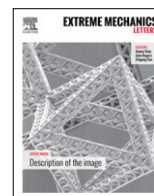


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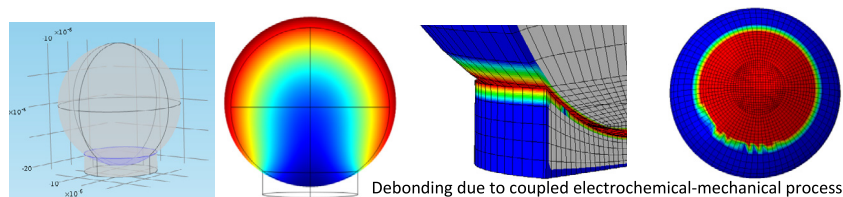
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Debonding at the interface between active particles and PVDF binder in Li-ion batteries

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GRAPHICAL ABSTRACT



HIGHLIGHTS

- Studied debonding at the interface between particle and binder in a battery.
- Integrated electrochemical–mechanical model and cohesive zone model.
- Debonding is more likely to happen as the particle size and C-rate decrease.
- The trend of debonding is opposite to that of fracture inside a particle.
- Debonding is closely related to the total amount of lithium intercalation.

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ABSTRACT

Mechanical failure in the electrode is one of major reasons for capacity fade. In this study we focus on inter-particle fracture, specifically the debonding at the interface between the graphite particle and binder. We integrate the electrochemical–mechanical model and the cohesive zone model to investigate the interfacial debonding during lithium intercalation. We found that the mechanism of fracture at the particle/binder interface is different from that inside a particle. The debonding at the interface is caused by the expansion of the particle that is closely related to the total amount of lithium intercalation, while the fracture inside a particle is caused by the gradient of lithium concentration. As a result, debonding at the interface is more likely to occur as the particle size and C-rate decrease, which is opposite to the trend of fracture inside a particle that is more likely to occur as the particle size and C-rate increase. This understanding of debonding mechanism can provide insight into capacity fade and guide the development of more robust electrodes.

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1. Introduction

Fracture in the electrode is known as one of the major causes for degradation in Li-ion batteries [1–3]. The effects include loss of electric contact, isolation of active materials from the conductive matrix, and increased cell internal resistance. Fracture also increases the exposure of active materials to the electrolyte, accelerating side-reactions and capacity fade. The fracture behavior inside a single particle has been addressed by several works. Zhao et al. studied the effect of particle size and charging rate on fracturing in an electrode particle and predicted the critical fracture conditions by the energy release rate [4]. Woodford et al. constructed a map showing the safe parameter regimes against fracture, which depended on the C-rates, particle size and fracture toughness of the material [5]. Bhandakkar et al. calculated the critical electrode size to avoid crack nucleation in a cylindrical electrode particle [6]. Zhu et al. evaluated the effects of current density, particle size and particle aspect ratio on crack initiation using the extended finite element method [7]. These studies have revealed the critical conditions that determine whether a crack will grow or not. Furthermore, Grantab et al. investigated the progressive propagation of a crack during cycling in a graphite particle [8] or in a silicon nanowire [9].

However, inter-particle fracture in electrode materials has rarely been addressed in theoretical studies. In an electrode, particles are connected together by the binder. Two scenarios can possibly happen for inter-particle fracture depending on the locations: fracture in the middle of binder or at the particle/binder interface. Recently, atomistic simulations have shown that the cohesive strength at the interface is weaker than that inside the binder, suggesting that inter-particle fracture is more likely to happen at the particle/binder interface [10].

In this paper, we study the debonding behavior between a graphite particle and a PVDF (polyvinylidene fluoride) binder associated with the electrochemical–mechanical processes during the operation of a Li-ion battery. Graphite particles and PVDF are commonly used as the anode material and the binder in current commercial batteries. To capture fracture at the interface, we have developed an electrochemical–mechanical model that incorporates the cohesive zone approach. First, diffusion-induced stresses are calculated by the electrochemical–mechanical model. Then, the lithium concentration profile is exported to the cohesive zone model by thermal stress analogy. Since the cohesive zone model can describe crack initiation and its propagation, the simulation results can provide quantitative insight into the debonding process during lithium intercalation and capture the creation of new interfaces that are exposed to the electrolyte.

In the cohesive zone model, the traction–separation curve is an important input. We have implemented the parameterized traction–separation curve obtained from atomistic calculations as input data for the cohesive zone model. Our study shows that the fracture mechanism at the particle/binder interface is significantly different from that inside a particle: the interfacial fracture is more likely to happen at small particle sizes and low C-rates, a trend that is opposite to that inside a particle. This paper is

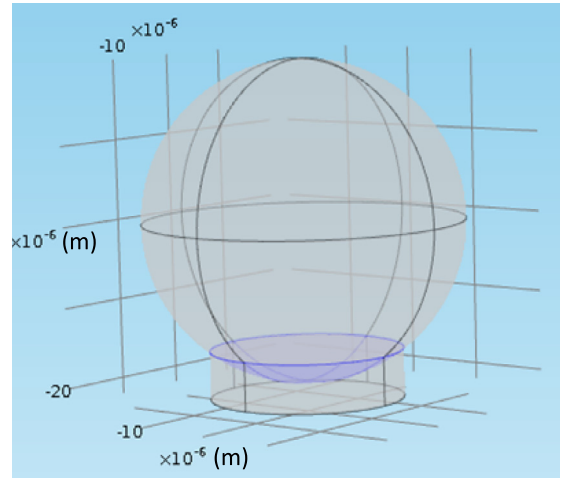


Fig. 1. The model system of a spherical graphite particle attached to a cylindrical PVDF binder.

structured as follows. First, the theoretical framework for the calculation of lithium diffusion and the simulation of debonding at the interface are described. Then, simulation results with the electrochemical–mechanical model and the cohesive zone approach are shown. Finally, the conclusion is given at the end.

2. Method

We consider a spherical graphite particle attached to a cylindrical PVDF binder, as shown in Fig. 1. A two-step method is used to study the interfacial debonding due to lithium intercalation. First, we calculate the diffusion-induced stress in the particle by coupling the stress field and Li-ion diffusion. Second, we export the calculated lithium concentration profile at a specific state of charge and apply it to the cohesive zone model to examine the debonding behavior.

2.1. Electrochemical–mechanical model

The stress–strain relation in the particle can be written as [11,12]

$$\varepsilon_{ij} = \frac{1}{E} \left[(1 + \nu)\sigma_{ij} - \nu\sigma_{kk}\delta_{ij} \right] + \frac{c\Omega}{3}\delta_{ij}, \quad (1)$$

where ε_{ij} are strain components, E is Young's modulus, ν is Poisson's ratio, σ_{ij} are stress components, δ_{ij} is the Kronecker delta, c is the Li-ion concentration and Ω is partial molar volume of Li-ions. Comparing to the regular stress–strain relation, the last term in Eq. (1) is additional, which accounts for the strain due to Li-ion intercalation. In the binder region the regular stress–strain relation is applied. The mechanical boundary condition is given by constraining the displacements at the bottom of the binder, as shown in Fig. 1.

The gradient of chemical potential drives Li-ion diffusion. Both concentration and stress contribute to the chemical potential, leading to a diffusion flux of \mathbf{J} ,

$$\mathbf{J} = -D \left(\nabla c - \frac{\Omega c}{RT} \nabla \sigma_h \right), \quad (2)$$

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