Modeling Electrified Interfaces in Batteries

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Electrochemical interfaces are essential components in electrochemical energy conversion and storage devices; and the electric double layer (EDL) is a fundamental concept used to describe the electrified interfaces. However, the century-old EDL concept, rooted in continuum approaches, lacks atomistic details. Additionally, the classical Poisson–Boltzmann EDL model, originally designed for fully solvated ions, becomes inadequate for the new and emerging electrolytes in batteries, such as high concentration liquid electrolytes (HCE), localized high concentration liquid electrolytes (LHCE), and solid electrolytes (SE). **This presentation** aims to tackle the challenges associated with constructing predictive models of charged interfaces, specifically focusing on Li-metal/electrolyte interfaces in batteries. At these interfaces, two critical charge transfer reactions unfold: the desired ion transfer reaction of lithium during each charge/discharge cycle and a set of electron transfer reactions leading to the undesirable chemical decomposition of the electrolyte and the formation and growth of the solid electrolyte interphase (SEI). To address these challenges, we start from density functional theory (DFT)-informed continuum models to discuss the electric potential alignments in both full cell and half-cell configurations, encompassing both solid and liquid electrolytes. [1,2] After determining the surface charge state at Li-plating, an interactive Molecular Dynamics - DFT - data statistics model is developed to analyze the reduction reactions of multicomponent electrolytes within the EDL. The interplay among cations, anions, and various solvent species with a charged surface at different temperatures collectively influences the EDL structure and, consequently, the composition of the SEI.[3] Moreover, we will introduce a novel EDL model in SE and SEI by solving the DFT-informed Poisson–Fermi–Dirac equation, showcasing its utility in interlayer thickness design.[4] Additionally, the presentation will explore the EDL structures in electrolytes with heterogeneous structures, such as LHCE.[5] These predictions are supported by experimental measurements. They not only provide valuable insights but also offer guidance and toolset for the direct design of interfaces in batteries.

[1]Energy & Environmental Science 12 (4), 1286-1295 (2019); [2]Physical review letters 122 (16), 167701 (2019); [3]Journal of the American Chemical Society 145 (4), 2473-2484 (2023); [4]Nature Computational Science 1, 212–220 (2021); [5]Nature Materials 22, 1531–1539 (2023).

Speaker Bio:

Dr. Yue Qi is the Joan Wernig Sorensen Professor of Engineering at Brown University. She obtained her B.S in Materials Science and Engineering and Computer Science at Tsinghua University in 1996. After receiving her Ph.D. from Caltech, she spent 12 years working at the General Motors R&D Center. At GM, she developed multi-scale models starting from the atomistic level to solve engineering problems related to lightweight alloys, fuel cells, and batteries. She transitioned from industry to academia in 2013 and served on the faculty in the Chemical Engineering and Materials Science Department at Michigan State University (MSU) till 2020. Professor Qi and her “Materials Simulation for Clean Energy” Lab develop multi-scale simulation methods to design materials and interfaces that are critically important for an energy-efficient and sustainable future. She has received several awards for her research, including the co-recipient of the 1999 Feynman Prize in Nanotechnology for Theoretical Work for her Ph.D. work; three GM Campbell awards for fundamental research on various topics while working in GM; and the 2017 Minerals, Metals & Materials Society (TMS) Brimacombe Medalist Award for her contributions in multidisciplinary computational materials science.