Electronic Supplementary Information

“High-Throughput and Data Driven Strategies for the Design of Deep-Eutectic Solvent Electrolytes”

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Table S1: Concentrated stock solution information for all Hydrogen Bond Donors prepared. There were prepared by hand by dissolving the indicated amount [g] of HBD species and mixed with 25mL of the selected solvent. If higher volumes were needed, the required mass for each component was linearly scaled to maintain the indicated concentration

HBD	MW $[g/mol]$	Solvent	HBD Max Conc [M]	Mass $[g]$ for 25mL solution
Ethylene Glycol	62.07	Water	7	10.86
Glycerol	92.09	Water	7	16.12
Acetamide	59.07	Water	7	10.34
N,N'-dimethylurea	88.11	Water	7	15.42
3-phenylpropionic acid	150.17	Ethanol	3	11.26
Urea	60.06	Water	7	10.51
L-serine	105.09	Water	2	5.25
4-amino triazole	84.08	Water	4	8.41
Xylitol	152.15	Water	3	11.41
Phenylacetic acid	136.15	Ethanol	3	10.21

Table S2: Concentrated stock solution information for all Quaternary Ammonium Salts prepared. There were prepared by hand by dissolving the indicated amount [g] of QAS species and mixed with 25mL of the selected solvent. If higher volumes were needed, the required mass for each component was linearly scaled to maintain the indicated concentration

QAS	MW $[g/mol]$	Solvent	HBA Max Conc (M)	Mass $[g]$ for 25mL solution
Choline Chloride	139.62	Water	4	13.96
Acetylcholine Chloride	181.66	Water	4	18.17
Tetraethylammonium Chloride	165.7	Water	3	12.43
Tetrapropylammonium Bromide	266.26	Water	2	13.31
Tetraethylammonium Iodide	257.16	Water	1	6.429

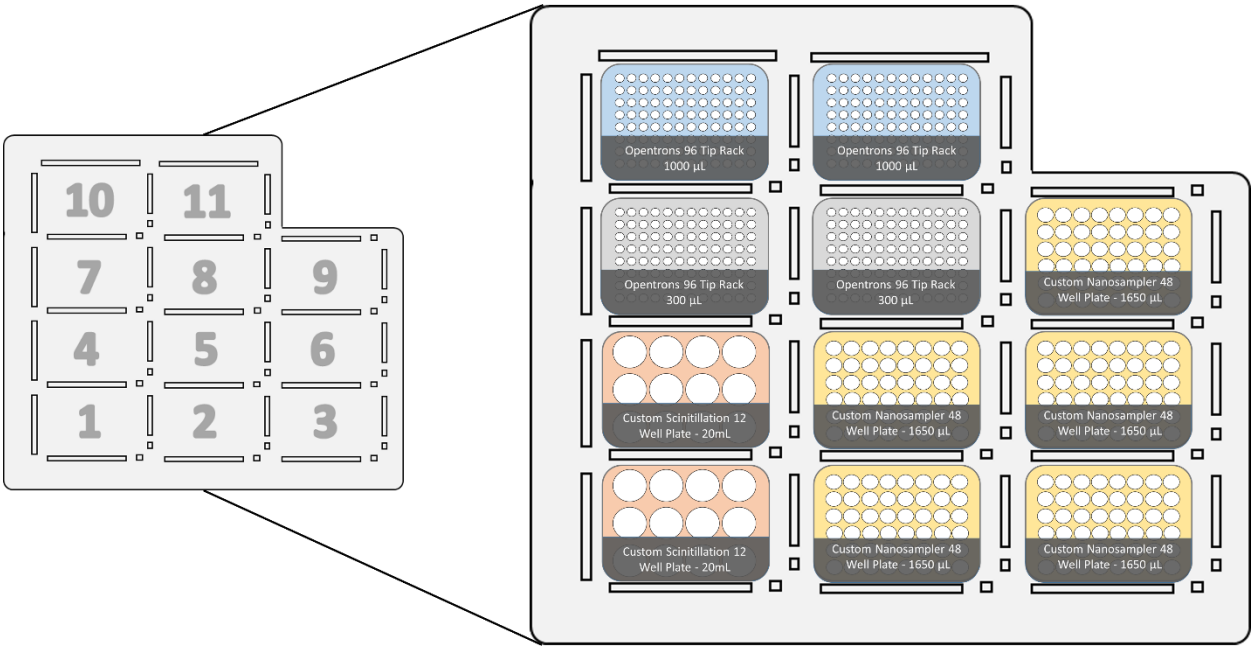

Figure S1: Example OT-2 Deck configuration used to prepare the DES samples. Note that only 240 samples, which correspond to 2 quaternary ammonium salts and all of their combinations with the 10 hydrogen bond donors, can be made in the same robot.

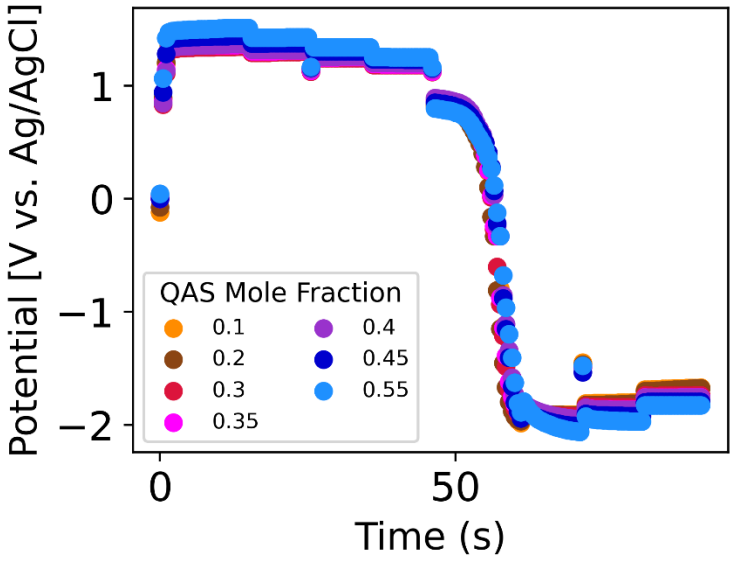

Figure S2: Example Chronopotentiometry measurements for all combination of DES between Choline Chloride and Ethylene Glycol. The current hold steps are as follows: 1,0.75,0.5,0.25 mA and they negative counterparts. Each current hold is maintained for 10s, while the very first positive and negative currents are held for 15s as conditioning steps.

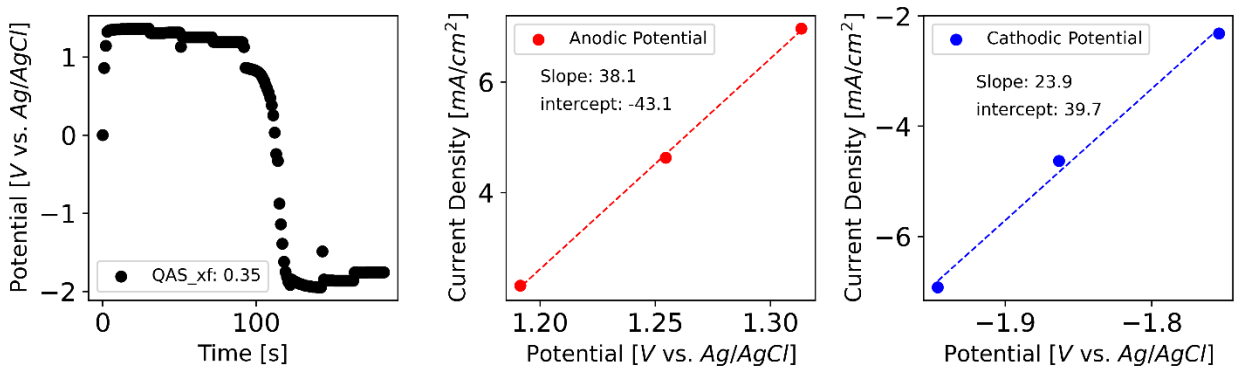

Figure S3: Example Chronopotentiometry analysis for the extraction of the Anodic and Cathodic Potentials for the DES formed by Choline Chloride and Ethylene Glycol (0.35: 0.65 molar fraction) sample. Left) Overall Chronopotentiometry measurement. The experimental parameters are indicated in Figure S2, as well as in the accompanying publication. Center) Average of the last 2 seconds for each of the positive current steps (excluding the conditioning step). The anodic potential can be calculated as follows: $E_a = -\frac{intercept}{slope}$. Right) Average of the last 2 seconds for each of the negative current steps (excluding the conditioning step). The cathodic potential can be calculated as follows: $E_c = -\frac{intercept}{slope}$.

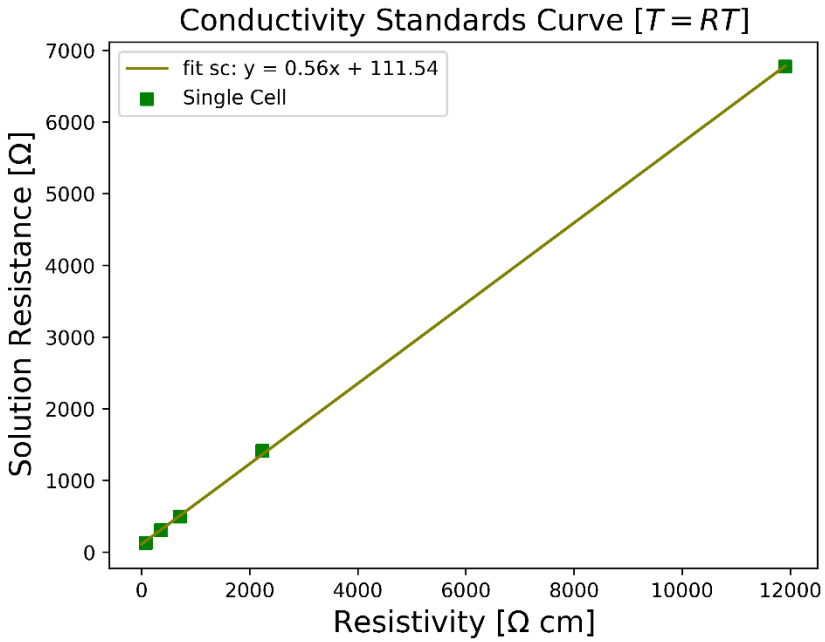

Figure S4: Calibration curve to convert the measured solution Ohmic Resistance obtained from the electrochemical impedance spectroscopy and the Metrohm-DropSens DRP-11L SPEs. This calibration was obtained using conductivity standards of 84 $\mu S/cm$, 443 $\mu S/cm$, 1413 $\mu S/cm$, 2764 $\mu S/cm$, and 12880 $\mu S/cm$.

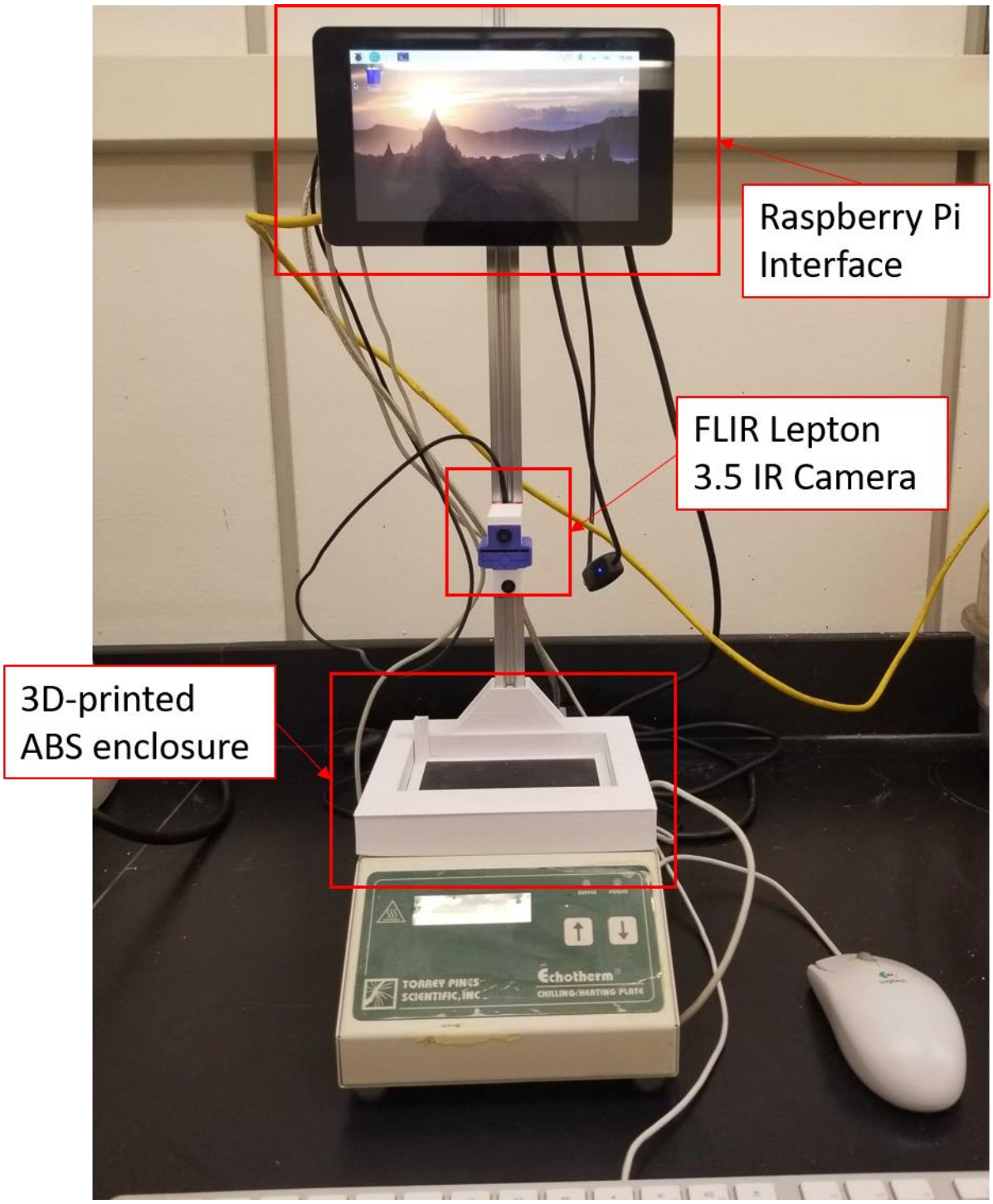

Figure S5: PhasIR Hardware System. This is composed by a Raspberry Pi Interface, an IR Camera, a 3D printed plate enclosure and a Peltier plate. For the current publication, a VWR Advanced Hot Plate Stirrer was used instead of Echotherm Peltier plate found in the image above. The heating source was changed to allow for a broader temperature range.

Table S3: Molar composition tested between each QAS and HBD combination.

QAS_{x_f}	HBD_{x_f}
0.1	0.9
0.2	0.8
0.3	0.7
0.35	0.65
0.4	0.6
0.45	0.55
0.5	0.5
0.55	0.45
0.6	0.4
0.7	0.3
0.8	0.2
0.9	0.1

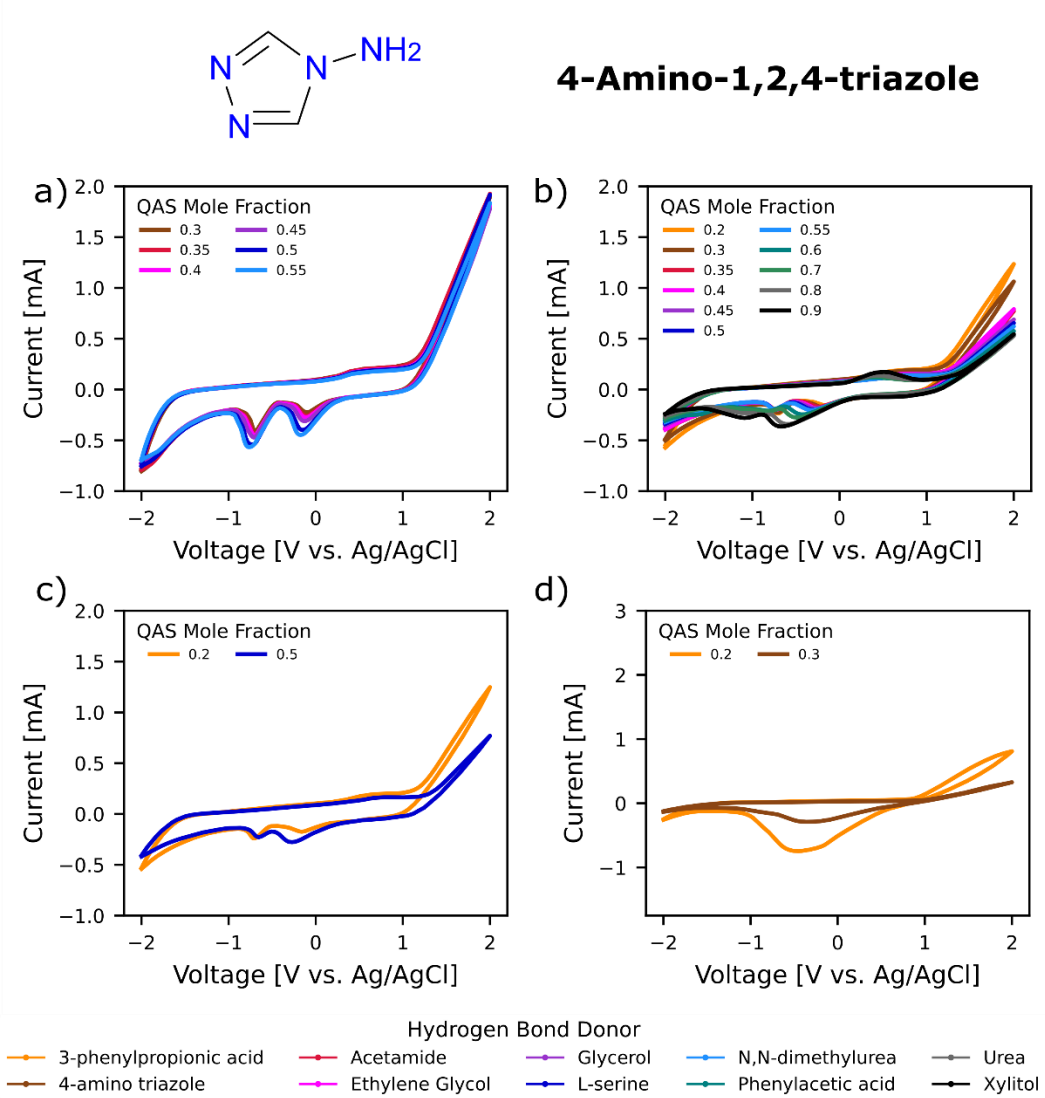

Figure S6: Cyclic voltammograms of 4-Amino-4H,1,2,4triazole-based DES as a function of QAS mole fraction. For all measurement, the potential was varied from $-2V$ and $2V$ with a scan rate of 100 mV/s and a total of 4 cycles per sample to ensure equilibration. a) Choline Chloride, b) Acetylcholine Chloride, c) Tetraethylammonium Chloride, d) Tetrapropylammonium Bromide

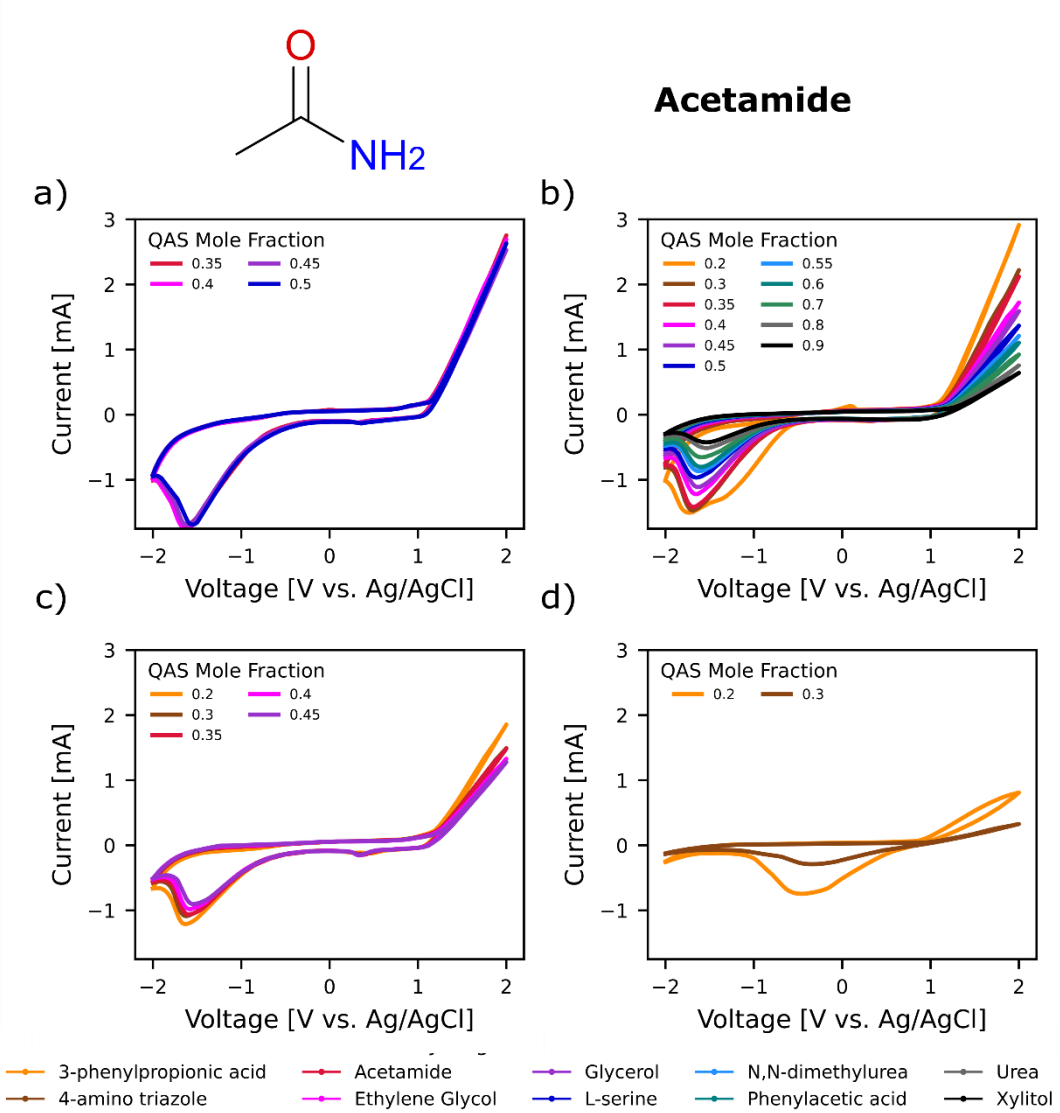

Figure S7: Cyclic voltammograms of Acetamide-based DES as a function of QAS mole fraction. For all measurement, the potential was varied from $-2V$ and $2V$ with a scan rate of $100\,mV/s$ and a total of 4 cycles per sample to ensure equilibration. a) Choline Chloride, b) Acetylcholine Chloride, c) Tetraethylammonium Chloride, d) Tetrapropylammonium Bromide

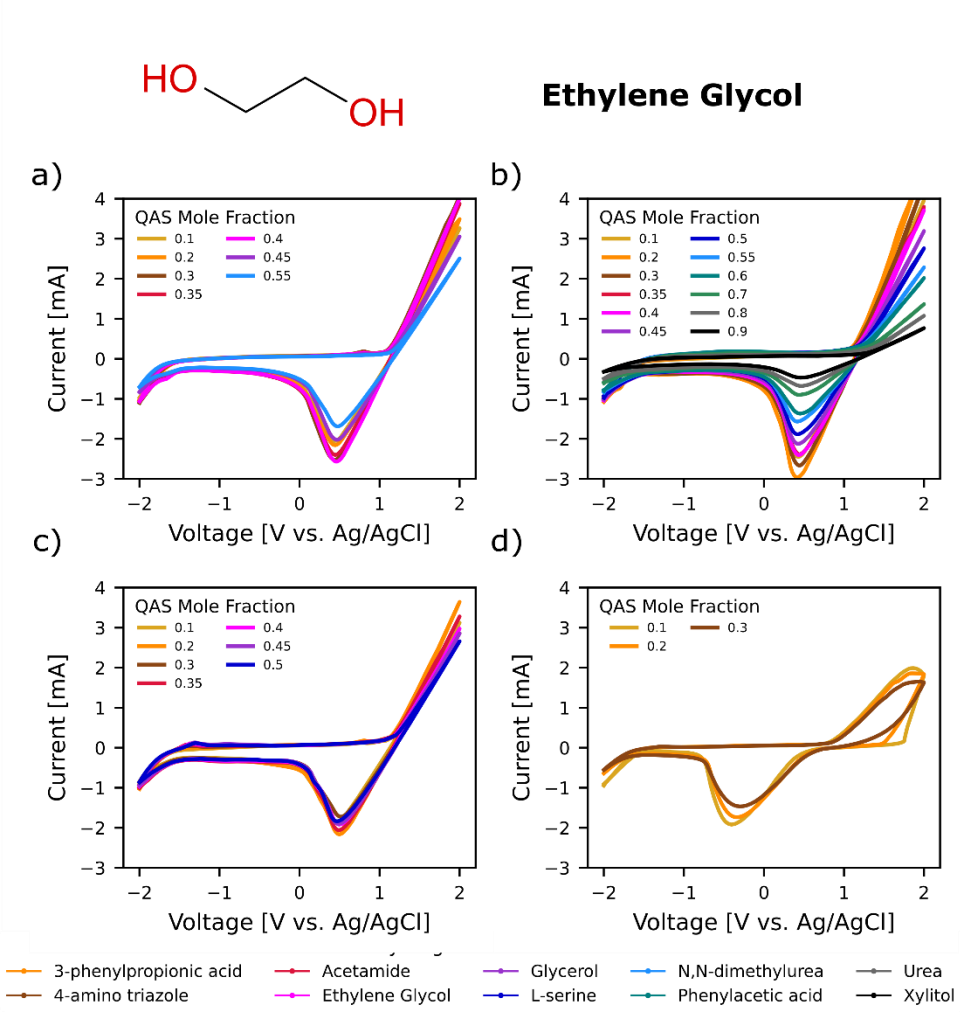

Figure S8: Cyclic voltammograms of Ethylene Glycol-based DES as a function of QAS mole fraction. For all measurement, the potential was varied from $-2V$ and $2V$ with a scan rate of $100\ mV/s$ and a total of 4 cycles per sample to ensure equilibration. a) Choline Chloride, b) Acetylcholine Chloride, c) Tetraethylammonium Chloride, d) Tetrapropylammonium Bromide

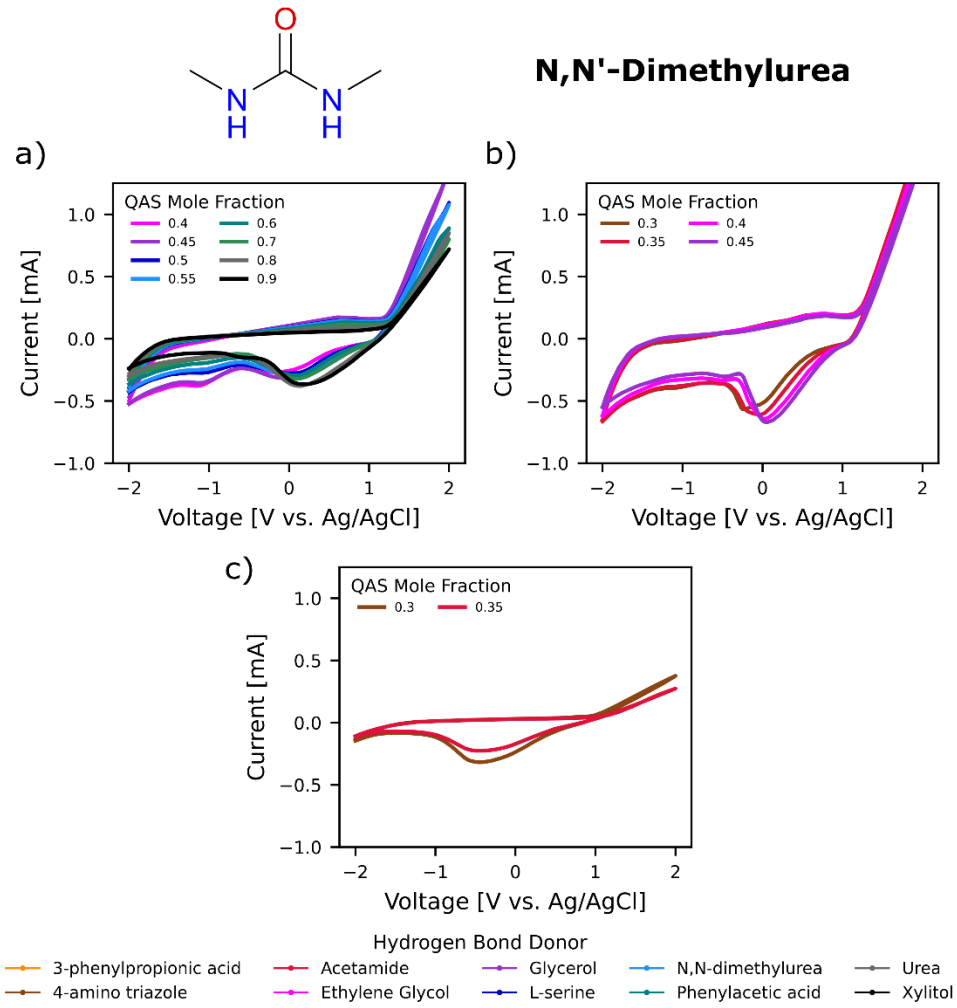

Figure S9: Cyclic voltammograms of N,N'-Dimethylurea-based DES as a function of QAS mole fraction. For all measurement, the potential was varied from $-2V$ and $2V$ with a scan rate of 100 mV/s and a total of 4 cycles per sample to ensure equilibration. a) Acetylcholine Chloride, b) Tetraethylammonium Chloride, c) Tetrapropylammonium Bromide. No combinations with Choline Chloride resulted in a liquid mixture at room temperature.

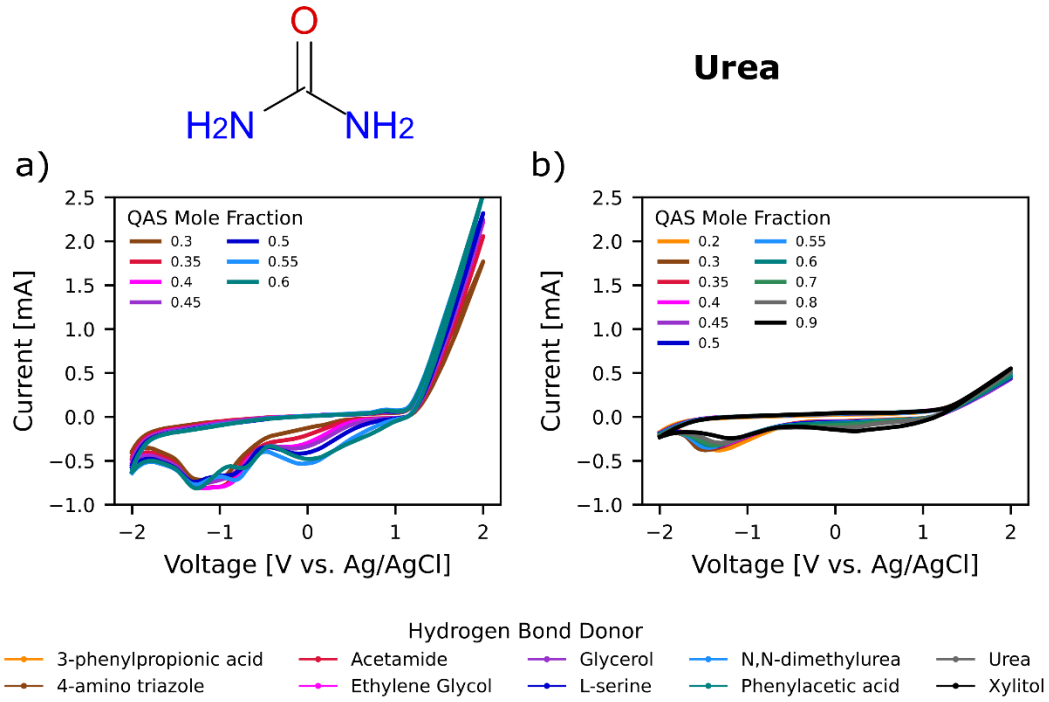

Figure S10: Cyclic voltammograms of Urea-based DES as a function of QAS mole fraction. For all measurement, the potential was varied from $-2V$ and $2V$ with a scan rate of 100 mV/s and a total of 4 cycles per sample to ensure equilibration. a) Choline Chloride, b) Acetylcholine Chloride. No combination with Tetraethylammonium Chloride and Tetrapropylammonium Bromide resulted in a liquid mixture at room temperature.

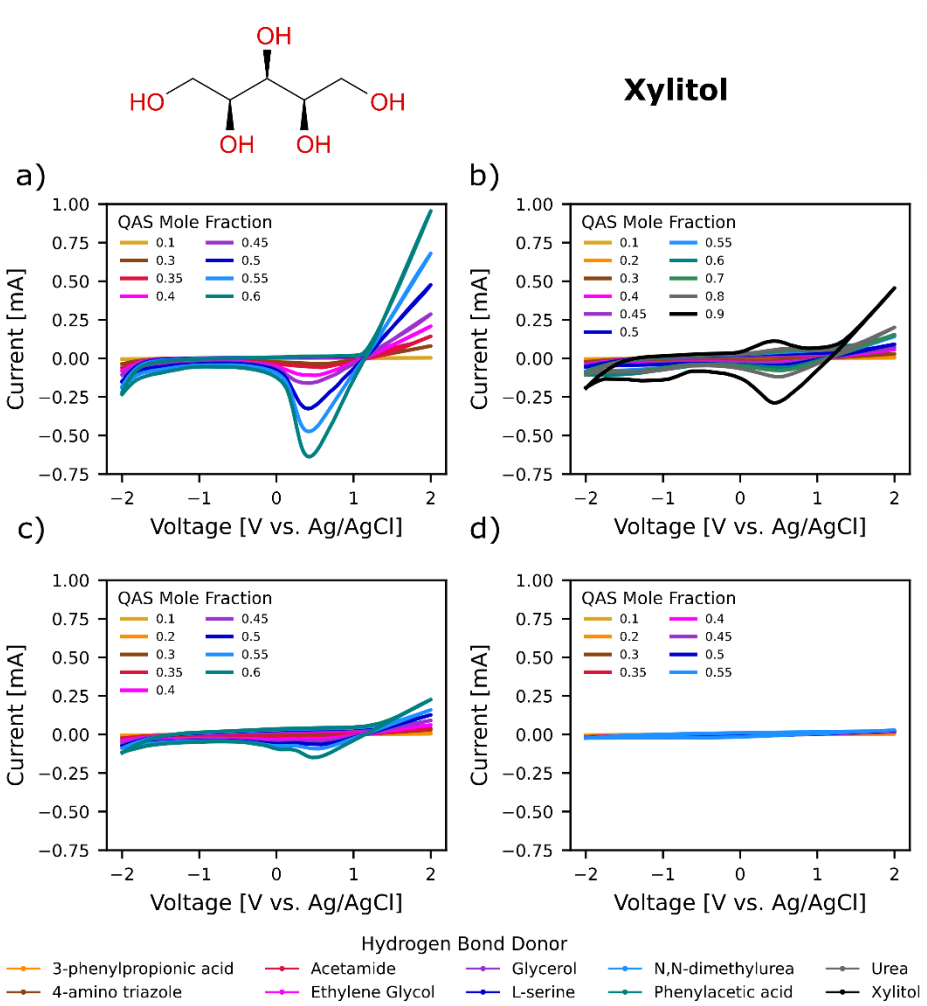

Figure S11: Cyclic voltammograms of Xylitol-based DES as a function of QAS mole fraction. For all measurement, the potential was varied from $-2V$ and $2V$ with a scan rate of $100\,mV/s$ and a total of 4 cycles per sample to ensure equilibration. a) Choline Chloride, b) Acetylcholine Chloride, c) Tetraethylammonium Chloride, d) Tetrapropylammonium Bromide

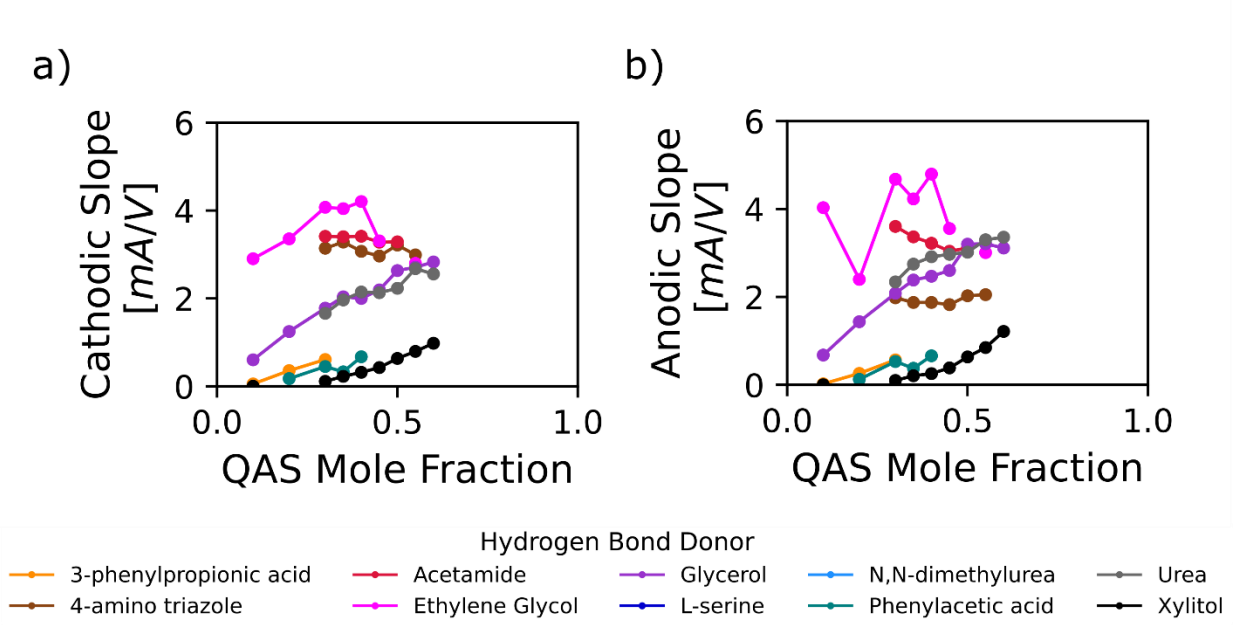

Figure S12: Limiting slope extracted from the cyclic voltammograms of Choline Chloride. The trend of these limiting steps seems to be similar to the conductivity data for the sample DES formulation, Figure 7a. a) Cathodic slope. b) Anodic slope

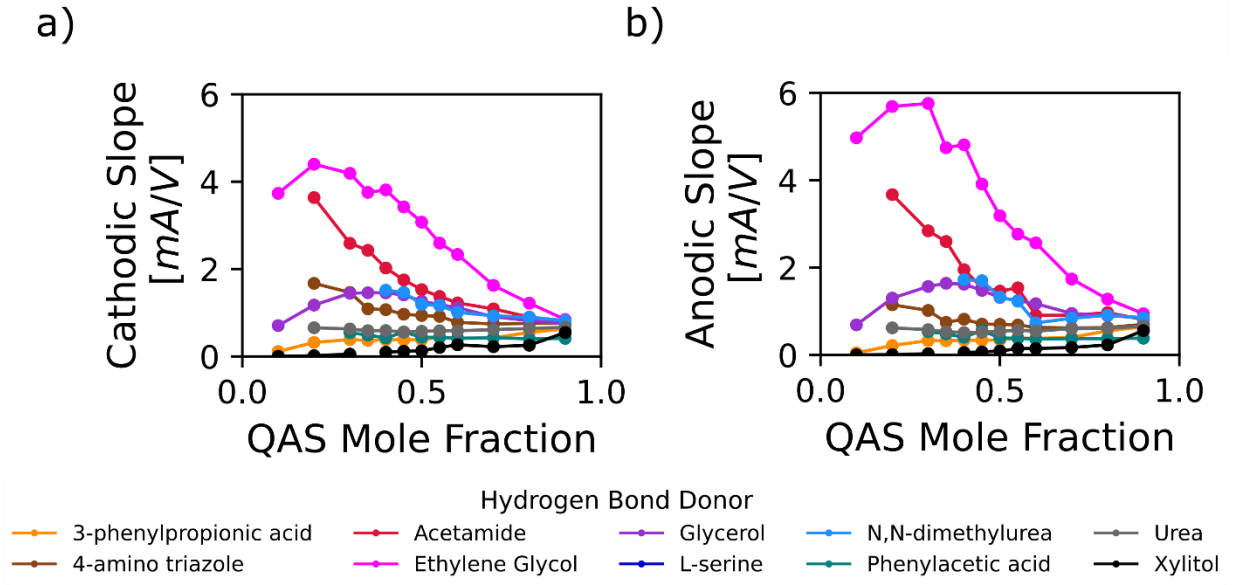

Figure S13: Limiting slope extracted from the cyclic voltammograms of Acetylcholine Chloride. The trend of these limiting steps seems to be similar to the conductivity data for the sample DES formulation, Figure 7b. a) Cathodic slope. b) Anodic slope

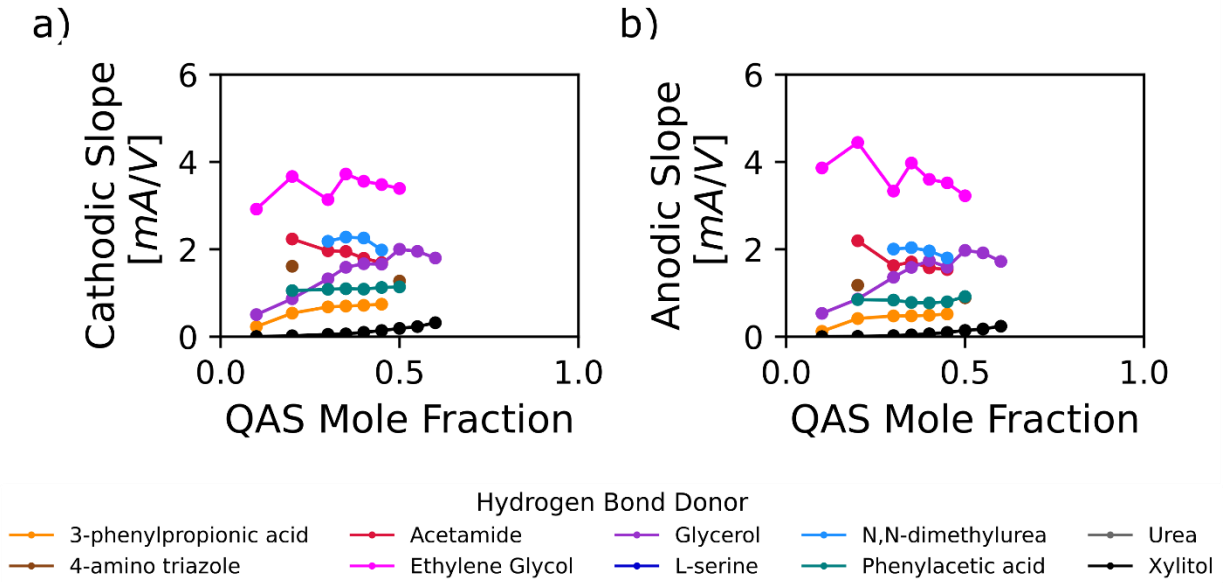

Figure S14: Limiting slope extracted from the cyclic voltammograms of Tetraethylammonium Bromide. The trend of these limiting steps seems to be similar to the conductivity data for the sample DES formulation, Figure 7c. a) Cathodic slope. b) Anodic slope

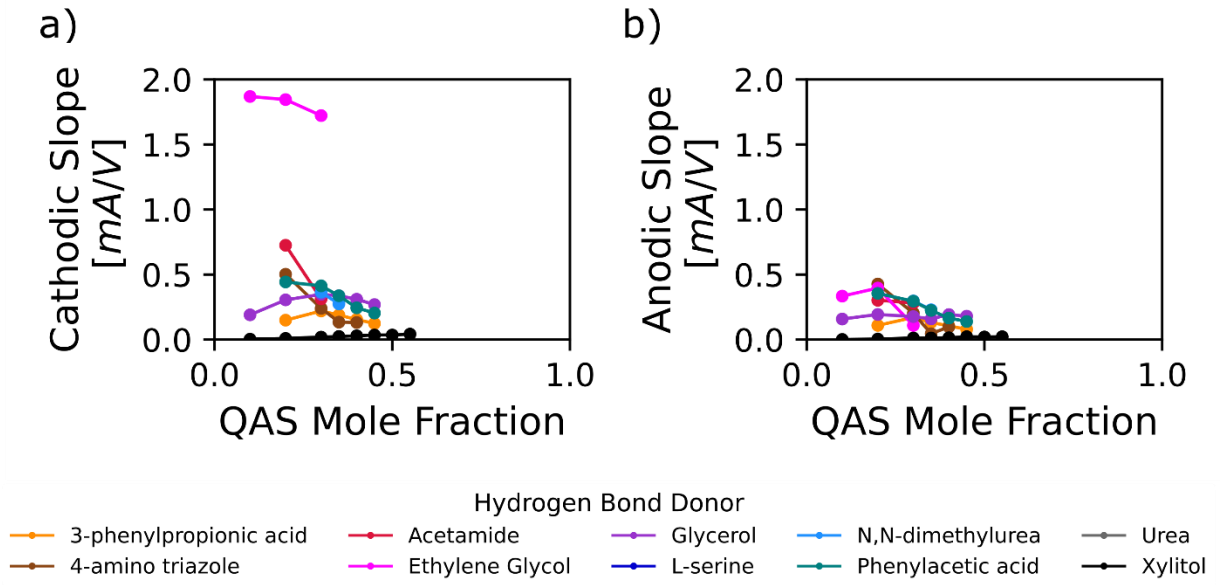

Figure S15: Limiting slope extracted from the cyclic voltammograms of Tetrapropylammonium Bromide. The trend of these limiting steps seems to be similar to the conductivity data for the sample DES formulation, Figure 7d. a) Cathodic slope. b) Anodic slope

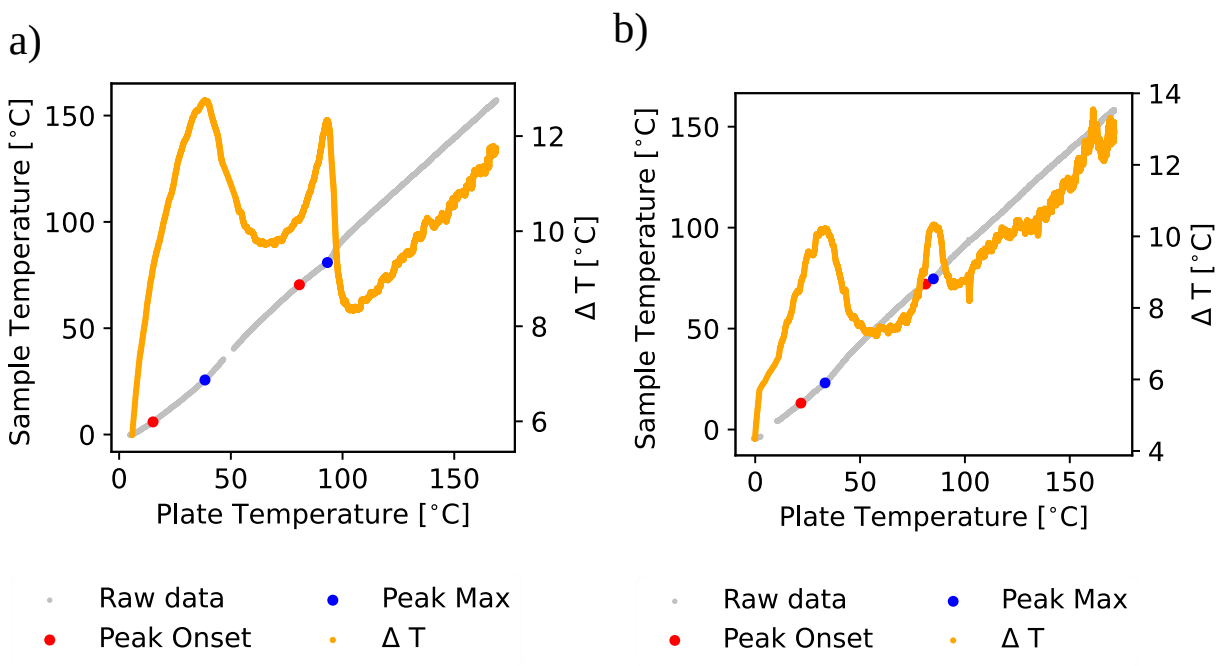

Figure S16: Melting point determination for the 60 $mol\%$ Choline Chloride with Acetamide (a) and 35 $mol\%$ Choline Chloride with Phenylacetic Acid obtained using the PhasIR system. The melting point is defined as the onset (red marker) of the feature in the $\Delta T = T_{plate} - T_{sample}$ curve (yellow). a) The first peak can be attributed to water present in the sample as the peak onset is found around 0 ℃. b) The water feature in this case is the shoulder in the ΔT curve in the lower temperature range. The peak at higher temperatures ($\approx$ 75℃) is most likely some pure Phenylacetic acid.

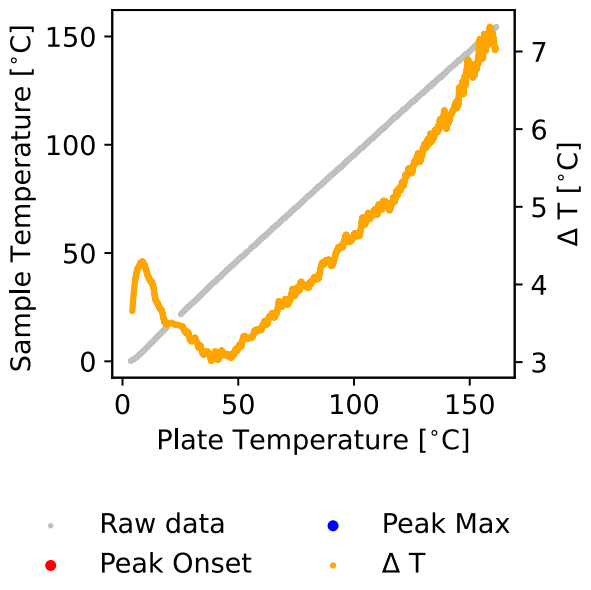

Figure S17: Melting point determination for the 40 $mol\%$ Choline Chloride with Urea obtained using the PhasIR system. The melting point is defined as the onset (red marker) of the feature in the $\Delta T = T_{plate} - T_{sample}$ curve (yellow). In this case, the ΔT curve does not show any significant feature, besides one with possible onset around 0℃ attributed to residual water, and therefore no phase transition was extracted from this curve.

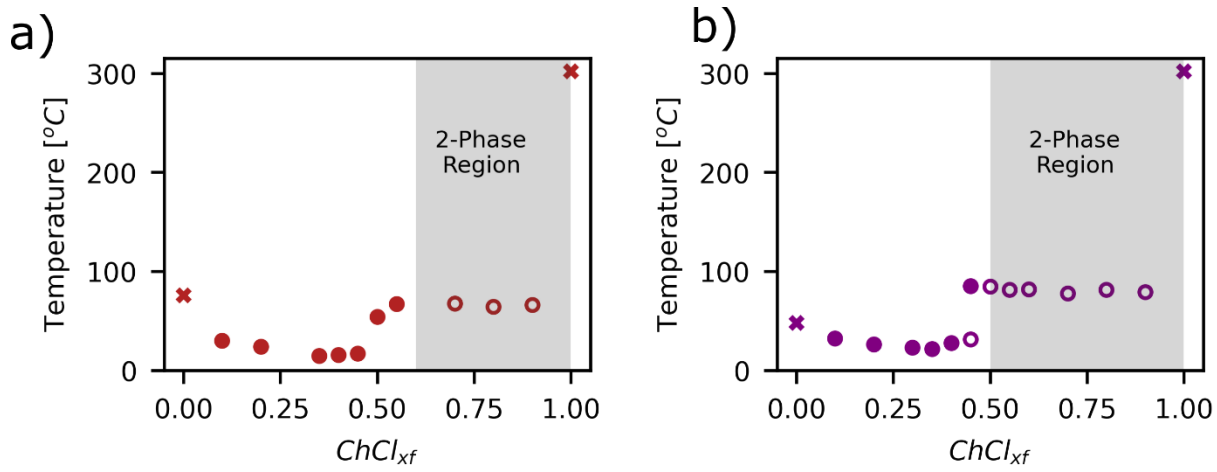

Figure S18: Phase diagrams for Phenylacetic Acid (a) and 3-Phenylproprionic Acid (b). The filled circular markers indicate measurements obtained using the PhasIR system, while the unfilled circles represent possible states of incongruent melting. The x-shaped markers indicate literature values for the pure DES components obtained from their corresponding MSDS.

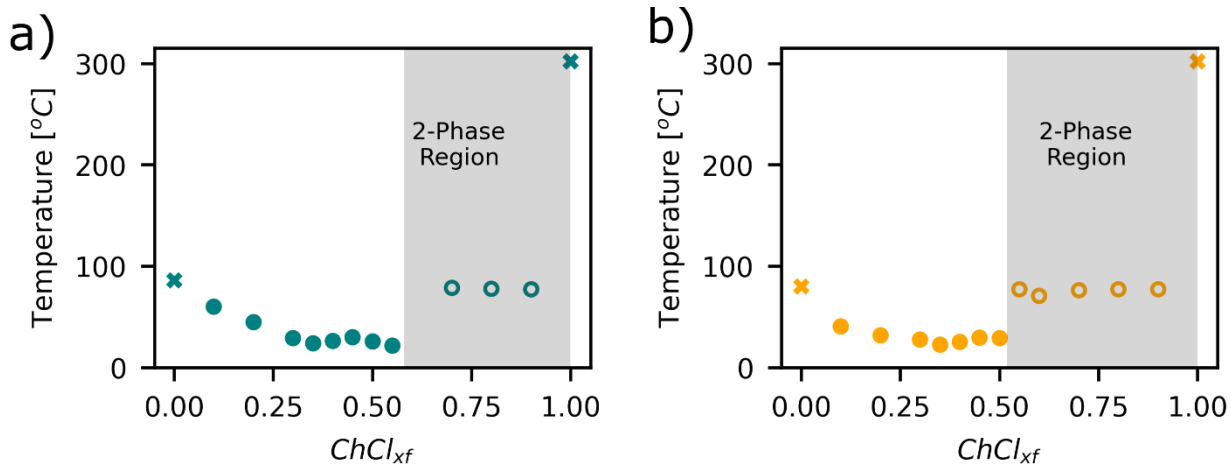

Figure S19: Phase diagrams for 4-Amino-4H,1,2,4triazole (a) and Acetamide (b). The filled circular markers indicate measurements obtained using the PhasIR system, while the unfilled circles represent possible states of incongruent melting. The x-shaped markers indicate literature values for the pure DES components obtained from their corresponding MSDS.

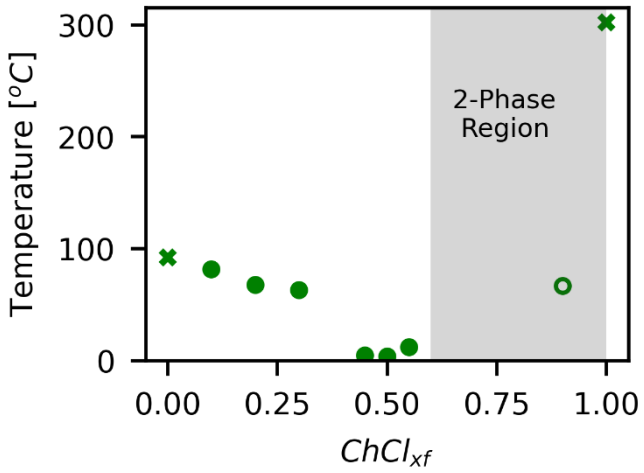

Figure S20: Phase diagrams for Xylitol. The filled circular markers indicate measurements obtained using the PhasIR system, while the unfilled circles represent possible states of incongruent melting. The x-shaped markers indicate literature values for the pure DES components obtained from their corresponding MSDS.

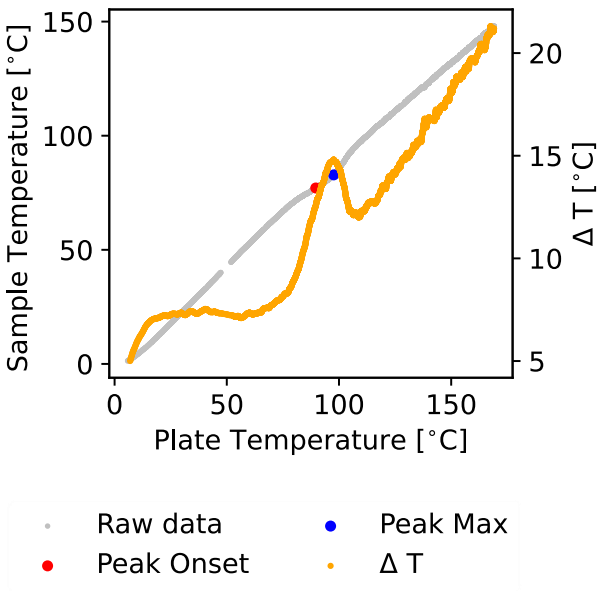

Figure S21: Melting point determination for the 80 $mol\%$ Choline Chloride with Acetamide obtained using the PhasIR system. The melting point is defined as the onset (red marker) of the feature in the $\Delta T = T_{plate} - T_{sample}$ curve (yellow). This shows an example of incongruent melting, as the no feature was expected to be present in this temperature range due to the higher content of QAS. In this case, the feature could be potentially attributed to pure acetamide melting point (80℃).

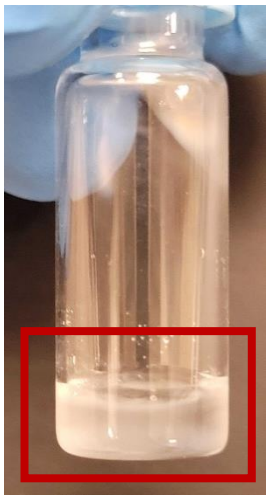

Figure S22: Sample composed of the 80 $mol\%$ Choline Chloride with Acetamide. The sample was heated using a hotplate and formed an opaque solution, which is a sign of formation of two phases.