**Reconstructing 3D nanoarchitecture** **of solid-liquid interface – from atomic scale electrical double layer to solid-electrolyte interphase**

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We use advanced scanning probe microscopy to explore one of the most enigmatic and, arguably, most important parts of rechargeable batteries – the solid electrolyte interphase (SEI) layer, its dynamics and, significantly, its nanoscale structure and composition that hold clues to high-performing and safe energy storage. We linked electrochemical AFM and a recently developed 3D nano-rheology scanning probe microscopy to study *in-situ* the dynamic formation of SEI starting from a few Å thick electrical double-layer (EDL) to the full 3D nanostructure of SEI. Nanoscale rheology maps allow to quantify in real-time 3D distribution of various components in the as formed SEI layer, revealing the dominant role of a solvent-guided molecular-scale EDL structure. Instigated by energy storage applications, this study establishes a new generic approach allowing to explore the nanoscale internal structure of soft materials via 3D mapping of their local nanorheological properties – with potential applications ranging from biological objects to artificial biomaterials to industrial coatings.

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