

Supporting Information for

**Halide-free deep eutectic solvents with low viscosity and corrosion
for efficient SO₂ capture and conversion under environmental
conditions**

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Experimental section

Materials

1-Ethyl-3-methylimidazolium acetate (EmimOAc), 1-ethylimidazole, 1-butylimidazole and glyoxaline (Im) were purchased from Adamas Chemical Co. Ltd. China. 2-aminopyridine (2-AmPy) were purchased from Energy Chemical Co. Ltd. 1-Methylimidazole were purchased from Aladdin Biochemical Technology Co. Ltd. glacial acetic acid (HOAc) were purchased from Titan Scientific Co. Ltd. All chemicals are analytical grade and used directly without any treatment unless otherwise stated. SO₂ (99.99vt.%) and N₂ (99.999vt.%) were purchased from Guiyang Shenjian Gas Center, Guiyang, China. The different SO₂ partial pressure gases were obtained using N₂ as the balance gas.

Preparation of PILs and DESs

Typically, 0.1 mol of N-alkylimidazole was dissolved in 30 mL of ethyl acetate in a 250 mL flask, and then an equimolar amount of glacial acetic acid was slowly dripped into the flask under stirring. The reaction system was reacted at 60°C for 24 h. The solvent was removed by vacuum evaporation at the end of the reaction. The remaining product was dried under vacuum at 60°C for 24 h to get proton ionic liquid used as hydrogen bond acceptor (HBA). DESs were preparation by mixing HBA and HBD in a molar ratio of 1:1, and the mixture was heated in an oven at 80°C until a homogeneous liquid was formed. All DESs were vacuum dried at 60°C for 24 hours prior to use.

Characterizations and instruments

The chemical structures of DESs were confirmed by NMR (JEOL FT-400) and FTIR (Nicolet iS50 infrared spectrometer) (see Figure S2-S10). Thermal gravimetric (TG) traces were recorded on a PerkinElmer Pyris 1 TGA with a scanning rate of 10 °C·min⁻¹ under N₂ atmosphere. Densities of DESs were collected using a 10 cm³ capillary pycnometer with a precision of 0.0001 g·cm⁻³. The volume of pycnometer was calibrated as a function of temperature using doubly distilled water at desired temperature. Viscosities of DESs were measured using an Ubbelohde, it was calibrated as a function of temperature with doubly distilled water and HPLC grade ethanol, respectively.

Absorption and desorption of SO₂

Absorption and desorption of SO₂ in DESs were conducted using the weighing method on U-shape device.¹ As a typical run, approximate 1.0 g of DES was added into an absorption tube, which was then immersed in a thermostatic water bath at 20°C. Subsequently, SO₂ at a flow rate of 100 mL·min⁻¹ was continuously bubbled through the solvent in the absorption tube. The quantity of SO₂ absorbed was measured at regular intervals during the process of absorption by an electronic balance with an accuracy of ± 0.1 mg. Desorption experiments were performed by passing N₂ 100 mL·min⁻¹ into SO₂-saturated DES at 80°C. The cycling performance of the DES was evaluated by measuring the mass of the absorption tube at regular intervals during the desorption process.

Cycloaddition of SO₂ with epoxides

Catalytic cycloaddition reactions were carried out using 25 mL Schlenk tubes on a heating plate. In a typical experiment, the epoxides (3 mmol) and DESs (0.3 mmol) were placed into Schlenk tubes without the addition of any solvent. The reactants were frozen with liquid nitrogen and the air was pumped out of the tubes by a water pump and then SO₂ was poured into the tubes. Subsequently, the Schlenk tube was placed on a heating plate at 30°C for 24 h. After the reaction, ethyl acetate and deionized water were added to the Schlenk tube and the organic layer was extracted. Yield and selectivity determined by gas chromatography (Shimadzu GC-2014C).

DFT calculation

All the structures were fully optimized with the B3LYP method based on DFT using the 6-31 G(d) basis to optimize for all atoms. Initially, the DES was optimized and then the interaction between the SO/SO₂ and DES was calculated. The bond angles and bond lengths of the catalysts and reactants were expressed in degrees (°) and angstroms (Å), respectively. All calculations were performed with the Gaussian 16 programs. Previous reports have provided considerable evidence for the accuracy of the calculations.^{2,3}

Corrosion experiment (weight loss method)

Six uniform Q235 iron spheres (8 mm diameter) were polished with sandpaper, sequentially cleaned in deionized water and anhydrous ethanol, and dried at 80°C in a blast drying oven. Two small bottles were filled with 2mL of DES ([EmimOAc][2-AmPy], [EmimBr][2-AmPy]) and 0.1 mL of deionized water. The dried spheres were divided into two groups of three. Each group was weighed (initial masses recorded as $m_{\text{OAc-0}}$ and $m_{\text{Br-0}}$) and placed into a corresponding bottle for corrosion testing. After corrosion, the spheres were rinsed with deionized water and anhydrous ethanol, then ultrasonically cleaned for 20 min in 20 mL of 0.5 mol·L⁻¹ nitric acid solution. Following another rinse with deionized water and anhydrous ethanol, the spheres were dried again at 80°C. Their final masses were recorded as $m_{\text{OAc-1}}$ and $m_{\text{Br-1}}$, respectively. The mass loss corrosion rate for each groups of three was calculated as:

$$CR_m = \frac{m_{x-0} - m_{x-1}}{t} \quad (S1)$$

Eq. (1) where CR_m is the mass corrosion rate, g·a⁻¹; x=OAc or Br; t is the immersion time, a.

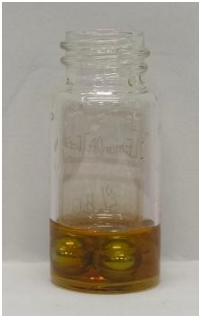


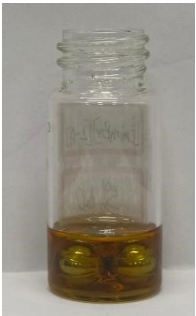


Table S1. Density and viscosity of investigative DESs at 293.2 K under atmospheric pressure.

Entry	DESs	Density (g·cm ⁻³)	Viscosity (cP)
1	[MimOAc][2-AmPy]	1.0895	12.1
2	[EimOAc][2-AmPy]	1.0685	11.8
3	[BimOAc][2-AmPy]	1.0162	14.3
4	[EmimOAc][2-AmPy]	1.1178	151.2
5	[MimOAc][Im]	1.0871	12.1
6	[EimOAc][Im]	1.0400	11.8
7	[BimOAc][Im]	1.0013	16.3
8	[EmimOAc][Im]	1.1041	46.7

Table S2. Comparison of SO₂ absorption capacity in DESs.

Entry	Absorbent	Temperature(K)	SO ₂ absorption(g SO ₂ /g DES)	Ref.
1	[MimOAc][2-AmPy]	293.15	1.1298	This work
2	[EimOAc][2-AmPy]	293.15	1.0671	This work
3	[BimOAc][2-AmPy]	293.15	0.9767	This work
4	[EmimOAc][2-AmPy]	293.15	1.1569	This work
5	[MimOAc][Im]	293.15	1.1279	This work
6	[EimOAc][Im]	293.15	1.0996	This work
7	[BimOAc][Im]	293.15	0.9416	This work
8	[EmimOAc][Im]	293.15	1.2514	This work
9	[Emim]Cl/DMU (2:1)	293.15	1.22	4
10	[Emim]Cl/MU (2:1)	293.15	1.09	4
11	EmimCl-EG (2:1)	293.15	1.15	5
12	EmimCl-EG (1:1)	293.15	1.03	5
13	NMP-2-Pyr (1:1)	298.15	0.89	6
14	Eim-Im (1:1)	298.15	1.02	6
15	[MImH]Cl/Im (1:1)	293.15	1.13	2
16	[ElmH]Cl/2-MIm (1:1)	293.15	1.28	2
17	[EmimCl][2-AmPy]	293.15	1.19	3
18	[EmimBr][2-AmPy]	293.15	1.04	3
19	AmimCl/2-NH ₂ Py (2:1)	293.15	1.26	7
20	AmimCl/2-NH ₂ Py (1:1)	293.15	1.18	7
21	EmimCl: 2-NH ₂ Py (7:1)	298.15	1.24	8
22	EmimCl: 3-OHPy (7:1)	298.15	1.21	8
23	EmimCl:SN (1: 1)	293.15	1.13	9
24	EmimCl:SN (1: 2)	293.15	0.96	9
25	TEACl-Im (1:3)	293.15	1.25	10

Table S3. Comparison of corrosion experiments.

Entry	September 25, 2024		June 9, 2025		
	Photo	Mass(g)	Photo	Photo of after processing	Mass(g)
[EmimOAc][2-AmPy]		6.3138			6.3130
[EmimBr][2-AmPy]		6.2830			6.1101

Conditions: DES (2 mL), H₂O (0.1 mL), 3 iron balls (Q235), environmental conditions. Mass loss is determined by an analytical balance with an accuracy of ± 0.0001 g.

Table S4. Comparison of SO₂ absorption and conversion capacity in DESs/precursor.

Entry	DESs or precursor	SO ₂ absorption(g SO ₂ /g DES) ^a	SO ₂ absorption(mol SO ₂ /mol DES)	Yield(%) ^b
1	[EmimOAc][Im]	1.2514	4.6	93
2	EmimOAc	1.1632	3.1	80
3	[EmimOAc][Im]	1.0996	3.8	95
4	1-Ethylimidazole	1.3996	2.1	89

^aSO₂ absorption was carried out at 20 °C with a flow rate of 100 mL·min⁻¹. ^bStyrene oxide (3 mmol), catalyst (0.3 mmol), 60 °C, 5 h, SO₂ (1.0 bar).

Table S5. The coordinates of the optimized structures.

SO ₂			
S	0.0000000	0.0000000	0.4389631
O	0.0000000	-1.3513296	-0.4389631
O	0.0000000	1.3513296	-0.4389631
styrene oxide (SO)			
C	2.1612168	1.0233197	-0.0459134
C	0.7969846	1.3089849	0.0693609
C	-0.1454958	0.268947	0.1184317
C	0.2980016	-1.0630772	0.0710644
C	1.6620246	-1.3468703	-0.0399608
C	2.5970957	-0.3057027	-0.1029199
H	2.881054	1.8348389	-0.0849513
H	0.4622942	2.3417039	0.120908
H	-0.4327568	-1.860794	0.1485824
H	1.9963971	-2.3792135	-0.0695491
H	3.656058	-0.5281988	-0.1875605
C	-1.5960019	0.591104	0.2067273
C	-2.5848774	0.0533479	-0.7668721
O	-2.5166858	-0.515957	0.608573
H	-1.836116	1.5404403	0.6809671
H	-2.2363007	-0.6127137	-1.5507133
H	-3.4908324	0.6112727	-0.9857753
[EimOAc][Im]			
C	0.7762774	0.5728656	0.6842404
C	0.4222753	2.6142884	-0.048931
C	1.7716786	2.3900022	-0.1416003
N	1.9874289	1.0892873	0.3206629
H	0.6487599	-0.4201174	1.0757832
H	-1.5947126	1.3121758	0.5282042
H	-0.1423996	3.4899014	-0.3117372
H	2.5681128	3.0182315	-0.4912712
C	3.2664882	0.3210856	0.354544
H	3.6786078	0.3762096	1.3673594
H	2.9987288	-0.7156517	0.1188539
C	4.2833845	0.8489323	-0.6674466
H	4.6013543	1.8708965	-0.4371049
H	5.1692757	0.2064223	-0.6463465
H	3.8616414	0.8218276	-1.6764561
N	-0.1907983	1.4689787	0.4655926
C	-3.3980212	0.4220257	-0.1393648
O	-2.7265355	1.3459367	0.521365

O	-2.9216267	-0.5958722	-0.6961129
C	-4.8932249	0.703723	-0.174113
H	-5.2711937	0.7787473	0.8502478
H	-5.0703179	1.6677359	-0.6613247
H	-5.4053398	-0.0931901	-0.7119511
C	-0.3561333	-2.6019262	0.7970819
C	0.9957198	-2.878767	0.7795647
C	0.5472984	-1.8516874	-1.0871147
N	-0.6352884	-1.9476245	-0.397026
H	-1.5397243	-1.4546042	-0.6120893
H	-1.1183385	-2.814897	1.5251985
H	1.5840945	-3.3918847	1.5198989
H	0.623302	-1.3706284	-2.045829
N	1.567085	-2.3999192	-0.4084602

[EimOAc][Im] + SO + SO ₂			
C	-0.7182742	2.629922	-0.6945859
C	-1.6260106	2.5215209	-2.749567
C	-1.194777	3.802162	-2.5502155
N	-0.6372609	3.8486324	-1.2633625
H	-0.3845286	2.3077706	0.3433988
H	-1.5012086	0.8113923	-1.340731
H	-2.1092438	2.0737831	-3.5976503
H	-1.2368567	4.6629167	-3.1911704
C	-0.0173866	5.0252497	-0.5777247
H	-0.7174012	5.8609372	-0.6654883
H	0.0862403	4.7247636	0.4766258
C	1.3458575	5.3666975	-1.1983678
H	1.2577296	5.6226727	-2.259982
H	1.7741086	6.2237797	-0.6689274
H	2.0300823	4.5198514	-1.0926741
N	-1.3205333	1.8130844	-1.5836171
C	0.189982	2.677272	2.5850022
O	0.235032	3.8904741	2.2575572
O	-0.0688192	1.6859843	1.7510481
C	0.4561054	2.2506039	4.0326325
H	1.3503831	1.6184865	4.0663374
H	0.5999089	3.1309544	4.6592113
H	-0.3838899	1.6533907	4.4023421
C	1.7802372	-0.4736942	0.4041343
C	3.0918077	-0.8762194	0.3446375
C	2.0023337	-2.1502447	-1.0721395
N	1.0945375	-1.2911654	-0.5030241
H	0.0931525	-1.2461149	-0.6889892

H	1.2755667	0.2906741	0.9820791
H	3.9625765	-0.5323571	0.8753001
H	1.7464074	-2.8955836	-1.8034179
N	3.2163576	-1.9168085	-0.5762426
C	-5.3896896	-4.186476	0.0072639
C	-5.9342027	-3.0221483	0.5541219
C	-5.1704701	-1.8556782	0.6185311
C	-3.8522127	-1.8533178	0.1468479
C	-3.3117666	-3.0187151	-0.4090456
C	-4.0788886	-4.1805069	-0.4778009
H	-5.9842701	-5.090723	-0.0455958
H	-6.9517043	-3.0208147	0.9266034
H	-5.5968188	-0.9518323	1.0411643
H	-2.3023903	-2.990096	-0.7974647
H	-3.6576336	-5.0793875	-0.9123458
C	-3.0269889	-0.6227424	0.2611517
C	-1.7514528	-0.625044	1.0250374
O	-1.7190316	-0.6576893	-0.5270368
H	-3.5466117	0.3263655	0.1596921
H	-1.4392581	-1.5784327	1.4388865
H	-1.3106162	0.2816875	1.4511662
S	5.6291676	-2.5847291	-0.5947467
O	5.923756	-1.4808905	0.5117783
O	5.5843962	-4.0624368	-0.0256192

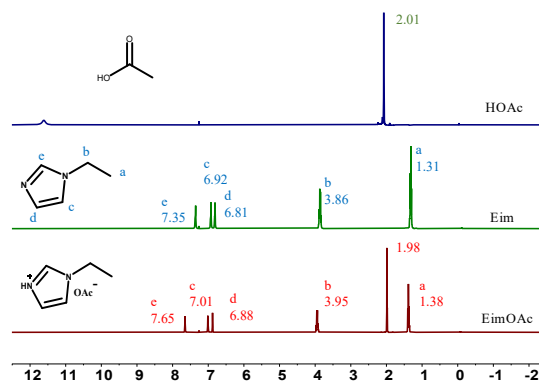


Fig. S1 ^1H NMR spectrum of HOAc, 1-ethylimidazole and EimOAc with an internal reference in CDCl_3 .

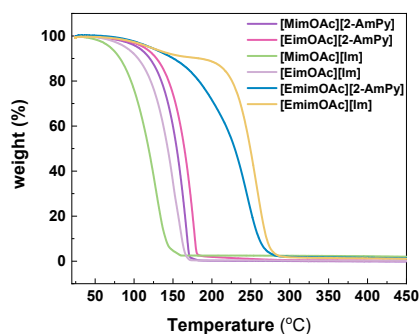


Fig. S2 TGA curves for six DESs with a $10\text{ }^{\circ}\text{C}\cdot\text{min}^{-1}$ heating rate to $450\text{ }^{\circ}\text{C}$ under N_2 atmosphere.

[MimOAc][2-AmPy]: ^1H NMR (400 MHz, $\text{DMSO-}D_6$) δ = 7.90 – 7.81 (m, 1H), 7.56 (s, 1H), 7.36 – 7.26 (m, 1H), 7.08 (s, 1H), 6.87 (s, 1H), 6.41 (dt, J = 8.0, 4.3 Hz, 2H), 6.16 – 5.56 (m, 2H), 3.60 (d, J = 2.7 Hz, 3H), 1.88 (d, J = 2.6 Hz, 3H) ppm. ^{13}C NMR (101 MHz, $\text{DMSO-}D_6$) δ = 172.74 (s), 160.23 (s), 148.00 (s), 137.57 (s), 128.78 (s), 121.19 (s), 112.33 (s), 108.59 (s), 33.31 (s) ppm.

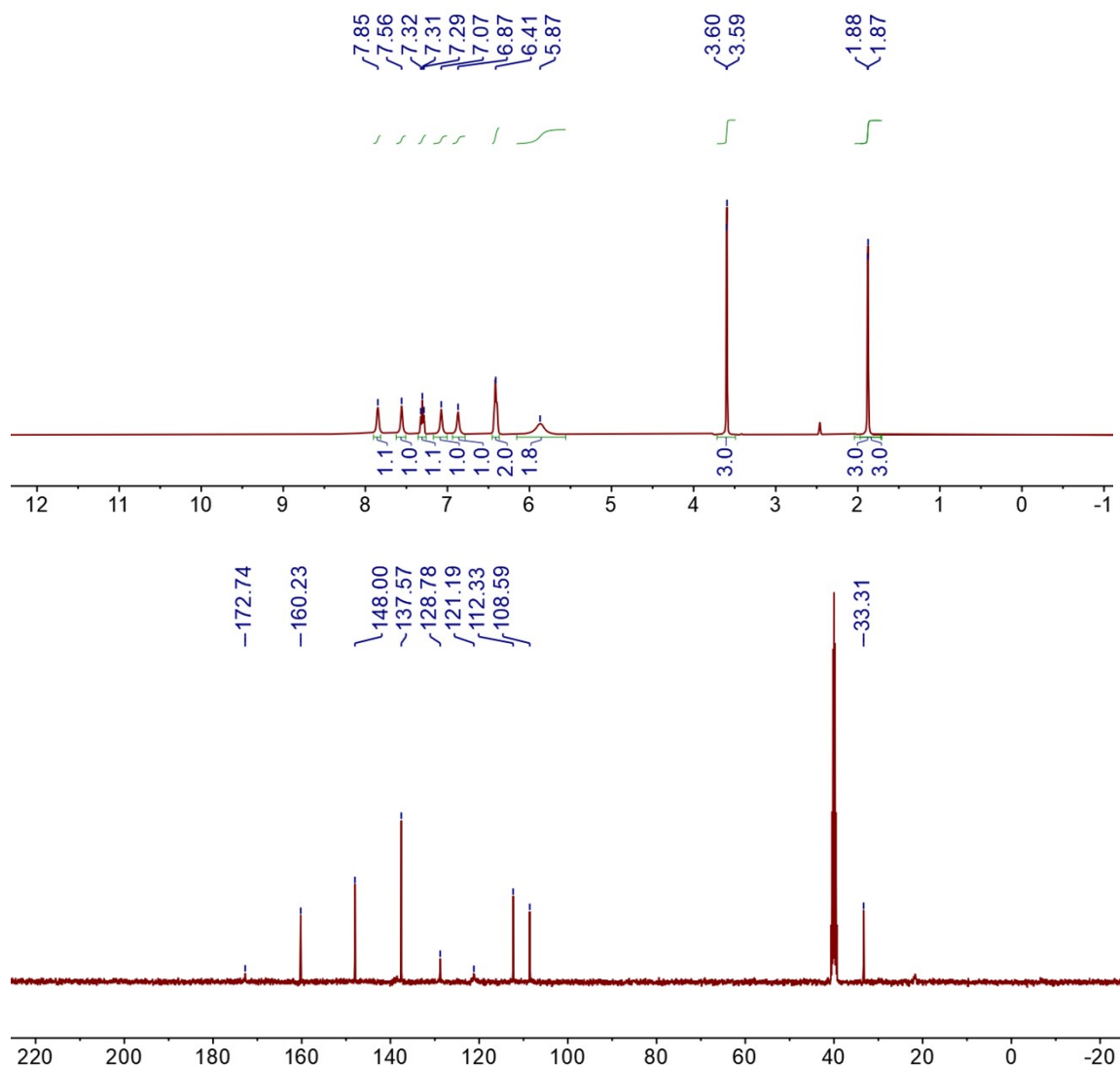


Fig. S3 ^1H and ^{13}C NMR spectra of [MimOAc][2-AmPy] in $(\text{CD}_3)_2\text{SO}$.

[EimOAc][2-AmPy]: ^1H NMR (400 MHz, DMSO- D_6) δ = 7.84 (qd, J = 4.7, 3.1 Hz, 1H), 7.62 (d, J = 4.6 Hz, 1H), 7.31 (dt, J = 7.8, 5.2, 2.2 Hz, 1H), 7.13 (d, J = 5.0 Hz, 1H), 6.87 (d, J = 4.6 Hz, 1H), 6.41 (dd, J = 7.6, 4.9 Hz, 2H), 5.90 (s, 2H), 4.05 – 3.76 (m, 2H), 1.95 – 1.76 (m, 3H), 1.27 (qd, J = 5.9, 2.6 Hz, 3H) ppm. ^{13}C NMR (101 MHz, DMSO- D_6) δ = 172.90 (s), 160.21 (s), 147.91 (s), 137.61 (s), 137.19 (s), 128.58 (s), 119.44 (s), 112.32 (s), 108.63 (s), 41.49 (s), 21.70 (s), 16.81 (s) ppm.

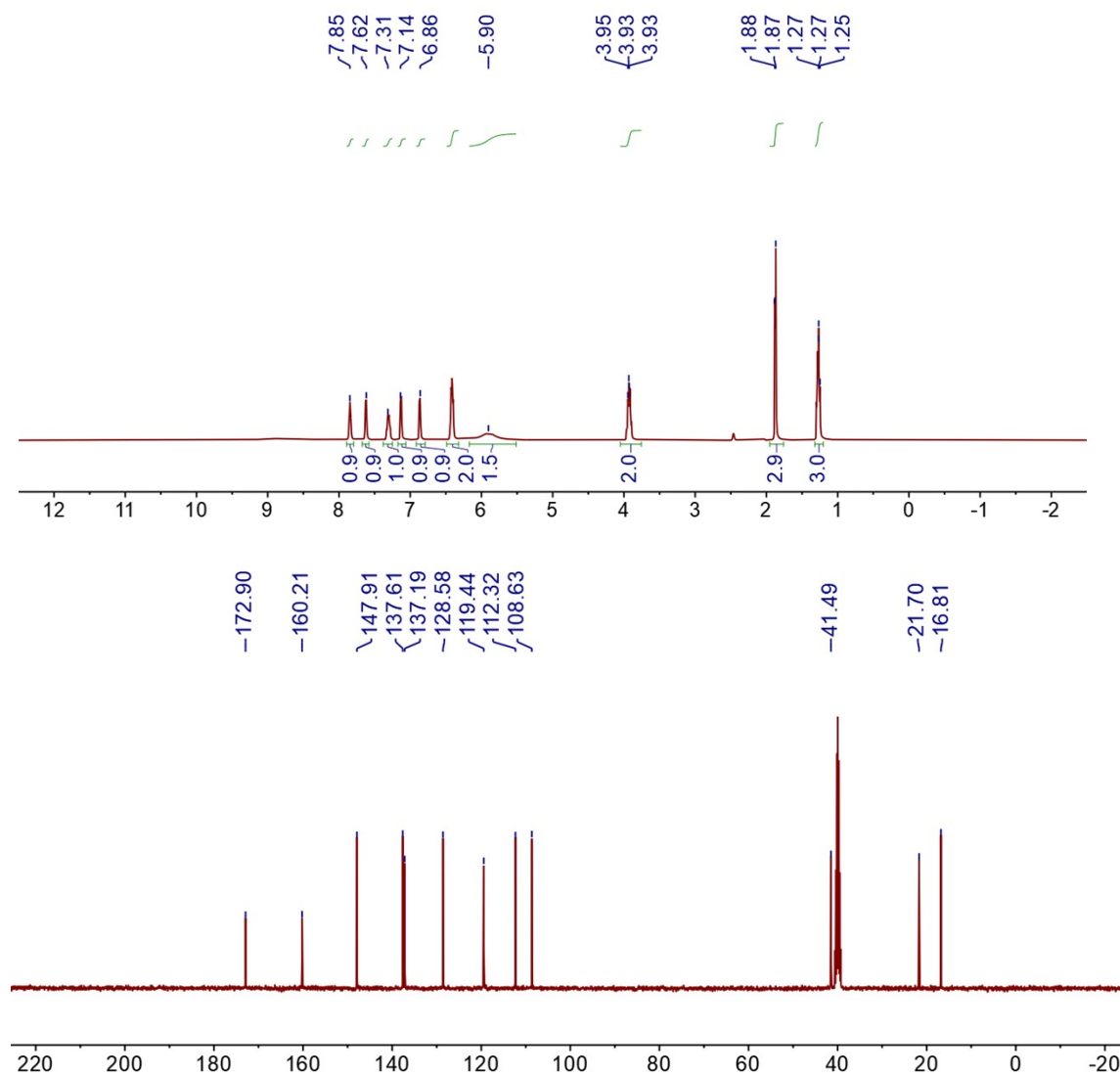


Fig. S4 ^1H and ^{13}C NMR spectra of [EimOAc][2-AmPy] in $(\text{CD}_3)_2\text{SO}$.

[BimOAc][2-AmPy]: ^1H NMR (400 MHz, $\text{DMSO-}D_6$) δ = 7.88 (dd, J = 4.8, 1.8 Hz, 1H), 7.63 (s, 1H), 7.44 – 7.28 (m, 1H), 7.15 (d, J = 1.4 Hz, 1H), 6.89 (d, J = 1.3 Hz, 1H), 6.56 – 6.24 (m, 2H), 5.90 (s, 2H), 3.94 (d, J = 7.2 Hz, 2H), 1.91 (s, 3H), 1.66 (p, J = 7.3 Hz, 2H), 1.20 (dq, J = 14.7, 7.3 Hz, 2H), 0.86 (t, J = 7.4 Hz, 3H) ppm. ^{13}C NMR (101 MHz, $\text{DMSO-}D_6$) δ = 172.79 (s), 160.24 (s), 148.01 (s), 137.67 (s), 137.54 (s), 128.63 (s), 119.78 (s), 112.29 (s), 108.57 (s), 46.19 (s), 33.13 (s), 21.67 (s), 19.65 (s), 13.86 (s) ppm.

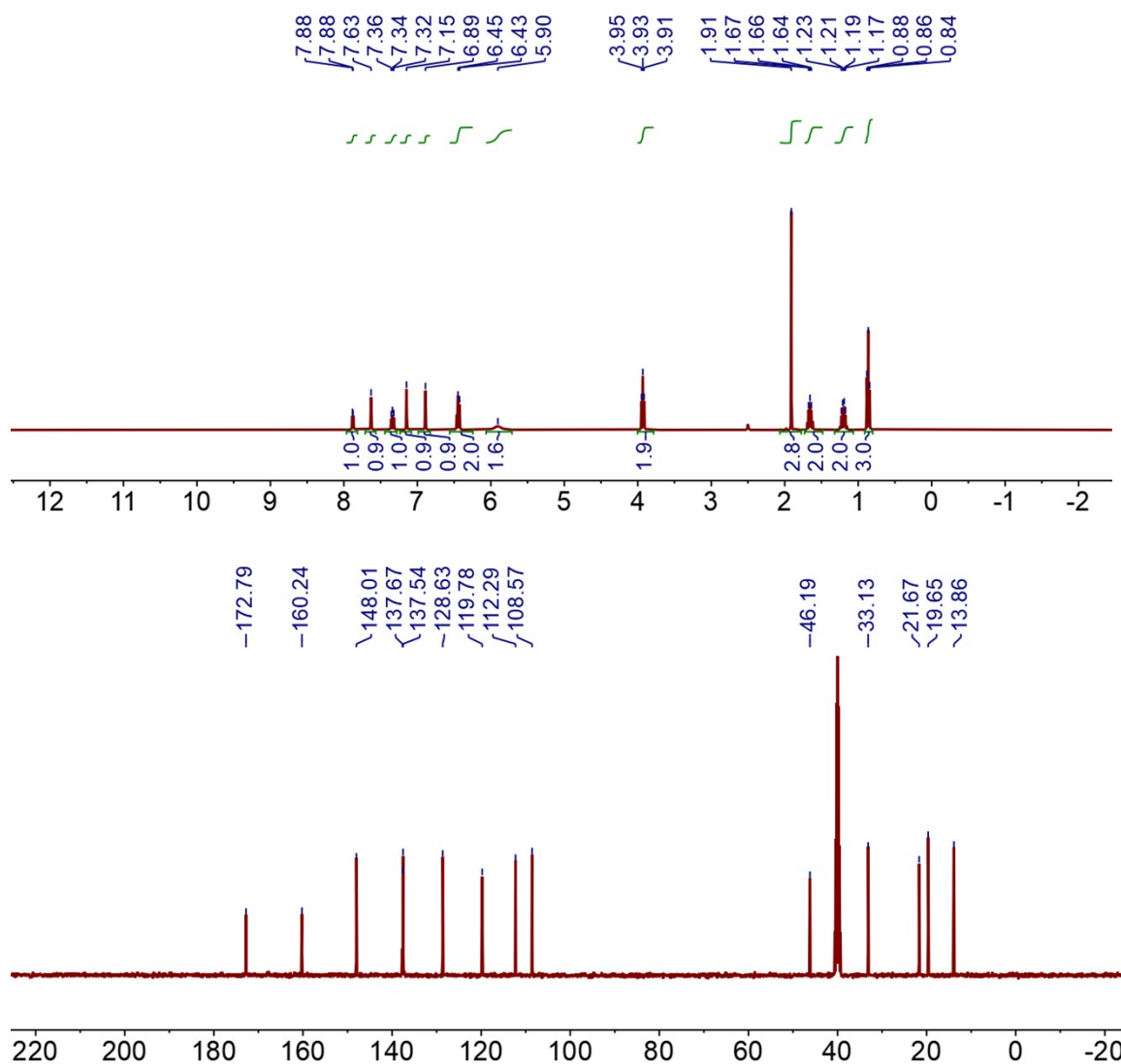


Fig. S5 ^1H and ^{13}C NMR spectra of [BimOAc][2-AmPy] in $(\text{CD}_3)_2\text{SO}$.

[EmimOAc][2-AmPy]: ^1H NMR (400 MHz, DMSO- D_6) δ = 9.82 (s, 1H), 7.82 (dd, J = 5.1, 2.0 Hz, 1H), 7.81 – 7.78 (m, 1H), 7.71 (t, J = 1.8 Hz, 1H), 7.28 (ddd, J = 8.7, 7.0, 2.0 Hz, 1H), 6.43 – 6.35 (m, 2H), 5.92 (s, 2H), 4.17 (q, J = 7.3 Hz, 2H), 3.82 (s, 3H), 1.53 (s, 3H), 1.36 (t, J = 7.3 Hz, 3H) ppm. ^{13}C NMR (101 MHz, DMSO- D_6) δ = 173.47 (s), 160.38 (s), 148.21 (s), 137.65 (s), 137.39 (s), 124.02 (s), 122.45 (s), 112.14 (s), 108.48 (s), 44.48 (s), 36.06 (s), 26.80 (s), 15.74 (s) ppm.

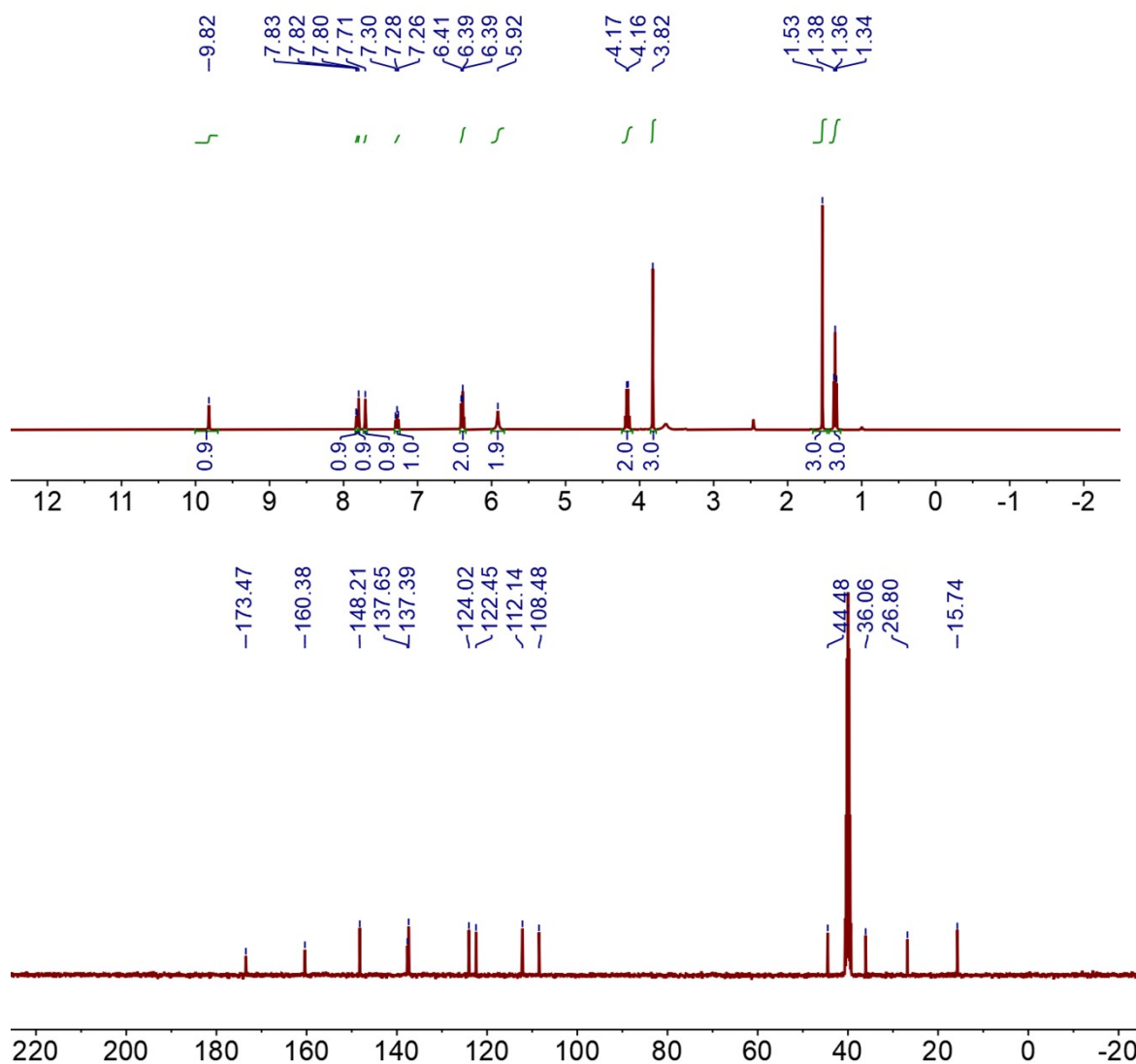


Fig. S6 ^1H and ^{13}C NMR spectra of [EmimOAc][2-AmPy] in $(\text{CD}_3)_2\text{SO}$.

[MimOAc][Im]: ^1H NMR (400 MHz, $\text{DMSO-}D_6$) δ = 9.39 (s, 2H), 7.67 (d, J = 6.3 Hz, 1H), 7.56 (d, J = 6.2 Hz, 1H), 7.06 (d, J = 6.1 Hz, 1H), 7.01 (d, J = 6.5 Hz, 2H), 6.86 (d, J = 6.4 Hz, 1H), 3.60 (s, 3H), 1.87 (d, J = 6.3 Hz, 3H) ppm. ^{13}C NMR (101 MHz, $\text{DMSO-}D_6$) δ = 172.82 (s), 138.35 (s), 135.70 (s), 128.64 (s), 122.14 (s), 121.06 (s), 33.29 (s), 21.74 (s) ppm.

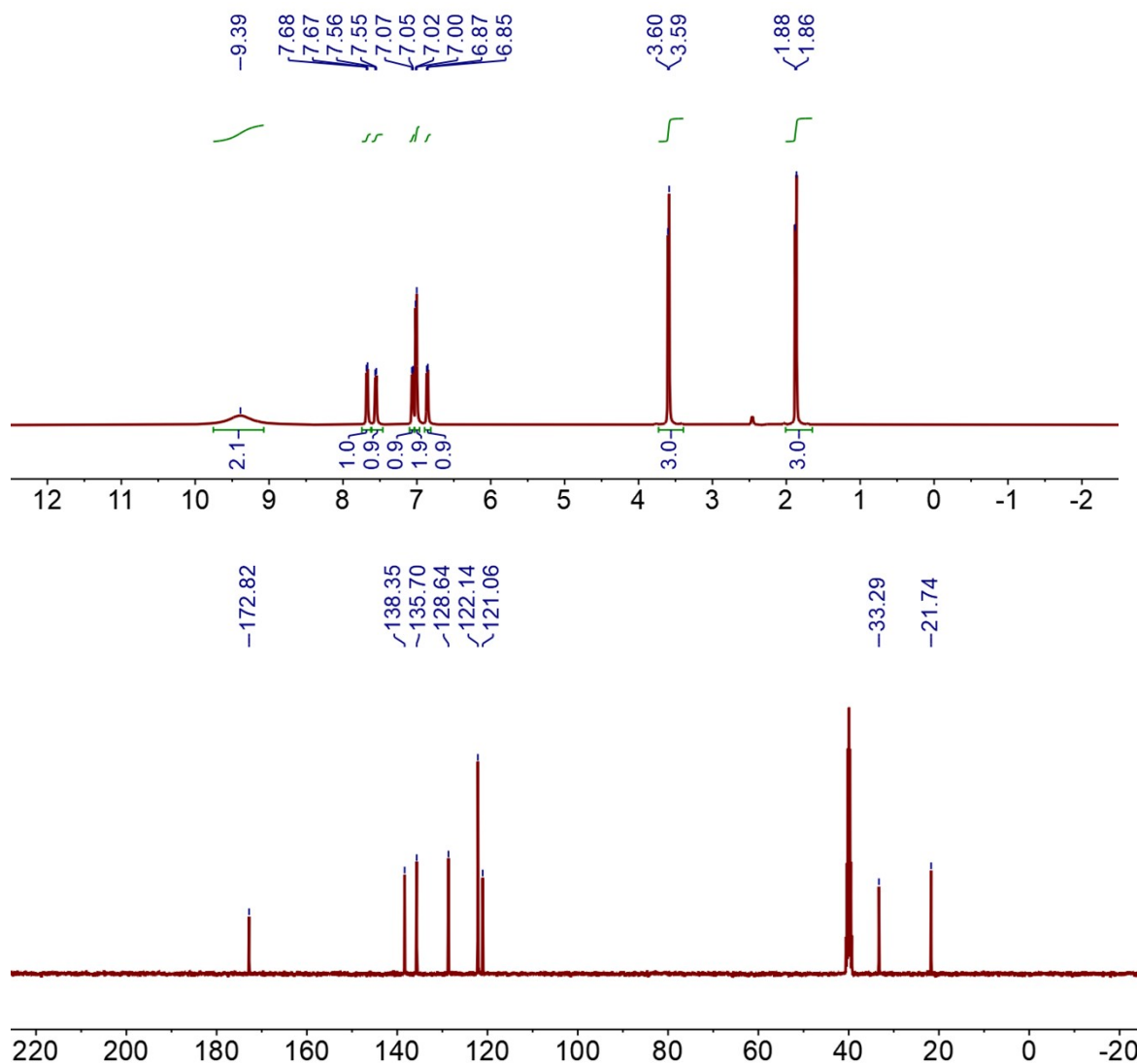


Fig. S7 ^1H and ^{13}C NMR spectra of [MimOAc][Im] in $(\text{CD}_3)_2\text{SO}$.

[EimOAc][Im]: ^1H NMR (400 MHz, $\text{DMSO-}D_6$) δ = 10.45 (s, 2H), 7.66 (s, 1H), 7.62 (s, 1H), 7.13 (s, 1H), 7.01 (d, J = 1.9 Hz, 2H), 6.86 (s, 1H), 3.93 (qd, J = 7.3, 1.7 Hz, 2H), 1.87 (s, 3H), 1.28 (td, J = 7.3, 1.7 Hz, 3H) ppm. ^{13}C NMR (101 MHz, $\text{DMSO-}D_6$) δ = 172.78 (s), 137.20 (s), 135.69 (s), 128.60 (s), 122.14 (s), 119.44 (s), 41.48 (s), 21.73 (s), 16.81 (s) ppm.

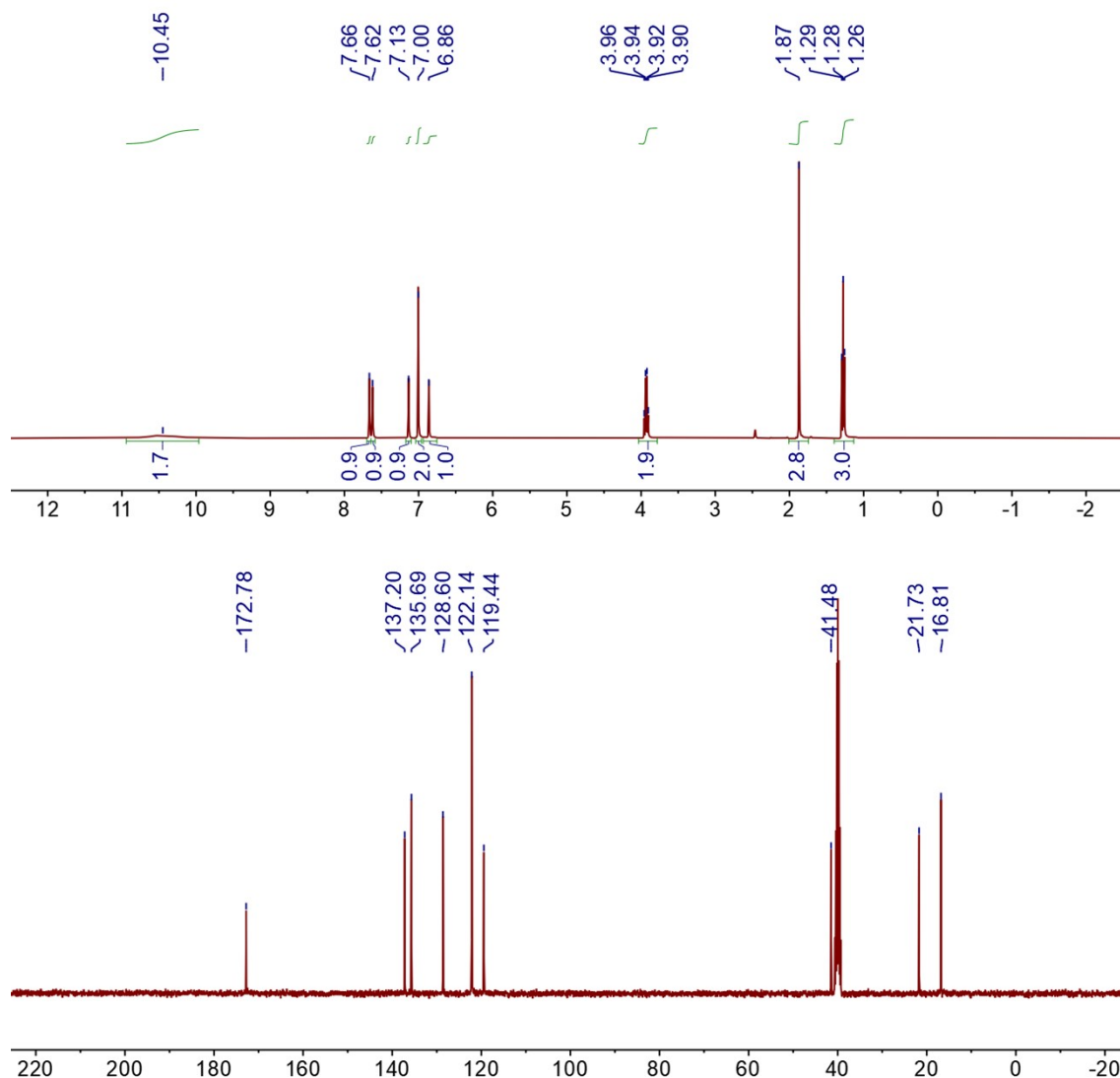


Fig. S8 ^1H and ^{13}C NMR spectra of [EimOAc][2-AmPy] in $(\text{CD}_3)_2\text{SO}$.

[BimOAc][Im]: ^1H NMR (400 MHz, $\text{DMSO-}D_6$) δ = 7.00 (s, 1H), 3.90 (td, J = 7.1, 1.6 Hz, 1H), 1.87 (d, J = 1.7 Hz, 1H), 1.76 – 1.52 (m, 1H), 1.27 – 1.04 (m, 1H), 0.83 (td, J = 7.3, 1.5 Hz, 1H) ppm. ^{13}C NMR (101 MHz, $\text{DMSO-}D_6$) δ = 172.73 (s), 137.68 (s), 135.68 (s), 128.62 (s), 122.15 (s), 119.79 (s), 46.19 (s), 33.13 (s), 21.70 (s), 19.65 (s), 13.86 (s) ppm.

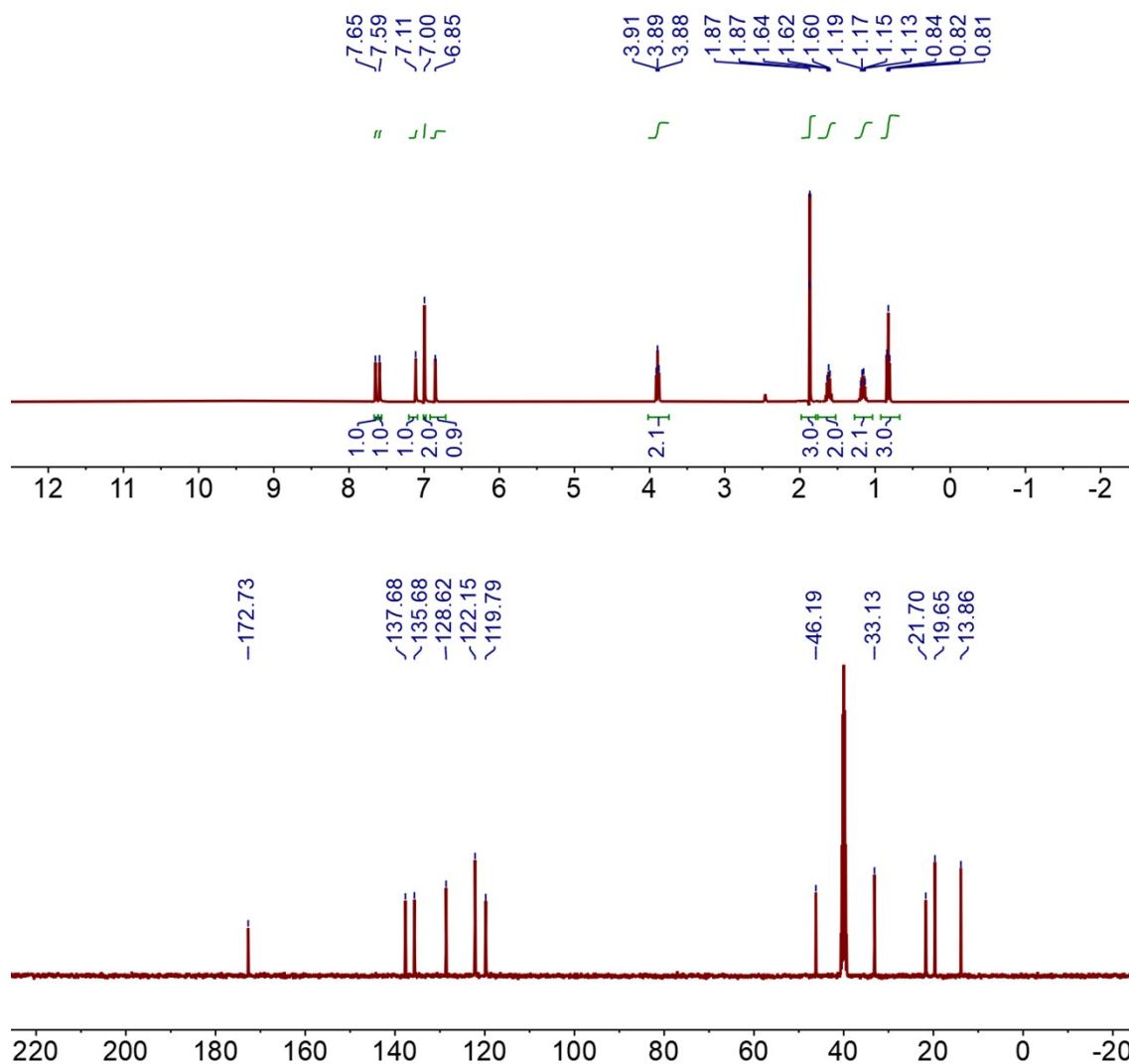


Fig. S9 ^1H and ^{13}C NMR spectra of [BimOAc][2-AmPy] in $(\text{CD}_3)_2\text{SO}$.

[EmimOAc][Im]: ^1H NMR (400 MHz, DMSO- D_6) δ = 9.57 (d, J = 3.9 Hz, 1H), 7.82 – 7.75 (m, 1H), 7.74 – 7.66 (m, 1H), 7.52 (d, J = 4.0 Hz, 1H), 6.92 (d, J = 4.0 Hz, 2H), 4.15 (qd, J = 7.3, 4.1 Hz, 2H), 3.81 (d, J = 4.1 Hz, 3H), 1.58 (d, J = 4.2 Hz, 3H), 1.36 (td, J = 7.3, 4.1 Hz, 3H) ppm. ^{13}C NMR (101 MHz, DMSO- D_6) δ = 137.52 (s), 135.93 (s), 124.01 (s), 122.44 (s), 44.46 (s), 35.94 (s), 26.11 (s), 15.63 (s) ppm.

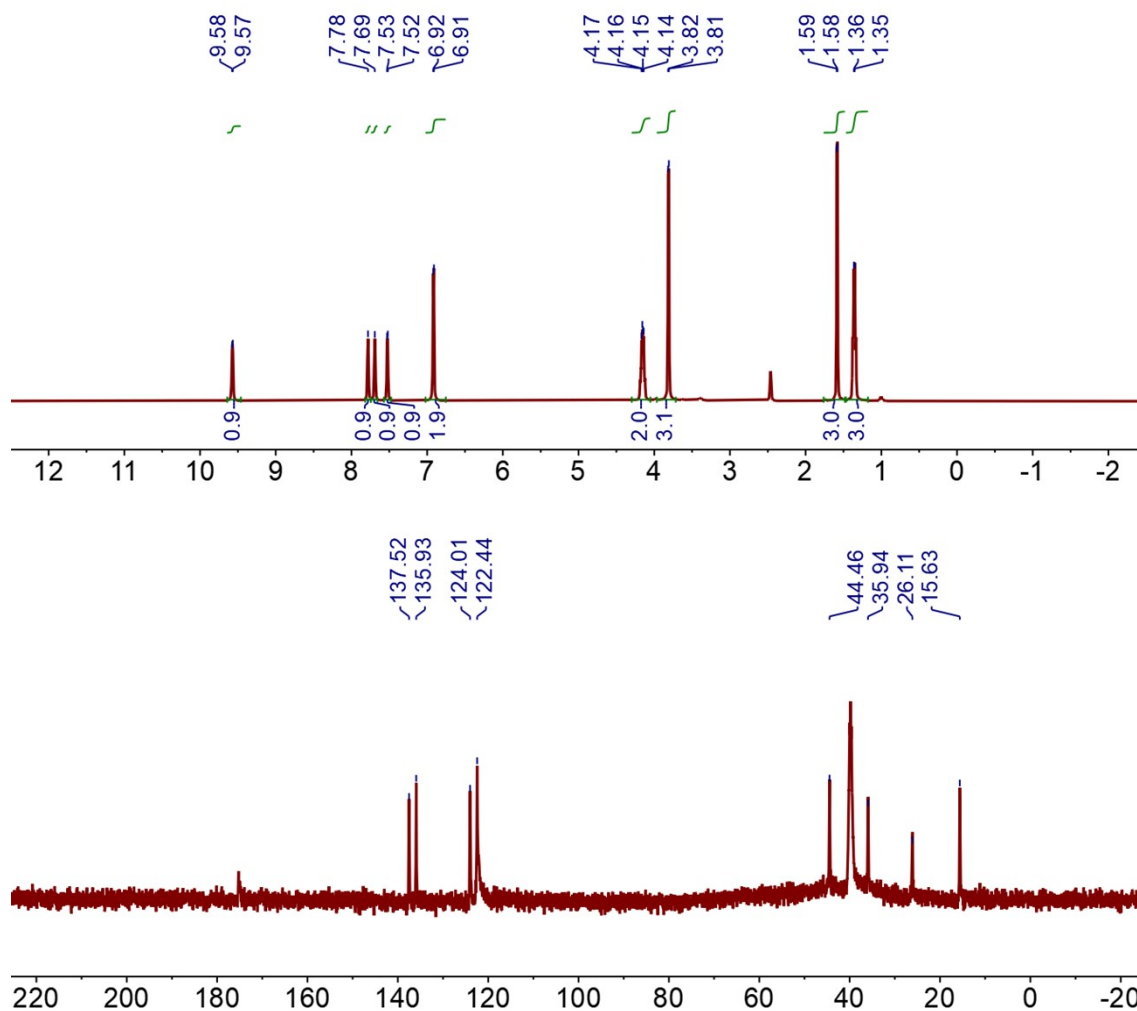


Fig.S10 ^1H and ^{13}C NMR spectra of [EmimOAc][2-AmPy] in $(\text{CD}_3)_2\text{SO}$.

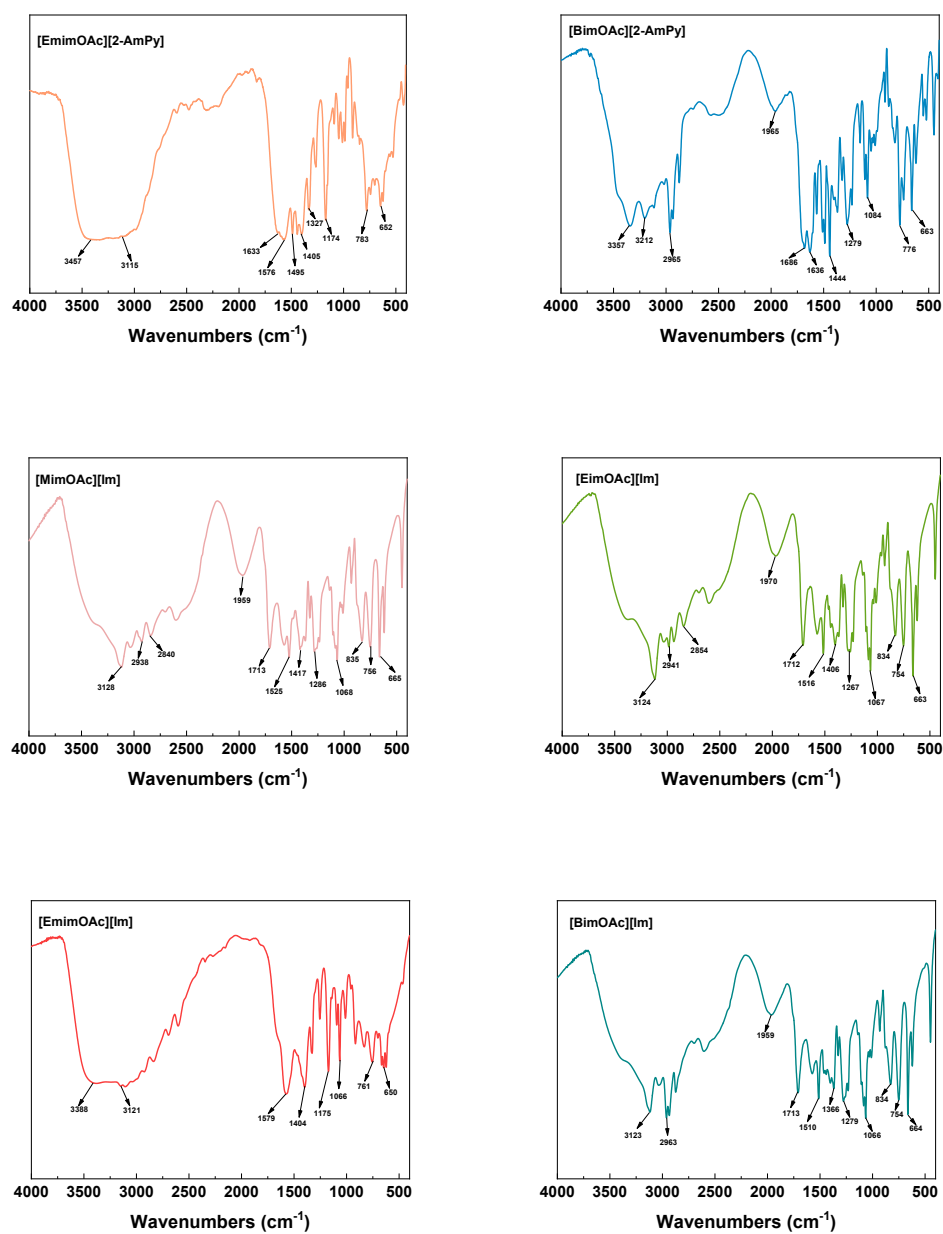


Fig. S11 FTIR spectra of the eight prepared DESs.

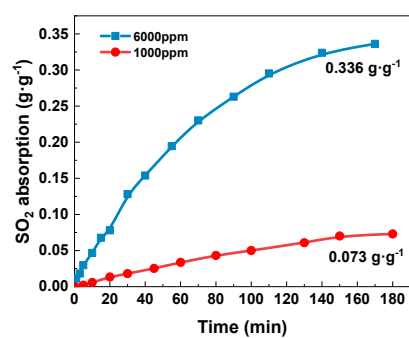


Fig. S12 Absorption of 6000 and 1000 ppm SO₂ in [EimOAc][Im] at 20 °C

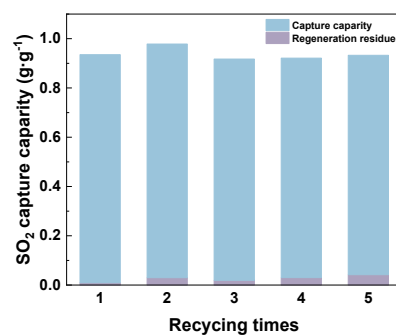


Fig. S13 Five consecutive cycles of SO₂ absorption and desorption in [EimOAc][Im]. Absorption temperature: 30 °C, and desorption temperature: 80 °C

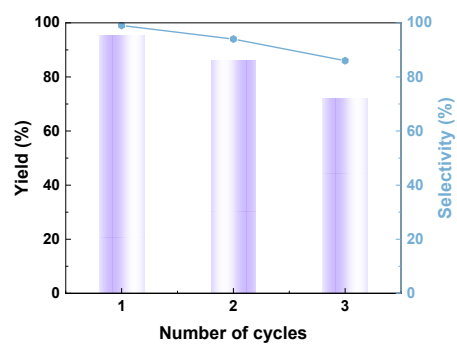


Fig. S14 Recyclability of [EimOAc][Im]. Reaction condition: 3 mmol of SO. 0.1 g of [EimOAc][Im], 30 °C, 24 h.

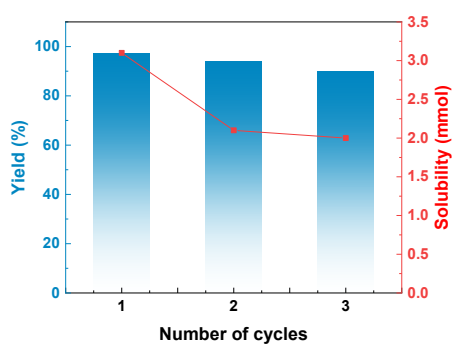


Fig. S15 Reactive regeneration of [EimOAc][Im]. Reaction condition: SO with equal moles to SO₂. 0.5 g of [EimOAc][Im], 60 °C, 5 h.

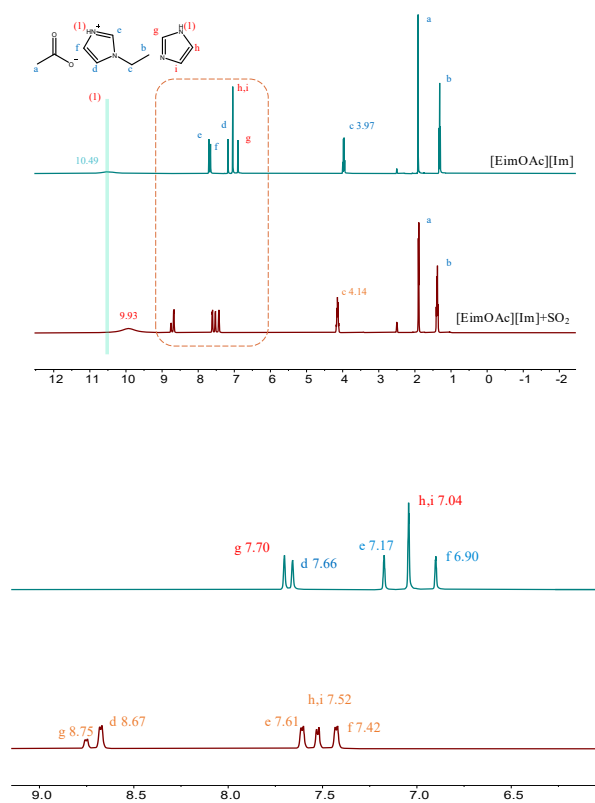


Fig. S16 ¹H NMR spectrum of [EimOAc][Im] before and after capturing SO₂ with an internal reference in (CD₃)₂SO.

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