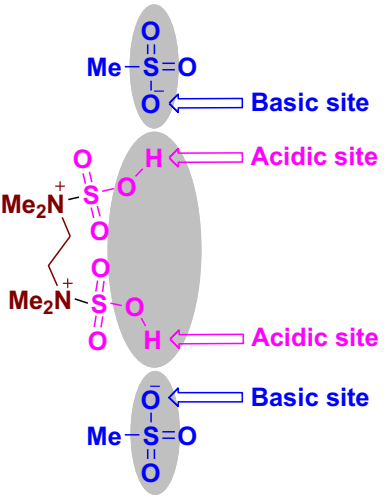
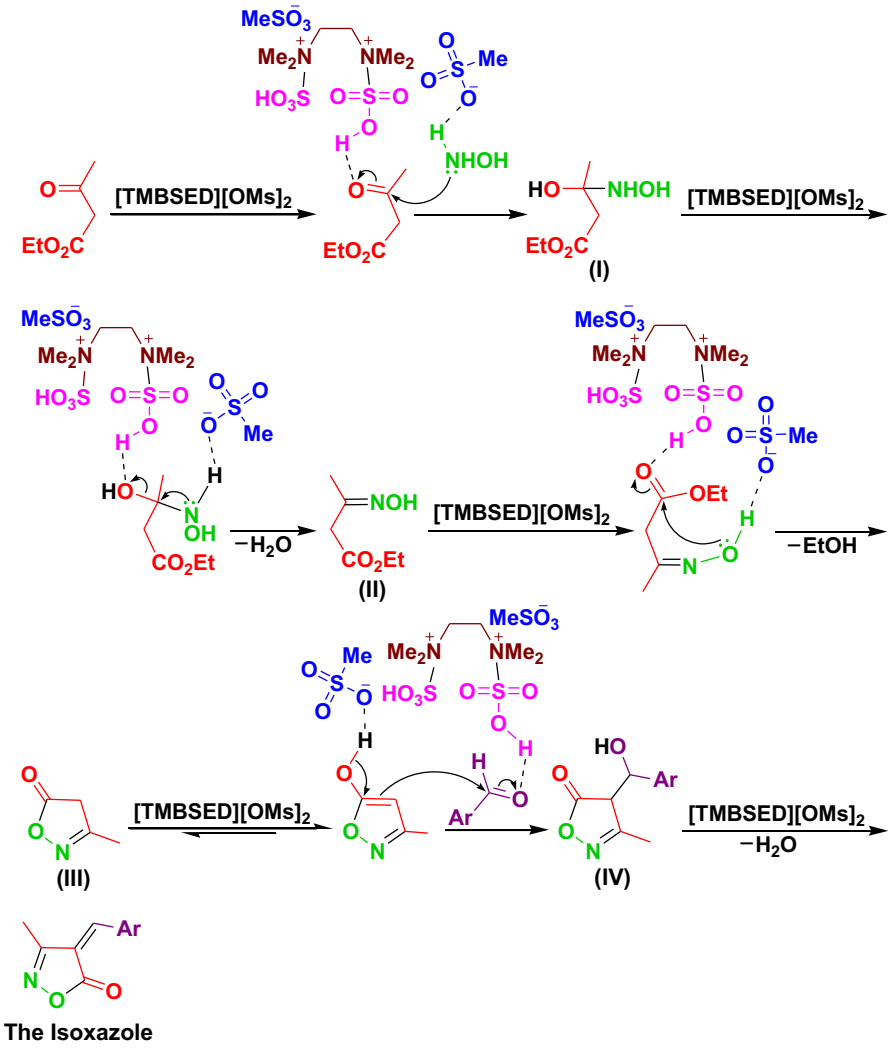


Fig. 7 Acidic and basic sites of $[TMBSED][OMs]_2$

reaction conditions (the catalyst mol% and temperature), the reaction of ethyl acetoacetate (1 mmol) with hydroxylamine hydrochloride (1 mmol) and benzaldehyde (1 mmol) was studied in the presence of 5, 10 and 15 mol% of $[TMBSED][OMs]_2$ in the range of 60–80 °C in solvent-free conditions (Scheme 2). The best results were obtained when 10 mol% of the catalyst was employed at 70 °C (time: 40 min; yield: 93%).

After choosing the best mol% of $[TMBSED][OMs]_2$ and temperature, these conditions were utilized for the condensation of ethyl acetoacetate with



Scheme 3 Proposed mechanism for the synthesis of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones

hydroxylamine hydrochloride and different arylaldehydes; the results are illustrated in Table 4. According to these results, $[TMBSED][OMs]_2$ was highly effective and a general catalyst for the synthesis, because all the aldehydes, including benzaldehyde and arylaldehydes bearing various substituents on the ortho, meta and para positions, afforded the relevant 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones with high yields in relatively short times.

Our novel ionic liquid is a dual-functional catalyst, because it has both acidic and basic sites (the SO_3H group is acidic and the mesylate is basic); moreover, there are two acidic sites, and two basic sites in the catalyst (Fig. 7). Consequently, $[TMBSED][OMs]_2$ can in particular act as a highly effective and general catalyst for

reactions which need to be both acidic and basic catalysts simultaneously; e.g., the production of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones. This issue has been shown in the reaction mechanism (Scheme 3). Two acidic and two basic sites of the ionic liquid can simultaneously catalyze the reaction.

Conclusions

We have introduced *N*,*N*,*N*′,*N*′-tetramethyl-*N*,*N*′-bis(sulfo)ethane-1,2-diaminium mesylate as a novel and homogeneous catalyst for the production of 3-methyl-4-arylmethylene-isoxazole-5(4*H*)-ones. The merits of the presented protocol consist of effectiveness, generality, relatively short reaction times, high yields, purifying the products by a non-chromatography method (crystallization only), broad range of substrate applicability, clean reaction profile, simple experimental procedure, low cost, simple synthesis of the catalyst from available reactants, and dual-functionality of the catalyst.

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