Abstract

Suppressing dendritic growth in metal anode based rechargeable batteries is one of the major research focus for critical commercial applications. In this regard, modulation of charging strategy, among other methods, has gained increasing attention in recent days. The present thesis delves into the understanding of the transient behavior of the growing electrode-electrolyte interface under a variable applied voltage instead of a constant DC. Herein, the evolving interface has been tracked in real-time and characterized by image analysis to quantify the velocity of the growing front, height of electrodeposits, and the extent of the interfacial unevenness. The work begins with the application of different shaped voltage ripples (such as sinusoidal and square), followed by the application of voltage pulses with a varied range of commercial pulses parameters, namely, duty cycle and frequency. Furthermore, a fundamental question is probed whether the simulation results from an atomistic consideration can be directly compared to the experimentally obtained results because of the existence of a large scale gap. To answer this, a dynamically equivalent atomistic paradigm has been established, wherein the ions are in identical harmony as in continuum. The thesis also examines electrodeposition from the nucleation point of view. Nucleation induced island formation in electrodeposition has been explored while applying voltage pulses of varying duty cycle and frequency along with the different triangular pulse waveforms. The work evaluates the limits of duty cycle till which pulses generate almost identical island density (in spite of having periodic OFF-time in pulses) with the DC voltage being the same as the pulse peak voltage. Additionally, the effects of the application of different triangular pulses instead of square pulses on the island density are also explored with relevant explanations using a first principle based approach. The thesis provides new insights to the fundamental aspects of ionic transport in solution, the interfacial dynamics at the electrode-electrolyte boundary, and nucleation-induced island formation.

Keywords

Interface, Dendrites, Real-Time Visualization, Growth Front Velocity, Tortuosity, DC Ripples, Duty Cycle, Frequency, Transition Time, Continuum, Atomistic, Nucleation, Island Density, Rectangular Pulses, Triangular Waveforms