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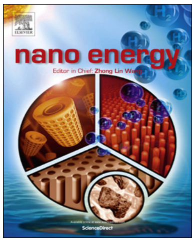

REVIEW

Nanoscale silicon as anode for Li-ion batteries: The fundamentals, promises, and challenges

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Abstract
Silicon (Si), associated with its natural abundance, low discharge voltage vs. Li/Li^+, and extremely high theoretical capacity ($\sim$4200 mAh g^{-1},), has been extensively explored as anode for lithium ion battery. One of the key challenges for using Si as anode is the large volume change upon lithiation and delithiation, which causes a fast capacity fading. Over the last few years, dramatic progress has been made for addressing this issue. In this paper, we review the progress towards tailoring of Si as anode for lithium ion battery. The paper is organized such that it covers the fundamentals, the promises offered by nanoscale designs, and the challenges that remained to be addressed to allow the application of Si based materials as high capacity anode for lithium ion batteries.

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Introduction

Advantages and disadvantages of Si-based anode

Li-ion batteries (LIB) appear to be tangible items of our daily life as they are indispensably used for portable electronics, electric transport, and grid energy storage [1]. In a conventional Li-ion battery, the anode is composed of graphite and the cathode is composed of $LiCoO_2$. However, these conventional electrode materials suffers from low capacity, high cost and limited power output. In order to meet the increasing demand of higher energy density, cost effectiveness and long-term cycle life for Li-ion batteries, new electrode materials must be discovered and implanted for battery applications. Silicon is one of the most promising candidate materials as anode for lithium ion battery, potentially offering of high capacity for modern Li-ion batteries. Si possesses a capacity of 4200 $mAh\,g^{-1}$, which is about ten times of conventional graphite anode (372 $mAhg^{-1}$). Meanwhile, Si anode has a lithiation voltage plateau at 0.2–0.3 V vs. Li/Li^+ than the graphite anode ($<$0.1 V vs. Li/Li^+), which could avoid the undesired lithium plating and potential dendrite formation, alleviating the safety concern of the corresponding LIB system [2-14]. Therefore, Si appears to be a very promising anode material for the application in next generation electric vehicles (EVs) or hybrid electric vehicles (HEVs) which require to be powered by LIB with both higher gravimetric/volumetric energy density and less safety concern.

Silicon is abundant in earth and large-scale production of silicon based materials has been well realized for decades. However, translating the Si-based materials into industry scale usage as anode for lithium ion battery is not a straight forward process. A range of challenges needs to be tackled in the path for the industrial application of silicon-based materials as anode. The biggest challenge is the large volume change during lithiation of Si ($\sim$280%) [15-31]. Phenomenologically, such a large volume change will lead to a number of consequences, typically including cracking and fracturing of the Si anode, destruction and regeneration of solid electrolyte interphase (SEI) layer on the fractured surfaces, and degradation of electronic conductivity. These factors and their consequent effect on the electrochemical properties are now generally termed as chemi-mechanical effect, which directly contributes to the fast capacity fading of the battery system.

To address the chemi-mechanical effect of Si based materials as anode for lithium ion battery, innovative nanostructure designing concepts have been emerged over the last few years. These nanostructural designing concepts set the foundation for optimized tailoring Si based material as high capacity anode for lithium ion battery. In addition, development of in-situ TEM over the last few years enables the direct visualization of the structural and chemical evolution of the nanostructured electrode materials under dynamic operating condition. The in-situ TEM technique has been widely applied to study the lithiation characteristics and failure/functional mechanisms of different Si based electrode materials, such as Si nanotube, Si nanowire, Si-carbon composite and porous Si anodes. The information captured using in-situ TEM experiments provides valuable insights for optimization of Si-based anodes with enhanced performances. This paper summaries and reviews recent progress on the fundamental understanding of Si as anode for lithium ion battery, including the basic knowledge, the nanoscale designing concepts, and challenges that need to