Supporting Information for

A Computational Study Of The Properties Of Acetonitrile/Water-In-Salt Hybrid Electrolytes As Electrolytes For Supercapacitors

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Transport properties and the corresponding standard deviations

Table S1: Diffusion coefficients ($\mu m^{-2}\ s^{-1}$) of molecular species for all electrolytes investigated at both concentrations. The presented errors are the standard deviations of properties mean.

D ($\mu m^{-2}\ s^{-1}$)	**Water**	**Li**	**TFSi**	**ACN**
Normal/Pure	300.2±6.4	138.7±9.6	78.3±8.9	---
Normal/ACN3	135.1±5.1	72.9±9.1	43.3±7.5	105.5±6.2
Normal/ACN5	154.7±3.3	70.6±6.6	38.3±4.6	89.5±5.5
Normal/ACN7	171.6±4.4	80.5±12.4	42.2±5.8	92.8±12.3
AWiSE/Pure	17.5±0.6	6.7±0.4	3.2±0.3	---
AWiSE/ACN3	77.2±6.1	67.2±7.5	54.2±4.7	128.8±6.0
AWiSE/ACN5	40.5±2.5	29.7±2.3	18.9±1.2	48.3±2.7
AWiSE/ACN7	38.5±1.7	27.2±2.0	16.2±1.0	43.6±4.1

Table S2: Viscosity (m Pa s) and ionic conductivity ($S\ m^{-1}$) for all electrolytes investigated at both concentrations. The presented errors are the standard deviations of properties mean.

Systems	**η (m Pa s)**		**σ ($S\ m^{-1}$)**	
	Normal	**AWiSE**	**Normal**	**AWiSE**
Pure	0.63±0.01	31.6±0.5	6.95±0.56	0.60±0.08
ACN3	1.10±0.02	1.0±0.0	2.80±0.53	3.96±0.81
ACN5	3.06±0.06	4.5±0.1	2.95±0.78	2.04±0.77
ACN7	1.02±0.02	2.9±0.0	3.94±1.10	1.94±0.85

Electrostatic properties

The electrostatic properties of the investigated supercapacitors are discussed in terms of the electrostatic potential profile that can be obtained by the Poisson equation. For this purpose, the distribution of electrostatic charges over the entire supercapacitor is integrated twice (see Figure S1). In these charge distributions, note that the first peak in each profile refers to the charge induced in the electrode in contact with the electrolyte. Much lower charges were induced on the outermost electrodes and, for the sake of clarity, are not shown in the graph.

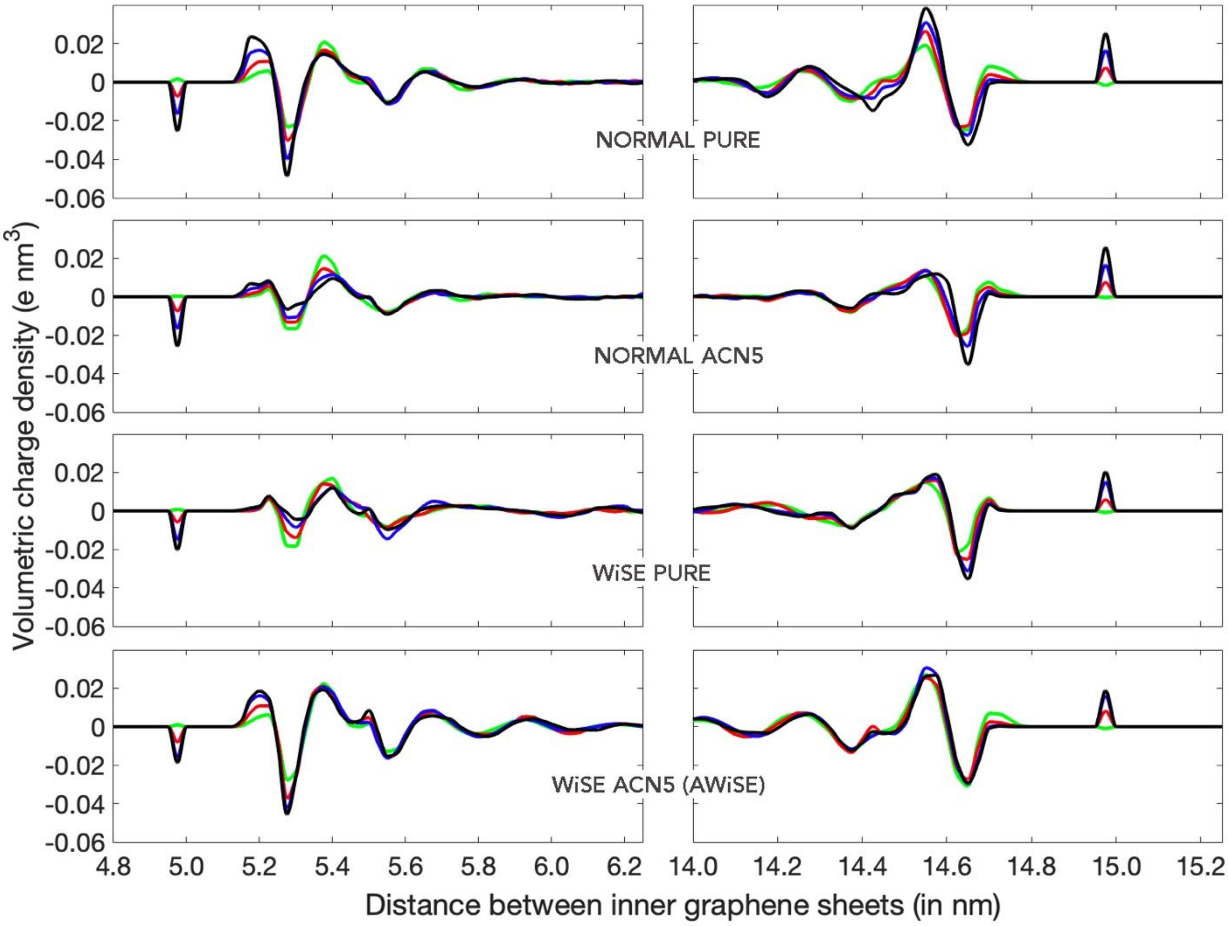

Figure S1: Volumetric charge density profile (e nm-3) as a function of the separation of the electrodes for the four types of supercapacitors investigated. 0V, green curve; 1V, red curve; 2V, blue curve; and 3V, black curve. The graphs on the left show the cartoons in the electric double layer near the negative electrode and those on the right show the cartoons near the positive electrode.

Tabela S3: Densidade superficial de carga ($\mu C\ cm^{-2}$) nos eletrodos, para cada supercapacitor em função da voltagem aplicada (V).

Voltage (V)	NORMAL-PURE	NORMAL-ACN5	AWiSE-PURE	AWiSE-ACN5
	POSITIVE ELECTRODE			
0.0	0.6	0.2	0.3	0.0
0.5	1.3	1.4	1.7	1.3
1.0	2.6	2.6	3.0	2.1
1.5	4.3	4.4	4.5	4.1
2.0	5.7	5.8	6.1	5.2
2.5	7.0	7.5	7.1	7.0
3.0	8.8	9.1	8.2	8.1
	NEGATIVE ELETRODE			
0.0	-0.6	-0.2	-0.2	-0.3
0.5	-1.3	-1.4	-1.6	-1.2
1.0	-2.6	-2.6	-2.9	-2.3

1.5	-4.3	-4.4	-4.4	-4.1
2.0	-5.7	-5.8	-6.1	-5.2
2.5	-7.1	-7.5	-7.1	-7.0
3.0	-8.8	-9.1	-8.2	-8.0

Figure 11 shows the electrostatic potential profiles through the supercapacitor. We can observe that all of them present typical behavior with strong oscillation in the double electric layer and having the same signal of the charge induced in each electrode. From each of these potentials we obtained the potential drop between the electrodes, obtained as the difference between the potential in the positive electrode and the potential in the negative electrode, i.e.:

$$\Delta\Phi = \Delta\Phi(+) - \Delta\Phi(-)$$

Such values for $\Delta\Phi$ already include the correction for the potential zero charge, which, as the name says, is the residual potential that arises even in the absence of an external voltage applied to the supercapacitor.

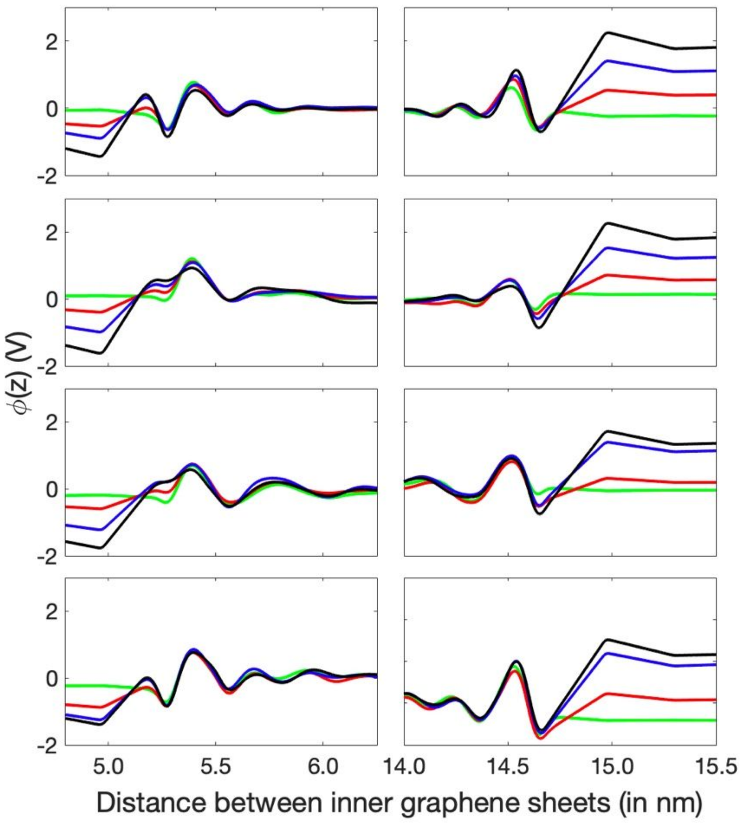

Figure 11: Profile of electrostatic potential (V) as a function of the separation of the electrodes for the four types of supercapacitors investigated. 0V, green curve; 1V, red curve; 2V, blue curve; and

3V, black curve. The graphs on the left show the charges in the double layer near the negative electrode and those on the right show the charges near the positive electrode.

The value of the drop potential across the supercapacitors is shown in Table 6. Again, we observed that, in general, it was observed that the potential modulus in each electrode, $\Delta\Phi(+)$ and $\Delta\Phi(-)$, grows linearly with the increase in the applied external voltage. The values calculated for $\Delta\Phi$ already include the correction for the potential zero charge (PZC), which, as the name says, is the residual potential that arises even in the absence of an external voltage applied to the supercapacitor.

Tabela 6: Drop potential across the supercapacitors (in V) as function of applied voltage (V).

	NORMAL-PURO			NORMAL-ACN5		
Voltagem (V)	$\Delta\Phi(-)$	$\Delta\Phi(+)$	$\Delta\Phi$	$\Delta\Phi(-)$	$\Delta\Phi(+)$	$\Delta\Phi$
0.0	0.0	0.0	0.0	0.0	0.0	0.0
0.5	-0.3	0.4	0.8	-0.2	0.3	0.5
1.0	-0.5	0.8	1.3	-0.5	0.6	1.1
1.5	-0.7	1.4	2.1	-0.8	1.0	1.8
2.0	-0.9	1.6	2.5	-1.1	1.4	2.5
2.5	-1.1	2.1	3.2	-1.4	1.8	3.2
3.0	-1.4	2.5	3.9	-1.7	2.1	3.8
	AWiSE-PURO			AWiSE-ACN5		
Voltagem (V)	$\Delta\Phi(-)$	$\Delta\Phi(+)$	$\Delta\Phi$	$\Delta\Phi(-)$	$\Delta\Phi(+)$	$\Delta\Phi$
0.0	0.0	0.0	0.0	0.0	0.0	0.0
0.5	-0.4	0.4	0.8	-0.3	0.3	0.6
1.0	-0.6	0.6	1.3	-0.5	0.6	1.1
1.5	-0.8	1.1	2.0	-0.9	0.9	1.9
2.0	-1.0	1.6	2.6	-1.2	1.3	2.6
2.5	-1.2	1.9	3.1	-1.5	1.7	3.2
3.0	-1.4	2.3	3.7	-1.8	2.0	3.8