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Double layer capacitance of ionic liquids for electrolyte gating of ZnO thin film transistors and effect of gate electrode.

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Supplementary Information

Analysis of the electrochemical impedance spectroscopy (EIS)

The Warburg impedance element represents the contribution to the impedance of semi-infinite diffusion mechanism to/from a flat electrode. It contributes equally to the real and imaginary parts of impedance i.e.:

$$Z_W = \frac{A_W}{\omega^{0.5}}(1 - j) \quad (1)$$

where ω is the angular frequency of the AC voltage signal, j is the imaginary unit, A_W the Warburg coefficient, depending on diffusion coefficient of ionic species, *D*, and their surface concentration.

The Constant Phase Element (CPE) is used to represent the response of non-ideal and non-homogeneous systems[1]. The expression for such an impedance is given by:

$$Z_{CPE} = \frac{1}{Q(j\omega)^n} \quad (2)$$

Depending on the value of n (ranging from -1 to 1) this element may in turn represent a non ideal capacitor, an inhomogeneous resistance or either an inhomogeneous diffusion mechanism. If n=1 the CPE becomes an ideal capacitor, i.e. Q = C. Similarly, if n = 0 then Q = 1/R (being R an ideal resistance), or with n=0.5 the Z_{CPE} reduces to the ideal Warburg diffusion impedance.

For comparison purposes a capacitance *C(ω)* can be calculated from Z_{CPE} provided that n≈1. Under such a condition the following equivalence holds:

$$|Z_{CPE}| = \frac{1}{Q\omega^n} = \frac{1}{C(\omega)\omega^1}$$
(3)

where $|Z_{CPE}|$ being the modulus of Z_{CPE}, one can write:

$$C(\omega) = Q\omega^{n-1} \tag{4}$$

The impedance of the Warburg Short (W_s) diffusion element can be written as

$$Z_W = \frac{W_{sr}}{\omega^{0.5}}(1 - j)tanh\left(W_{sc}\,(j\omega)^{0.5}\right) \tag{5}$$

and represents a finite-length diffusion mechanism with transmissive boundary[2].

W_{Sr} is equal to Warburg coefficient and W_{SC} = d/$D^{0.5}$, where d is the Nernst diffusion layer thickness.

Table S1 shows the impedance parameters obtained after fitting the data with the two equivalent circuit models presented in the manuscript.

Table S1: Best-fit Impedance parameters

	[bmim][PF$_6$]		[bmim][BF$_4$]	
Fitted parameters	At 0V	At -1.5V	At 0V	At -1.5V
R1(ohm)	333.7	191.1	87.8	85.0
R2(ohm)	4.3×10^5	4.04×10^5	1.2×10^5	1.2×10^5
CPE1	1.2×10^{-7}	3.8×10^{-7}	4.0×10^{-7}	6.8×10^{-7}
CPE2	1.7×10^{-7}	--------	2.1×10^{-7}	--------
Ws(ohm)	-------	4.7×10^5	---------	1.6×10^5
n1	0.9	0.9	0.9	0.9
n2	0.7	---------	0.5	--------

References

[1] Barsoukov, E. & Macdonald, J. R. *Impedance Spectroscopy. Impedance Spectroscopy: Theory,*

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[2] Bisquert, J., Garcia-Belmonte, G., Fabregat-Santiago, F. & Bueno, P. R. Theoretical models for ac impedance of finite diffusion layers exhibiting low frequency dispersion. *J. Electroanal. Chem.* **475,** 152–163 (1999).