

# Visualizing Live-formation of Interphase at Atomic Scale

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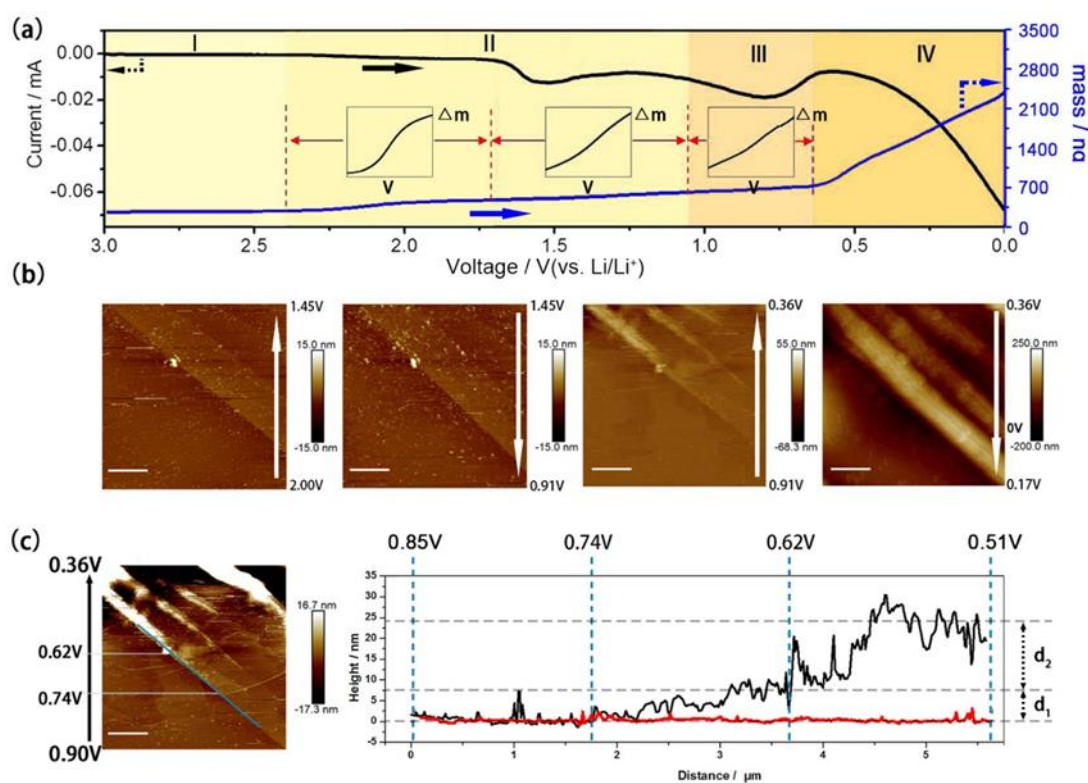
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The solid-electrolyte-interphase (SEI) in Li-ion battery has been considered the most important but least understood and elusive component. In this work, we achieved the quantitative and atomistic observation of the live-formation of interphase on graphitic electrode in non-aqueous electrolyte by using atomic force microscope (AFM) and electrochemical quartz crystal microbalance (EQCM) for the first time. Meanwhile, we found that while co-intercalation of solvated  $\text{Li}^+$  directs the formation of SEI at edge-sites of graphite, there is no apparent staging for the co-intercalation process. We also accurately weighed the mass accumulation during the formation of SEI, and confirmed for the first time the generation of LEDC as the main SEI component. Most interestingly, we found that this nascent interphase, after its initial formation, is re-oxidizable upon the reversal of electrode potential in the first de-lithiation process, whose product is again accurately weighed. These findings rewrite the laws governing the formation chemistry, mechanism and properties of SEI, which dictates the reversibility, rate-capability and energy output of the advanced batteries.



**Figure 1.** (a) CV scan (black) of the fresh graphite electrode in 1 mol/L  $\text{LiPF}_6$  EC/DMC at 1 mV/s from OCV (3.0 V) to 0.0 V, and the simultaneous EQCM responses (blue) recorded; (b) AFM topological imaging of HOPG during the CV scan, where the long white arrows indicate

the scan directions; and (c) AFM imaging of first SEI formation scanning from 0.90 V to 0.36 V and the height distribution of SEI.