



Effect of prestress on the stability of electrode–electrolyte interfaces during charging in lithium batteries



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ABSTRACT

We formulate a model of the growth of electrode–electrolyte interfaces in lithium batteries in the presence of an elastic prestress. The model accounts for the kinetics of Li^+ transport through a solid electrolyte and, within the interface, for the kinetics of Li^+ adsorption by the anode, electrostatics, and the elastic field. We specifically account for the effect of the elastic field through an asymptotic analysis of a nearly flat interface between two semi-infinite elastic bodies. We use the model as a basis for assessing the effect of prestress on the stability of planar growth and the potential of prestress as a means of suppressing the formation of deleterious dendrites. We present a linear stability analysis that results in explicit analytical expressions for the dependence of growth rates, and of the critical unstable wavelength for the interfacial roughening, on the state of prestress and on fundamental parameters such as surface diffusivities, surface energy, deposition kinetics, and elastic moduli. Finally, we examine the model in the light of experimental observations concerned with the effect of applied pressure on a lithium/dioxolane-dimethoxy ethane electrolyte systems. With reasonable choices of parameters and some calibration, the model accounts for the observation that a modest applied pressure indeed results in a substantial reduction in the roughening of the lithium surface during cycling.

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1. Introduction

The present work is concerned with the assessment of the effect of prestress on the stability of electrode–electrolyte interfaces in lithium batteries and, in particular, of the potential of prestress as a means of suppressing the formation of deleterious dendrites. Lithium compounds are widely used as a electrode materials for rechargeable batteries due to the high electropositivity and low weight of metallic lithium. During battery operation, lithium at the anode, the negative electrode, gives up electrons to become Li^+ , which dissolves into the electrolyte. The electrons move through the electrical circuit to the positive electrode, the cathode, as shown in Fig. 1. In a recharging battery this process is reversed and lithium receives current from an external generator. In order to avoid confusion, we will refer to the electrode on the left of Fig. 1 as the negative electrode and to the one on the right as the positive electrode, since this characterization is correct during both charging and discharging cycles.

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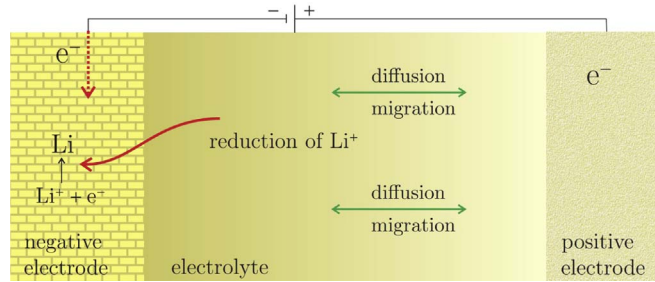


Fig. 1. Charging process at the lithium anode.

Lithium cations, during recharge, move from the positive electrode to the negative one. If the lithium deposition is not uniform across the electrode surface, it can form protrusions, and even dendrites, that eventually may short the battery (Bhattacharyya et al., 2010; Chandrashekar et al., 2012; Harry et al., 2014). Because reactions between the lithium metal and the electrolytes are exothermic, large surface areas also pose risk of overheating, or thermal runaway (von Sacken et al., 1995; Tobishima and Yamaki, 1999). Dendrites at the lithium electrode have been reported to grow from one electrode to the other through the electrolyte (Akolkar, 2013; Monroe and Newman, 2003; Winter et al., 2001; Despic and Popov, 1972; Armand, 1983). One of the mechanisms that trigger and mediate dendrite formation is thought to be the localized current surge at dendrite tips (Christensen et al., 2011). The size of dendrites ranges between 2 and 2000 μm depending on the material system, cf. Figs. 2 and 3.

Dendritic lithium deposits on the battery electrode after being subjected to multiple charges and recharges were first observed by Epelboin et al. (1980) and later on by Yoshimatsu et al. (1988) and others Dolle et al. (2002). reported dendritic growth of the interface on lithium polymer batteries and confirmed direct correlation between current density and dendrite formation. Other authors (Peled, 1979; Chazalviel, 1990; Sundstrom and Bark, 1995; Kanamura et al., 1996; Xu, 2004; Yamaki et al., 1998) studied the deposition mechanisms of metals in non-aqueous battery systems Barton and Bockris (1962) and Diggle et al. (1969). put forth the first comprehensive model of dendrite growth. Improvement in lithium cycling efficiency using additives is discussed in other works (Saito et al., 1997; Eweka et al., 1997; Richardson and Chen, 2007).

Studies with a more mechanical focus include those of Ely and Edwin Garcia (2013), Nishida et al. (2013), and Monroe and Newman (2003). The latter study conclude that interfacial roughening is mechanically suppressed when the separator shear modulus is about twice that of lithium Mikhaylik et al. (2010). Experimentally confirmed that application of pressure between the electrodes lowers the roughness of the lithium anode, an effect first posited by Hirai et al. (1994). However, a fundamental understanding of the effect of pressure and, more generally, prestress on the stability of lithium–electrolyte interfaces appears to be lacking at present, which hampers the practical exploitation of prestress in battery design.

The objective of the present study is to ascertain conditions under which an electrode–electrolyte interface grows in a planar geometry and how such conditions are influenced by the elastic field of built-in prestress. Of particular interest is to elucidate the manner in which elastic prestress can be utilized in the design of lithium batteries in order to eliminate dendrite formation and its deleterious effects during the lifetime of the cell. We base our analysis on a model of interfacial growth that accounts for the kinetics of Li^+ transport through the solid electrolyte (e.g., LIPON) and within the interface, the kinetics of Li^+ adsorption by the anode, electrostatics, and the elastic field. These fields are coupled due to pressure-assisted diffusion, swelling of the intercalated electrolyte, and the Maxwell stress of the electrostatic field, and contribute jointly to the chemical potential driving the motion of interface. In particular, the elastic field is sensitive to the shape of the interface,

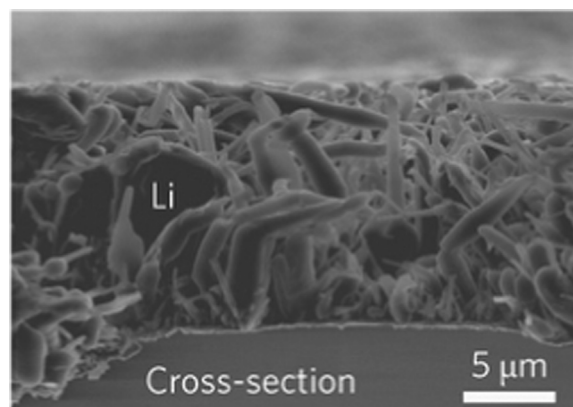


Fig. 2. SEM characterization of lithium deposition on a copper substrate: at the electrode ramified growth of mossy lithium/dendrites is clearly observed. Photograph from Zheng et al. (2014).

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