Supporting Information for "Theory-Augmented Informatics of Ionic Liquid Electrolytes for Co-Design with Nanoporous Electrode Materials"

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S1 Computational Methods

Room temperature ionic liquids were modeled using planewave density functional theory (DFT) as implemented in the PWscf code in the open-source Quantum ESPRESSO software suite.[1–3] The ionic cores were represented with scalar-relativistic pseudopotentials sourced from the SG15 Optimized Norm-Conserving Vanderbilt (ONCV) pseudopotential library.[4–6] All calculations were performed with a kinetic energy cutoff of 80 Ry and a charge density cutoff of 320 Ry. Periodic interactions were corrected for using the reciprocal space Martyna-Tuckerman correction in a cubic cell with an edge length that was set to be 4-5× the largest interatomic distance in the molecular ion. All calculations were spin-polarized and performed with a single k-point centered at Γ in the Brillouin zone. The electronic interactions were described using the hybrid PBE0 exchange-correlation functional to mitigate possible self-interaction errors in the DFT calculations.[7] The structure of each molecular ion was optimized in vacuum prior to performing Δ-SCF[8] calculations on the fixed optimized geometries to obtain the HOMO and LUMO levels of each ion. Within the Δ-SCF procedure, the HOMO level is obtained as the negative of the ionization potential

$$\epsilon_{\mathrm{HOMO}} = -IP = -(E_{N-1} - E_N), \tag{S1}$$

and the LUMO level is obtained as the negative of the electron affinity

$$\epsilon_{\mathrm{LUMO}} = -EA = -(E_N - E_{N+1}), \tag{S2}$$

where E_N is the DFT total energy of a system containing N electrons. The frontier orbital energy levels for each cation and anion considered in this work are presented below in Table S3 and Table S4.

The ESW values were computed from DFT as the difference between the anodic limit V_{AL} and cathodic limit V_{CL} of the RTIL that correspond to the onset of oxidation and reduction, respectively. These potential limits are approximated by the frontier orbitals of isolated RTIL ions in vacuum, following the approach described in Ref .[9]:

$$V_{\mathrm{AL}} = \min\left(\left\{\frac{-\varepsilon^i_{\mathrm{HOMO}}}{e}\right\}\right) \tag{S3}$$

$$V_{\mathrm{CL}} = \max\left(\left\{\frac{-\varepsilon^i_{\mathrm{LUMO}}}{e}\right\}\right) \tag{S4}$$

$$\Delta V_{ESW} = V_{\mathrm{AL}} - V_{\mathrm{CL}} \tag{S5}$$

where i denotes a cation or anion, and ϵ_{HOMO} and ϵ_{LUMO} denote the highest occupied and lowest unoccupied molecular orbital energy levels, and e is the unsigned elementary charge. Within this approximation, the ESW is defined as the energy difference between the cation LUMO and anion HOMO levels. We note that while the vacuum-based approximation employed herein is sufficient for recovering experimental trends in the ESW of RTILs, such results are only semi-quantitatively accurate.[9] While improved quantitative accuracy

may be obtained by directly simulating the bulk RTIL environment *via* first principles molecular dynamics, the high computational cost of such simulations severely limits the number of RTIL compositions that could be analysed for screening purposes.

S2 Molecular dataset

In what follows we include the following sets of tables and figures describing the molecular dataset:

- Table S1: SMILES strings for the select set of cations considered in this work.
- Table S2: SMILES strings for the select set of anions considered in this work.
- Figure S1: 2D molecular depictions of the select set of cations considered in this work.
- Figure S2: 2D molecular depictions of the select set of anions considered in this work.
- Figure S3: 3D rendered depictions of the down-selected set of cations appearing in Fig. 3.
- Figure S4: 3D rendered depictions of the down-selected set of anions appearing in Fig. 3.
- Table S3: Frontier orbital energy levels of the select set of cations considered in this work.
- Table S4: Frontier orbital energy levels of the select set of anions considered in this work.
- Figure S5: Theoretical electrochemical stability windows computed at three different levels of theory for a select set of RTIL compositions.
- Figure S6: Theoretical electrochemical stability windows computed from the frontier orbital energy levels of isolated cations and anions in vacuum.
- Table S5: Data presented in Fig. 2a describing RTIL performance at the flat interface limit.
- Table S6: Data presented in Fig. 2b describing RTIL performance confined in nanopores with a mean pore size of 10.0 Å
- Table S7: Data presented in Fig. 2c describing RTIL performance confined in nanopores with a mean pore size of 7.5 Å

Table S1: SMILES strings for the select set of cations considered in this work.

Index	**Cation**	**SMILES**
1	(3-aminopropyl)tributylphosphonium	CCCC[P+](CCCC)(CCCC)CCCN
2	(3-carboxypropyl)trimethylammonium	C[N+](C)(C)CCCC(=O)O
3	(buten-1-yl)triethylphosphonium	CC/C=C/[P+](CC)(CC)CC
4	(ethoxymethyl)triethylphosphonium	CCOC[P+](CC)(CC)CC
5	1,1,3,3-tetramethylguanidinium	[H]/N=C(/N(C)C)[NH+](C)C
6	1,1-dimethylpyrrolidinium	C[N+]1(C)CCCC1
7	1,2-diethylpyridinium	CCc1cccc[n+]1CC
8	1,2-dimethyl-3-propylimidazolium	CCCn1cc[n+](C)c1C
9	1,3-diethylimidazolium	CCn1cc[n+](CC)c1
10	1,3-dimethylimidazolium	Cn1cc[n+](C)c1
11	1,3-dimethylpyridinium	Cc1ccc[n+](C)c1
12	1-(2,3-dihydroxypropyl)-1-methylpyrrolidin-1-ium	C[N+]1(C[C@H](O)CO)CCCC1
13	1-(2-(2-methoxyethoxy)ethyl)-3-methylimidazolium	COCCOCC[n+]1ccn(C)c1
14	1-(2-azido-3-bromopropyl)-3-methyl-3H-imidazol-1-ium	Cn1cc[n+](C[C@H](CBr)[N-][N+]#N)c1
15	1-(2-chloroethyl)-3-methylimidazolium	Cn1cc[n+](CCCl)c1
16	1-(2-cyanoethyl)-3-(2-hydroxyethyl)-1H-imidazolium	N#CCCn1cc[n+](CCO)c1
17	1-(2-cyanoethyl)-3-hexyl-1H-imidazol-3-ium	CCCCCCn1cc[n+](CCC#N)c1
18	1-(2-cyanoethyl)imidazolium	N#CCC[n+]1cc[nH]c1
19	1-(2-ethoxy-2-oxoethyl)-pyridinium	CCOC(=O)C[n+]1ccccc1
20	1-(2-ethoxyethyl)-1-methylpyrrolidinium	CCOCC[N+]1(C)CCCC1
21	1-(2-hydroxyethyl)-1-methylpyrrolidin-1-ium	C[N+]1(CCO)CCCC1
22	1-(2-hydroxyethyl)-3-methylimidazolium	Cn1cc[n+](CCO)c1
23	1-(2-hydroxyethyl)-pyridinium	OCC[n+]1ccccc1
24	1-(2-methoxyethyl)-1-methylpiperidinium	COCC[N+]1(C)CCCCC1
25	1-(2-methoxyethyl)-1-methylpyrrolidinium	COCC[N+]1(C)CCCC1
26	1-(2-methoxyethyl)-3-methyl-imidazolium	COCC[n+]1ccn(C)c1
27	1-(3-cyanopropyl)-3-methylimidazolium	C[n+]1ccn(CCCC#N)c1
28	1-(3-hydroxypropyl)pyridinium	OCCC[n+]1ccccc1
29	1-(3-methylbutyl)-3-methylimidazolium	CC(C)CC[n+]1ccn(C)c1
30	1-butyl-3-methylimidazolium	CCCCN1C=C[N+](=C1)C
31	1-ethyl-3-methylimidazolium	CCN1C=C[N+](=C1)C
32	tetrabutylphosphonium	CCCC[P+](CCCC)(CCCC)CCCC
33	ethylammonium	CC[NH3+]

Table S2: SMILES strings for the select set of anions considered in this work.

Index	Anion	SMILES
1	(S)-2-amino-4-carboxybutanoate	N[C@@H](CCC(=O)O)C(=O)[O-]
2	1,1,2,2,2-pentafluoro-N-[(pentafluoroethyl)sulfonyl]ethanesulfonamide	O=S(=O)(N=[S@](=O)([O-])C(F)(F)C(F)(F)F)C(F)(F)C(F)(F)F
3	1,1,2,2-tetrafluoroethanesulfonate	O=S(=O)([O-])C(F)(F)C(F)F
4	1,1,2-trifluoro-2-(perfluoroethoxy)ethanesulfonate	O=S(=O)([O-])C(F)(F)[C@@H](F)OC(F)(F)C(F)(F)F
5	1,1,2-trifluoro-2-(trifluoromethoxy)ethanesulfonate	O=S(=O)([O-])C(F)(F)[C@H](F)OC(F)(F)F
6	1,2,3-triazolide	c1[cH-]nnn1
7	1,2,4-triazolide	c1n[cH-]nn1
8	1-butanesulfonate	CCCCS(=O)(=O)[O-]
9	2-(bis(2-hydroxyethyl)amino)ethanesulfonate	O=S(=O)([O-])CCN(CCO)CCO
10	2-aminoethanesulfonate	NCCS(=O)(=O)[O-]
11	2-cyanopyrrolide	N#Cc1[cH-]ccn1
12	2-hydroxy-3-morpholinopropanesulfonate	O=S(=O)([O-])C[C@@H](O)CN1CCOCC1
13	3-sulfobenzoate	O=C([O-])c1cccc(S(=O)(=O)O)c1
14	4,5-dichloroimidazolide	Clc1n[cH-]nc1Cl
15	4,5-dicyanoimidazolide	N#Cc1n[cH-]nc1C#N
16	4-nitroimidazolide	[O-]N([O-])C1=NC=[N+]=C1
17	L-alaninate	C[C@H](N)C(=O)[O-]
18	L-asparaginate	C(C(C(=O)[O-])N)C(=O)N
19	L-cysteinate	N[C@@H](CS)C(=O)[O-]
20	L-leucinate	CC(C)C[C@@H](N)C(=O)[O-]
21	L-lysinate	NCCCC[C@@H](N)C(=O)[O-]
22	L-prolinate	O=C([O-])[C@H]1CCCN1
23	L-serinate	N[C@@H](CO)C(=O)[O-]
24	L-threoninate	C[C@@H](O)[C@@H](N)C(=O)[O-]
25	L-valinate	CC(C)[C@@H](N)C(=O)[O-]
26	acetate	CC(=O)[O-]
27	azide	N#[N+][N-2]
28	benzenesulfonate	Cc1ccc(S(=O)(=O)[O-])cc1
29	benzoate	O=C([O-])c1ccccc1
30	bicarbonate	O=C([O-])O
31	bis(fluorosulfonyl)amide	[N-](S(=O)(=O)F)S(=O)(=O)F
32	bis(trifluoromethanesulfonyl)imide	O=S(=O)(N=[S@](=O)([O-])C(F)(F)F)C(F)(F)F
33	bisulfate	O=S(=O)([O-])O
34	bromide	[Br-]

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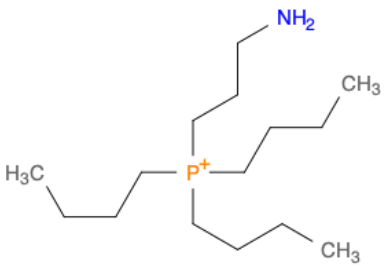
Table S2 – *Continued from previous page*

Index	Anion	SMILES
35	butanoate	CCCC(=O)[O-]
36	butylphosphonate	CCCCP(=O)(O)[O-]
37	butylsulfate	CCCCOS(=O)(=O)[O-]
38	chloride	[Cl-]
39	decanoate	CCCCCCCCCC(=O)[O-]
40	dicyanamide	N#C[N-]C#N
41	diethylphosphate	CCOP(=O)([O-])OCC
42	dihydrogenphosphate	O=P([O-])(O)O
43	dimethylphosphate	COP(=O)([O-])OC
44	ethylphosphonate	CCP(=O)(O)[O-]
45	ethylsulfate	CCOS(=O)(=O)[O-]
46	formate	O=C[O-]
47	glycinate	NCC(=O)[O-]
48	glycolate	O=C([O-])CO
49	heptachlorodialuminate	Cl[Al-](Cl)(Cl)[Cl+][Al-](Cl)(Cl)Cl
50	hexafluorophosphate	F[P-](F)(F)(F)(F)F
51	hexanoate	CCCCCC(=O)[O-]
52	imidazolide	C1=CN=C[N-]1
53	isobutyrate	CC(C)C(=O)[O-]
54	lactate	C[C@@H](O)C(=O)[O-]
55	methanesulfonate	CS(=O)(=O)[O-]
56	methylphosphonate	CP(=O)([O-])[O-]
57	methylsulfate	COS(=O)(=O)[O-]
58	nitrate	O=[N+]([O-])[O-]
59	octanoate	CCCCCCCC(=O)[O-]
60	octylphosphonate	CCCCCCCCP(=O)([O-])[O]
61	pentanoate	CCCCC(=O)[O-]
62	perchlorate	O=Cl(=O)(=O)[O-]
63	perfluorobutanesulfonate	O=S(=O)([O-])C(F)(F)C(F)(F)C(F)(F)C(F)(F)F
64	perfluoropentanoate	O=C([O-])C(F)(F)C(F)(F)C(F)(F)C(F)(F)F
65	propanoate	CCC(=O)[O-]
66	salicylate	O=C([O-])c1ccccc1O
67	taurate	NCCS(=O)(=O)[O-]
68	tetrachloroaluminate	[Al-](Cl)(Cl)(Cl)Cl
69	tetracyanoborate	N#C[B-](C#N)(C#N)C#N

Continued on next page

Table S2 – *Continued from previous page*

Index	**Anion**	**SMILES**
70	tetrafluoroborate	F[B-](F)(F)F
71	tetrazolide	[cH-]1nnnn1
72	thiocyanate	N#C[S-]
73	tosylate	Cc1ccc(S(=O)(=O)[O-])cc1
74	trifluoro(perfluoroethyl)borate	F[B-](F)(F)C(F)(F)C(F)(F)F
75	trifluoro(perfluoropropyl)borate	F[B-](F)(F)C(F)(F)C(F)(F)C(F)(F)F
76	trifluoroacetate	O=C([O-])C(F)(F)F
77	trifluoromethanesulfonate	O=S(=O)([O-])C(F)(F)F



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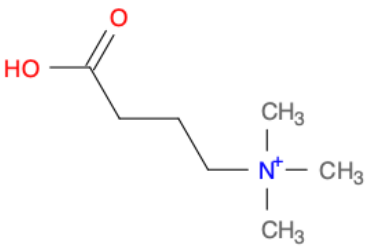

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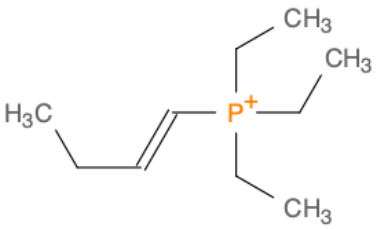

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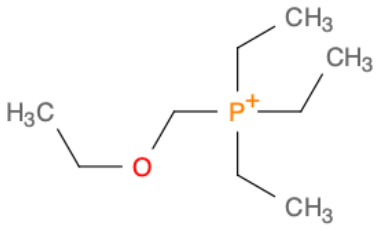

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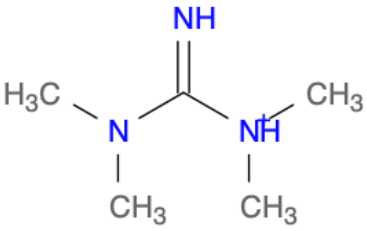

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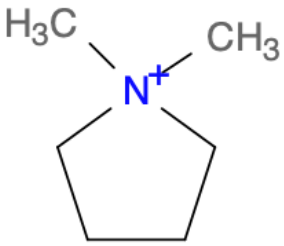

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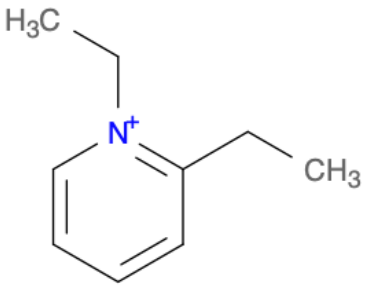

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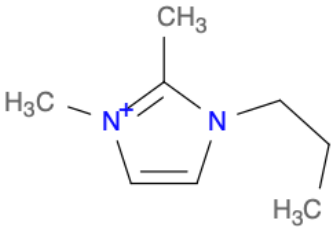

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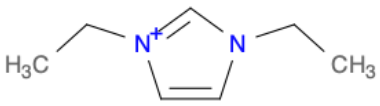

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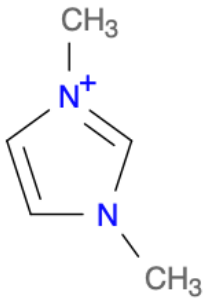
Figure S1: 2D molecular depictions of the select set of cations considered in this work.



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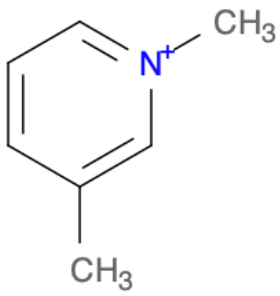

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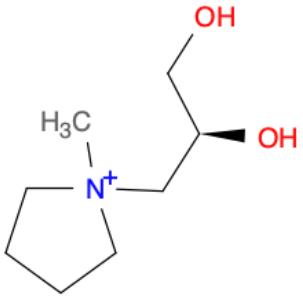

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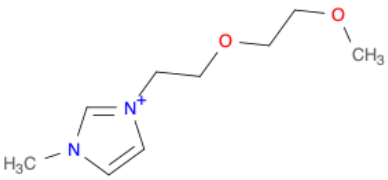

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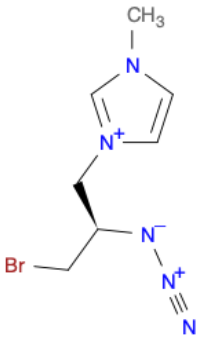

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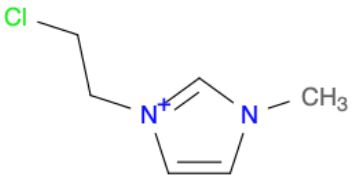

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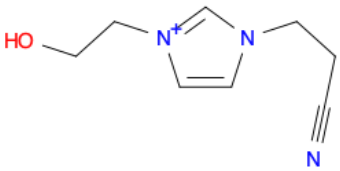

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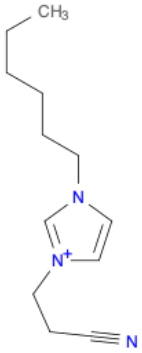

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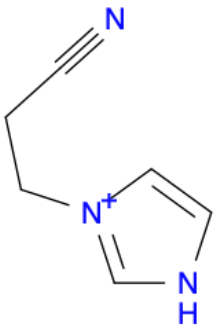

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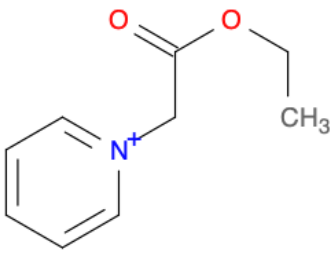
Figure S1: 2D molecular depictions of the select set of cations considered in this work.



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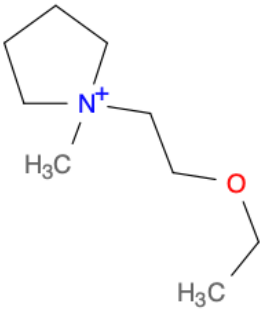

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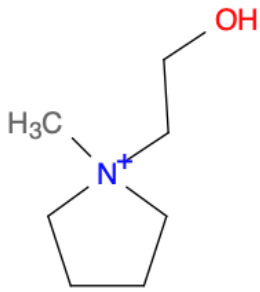

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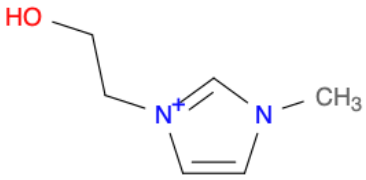

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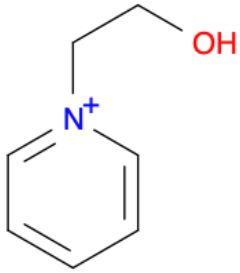

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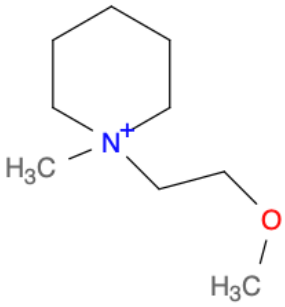

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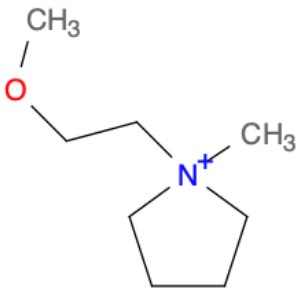

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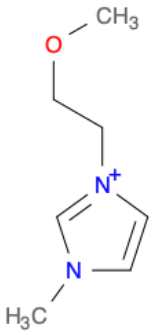

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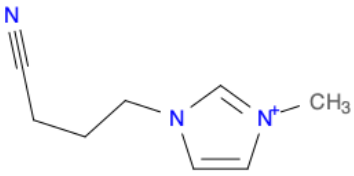
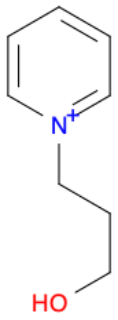


Cation 27

Figure S1: 2D molecular depictions of the select set of cations considered in this work.



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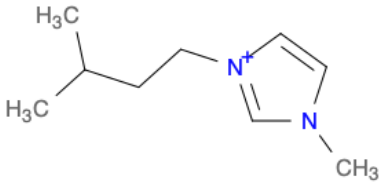

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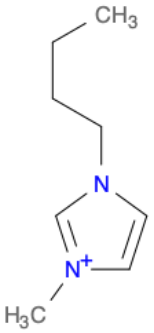

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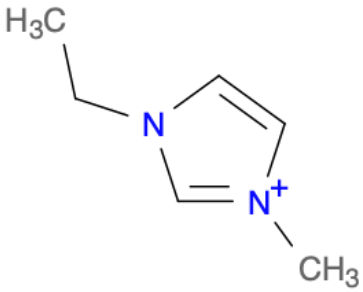
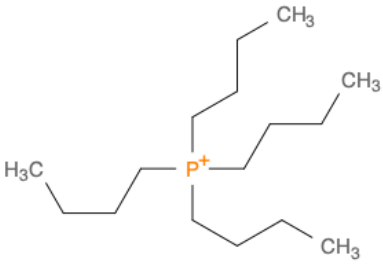
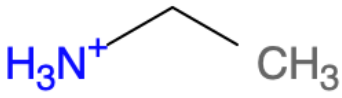


Cation 31

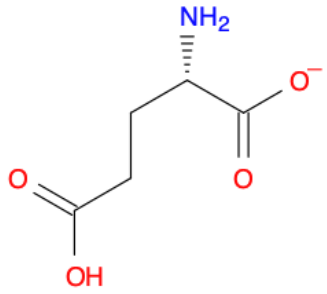


Cation 32



Cation 33

Figure S1: 2D molecular depictions of the select set of cations considered in this work.



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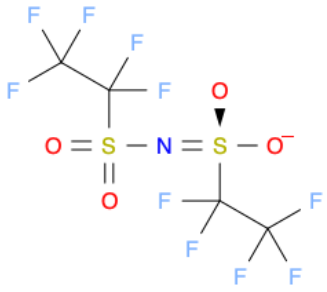

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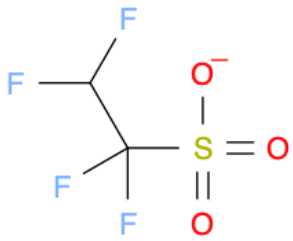

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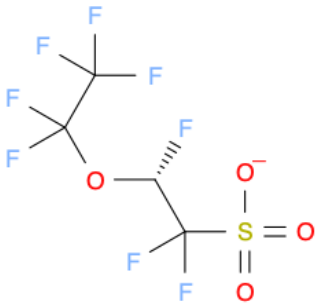

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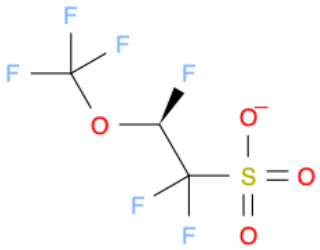

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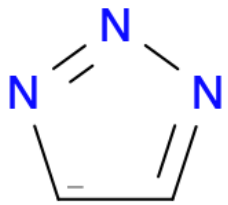

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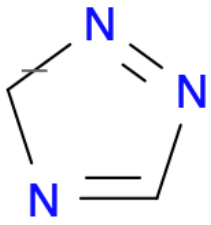

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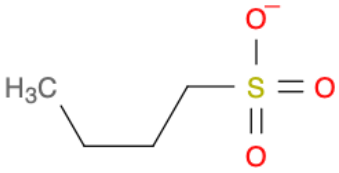

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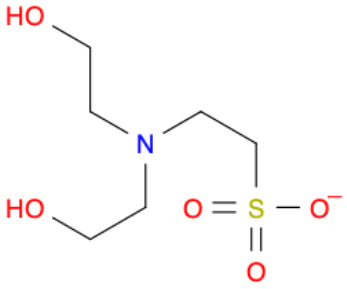
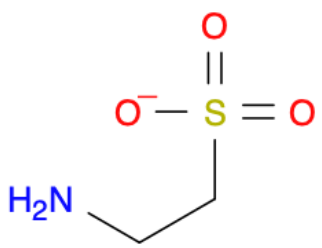


Anion 9

Figure S2: 2D molecular depictions of the select set of anions considered in this work.



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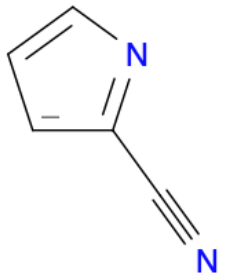

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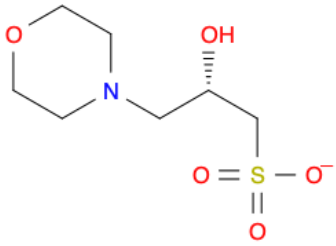

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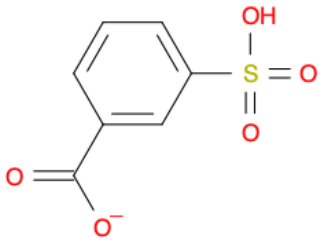

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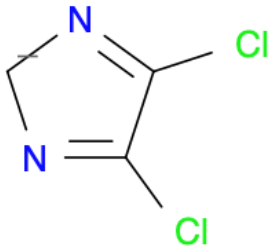

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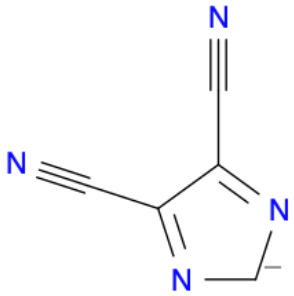

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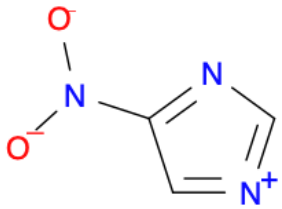

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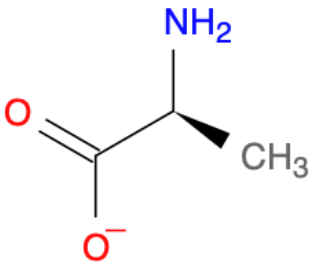

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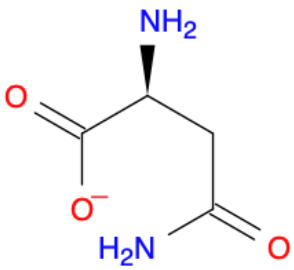
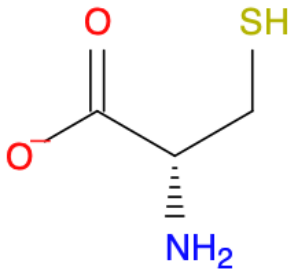


Anion 18

Figure S2: 2D molecular depictions of the select set of anions considered in this work.



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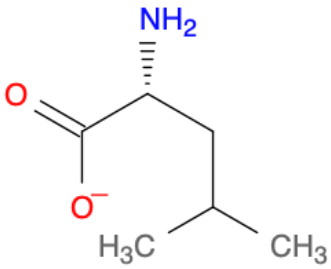

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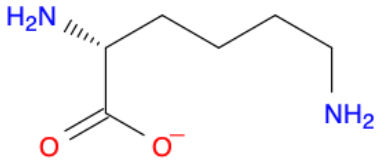

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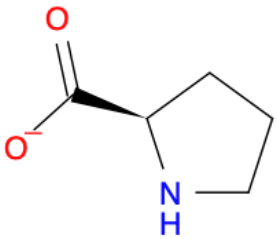

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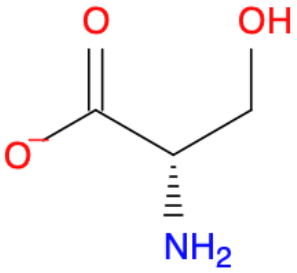

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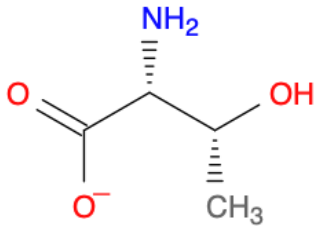

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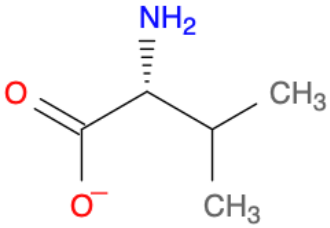

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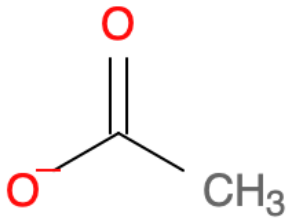
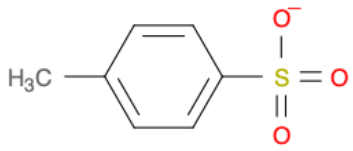


Anion 26

Anion 27

Figure S2: 2D molecular depictions of the select set of anions considered in this work.



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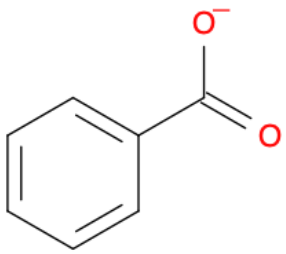

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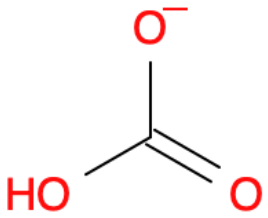

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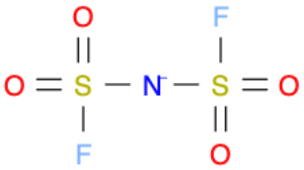

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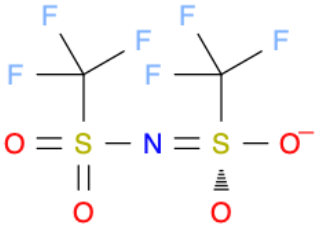

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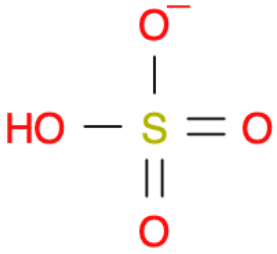
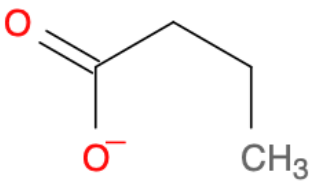


Anion 33

Anion 34



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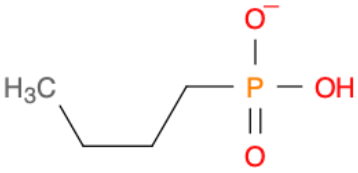
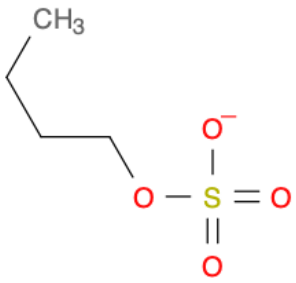


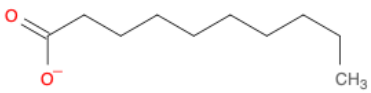
Anion 36

Figure S2: 2D molecular depictions of the select set of anions considered in this work.



Anion 37

Anion 38



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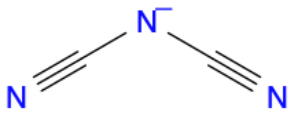

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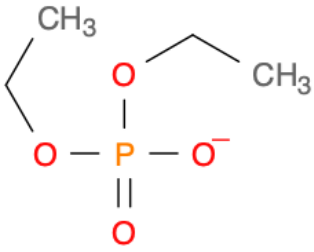

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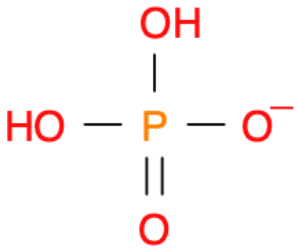

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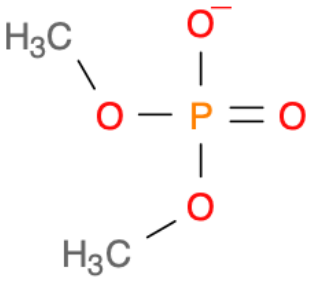

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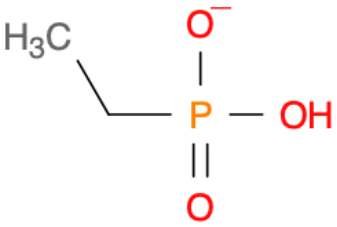

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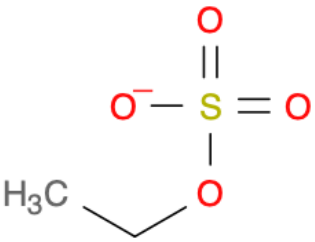
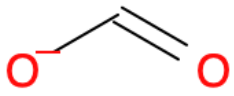


Anion 45

Figure S2: 2D molecular depictions of the select set of anions considered in this work.



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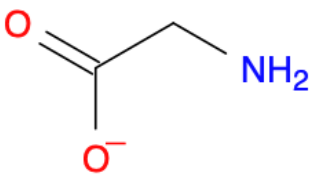

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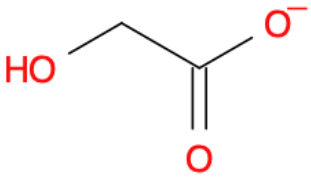

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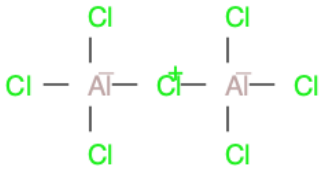

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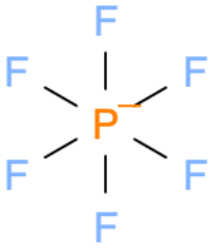

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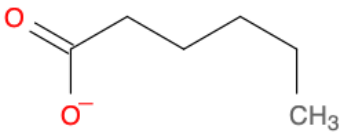
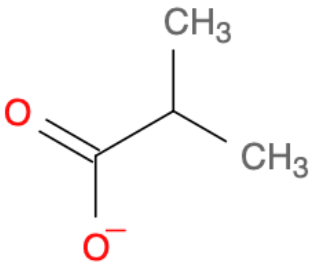


Anion 51

Anion 52



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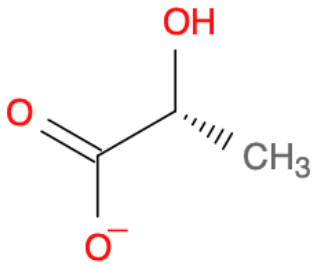
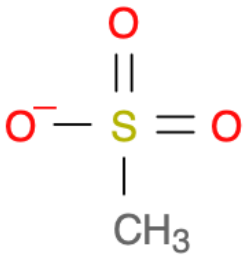


Anion 54

Figure S2: 2D molecular depictions of the select set of anions considered in this work.



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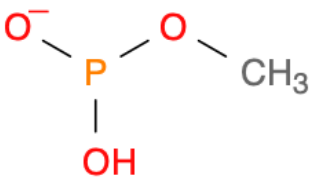
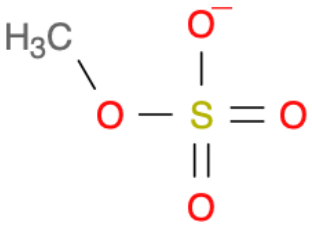


Anion 56



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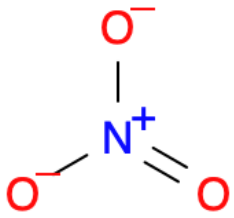

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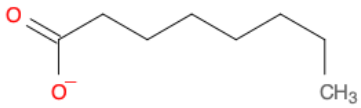

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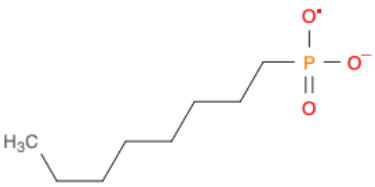

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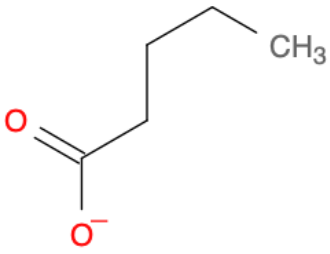

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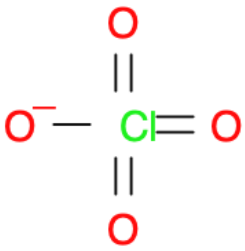

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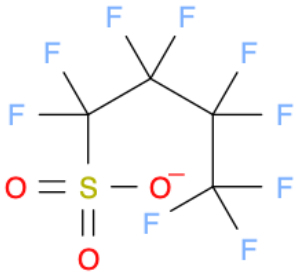
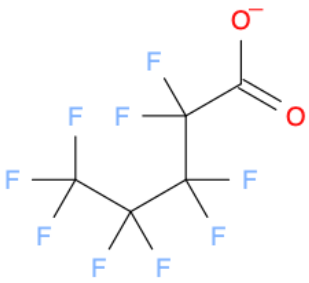


Anion 63

Figure S2: 2D molecular depictions of the select set of anions considered in this work.



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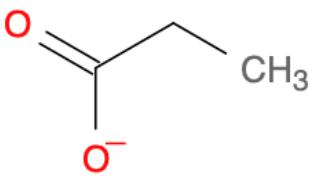

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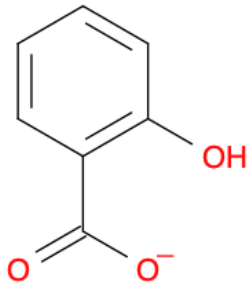

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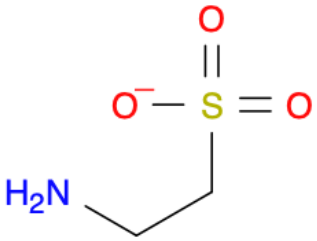

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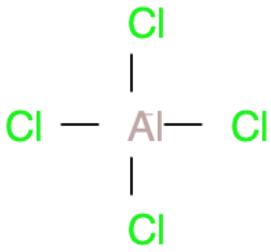

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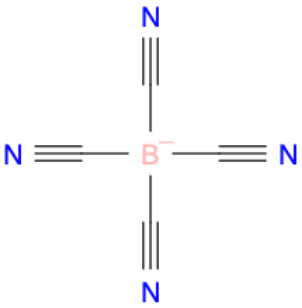

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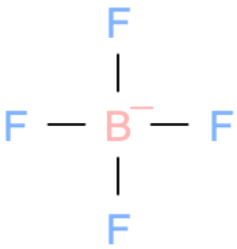

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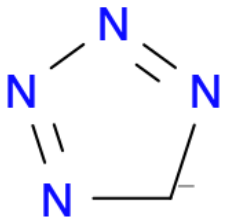

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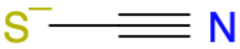
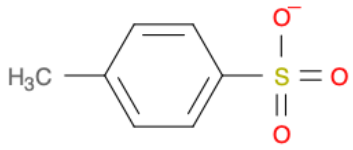


Anion 72

Figure S2: 2D molecular depictions of the select set of anions considered in this work.



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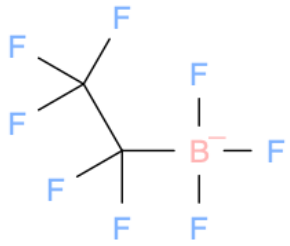

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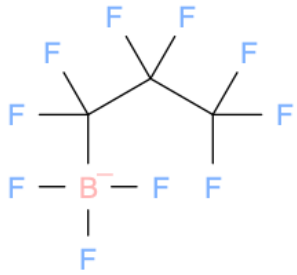

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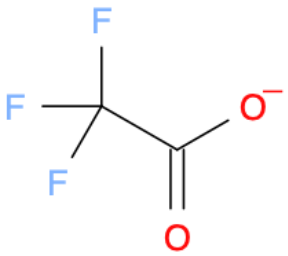

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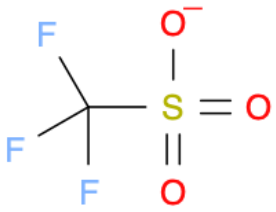
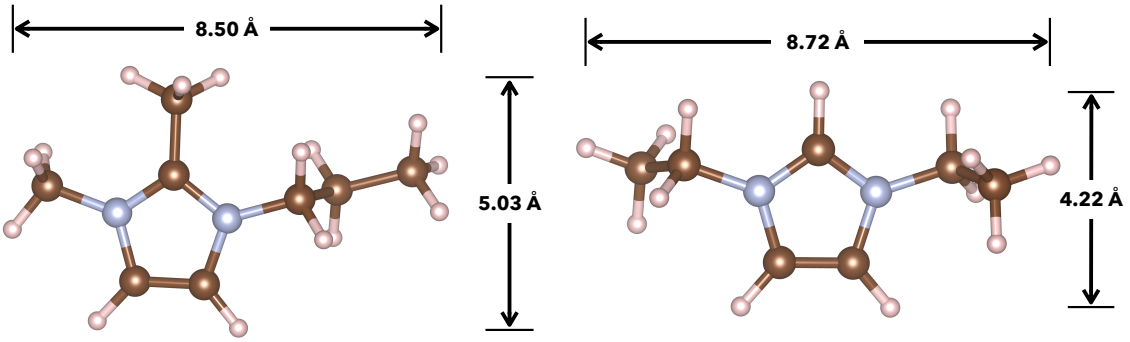
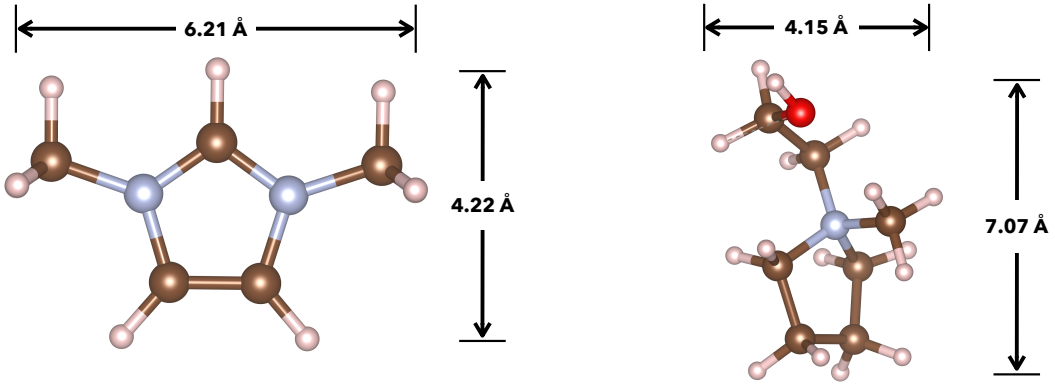


Anion 77

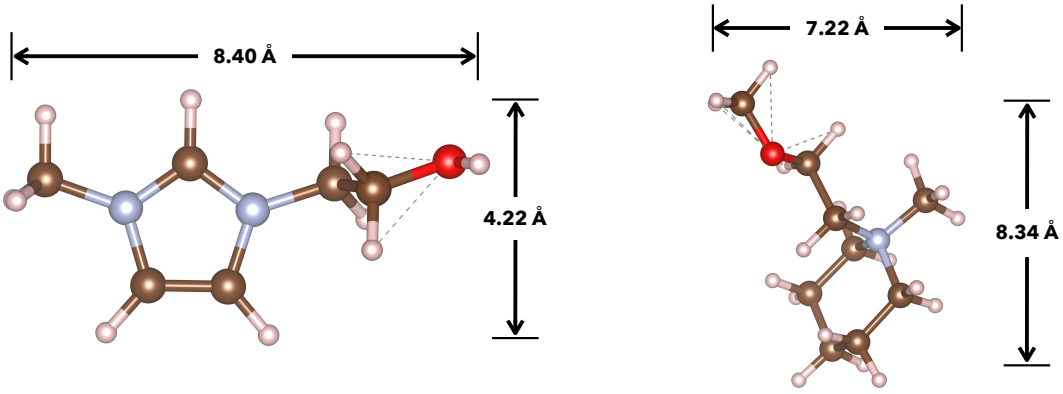
Figure S2: 2D molecular depictions of the select set of anions considered in this work.



Cation 8 Cation 9

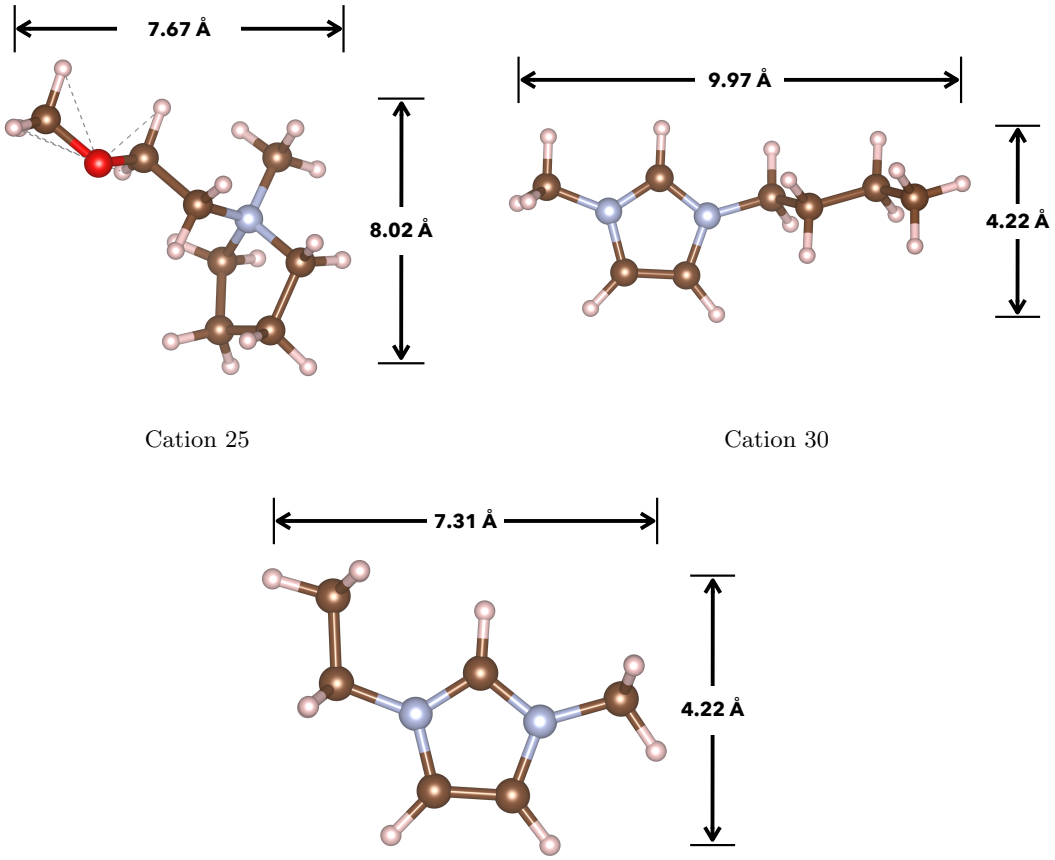


Cation 10 Cation 21



Cation 22 Cation 24

Figure S3: 3D rendered depictions of the down-selected set of cations appearing in Fig. 3.

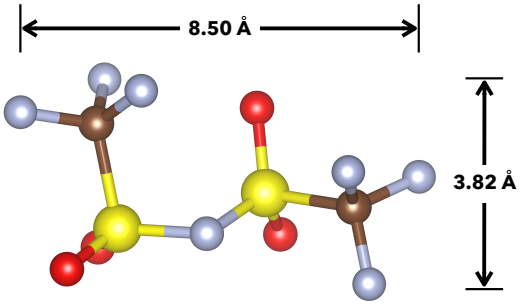


Cation 25

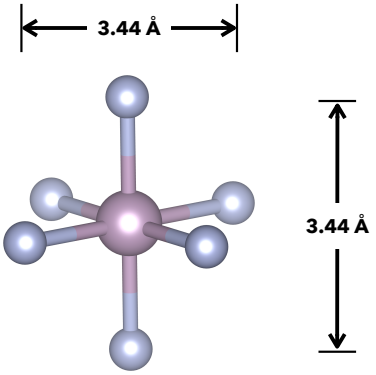
Cation 30

Cation 31

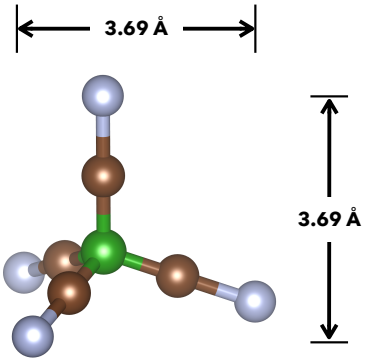
Figure S3: 3D rendered depictions of the down-selected set of cations appearing in Fig. 3.



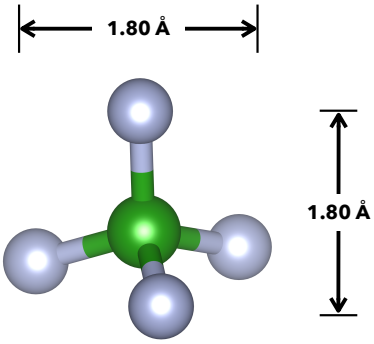
Cation 8



Cation 9



Cation 10



Cation 21

Figure S4: 3D rendered depictions of the down-selected set of anions appearing in Fig. 3.

Table S3: Frontier orbital energy levels of the select set of cations considered in this work.

Index	**Cation**	**HOMO level [eV]**	**LUMO level [eV]**	**Gap [eV]**
1	(3-aminopropyl)tributylphosphonium	−11.77	−2.49	9.28
2	(3-carboxypropyl)trimethylammonium	−13.52	−2.98	10.54
3	(buten-1-yl)triethylphosphonium	−13.38	−3.12	10.26
4	(ethoxymethyl)triethylphosphonium	−13.43	−2.72	10.70
5	1,1,3,3-tetramethylguanidinium	−13.07	−3.34	9.72
6	1,1-dimethylpyrrolidinium	−16.06	−2.92	13.14
7	1,2-diethylpyridinium	−14.00	−4.46	9.54
8	1,2-dimethyl-3-propylimidazolium	−13.40	−2.95	10.44
9	1,3-diethylimidazolium	−13.96	−3.23	10.73
10	1,3-dimethylimidazolium	−14.34	−3.36	10.98
11	1,3-dimethylpyridinium	−14.39	−4.73	9.66
12	1-(2,3-dihydroxypropyl)-1-methylpyrrolidin-1-ium	−13.71	−2.72	10.99
13	1-(2-(2-methoxyethoxy)ethyl)-3-methylimidazolium	−11.71	−3.33	8.39
14	1-(2-azido-3-bromopropyl)-3-methyl-3H-imidazol-1-ium	−12.41	−3.52	8.89
15	1-(2-chloroethyl)-3-methylimidazolium	−13.74	−3.57	10.17
16	1-(2-cyanoethyl)-3-(2-hydroxyethyl)-1H-imidazolium	−13.58	−3.74	9.85
17	1-(2-cyanoethyl)-3-hexyl-1H-imidazol-3-ium	−13.07	−3.59	9.47
18	1-(2-cyanoethyl)imidazolium	−14.46	−3.91	10.56
19	1-(2-ethoxy-2-oxoethyl)-pyridinium	−13.90	−4.73	9.17
20	1-(2-ethoxyethyl)-1-methylpyrrolidinium	−13.00	−2.73	10.27
21	1-(2-hydroxyethyl)-1-methylpyrrolidin-1-ium	−14.45	−2.80	11.65
22	1-(2-hydroxyethyl)-3-methylimidazolium	−13.59	−3.37	10.22
23	1-(2-hydroxyethyl)-pyridinium	−14.04	−4.84	9.20
24	1-(2-methoxyethyl)-1-methylpiperidinium	−13.26	−2.71	10.55
25	1-(2-methoxyethyl)-1-methylpyrrolidinium	−13.33	−2.81	10.53
26	1-(2-methoxyethyl)-3-methyl-imidazolium	−12.91	−3.33	9.58
27	1-(3-cyanopropyl)-3-methylimidazolium	−13.75	−3.58	10.17
28	1-(3-hydroxypropyl)pyridinium	−13.46	−4.74	8.72
29	1-(3-methylbutyl)-3-methylimidazolium	−13.70	−3.21	10.48
30	1-butyl-3-methylimidazolium	−13.66	−3.25	10.42
31	1-ethyl-3-methylimidazolium	−14.14	−3.30	10.84
32	tetrabutylphosphonium	−13.25	−1.70	11.55
33	ethylammonium	−17.97	−4.05	13.92

Table S4: Frontier orbital energy levels of the select set of anions considered in this work.

Index	**Anion**	**HOMO level [eV]**	**LUMO level [eV]**	**Gap [eV]**
1	(S)-2-amino-4-carboxybutanoate	−3.72	2.41	6.13
2	1,1,2,2,2-pentafluoro-N-[(pentafluoroethyl)sulfonyl]ethanesulfonamide	−6.30	2.89	9.19
3	1,1,2,2-tetrafluoroethanesulfonate	−5.43	3.14	8.57
4	1,1,2-trifluoro-2-(perfluoroethoxy)ethanesulfonate	−5.58	2.69	8.27
5	1,1,2-trifluoro-2-(trifluoromethoxy)ethanesulfonate	−5.55	2.92	8.47
6	1,2,3-triazolide	−3.74	3.21	6.95
7	1,2,4-triazolide	−4.53	3.26	7.78
8	1-butanesulfonate	−5.09	2.31	7.40
9	2-(bis(2-hydroxyethyl)amino)ethanesulfonate	−4.79	2.22	7.02
10	2-aminoethanesulfonate	−4.44	2.54	6.98
11	2-cyanopyrrolide	−3.24	2.90	6.13
12	2-hydroxy-3-morpholinopropanesulfonate	−4.75	2.43	7.19
13	3-sulfobenzoate	−4.63	2.41	7.04
14	4,5-dichloroimidazolide	−4.87	3.18	8.05
15	4,5-dicyanoimidazolide	−5.71	2.88	8.60
16	4-nitroimidazolide	−4.17	2.94	7.11
17	L-alaninate	−3.50	2.98	6.49
18	L-asparaginate	−4.31	2.79	7.10
19	L-cysteinate	−3.78	2.98	6.76
20	L-leucinate	−3.53	2.42	5.95
21	L-lysinate	−3.53	2.03	5.57
22	L-prolinate	−3.36	2.66	6.02
23	L-serinate	−3.86	2.95	6.82
24	L-threoninate	−4.05	2.77	6.83
25	L-valinate	−3.54	2.61	6.16
26	acetate	−3.63	3.31	6.95
27	azide	−2.69	6.55	9.24
28	benzenesulfonate	−4.77	2.23	7.00
29	benzoate	−4.03	2.51	6.53
30	bicarbonate	−4.07	3.55	7.62
31	bis(fluorosulfonyl)amide	−6.52	4.04	10.56
32	bis(trifluoromethanesulfonyl)imide	−6.13	3.33	9.46
33	bisulfate	−4.93	3.44	8.37
34	bromide	−3.55	6.78	10.33

Continued on next page

Table S4 – *Continued from previous page*

Index	Anion	HOMO level [eV]	LUMO level [eV]	Gap [eV]
35	butanoate	−3.59	2.61	6.20
36	butylphosphonate	−4.65	2.19	6.84
37	butylsulfate	−4.82	2.17	6.99
38	chloride	−3.68	8.23	11.91
39	decanoate	−3.66	1.67	5.33
40	dicyanamide	−4.07	4.18	8.25
41	diethylphosphate	−4.63	2.41	7.04
42	dihydrogenphosphate	−4.82	3.25	8.07
43	dimethylphosphate	−4.65	2.74	7.39
44	ethylphosphonate	−4.63	2.67	7.31
45	ethylsulfate	−4.81	2.67	7.48
46	formate	−3.73	3.67	7.40
47	glycinate	−3.51	3.21	6.72
48	glycolate	−4.11	3.37	7.48
49	heptachlorodialuminate	−6.99	2.24	9.23
50	hexafluorophosphate	−7.85	4.60	12.45
51	hexanoate	−3.69	2.21	5.89
52	imidazolide	−3.94	3.17	7.11
53	isobutyrate	−3.65	2.89	6.54
54	lactate	−4.06	3.06	7.12
55	methanesulfonate	−4.39	3.17	7.57
56	methylphosphonate	−4.64	2.94	7.59
57	methylsulfate	−4.83	2.96	7.79
58	nitrate	−4.03	5.46	9.49
59	octanoate	−3.59	1.90	5.49
60	octylphosphonate	−4.65	1.66	6.31
61	pentanoate	−3.66	2.38	6.05
62	perchlorate	−5.49	5.36	10.85
63	perfluorobutanesulfonate	−5.54	2.93	8.47
64	perfluoropentanoate	−4.94	3.03	7.97
65	propanoate	−3.58	2.96	6.54
66	salicylate	−4.18	2.64	6.82
67	taurate	−4.44	2.54	6.98
68	tetrachloroaluminate	−6.17	3.08	9.24
69	tetracyanoborate	−7.74	3.01	10.75

Continued on next page

Table S4 – *Continued from previous page*

Index	**Anion**	**HOMO level [eV]**	**LUMO level [eV]**	**Gap [eV]**
70	tetrafluoroborate	−7.31	4.11	11.42
71	tetrazolide	−4.76	3.30	8.06
72	thiocyanate	−3.50	4.29	7.79
73	tosylate	−4.79	2.40	7.20
74	trifluoro(perfluoroethyl)borate	−6.43	3.59	10.02
75	trifluoro(perfluoropropyl)borate	−6.36	3.14	9.49
76	trifluoroacetate	−4.86	4.11	8.96
77	trifluoromethanesulfonate	−6.26	4.06	10.32

S3 Theoretical Electrochemical Stability Windows

S3.1 Trend validation across different levels of theory

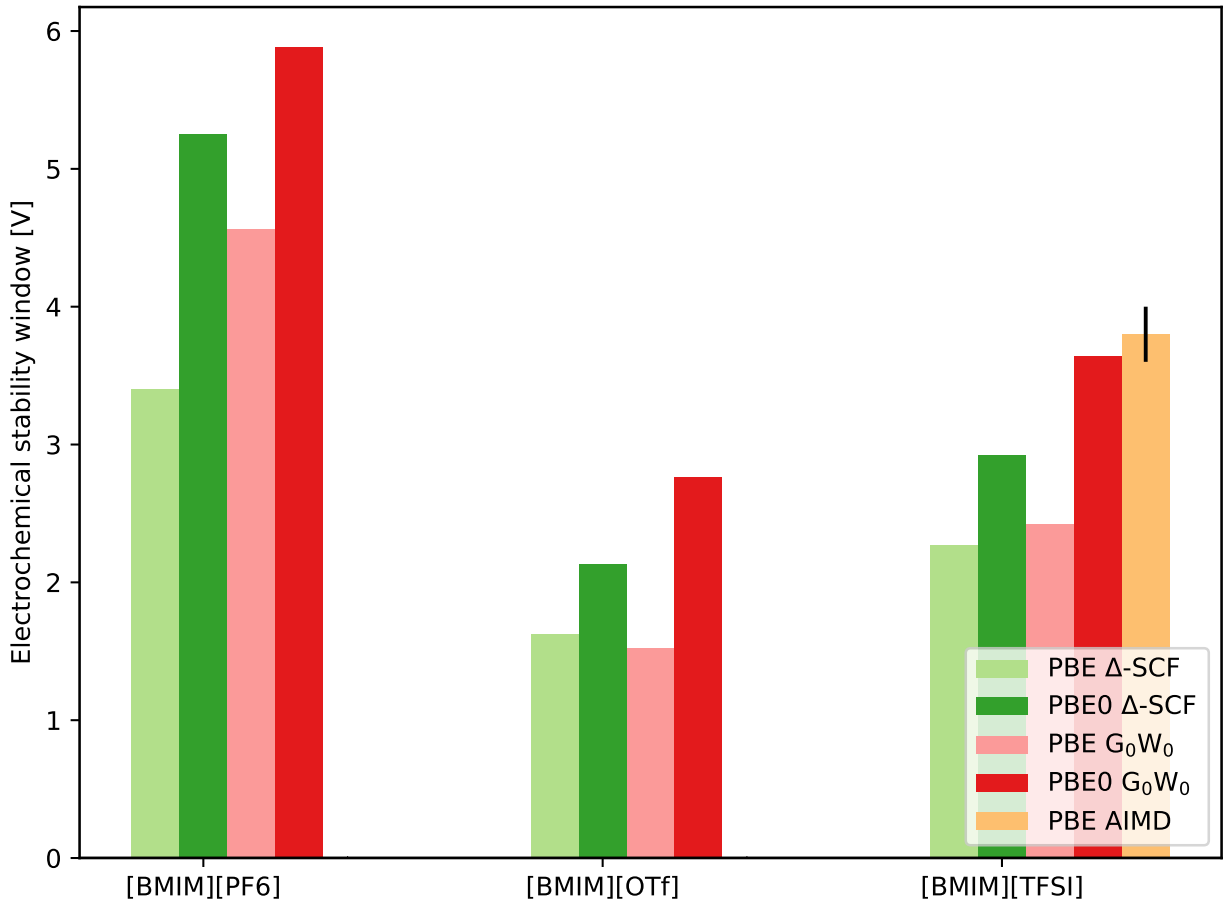


Figure S5: Theoretical electrochemical stability windows computed at three different levels of theory for a select set of RTIL compositions.

S3.2 Composition sampling

RTIL compositions were generated by considering cation-anion pairs from the set of ions listed in Table S1 and Table S2, and their associated ESWs were estimated using Eq. S5. In situations where the anion HOMO level was higher in energy than the cation LUMO level, we anticipate spontaneous charge transfer occurring between the cation and anion and assess the theoretical RTIL composition as being unstable. These results are presented below in Fig. S6. According to our analysis, pyridinium- and thiozolium-based RTILs are largely predicted to be unstable and undergo spontaneous charge transfer reactions. Imidizaolium-, pyrrolidinium-, phosphonium-, ammonium-, and sulphonium-based ionic liquids are generally predicted to be stable. Furthermore, consistent with the skewed anion HOMO level histogram and ESW histogram reported in Fig. 2a and Fig. 2b, respectively, we observe that few RTILs in our generated composition space exhibit ESWs > 3 V, and that anion identity appears to be the main determining factor. Here we observe several bright bands in Fig. S6 that correspond to RTILs composed with PF_6^-, $B(CN)_4^-$, and BF_4^-, all of which are commonly reported in the literature. As a final note, it is worthwhile to point out that our calculations exclude crucial environmental effects and may only be providing a semi-quantitative view of RTIL stability and trends in ESW. Enhanced quantitative accuracy can be achieved through more refined,

albeit computationally demanding techniques, such as the molecular dynamics approach proposed by Ong and co-workers.[9]

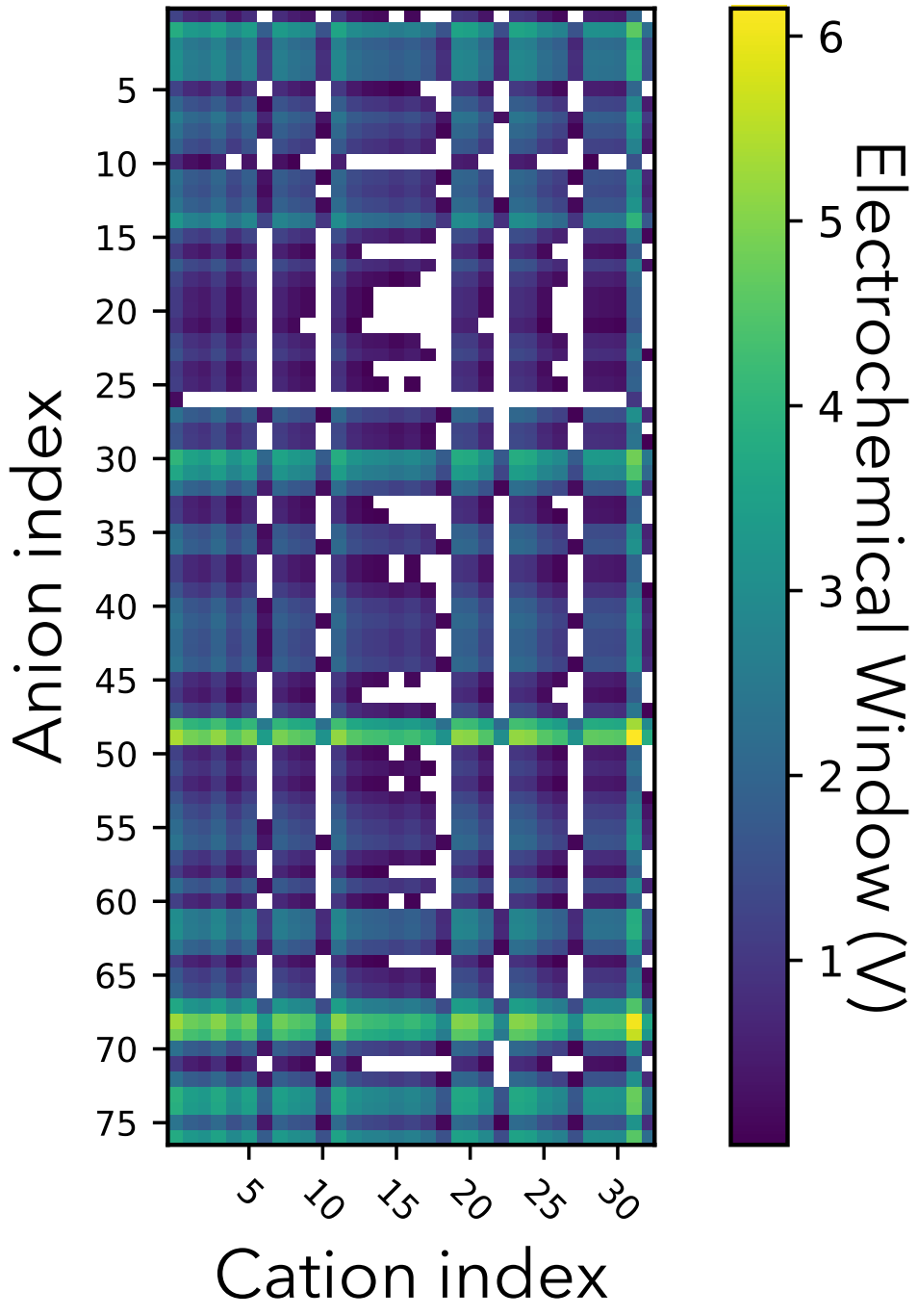


Figure S6: Theoretical electrochemical stability windows computed from the frontier orbital energy levels of isolated cations and anions in vacuum. Entries which appear white are compositions that have been predicted to be unstable.

S4 Pore Size-Dependent Figure of Merit

Table S5: Data presented in Fig. 2a describing RTIL performance at the flat interface limit.

Cation	**Anion**	λ_I [Å]	η [Pa·s]	ΔV_{ESW} [V]	G	$\Delta V_{ESW} \times G$ [V]	ζ
1-ethyl-3-methylimidazolium	methanesulfonate	7.46	1.60×10^{-1}	1.10	1.00	1.10	0.13
1-ethyl-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	7.46	3.44×10^{-2}	2.84	1.00	2.84	1.00
1-ethyl-3-methylimidazolium	tetrafluoroborate	7.46	3.88×10^{-2}	4.02	1.00	4.02	1.36
1-butyl-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	9.90	5.09×10^{-2}	2.89	1.00	2.89	0.88
1-butyl-3-methylimidazolium	trifluoroacetate	9.90	5.29×10^{-2}	1.61	1.00	1.61	0.48
1-butyl-3-methylimidazolium	tetrafluoroborate	9.90	1.08×10^{-1}	4.07	1.00	4.07	0.76
1-butyl-3-methylimidazolium	hexafluorophosphate	9.90	2.71×10^{-1}	4.61	1.00	4.61	0.21
1,3-dimethylimidazolium	bis(trifluoromethanesulfonyl)imide	7.39	3.81×10^{-2}	2.77	1.00	2.77	0.94
1-butyl-3-methylimidazolium	methylsulfate	9.90	2.13×10^{-1}	1.59	1.00	1.59	0.12
1-ethyl-3-methylimidazolium	methylsulfate	7.46	7.88×10^{-2}	1.54	1.00	1.54	0.37
1-ethyl-3-methylimidazolium	tetracyanoborate	7.46	1.80×10^{-2}	4.45	1.00	4.45	1.80
1,3-dimethylimidazolium	methylsulfate	6.23	7.17×10^{-2}	1.47	1.00	1.47	0.37
1-ethyl-3-methylimidazolium	ethylsulfate	7.46	9.78×10^{-2}	1.52	1.00	1.52	0.31
1-(2-methoxyethyl)-1-methylpyrrolidinium	bis(trifluoromethanesulfonyl)imide	8.03	5.51×10^{-2}	3.33	1.00	3.33	0.98
1-ethyl-3-methylimidazolium	bisulfate	7.46	1.63	1.63	1.00	1.63	0.00
1-ethyl-3-methylimidazolium	acetate	7.46	1.44×10^{-1}	0.34	1.00	0.34	0.05
1-ethyl-3-methylimidazolium	thiocyanate	7.46	2.45×10^{-2}	0.21	1.00	0.21	0.08
1-butyl-3-methylimidazolium	thiocyanate	9.90	5.65×10^{-2}	0.26	1.00	0.26	0.07
1-butyl-3-methylimidazolium	acetate	9.90	3.00×10^{-1}	0.39	1.00	0.39	0.01
1-butyl-3-methylimidazolium	dimethylphosphate	9.90	6.42×10^{-1}	1.40	1.00	1.40	0.00
1,2-dimethyl-3-propylimidazolium	bis(trifluoromethanesulfonyl)imide	8.46	9.28×10^{-2}	3.18	1.00	3.18	0.68
1,3-diethylimidazolium	bis(trifluoromethanesulfonyl)imide	8.71	3.08×10^{-2}	2.91	1.00	2.91	1.06
1-(2-hydroxyethyl)-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	8.38	8.69×10^{-2}	2.77	1.00	2.77	0.62
1-butyl-3-methylimidazolium	perchlorate	9.90	1.80×10^{-1}	2.24	1.00	2.24	0.22
1,2-diethylpyridinium	bis(trifluoromethanesulfonyl)imide	7.39	6.88×10^{-2}	1.67	1.00	1.67	0.44
tetrabutylphosphonium	2-(bis(2-hydroxyethyl)amino)ethanesulfonate	12.27	1.41×10^{1}	3.09	1.00	3.09	0.00
tetrabutylphosphonium	2-hydroxy-3-morpholinopropanesulfonate	12.27	7.31	3.05	1.00	3.05	0.00
1-ethyl-3-methylimidazolium	dimethylphosphate	7.46	2.70×10^{-1}	1.35	1.00	1.35	0.06
1-ethyl-3-methylimidazolium	L-alaninate	7.46	2.64×10^{-1}	0.21	1.00	0.21	0.01
1,3-dimethylimidazolium	acetate	6.23	8.70×10^{-2}	0.27	1.00	0.27	0.06
1-(2-hydroxyethyl)-1-methylpyrrolidin-1-ium	bis(trifluoromethanesulfonyl)imide	7.39	8.00×10^{-2}	3.34	1.00	3.34	0.79
1-(2,3-dihydroxypropyl)-1-methylpyrrolidin-1-ium	bis(trifluoromethanesulfonyl)imide	7.73	1.50	3.41	1.00	3.41	0.00
1-(2-hydroxyethyl)-pyridinium	bis(trifluoromethanesulfonyl)imide	7.91	1.09×10^{-1}	1.30	1.00	1.30	0.24
1-butyl-3-methylimidazolium	nitrate	9.90	1.65×10^{-1}	0.78	1.00	0.78	0.09
1-butyl-3-methylimidazolium	bicarbonate	9.90	7.37×10^{-2}	0.82	1.00	0.82	0.21
1-butyl-3-methylimidazolium	dihydrogenphosphate	9.90	1.26×10^{-1}	1.57	1.00	1.57	0.25
1-(2-hydroxyethyl)-3-methylimidazolium	tetrafluoroborate	8.38	1.15×10^{-1}	3.95	1.00	3.95	0.69
1,2-diethylpyridinium	ethylsulfate	7.11	3.25×10^{-1}	0.35	1.00	0.35	0.01
1,3-dimethylpyridinium	methylsulfate	6.35	1.29×10^{-1}	0.11	1.00	0.11	0.02
1-ethyl-3-methylimidazolium	diethylphosphate	7.46	4.10×10^{-1}	1.33	1.00	1.33	0.02
1-butyl-3-methylimidazolium	ethylsulfate	9.90	3.79×10^{-1}	1.57	1.00	1.57	0.03
1-butyl-3-methylimidazolium	L-serinate	9.90	5.42	0.62	1.00	0.62	0.00
tetrabutylphosphonium	L-cysteinate	12.27	2.95	2.08	1.00	2.08	0.00
tetrabutylphosphonium	L-prolinate	12.27	1.70	1.65	1.00	1.65	0.00

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Table S5 – *Continued from previous page*

Cation	**Anion**	λ_I [Å]	η [Pa·s]	ΔV_{ESW} [V]	G	$\Delta V_{ESW} \times G$ [V]	ζ
tetrabutylphosphonium	L-threoninate	12.27	7.40×10^{-1}	2.35	1.00	2.35	0.00
tetrabutylphosphonium	L-lysinate	12.27	7.40×10^{-1}	1.83	1.00	1.83	0.00
tetrabutylphosphonium	2-aminoethanesulfonate	12.27	1.02	2.74	1.00	2.74	0.00
tetrabutylphosphonium	L-serinate	12.27	1.14	2.16	1.00	2.16	0.00
1,2-dimethyl-3-propylimidazolium	tetrafluoroborate	8.46	3.77×10^{-1}	4.36	1.00	4.36	0.08
1-butyl-3-methylimidazolium	salicylate	9.90	4.10×10^{-1}	0.93	1.00	0.93	0.01
1-ethyl-3-methylimidazolium	trifluoroacetate	7.46	3.20×10^{-2}	1.56	1.00	1.56	0.56
1-(3-methylbutyl)-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	9.64	6.90×10^{-2}	2.92	1.00	2.92	0.76
1-(2-methoxyethyl)-1-methylpiperidinium	bis(trifluoromethanesulfonyl)imide	9.07	6.99×10^{-2}	3.42	1.00	3.42	0.89
(3-carboxypropyl)trimethylammonium	bis(trifluoromethanesulfonyl)imide	8.27	1.75	3.15	1.00	3.15	0.00
1-butyl-3-methylimidazolium	taurate	9.90	1.17	1.19	1.00	1.19	0.00
1,3-dimethylimidazolium	dimethylphosphate	6.23	2.91×10^{-1}	1.29	1.00	1.29	0.05
(3-aminopropyl)tributylphosphonium	L-valinate	12.05	8.05×10^{-1}	1.05	1.00	1.05	0.00
(3-aminopropyl)tributylphosphonium	L-alaninate	12.05	6.77×10^{-1}	1.01	1.00	1.01	0.00
(3-aminopropyl)tributylphosphonium	L-leucinate	12.05	1.23	1.04	1.00	1.04	0.00
1-butyl-3-methylimidazolium	L-alaninate	9.90	7.83×10^{-2}	0.26	1.00	0.26	0.06
1-butyl-3-methylimidazolium	(S)-2-amino-4-carboxybutanoate	9.90	8.30×10^{-2}	0.48	1.00	0.48	0.11
ethylammonium	bisulfate	4.21	1.28×10^{-1}	0.88	1.00	0.88	0.14
ethylammonium	lactate	4.33	8.03×10^{-1}	0.01	1.00	0.01	0.00
ethylammonium	glycolate	4.21	1.20	0.06	1.00	0.06	0.00
1-butyl-3-methylimidazolium	methanesulfonate	9.90	1.11×10^{-1}	1.15	1.00	1.15	0.21

Table S6: Data presented in Fig. 2b describing RTIL performance confined in nanopores with a mean pore size of 10.0 Å.

Cation	**Anion**	λ_I [Å]	η [Pa·s]	ΔV_{ESW} [V]	G	$\Delta V_{ESW} \times G$ [V]	ζ
1-ethyl-3-methylimidazolium	methanesulfonate	7.46	1.60×10^{-1}	1.10	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	7.46	3.44×10^{-2}	2.84	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	tetrafluoroborate	7.46	3.88×10^{-2}	4.02	0.00	0.00	0.00
1-butyl-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	9.90	5.09×10^{-2}	2.89	0.95	2.76	0.84
1-butyl-3-methylimidazolium	trifluoroacetate	9.90	5.29×10^{-2}	1.61	0.95	1.54	0.46
1-butyl-3-methylimidazolium	tetrafluoroborate	9.90	1.08×10^{-1}	4.07	0.95	3.88	0.72
1-butyl-3-methylimidazolium	hexafluorophosphate	9.90	2.71×10^{-1}	4.61	0.95	4.40	0.20
1,3-dimethylimidazolium	bis(trifluoromethanesulfonyl)imide	7.39	3.81×10^{-2}	2.77	0.00	0.00	0.00
1-butyl-3-methylimidazolium	methylsulfate	9.90	2.13×10^{-1}	1.59	0.95	1.51	0.11
1-ethyl-3-methylimidazolium	methylsulfate	7.46	7.88×10^{-2}	1.54	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	tetracyanoborate	7.46	1.80×10^{-2}	4.45	0.00	0.00	0.00
1,3-dimethylimidazolium	methylsulfate	6.23	7.17×10^{-2}	1.47	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	ethylsulfate	7.46	9.78×10^{-2}	1.52	0.00	0.00	0.00
1-(2-methoxyethyl)-1-methylpyrrolidinium	bis(trifluoromethanesulfonyl)imide	8.03	5.51×10^{-2}	3.33	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	bisulfate	7.46	1.63	1.63	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	acetate	7.46	1.44×10^{-1}	0.34	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	thiocyanate	7.46	2.45×10^{-2}	0.21	0.00	0.00	0.00
1-butyl-3-methylimidazolium	thiocyanate	9.90	5.65×10^{-2}	0.26	0.95	0.24	0.07
1-butyl-3-methylimidazolium	acetate	9.90	3.00×10^{-1}	0.39	0.95	0.37	0.01
1-butyl-3-methylimidazolium	dimethylphosphate	9.90	6.42×10^{-1}	1.40	0.95	1.34	0.00
1,2-dimethyl-3-propylimidazolium	bis(trifluoromethanesulfonyl)imide	8.46	9.28×10^{-2}	3.18	0.00	0.00	0.00
1,3-diethylimidazolium	bis(trifluoromethanesulfonyl)imide	8.71	3.08×10^{-2}	2.91	0.00	0.00	0.00
1-(2-hydroxyethyl)-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	8.38	8.69×10^{-2}	2.77	0.00	0.00	0.00
1-butyl-3-methylimidazolium	perchlorate	9.90	1.80×10^{-1}	2.24	0.95	2.14	0.21
1,2-diethylpyridinium	bis(trifluoromethanesulfonyl)imide	7.39	6.88×10^{-2}	1.67	0.00	0.00	0.00
tetrabutylphosphonium	2-(bis(2-hydroxyethyl)amino)ethanesulfonate	12.27	1.41×10^{1}	3.09	0.00	0.00	0.00
tetrabutylphosphonium	2-hydroxy-3-morpholinopropanesulfonate	12.27	7.31	3.05	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	dimethylphosphate	7.46	2.70×10^{-1}	1.35	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	L-alaninate	7.46	2.64×10^{-1}	0.21	0.00	0.00	0.00
1,3-dimethylimidazolium	acetate	6.23	8.70×10^{-2}	0.27	0.00	0.00	0.00
1-(2-hydroxyethyl)-1-methylpyrrolidin-1-ium	bis(trifluoromethanesulfonyl)imide	7.39	8.00×10^{-2}	3.34	0.00	0.00	0.00
1-(2,3-dihydroxypropyl)-1-methylpyrrolidin-1-ium	bis(trifluoromethanesulfonyl)imide	7.73	1.50	3.41	0.00	0.00	0.00
1-(2-hydroxyethyl)-pyridinium	bis(trifluoromethanesulfonyl)imide	7.91	1.09×10^{-1}	1.30	0.00	0.00	0.00
1-butyl-3-methylimidazolium	nitrate	9.90	1.65×10^{-1}	0.78	0.95	0.75	0.09
1-butyl-3-methylimidazolium	bicarbonate	9.90	7.37×10^{-2}	0.82	0.95	0.78	0.20
1-butyl-3-methylimidazolium	dihydrogenphosphate	9.90	1.26×10^{-1}	1.57	0.95	1.50	0.24
1-(2-hydroxyethyl)-3-methylimidazolium	tetrafluoroborate	8.38	1.15×10^{-1}	3.95	0.00	0.00	0.00
1,2-diethylpyridinium	ethylsulfate	7.11	3.25×10^{-1}	0.35	0.00	0.00	0.00
1,3-dimethylpyridinium	methylsulfate	6.35	1.29×10^{-1}	0.11	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	diethylphosphate	7.46	4.10×10^{-1}	1.33	0.00	0.00	0.00
1-butyl-3-methylimidazolium	ethylsulfate	9.90	3.79×10^{-1}	1.57	0.95	1.49	0.03
1-butyl-3-methylimidazolium	L-serinate	9.90	5.42	0.62	0.95	0.59	0.00
tetrabutylphosphonium	L-cysteinate	12.27	2.95	2.08	0.00	0.00	0.00
tetrabutylphosphonium	L-prolinate	12.27	1.70	1.65	0.00	0.00	0.00

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Table S6 – *Continued from previous page*

Cation	**Anion**	λ_I [Å]	η [Pa·s]	ΔV_{ESW} [V]	G	$\Delta V_{ESW} \times G$ [V]	ζ
tetrabutylphosphonium	L-threoninate	12.27	7.40×10^{-1}	2.35	0.00	0.00	0.00
tetrabutylphosphonium	L-lysinate	12.27	7.40×10^{-1}	1.83	0.00	0.00	0.00
tetrabutylphosphonium	2-aminoethanesulfonate	12.27	1.02	2.74	0.00	0.00	0.00
tetrabutylphosphonium	L-serinate	12.27	1.14	2.16	0.00	0.00	0.00
1,2-dimethyl-3-propylimidazolium	tetrafluoroborate	8.46	3.77×10^{-1}	4.36	0.00	0.00	0.00
1-butyl-3-methylimidazolium	salicylate	9.90	4.10×10^{-1}	0.93	0.95	0.89	0.01
1-ethyl-3-methylimidazolium	trifluoroacetate	7.46	3.20×10^{-2}	1.56	0.00	0.00	0.00
1-(3-methylbutyl)-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	9.64	6.90×10^{-2}	2.92	0.55	1.61	0.42
1-(2-methoxyethyl)-1-methylpiperidinium	bis(trifluoromethanesulfonyl)imide	9.07	6.99×10^{-2}	3.42	0.02	0.06	0.02
(3-carboxypropyl)trimethylammonium	bis(trifluoromethanesulfonyl)imide	8.27	1.75	3.15	0.00	0.00	0.00
1-butyl-3-methylimidazolium	taurate	9.90	1.17	1.19	0.95	1.14	0.00
1,3-dimethylimidazolium	dimethylphosphate	6.23	2.91×10^{-1}	1.29	0.00	0.00	0.00
(3-aminopropyl)tributylphosphonium	L-valinate	12.05	8.05×10^{-1}	1.05	0.00	0.00	0.00
(3-aminopropyl)tributylphosphonium	L-alaninate	12.05	6.77×10^{-1}	1.01	0.00	0.00	0.00
(3-aminopropyl)tributylphosphonium	L-leucinate	12.05	1.23	1.04	0.00	0.00	0.00
1-butyl-3-methylimidazolium	L-alaninate	9.90	7.83×10^{-2}	0.26	0.95	0.25	0.06
1-butyl-3-methylimidazolium	(S)-2-amino-4-carboxybutanoate	9.90	8.30×10^{-2}	0.48	0.95	0.46	0.11
ethylammonium	bisulfate	4.21	1.28×10^{-1}	0.88	0.00	0.00	0.00
ethylammonium	lactate	4.33	8.03×10^{-1}	0.01	0.00	0.00	0.00
ethylammonium	glycolate	4.21	1.20	0.06	0.00	0.00	0.00
1-butyl-3-methylimidazolium	methanesulfonate	9.90	1.11×10^{-1}	1.15	0.95	1.09	0.20

Table S7: Data presented in Fig. 2c describing RTIL performance confined in nanopores with a mean pore size of 7.5 Å.

Cation	**Anion**	λ_I [Å]	η [Pa·s]	ΔV_{ESW} [V]	G	$\Delta V_{ESW} \times G$ [V]	ζ
1-ethyl-3-methylimidazolium	methanesulfonate	7.46	1.60×10^{-1}	1.10	0.99	1.09	0.13
1-ethyl-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	7.46	3.44×10^{-2}	2.84	0.99	2.82	0.99
1-ethyl-3-methylimidazolium	tetrafluoroborate	7.46	3.88×10^{-2}	4.02	0.99	3.99	1.35
1-butyl-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	9.90	5.09×10^{-2}	2.89	0.00	0.00	0.00
1-butyl-3-methylimidazolium	trifluoroacetate	9.90	5.29×10^{-2}	1.61	0.00	0.00	0.00
1-butyl-3-methylimidazolium	tetrafluoroborate	9.90	1.08×10^{-1}	4.07	0.00	0.00	0.00
1-butyl-3-methylimidazolium	hexafluorophosphate	9.90	2.71×10^{-1}	4.61	0.00	0.00	0.00
1,3-dimethylimidazolium	bis(trifluoromethanesulfonyl)imide	7.39	3.81×10^{-2}	2.77	0.95	2.62	0.89
1-butyl-3-methylimidazolium	methylsulfate	9.90	2.13×10^{-1}	1.59	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	methylsulfate	7.46	7.88×10^{-2}	1.54	0.99	1.52	0.37
1-ethyl-3-methylimidazolium	tetracyanoborate	7.46	1.80×10^{-2}	4.45	0.99	4.41	1.79
1,3-dimethylimidazolium	methylsulfate	6.23	7.17×10^{-2}	1.47	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	ethylsulfate	7.46	9.78×10^{-2}	1.52	0.99	1.50	0.31
1-(2-methoxyethyl)-1-methylpyrrolidinium	bis(trifluoromethanesulfonyl)imide	8.03	5.51×10^{-2}	3.33	0.27	0.91	0.27
1-ethyl-3-methylimidazolium	bisulfate	7.46	1.63	1.63	0.99	1.62	0.00
1-ethyl-3-methylimidazolium	acetate	7.46	1.44×10^{-1}	0.34	0.99	0.33	0.05
1-ethyl-3-methylimidazolium	thiocyanate	7.46	2.45×10^{-2}	0.21	0.99	0.20	0.08
1-butyl-3-methylimidazolium	thiocyanate	9.90	5.65×10^{-2}	0.26	0.00	0.00	0.00
1-butyl-3-methylimidazolium	acetate	9.90	3.00×10^{-1}	0.39	0.00	0.00	0.00
1-butyl-3-methylimidazolium	dimethylphosphate	9.90	6.42×10^{-1}	1.40	0.00	0.00	0.00
1,2-dimethyl-3-propylimidazolium	bis(trifluoromethanesulfonyl)imide	8.46	9.28×10^{-2}	3.18	0.01	0.05	0.01
1,3-diethylimidazolium	bis(trifluoromethanesulfonyl)imide	8.71	3.08×10^{-2}	2.91	0.00	0.00	0.00
1-(2-hydroxyethyl)-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	8.38	8.69×10^{-2}	2.77	0.03	0.08	0.02
1-butyl-3-methylimidazolium	perchlorate	9.90	1.80×10^{-1}	2.24	0.00	0.00	0.00
1,2-diethylpyridinium	bis(trifluoromethanesulfonyl)imide	7.39	6.88×10^{-2}	1.67	0.95	1.58	0.41
tetrabutylphosphonium	2-(bis(2-hydroxyethyl)amino)ethanesulfonate	12.27	1.41×10^{1}	3.09	0.00	0.00	0.00
tetrabutylphosphonium	2-hydroxy-3-morpholinopropanesulfonate	12.27	7.31	3.05	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	dimethylphosphate	7.46	2.70×10^{-1}	1.35	0.99	1.34	0.06
1-ethyl-3-methylimidazolium	L-alaninate	7.46	2.64×10^{-1}	0.21	0.99	0.21	0.01
1,3-dimethylimidazolium	acetate	6.23	8.70×10^{-2}	0.27	0.00	0.00	0.00
1-(2-hydroxyethyl)-1-methylpyrrolidin-1-ium	bis(trifluoromethanesulfonyl)imide	7.39	8.00×10^{-2}	3.34	0.95	3.16	0.75
1-(2,3-dihydroxypropyl)-1-methylpyrrolidin-1-ium	bis(trifluoromethanesulfonyl)imide	7.73	1.50	3.41	0.78	2.67	0.00
1-(2-hydroxyethyl)-pyridinium	bis(trifluoromethanesulfonyl)imide	7.91	1.09×10^{-1}	1.30	0.46	0.60	0.11
1-butyl-3-methylimidazolium	nitrate	9.90	1.65×10^{-1}	0.78	0.00	0.00	0.00
1-butyl-3-methylimidazolium	bicarbonate	9.90	7.37×10^{-2}	0.82	0.00	0.00	0.00
1-butyl-3-methylimidazolium	dihydrogenphosphate	9.90	1.26×10^{-1}	1.57	0.00	0.00	0.00
1-(2-hydroxyethyl)-3-methylimidazolium	tetrafluoroborate	8.38	1.15×10^{-1}	3.95	0.03	0.11	0.02
1,2-diethylpyridinium	ethylsulfate	7.11	3.25×10^{-1}	0.35	0.50	0.17	0.00
1,3-dimethylpyridinium	methylsulfate	6.35	1.29×10^{-1}	0.11	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	diethylphosphate	7.46	4.10×10^{-1}	1.33	0.99	1.32	0.02
1-butyl-3-methylimidazolium	ethylsulfate	9.90	3.79×10^{-1}	1.57	0.00	0.00	0.00
1-butyl-3-methylimidazolium	L-serinate	9.90	5.42	0.62	0.00	0.00	0.00
tetrabutylphosphonium	L-cysteinate	12.27	2.95	2.08	0.00	0.00	0.00
tetrabutylphosphonium	L-prolinate	12.27	1.70	1.65	0.00	0.00	0.00

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Table S7 – *Continued from previous page*

Cation	**Anion**	λ_I [Å]	η [Pa·s]	ΔV_{ESW} [V]	G	$\Delta V_{ESW} \times G$ [V]	ζ
tetrabutylphosphonium	L-threoninate	12.27	7.40×10^{-1}	2.35	0.00	0.00	0.00
tetrabutylphosphonium	L-lysinate	12.27	7.40×10^{-1}	1.83	0.00	0.00	0.00
tetrabutylphosphonium	2-aminoethanesulfonate	12.27	1.02	2.74	0.00	0.00	0.00
tetrabutylphosphonium	L-serinate	12.27	1.14	2.16	0.00	0.00	0.00
1,2-dimethyl-3-propylimidazolium	tetrafluoroborate	8.46	3.77×10^{-1}	4.36	0.01	0.06	0.00
1-butyl-3-methylimidazolium	salicylate	9.90	4.10×10^{-1}	0.93	0.00	0.00	0.00
1-ethyl-3-methylimidazolium	trifluoroacetate	7.46	3.20×10^{-2}	1.56	0.99	1.55	0.56
1-(3-methylbutyl)-3-methylimidazolium	bis(trifluoromethanesulfonyl)imide	9.64	6.90×10^{-2}	2.92	0.00	0.00	0.00
1-(2-methoxyethyl)-1-methylpiperidinium	bis(trifluoromethanesulfonyl)imide	9.07	6.99×10^{-2}	3.42	0.00	0.00	0.00
(3-carboxypropyl)trimethylammonium	bis(trifluoromethanesulfonyl)imide	8.27	1.75	3.15	0.07	0.21	0.00
1-butyl-3-methylimidazolium	taurate	9.90	1.17	1.19	0.00	0.00	0.00
1,3-dimethylimidazolium	dimethylphosphate	6.23	2.91×10^{-1}	1.29	0.00	0.00	0.00
(3-aminopropyl)tributylphosphonium	L-valinate	12.05	8.05×10^{-1}	1.05	0.00	0.00	0.00
(3-aminopropyl)tributylphosphonium	L-alaninate	12.05	6.77×10^{-1}	1.01	0.00	0.00	0.00
(3-aminopropyl)tributylphosphonium	L-leucinate	12.05	1.23	1.04	0.00	0.00	0.00
1-butyl-3-methylimidazolium	L-alaninate	9.90	7.83×10^{-2}	0.26	0.00	0.00	0.00
1-butyl-3-methylimidazolium	(S)-2-amino-4-carboxybutanoate	9.90	8.30×10^{-2}	0.48	0.00	0.00	0.00
ethylammonium	bisulfate	4.21	1.28×10^{-1}	0.88	0.00	0.00	0.00
ethylammonium	lactate	4.33	8.03×10^{-1}	0.01	0.00	0.00	0.00
ethylammonium	glycolate	4.21	1.20	0.06	0.00	0.00	0.00
1-butyl-3-methylimidazolium	methanesulfonate	9.90	1.11×10^{-1}	1.15	0.00	0.00	0.00

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