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Lithiation-Enhanced Charge Transfer and Sliding Strength at the Silicon-Graphene Interface: a First-Principles Study

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ABSTRACT

The application of silicon as ultrahigh capacity electrodes in lithium-ion batteries has been limited by a number of mechanical degradation mechanisms including fracture, delamination and plastic ratcheting, as a result of its large volumetric change during lithiation and delithiation. Graphene coating is one feasible technique to mitigate the mechanical degradation of Si anode and improve its conductivity. In this paper, first-principles calculations are performed to study the atomic structure, charge transfer and sliding strength of the interface between lithiated silicon and graphene. Our results show that Li atoms segregate at the (lithiated) Si-graphene interface preferentially, donating electrons to graphene and enhancing the interfacial sliding resistance. Moreover, the interfacial cohesion and sliding strength can be further enhanced by introducing single-vacancy defects into graphene. These findings provide insights that can guide the design of stable and efficient anodes of silicon/graphene hybrids for energy storage applications.

Keywords: lithium-ion batteries, silicon anode, graphene coating, interfacial sliding strength.

I. INTRODUCTION

Silicon has been proposed as a candidate anode material for lithium-ion batteries