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Stress evolution in elastic-plastic electrodes during electrochemical processes: A numerical method and its applications

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ABSTRACT

Monitoring in real time the stress state in high capacity electrodes during charge-discharge processes is pivotal to the performance assessment and structural optimization of advanced batteries. The wafer curvature measurement technique broadly employed in thin-film industry, together with stress analysis using the Stoney equation, has been successfully adopted to measure in situ the stress in thin film electrodes. How large plastic deformation or interfacial delamination during electrochemical cycles in such electrodes affects the applicability of Stoney equation remains unclear. Here we develop a robust electrochemical-mechanical coupled numerical procedure to investigate the influence of large plastic deformation and interfacial failure on the measured stress in thin film electrodes. We identify how the constitutive behavior of electrode materials and film-substrate interfacial properties affect the measured stress-capacity curves of electrodes, and hence establish the relationship of electrode material parameters with the characteristics of stress-capacity curves. Using Li-ions batteries as examples, we show that plastic deformation and interfacial delamination account for the asymmetric stress-capacity loops seen in in situ stress measurements. The methods used here, along with the finite-element code in the supplementary material, may be used to model the electrode behavior as a function of the state of charge.

1. Introduction

High charge capacity anode materials like Si and Sn have attracted great interests in recent years (Bourderau et al., 1999; Liang et al., 2014; Huggins, 1999; Winter and Besenhard, 1999). One major tradeoff arisen from the high capacity is the resulted high stress. Lithiation and de-lithiation in anode materials inevitably result in huge volume expansion and contraction, lead to high internal stresses and, thus, poor cycle life of high capacity anode batteries (Shi et al., 2016; Klinsmann et al., 2016; Zhang et al., 2016). Many strategies were proposed to prolong the service duration of anode materials. One popular approach was to optimize the microstructure of the materials (Szczech and Jin, 2011; Zhang et al., 2017) to alleviate internal stress induced by volume expansion. To examine the effectiveness of such structural optimization, we desire to have

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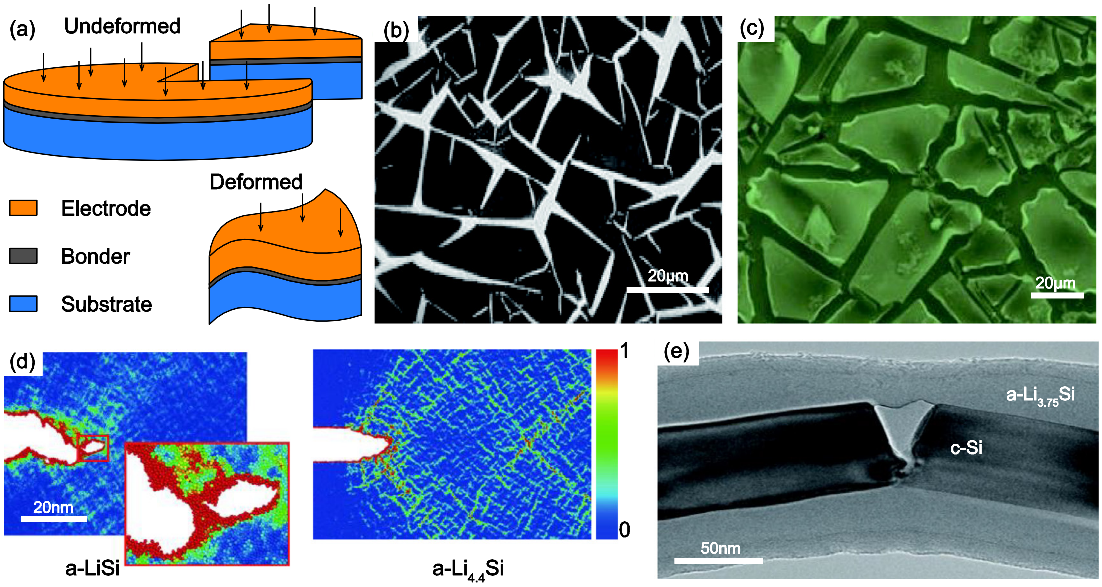

Fig. 1. Illustrations of high capacity battery electrodes undergone large deformation and interfacial failure: (a) A film-bonder-substrate system for in situ curvature monitoring. (b) Optical micrograph showing micro-cracks in amorphous silicon thin film electrode after charge and discharge. (c) SEM picture to show interfacial delamination and cracking in the thin film electrode after a certain amount of cycles (b and c, Courtesy of Yang et al., 2018). (d) Molecular dynamic simulations showed brittle to ductile transition, with cavitation dominated failure in low lithium concentration amorphous silicon (left) versus micro-shearing in high lithium concentration amorphous silicon (right, Courtesy of Ding et al., 2015). (e) Experimental observation on ductile deformation in amorphous silicon with high lithium concentration against brittle fracture in crystalline silicon (Courtesy of Wang et al., 2015).

an accurate and real time measurement of stress in high capacity electrodes (Sethuraman et al., 2010, 2012; Duan and Wei, 2017). Real time stress is also indispensable to better our understanding about the electrochemical-mechanical processes and is helpful for faithful model development for electrode materials.

Stresses in thin film electrodes may be measured by monitoring the bending of the electrode-substrate bilayer and using the Stoney equation. A typical battery setup for such in situ stress measurement is shown in Fig. 1a, which consists of a thin film, a bonder layer, and a substrate. Using this method, Sethuraman et al. (2010, 2012) showed how stress changes in Si- and graphite-based anode film materials during charging and discharging. Later on, Soni et al. (2011) and Haftbaradaran et al. (2012) used the same method to investigate the stress–capacity behavior of patterned electrode islands, aimed to illustrate the influence of interfacial failure between electrode islands and the substrate on capacity degradation (He et al., 2012). Recently, Pharr et al. (2016) measured stresses in sputter-deposited amorphous Ge thin films as the electrode material. The stress-capacity curves from different research groups all showed a hysteresis loop, which was caused by irreversible deformation during charge-discharge cycles. In addition, the electrode-substrate interface may be subjected to excessive deformation and may fail during cycling. According to the in situ tests, fracture and delamination of the film electrode from substrate, as shown in Fig. 1b, were broadly observed (e.g., Maranchi et al., 2006; Yang et al., 2018). Such interfacial deformation will inevitably influence the stress-capacity curve. Connecting the properties of the interface and the anode material with the measured curvature-capacity curve is hence crucial to determine the actual stress in the electrode material. More importantly, we may use the curvature-capacity curves to extract the interface and material properties.

A typical route to obtain material parameters in constitutive models is to compare the predicted stress-capacity response from the models with available experimental data. Following this strategy, different groups have developed constitutive models and the numerical methods for electrode materials to decipher the materials' properties from the stress-capacity response. For example, Bhandakkar et al. (2010) compared the stress-capacity curves of Sn films from numerical analysis and experimental data, and estimated the interfacial strength. Zhao et al. (2012) and Pharr et al. (2013) studied the mechanical properties of electrode materials using first-principle calculations, and compared their results with measured stress-capacity curves. Jia and Li (2016) and Wen et al. (2018) explored the influence of concentration-dependent elasticity on stresses in spherical particles and film-substrate structures, respectively. Zhao et al. (2011, 2012) and Brassart et al. (2013) reported how the electrode materials' plastic flow and shakedown were related to the characteristics in the stress-capacity curves. Bower et al. (2011) predicted the evolution of stress and potential for a one-dimension Li-ion half-cell with a Si anode by considering finite strain, plastic flow, and electric current flow. By taking an analogy between diffusion and heat conduction, Yang et al. (2014) presented a chemical-mechanical model to investigate the lithiation-induced phase transformation, morphological evolution, stress generation and fracture in crystalline silicon nanowires. Levitas and Attariani (2014) analyzed the anisotropic compositional expansion in elastic-plastic anode materials. The work by Anand (2012), Di Leo et al.