

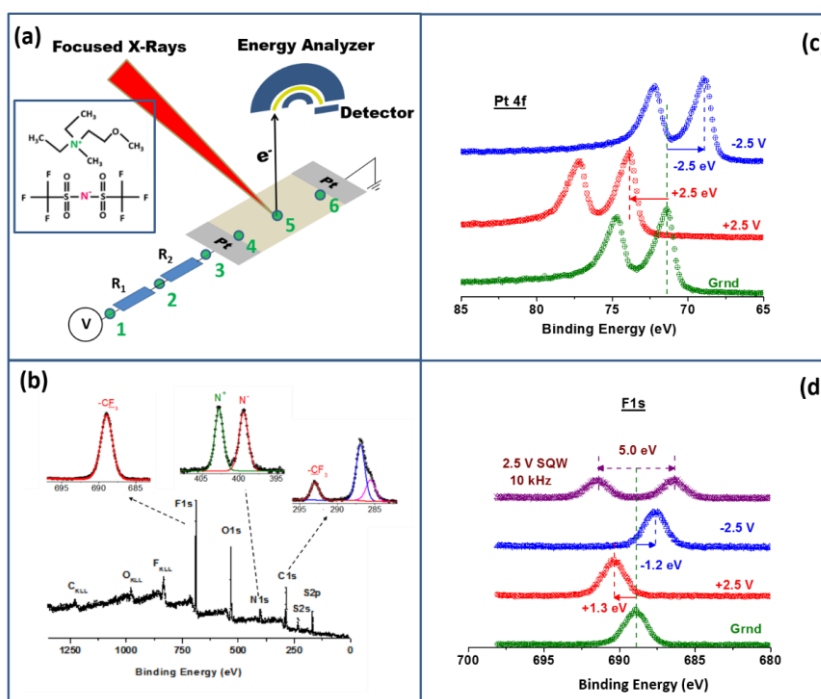
# Local Potential Determinations by XPS Provides the Missing Link about Charge Dynamics of Ionic Liquid Devices

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Ionic liquid materials show rich dynamic responses on electrified surfaces. A long-standing question is why these materials exhibit very different time responses, ranging from microseconds to several hours, which are attributed to the complex interplay of chemical and physical factors, including steric and molecular interactions. However, experimental observations and theoretical predictions do not always match in IL based devices. Particularly considering the overall electrode capacitance resulting from asymmetric ion size effects, the local voltage levels can show large differences, and drift-diffusion process of ions generally fails to account for them. To clarify this point, we have utilized a combination of electrochemical devices with different chemical make-up, geometry and dynamic XPS analysis and modelling, and were able to probe the spatiotemporal voltage profiles directly from the shifts in the binding energy positions of the ionic core levels, within IL based devices. Results from a number of single and mixed ILs, as well as different device geometries will be presented and discussed.



**Figure (a).** Schematics of the co-planer capacitor, containing optional and additional resistors ( $R_1$  and  $R_2$ ), for probing dynamics of charging of the ionic liquid as the electrolyte, by the XPS probe beam, under *operando* conditions, where both current and peak positions are recorded simultaneously at different locations (green dots), also indicated as 1, 2, 3, 4, 5 and 6. The function generator is connected to the source electrode to impose a DC, or an AC (SQW) excitation with a fixed or variable frequency. Inset shows the chemical formula of the IL. **(b).** Survey and region's XP spectra of the IL. **(c).** Pt4f XP Spectra, recorded at the source electrode and under 0, +2.5 and -2.5 V DC bias, and without the additional resistors. **(d).** F1s XP Spectra, recorded near the source electrode (position 4), under 0, +2.5 and -2.5 V DC bias, as well as under 2.5 V SQW excitation at 10 kHz.