

## Supporting Information

# Weighing the Surface Charge of an Ionic liquid

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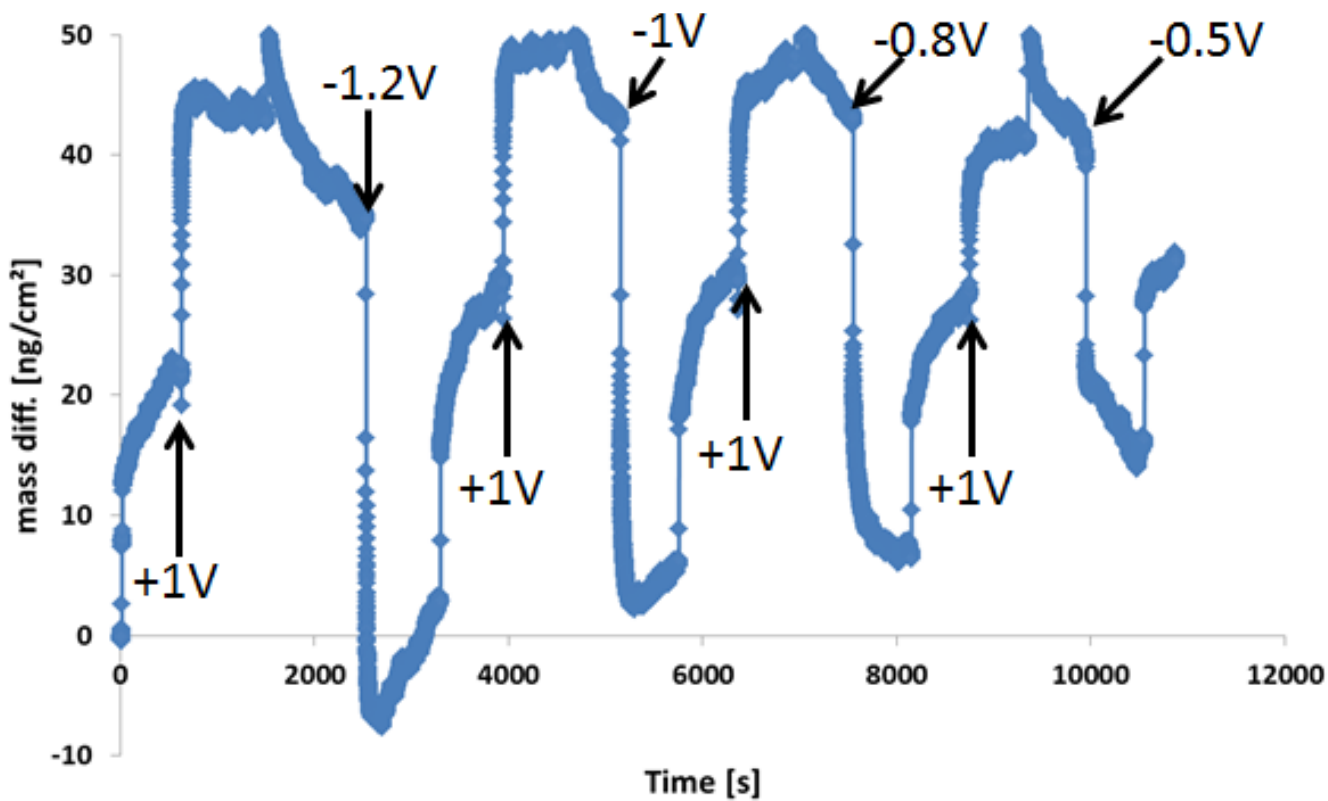
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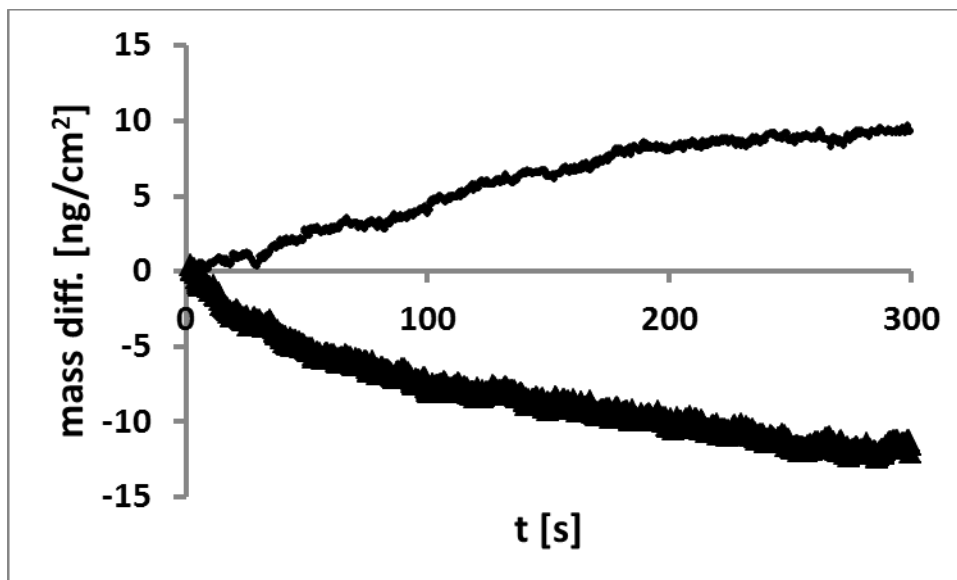
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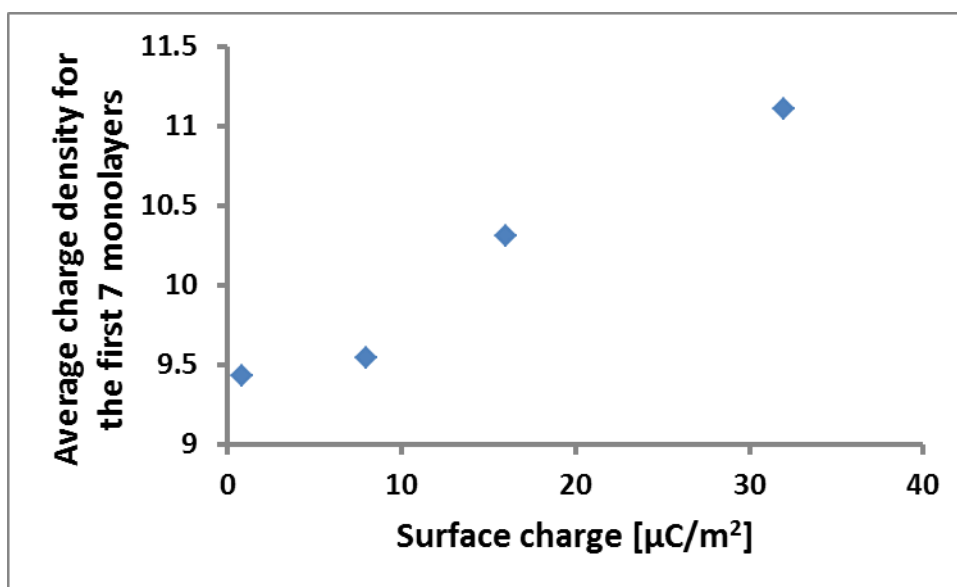
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SI 1. An entire load/unload cycle is shown here. The applied voltage is changed in a certain pattern: +V, 0V, -V, 0V,..., 0V . The positive potential is +1V in every case and the negative potential is systematically decreased -1.2V, -1V, -0.8V, -0.5V.



SI 2. Mass change plotted as a function of time when potential is turned off for +1V (thick line) and -1V (thin line). Equilibrium is reached at 250-300s.



SI 3. Data points extracted from Figure 2 in Ref<sup>1</sup> and plotted as an average charge density against surface charge.

In SI 3 data points for the charge density of the first seven monolayers were extracted<sup>1</sup> and averaged for four surface charges. The charge density hardly changes at low surface charges but encounters a clear transition and increases by 10 to 20% as the surface charge is further increased (in the transition from overscreening to crowding). If there were only a linear (incompressible) response to surface charge in SI 3 there would be no break in Figure 5. This strongly suggests that the clear change in gradient in Figure 5 corresponds to the changes in viscosity and density associated with the overscreening/crowding transition. A quantitative comparison is not possible due to the fact that 1) the simulations are performed on spheres of rather different size and with identical sized anions and cations so do not take into account the more complex packing behaviour of the real ions, nor their size/mass difference. 2) Both the *density* and *viscosity* affect the observed QCM response.

### Cleaning protocol

The QCM accessories were cleaned in two steps: Firstly, by sonicating them for 15 min in a 0.5 wt% solution of Extran MA 03 (Merck KGaA) and secondly by sonicating them for 15 min in water (all water used has a resistivity of 18.2 M $\Omega$ ·cm). All accessories were rinsed in copious amounts of water after each step.

### References

1. M. V. Fedorov and A. A. Kornyshev, *Electrochim Acta*, 2008, **53**, 6835-6840.