Supporting Information

The Electrical Double Layer Structure of Dicationic

Ionic Liquids at Onion-Like Carbon Surface

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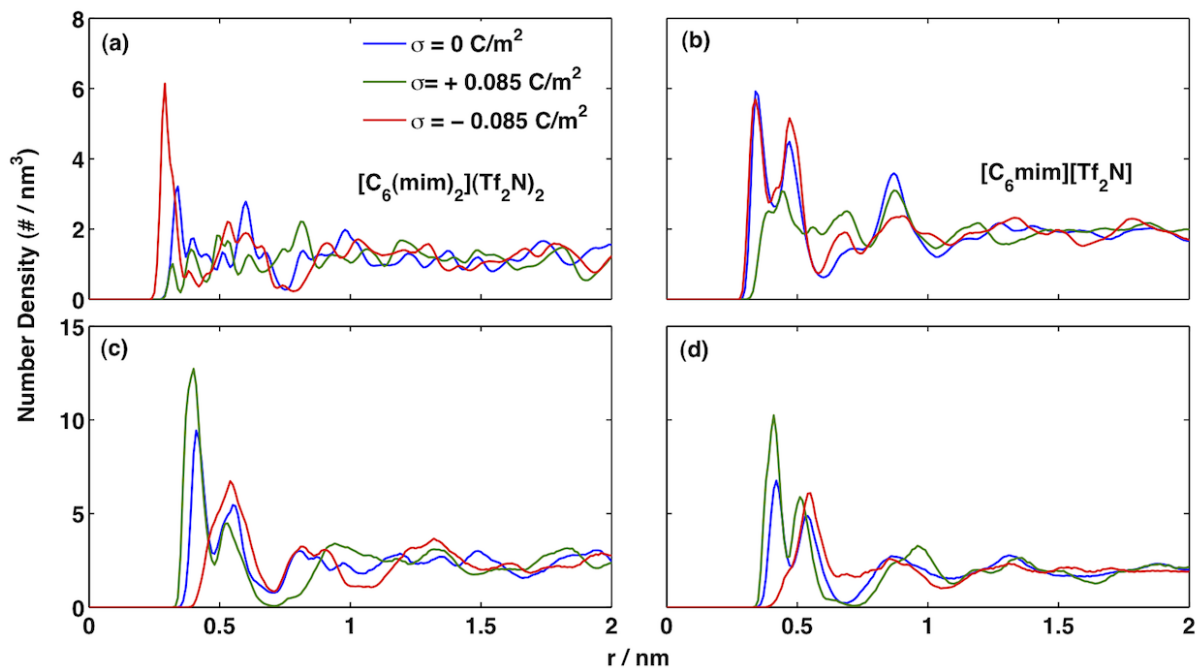[‖]School of Energy and Power Engineering, Huazhong University of Science and Technology, Wuhan 430074, China

Figure S1. Number density profile for dicationic ionic liquid (DIL) $[C_6(mim)_2](Tf_2N)_2$ and monocationic ionic liquid (MIL) $[C_6mim][Tf_2N]$. The top panels (a-b) are for cations and bottom panels (c-d) are for anions.
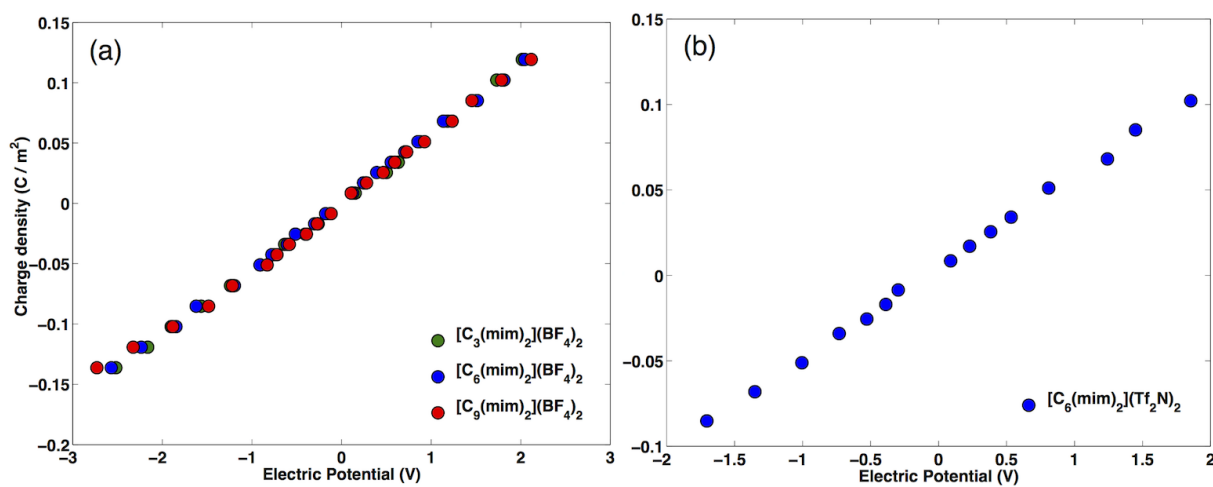

Figure S2. Electrode surface charge density as a function of electric potential for (a) DIL $[C_n(mim)_2](BF_4)_2$ (n=3, 6, 9) and (b) MIL $[C_6(mim)_2](Tf_2N)_2$.
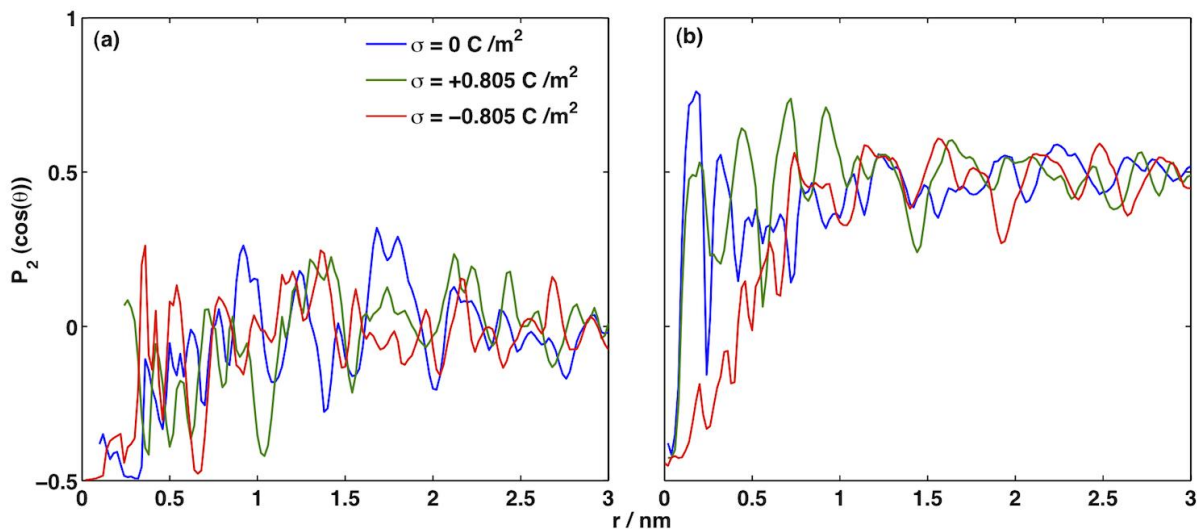
Figure S3. Orientational order parameter as a function of distance from OLC surface for the alkyl chain (a) and the imidazolium rings (b) in DIL $[C_6(mim)_2](Tf_2N)_2$. Panel (a) is for the angle formed by the linkage alkyl chain with respect to the surface normal of the OLC; panel (b) is for angle formed by the plane of imidazolium ring with respect to the surface normal of OLC.
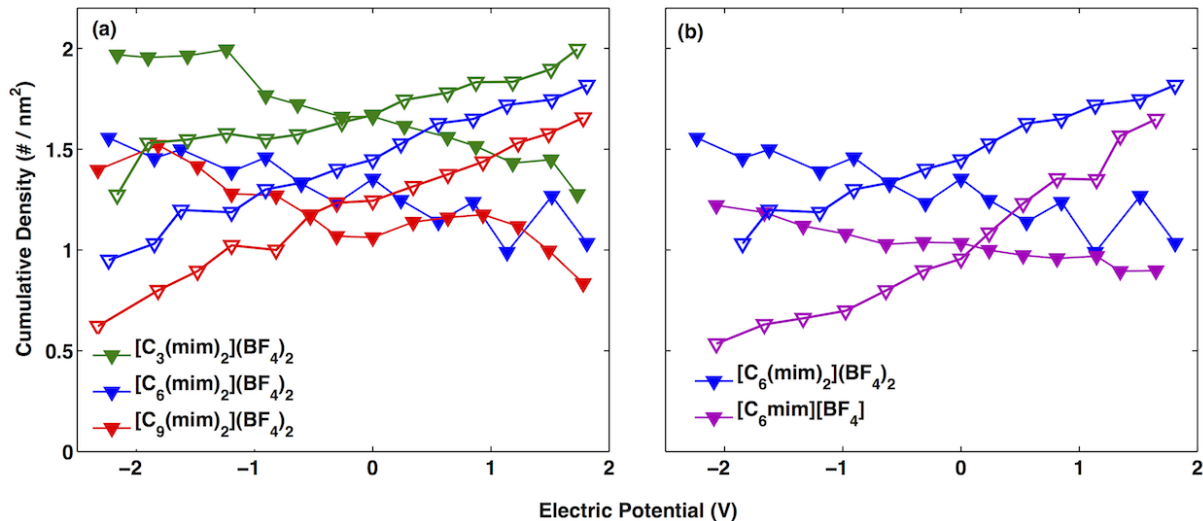

Figure S4. Cumulative density of charges in the electric double layer as a function of electric potential applied from MD simulation for $[C_n(mim)_2](BF_4)_2$ (a) and dicationic $C_6(mim)_2(BF_4)_2$ versus monocationic $[C_6mim][BF_4]$ (b), which is represented by the product of the cumulative number density and the charges carried by ions (i.e., cations and anions). The solid symbols denote cations and hollow symbols are for anions.
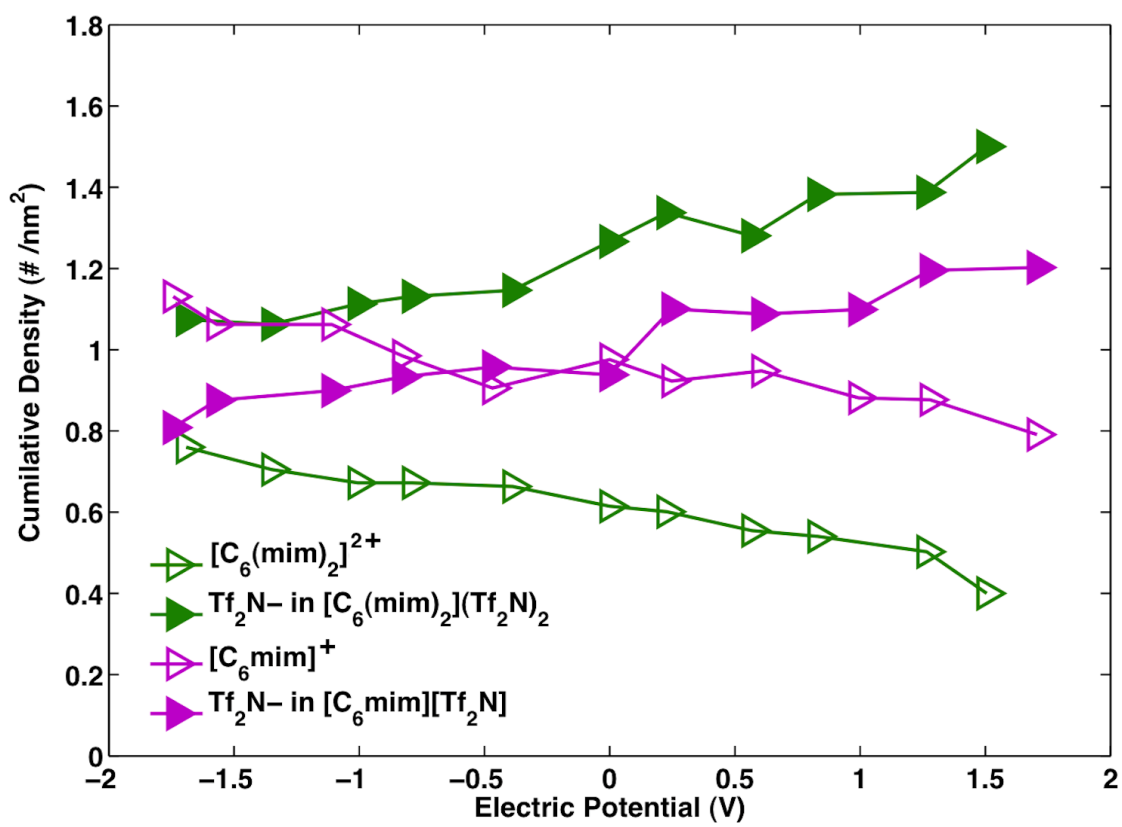
Figure S5. Cumulative density of ions center-of-mass in the electric double layer as a function of electric potential applied for dicationic $[C_6(mim)_2](Tf_2N)_2$ and monocationic $[C_6mim][Tf_2N]$.
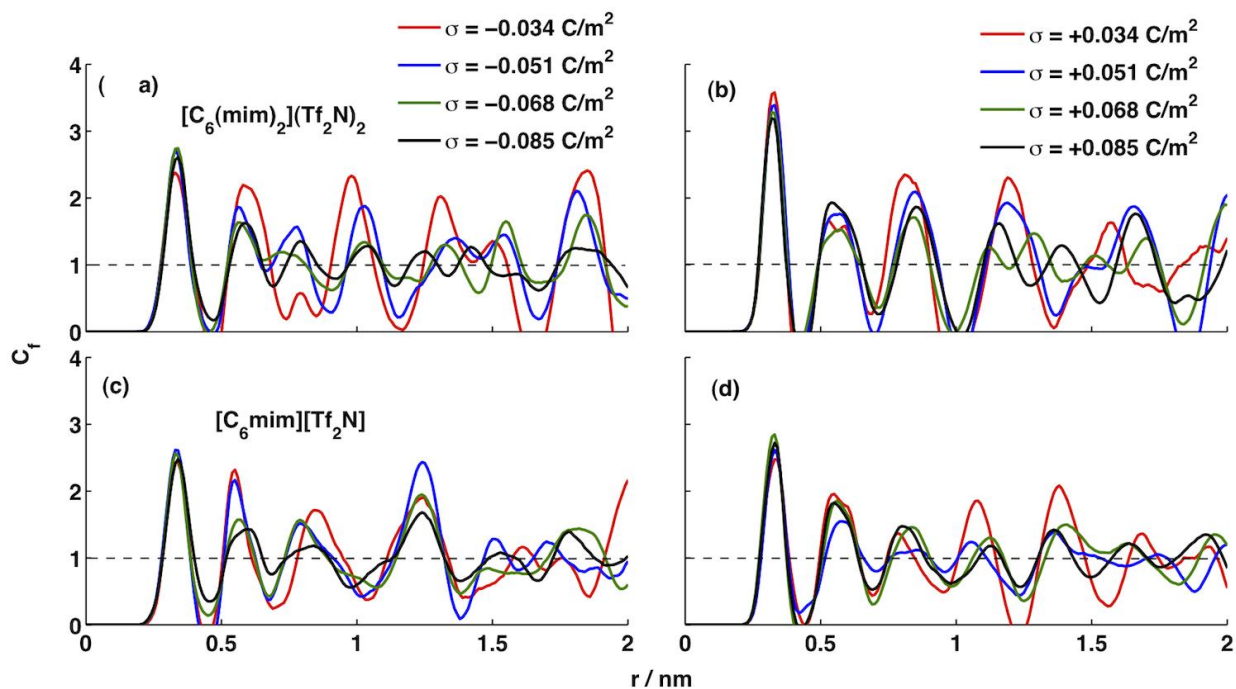
Figure S6. Charge screening factors for dicationic $[C_6(mim)_2](Tf_2N)_2$ and monocationic $[C_6mim][Tf_2N]$ at varying charge densities of electrode surface. The left panel is for negatively charged electrode with the electric potential within -1.5 ~ -0.5 V and the right panel is for positively charged electrode with the electric potential with 0.5 ~ 1.5 V.
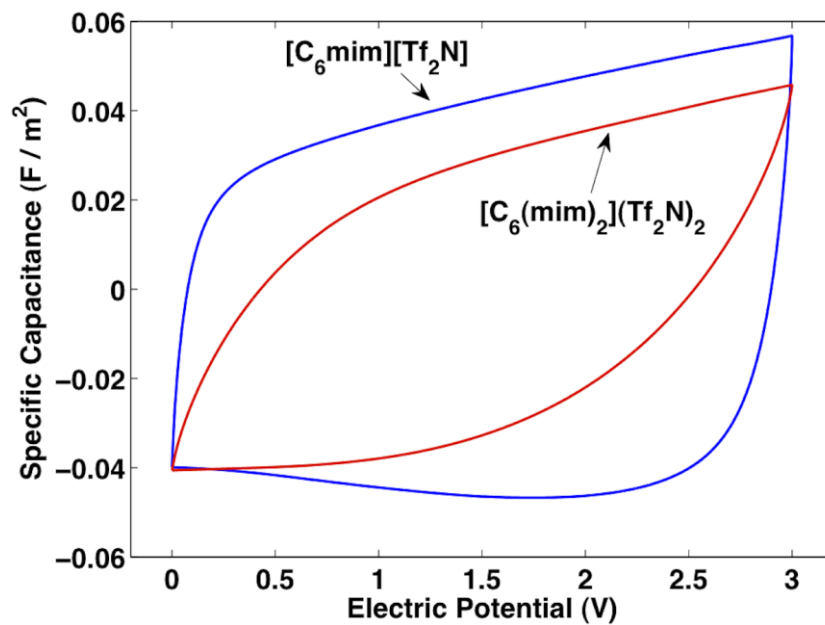

Figure S7. Cyclic voltammogram of OLC electrode in the electrolytes of dicationic $[C_6(mim)_2](Tf_2N)_2$ and monocationic $[C_6mim][Tf_2N]$ recorded at the scan rate of 50 mV/s.