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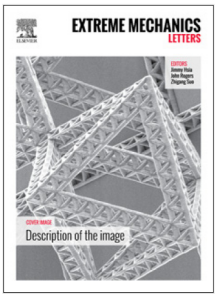

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Extreme Mechanics Letters

Fundamental Mechanisms of Fracture and its Suppression in Ni-rich Layered Cathodes: Mechanics-based Multiscale Approaches

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Ni-rich layered oxides have been identified as promising candidates for commercial cathodes in Li-ion batteries. However, the commercialization has been hindered by severe cyclic degradation and mechanical failure induced by severe phase transformations and fractures. To resolve these challenges by understanding their fundamental mechanisms, we present mechanics-based multiscale investigations to elucidate the fundamental mechanisms of mechanical failure including deformations and fractures. We have also suggested a practical solution to the failure, which involves enhancing electronic interactions between transition metal layers. The methodological framework for our investigations was developed from first-principles atomic calculations, electronic structure, thermodynamics and kinetics for combined phase transformation, phase field modeling, finite element methodology for mechanical deformation, and phase field crack modeling. Our practical solution addresses the electronic interactions that can be strengthened when O ions are reduced by substituting strongly oxidizing elements such as Ti. Our multiscale framework shows that the reduced O ions are responsible for higher fracture toughness, reduced volume changes, stable deformation, mitigated stress generation, and suppressed fractures. Thus, this study proposes a practical solution for the improvement and design of Ni-rich layered oxide cathode materials. Furthermore, the mechanics-based multiscale methodology employed herein could be applied to a number of other solid-state energy materials suffering from mechanical failures.

Keywords: Multiscale mechanics; First-principles calculation; Phase field fracture model; Finite element method, Ni-rich layered cathodes; Li-ion batteries