Quantitative Operando Mapping with Nanoscale Resolution of 3D Mechanical Properties of Surface Electrolyte Interphase in Li-Ion and Na-Ion Batteries When and Where



Apr 24, 2024 9:45am - 10:00am



Room 424, Level 4, Summit

Presenter

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Abstract

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A nanoscale thin but extremely important component of any rechargeable battery is the solid electrolyte interphase (SEI), a passivation layer that that defines the fundamental battery properties - capacity, cycle stability and safety. Local mechanical properties of SEI hold a clue to its performance, but their <i>operando</i> characterisation is very difficult as one has to probe nanoscale surface features in electrochemical environment that are also dynamically changing. Here, we report novel 3D nano-rheology microscopy (3D-NRM) that uses a tiny (subnm to few nm) lateral dithering of the sharp SPM tip at kHz frequencies to probe the minute sample reaction forces. By mapping the increments of the real and imaginary components of these forces, while the tip penetrates the soft interfacial layers, we obtain the true 3D

nanoscale structure of sub-mm thick layers. 3D-NRM already allowed us to elucidate key role of solvents in SEI formation and predict the conditions for building SEI for robust, safe and efficient Li-ion batteries [1]. We combine 3D NRM with magnetic excitation to investigate molecular-level solvation force spectroscopy and use molecular dynamics simulations to understand two morphologically dissimilar but chemically identical surfaces of typical carbon electrode material (basal and edge graphene planes) and different solvent-electrolyte systems (strong and weakly solvating electrolytes, as well as ionic liquid electrolyte). These approaches allowed us to get direct insight into the atomistic pictures for the underlying influence of cation's intercalation and solvation structures on the initial SEI formation.

Furthermore, here we explore the extension of these studies on smooth HOPG and inhomogeneous and rough copper anodes as sodium ion battery electrodes. Essentially, the new approach allows nanoscale characterisation of SEI with a few nm precision on the electrodes with 1000 nm roughness, and, importantly, to quantitatively evaluate the real and imaginary parts of the elastic moduli over the whole thickness of SEI layer. The observation of the change in moduli and the tip-surface distance helps to evaluate the growth of SEI as a function of the electrolyte, additives, electrode material and charge-discharge rate. Our understanding of these key interfacial structural factors in SEI formation allows targeting an electrochemically and mechanically robust surface passivation layer and guiding the development of efficient and safe rechargeable batteries. We believe that such evaluation of key interfacial nanomechanical properties of SEI will allow us to develop the electrochemically and mechanically robust SEI surface passivation layer and the development of efficient and safe rechargeable batteries.

| 1] Y Chen, W Wu, S Gonzalez-Munoz, L Forcieri, C Wells, SP Jarvis, F Wu, R Young, A Dey, M Isaacs, M Nagarathinam, RG Palgrave, N Tapia-Ruiz, OV Kolosov, Nature Comm 2023, 14, 1321.

Keywords

nanostructure | scanning probe microscopy (SPM)

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