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Chemomechanics of two-phase lithiation in amorphous silicon

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Abstract

Silicon electrodes for lithium-ion batteries exhibit a theoretical capacity ten times higher than that of graphite electrodes currently used in commercial systems. When lithiated/delithiated, silicon experiences stresses on the order of 1 GPa, a cause of poor cyclability and dissipation unfavorable to energy storage. These stresses affect the material from its very first lithiation, which proceeds through a two-phase process involving the progressive invasion of the pristine Si phase by an amorphous lithiated LixSi phase. Nonequilibrium continuum thermodynamics predicts that mechanical stresses affect the lithiation itself differently when the material is homogeneously lithiated (monophasic lithiation) than when it undergoes a two-phase lithiation [1]. When monophasic lithiation takes place, mechanical stresses affect lithium insertion through the chemical potential of the diffusing species (Larché-Cahn theory). In a first study we have investigated experimentally the coupling under homogeneous lithiation conditions by imposing an external mechanical loading on an amorphous silicon thin-film electrode at various lithiation rates and measuring its effect on the electrochemical response of the battery cell [2]. This mechanical loading was applied

by deforming elastically the steel substrate of the silicon thin film, which made it possible to directly probe the contribution of mechanics to the lithiation/delithiation processes. In this study, we showed that the instantaneous change in the electrochemical potential and its subsequent relaxation are well captured by a viscoplastic flow rule for the amorphous material. In the present work, we concentrate on the effect of an external mechanical load on the two-phase process that occurs during the first lithiation, both experimentally and through modeling. Our experiments show that when undergoing a phase transformation, the very same external mechanical loading as that applied during homogeneous lithiation leads to a very different response of the silicon electrode potential. While displaying a similar instantaneous change in potential, the subsequent voltage relaxation displays a very distinct pattern, which we attribute to the interplay between stress evolution and phase-boundary kinetics. To rationalize these observations, we resort to different modeling techniques aiming at describing the mechanisms underlying the phase boundary motion. First, an Arbitrary Lagrangian-Eulerian framework is developed to solve a sharp-interface model of the phase transformation in the one-dimensional setting of a thin film, and an implicit finite-element numerical implementation is formulated. This sharp-interface is then employed to calibrate a phase-field model inspired from [3], which is more amenable to simulations of complex phase-front geometries. Such a model offers an independent degree of freedom for the phase-front kinetics, one which is not available in the popular Cahn-Hilliard models. In this sense, the proposed phase-field model is a mere numerical regularization of the sharp-interface one, and numerical strategies are discussed.

Keywords:

Lithium-ion batteries Silicon electrode Free boundary problem Phase transformation Electro-chemo-mechanical coupling Elasto-visco-plastic material

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