Electrowetting of [EMIm][FSI] ionic liquid on HOPG: *In-situ* Electrochemical XPS probing

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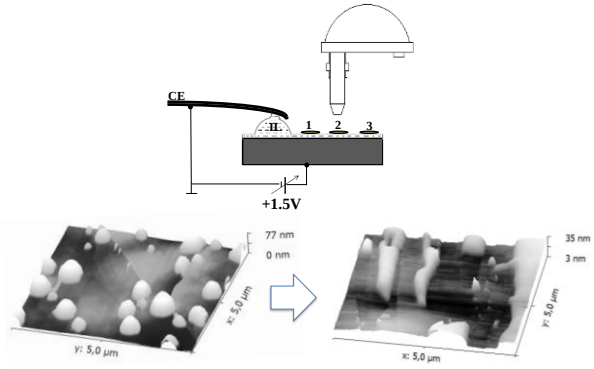

We report asymmetric electrowetting of [EMIM][FSI] ionic liquid on HOPG surface evidenced by the *in situ* electrochemical XPS analysis and confirmed by *in situ* SEM imaging and *ex situ* AFM imaging. The latter was performed under atmospheric pressure. The IL films were prepared via physical vapor deposition. Uncommon open design of the IL/electrode electrochemical system -with a macroscopic IL droplet (~∅1 mm) acted as a counter contact to the ultrathin ionic liquid film- allowed to perform *in situ* electrochemical XPS with lateral space resolution. At positive electrode potential the IL film undergoes transition from the “drop-on-layer” structure to the flat top layered structure and reversibly recovers the initial “drop-on-layer” structure upon switching back to cathodic polarization. The morphological transitions occur on the time scale of ~10-30min depending on the IL film thickness. Potential profile across the IL film on the nm scale can be inferred by mapping electrochemical shifts of the IL related XPS lines with lateral resolution along the IL film. Gauging potential distribution across the ionic-liquid/electrode interface is the indispensable tool to gain knowledge on the charge storage mechanisms in the electrical energy storage systems and about the forces controlling the basic functions of the microfluidics and lab-on-chip systems: droplet splitting, mixing, and dispensing.