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Computational multiobjective topology optimization of silicon anode structures for lithium-ion batteries

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HIGHLIGHTS

- Optimal silicon anode structures are designed using topology optimization methods.
- A multiobjective formulation considers compliance and electric conduction criteria.
- The trade-off between design objectives is explored using Pareto optimality.

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ABSTRACT

This study utilizes computational topology optimization methods for the systematic design of optimal multifunctional silicon anode structures for lithium-ion batteries. In order to develop next generation high performance lithium-ion batteries, key design challenges relating to the silicon anode structure must be addressed, namely the lithiation-induced mechanical degradation and the low intrinsic electrical conductivity of silicon. As such this work considers two design objectives, the first being minimum compliance under design dependent volume expansion, and the second maximum electrical conduction through the structure, both of which are subject to a constraint on material volume. Density-based topology optimization methods are employed in conjunction with regularization techniques, a continuation scheme, and mathematical programming methods. The objectives are first considered individually, during which the influence of the minimum structural feature size and prescribed volume fraction are investigated. The methodology is subsequently extended to a bi-objective formulation to simultaneously address both the structural and conduction design criteria. The weighted sum method is used to derive the Pareto fronts, which demonstrate a clear trade-off between the competing design objectives. A rigid frame structure was found to be an excellent compromise between the structural and conduction design criteria, providing both the required structural rigidity and direct conduction pathways. The developments and results presented in this work provide a foundation for the informed design and development of silicon anode structures for high performance lithium-ion batteries.

1. Introduction

The lithium-ion (Li-ion) battery is a highly successful secondary battery that is used to power electric cars, phones, laptops and other portable electronic devices. In order to meet the energy demands of today's multifunctional electronic devices, the Li-ion battery requires a significant performance enhancement in terms of capacity, energy density, and rate capability. One method to improve the Li-ion battery performance is to replace the traditional graphite anode with silicon. In addition to being an abundant, inexpensive, and sustainable material, silicon has the highest known theoretical specific capacity for Li-ion intercalation of 4200 mA h/g, over ten times greater than that of graphite [1,2]. Unfortunately, this excellent capacity comes at the expense of a 310% volume expansion and contraction of the silicon anode during lithium insertion and extraction [3,4], compared to the 6–10% volume expansion observed for a graphite anode [5]. This large change in volume causes severe detrimental effects that render the battery impractical for commercialization unless significant anode design changes are made.

In terms of adverse effects, the anode experiences extremely high compressive stresses upon lithiation due to the restrained

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volumetric expansion, resulting in pulverization of the active particles [6]. Furthermore, the volume contraction upon delithiation induces large tensile stresses that cause cracking and fracture of the anode structure, and therefore disconnected charge transport paths. These effects result in incomplete intercalation and a high irreversible capacity loss [7,8].

Furthermore, silicon is considered a semiconductor and, as such, has a low intrinsic electric conductivity. Therefore, silicon anode structures typically exhibit poor rate performance due to slow electron transport within the anode material [8]. Common methods to overcome the poor electronic conductivity of silicon include the use of conductive agents in powder-based structures [9,10], architectures featuring one-dimensional conductive pathways, and incorporating conductive coatings, such as copper or carbon [11–13]. These coatings provide continuous electric contact thereby improving the conductivity of the electrode. Additionally, the carbon or copper shell helps to suppress electrolyte decomposition and also constrain the silicon expansion, which minimizes particle fracture and therefore improves the cyclability of the structure.

In order to utilize high capacity silicon as a new anode material, several important design requirements must therefore be met. Firstly, the silicon anode structure must adequately accommodate the volume expansion upon lithiation, and reduce the associated induced mechanical stress. The design must also maximize electrical conduction through the structure to compensate for the low conductivity of silicon and ensure good rate capability of the battery.

Experimentalists have endeavoured to address these design requirements by investigating a range of silicon anode structures, such as nanoscale structures, porous structures, nanowire anodes, thin-films, and multiphase composite structures. Nanoscale structures are capable of sustaining very high stresses before pulverization or cracking due to the size-induced ductility of nanosilicon [14–16], thereby reducing the irreversible capacity loss and improving battery performance. Porous silicon anodes [17,18] utilize the void spaces in the structure to accommodate the volume expansion upon lithiation, while silicon nanowire structures [19–21] allow for rapid charge transport through one-dimensional electronic pathways and can withstand the large volume change without fracture. All of these structures are characterized by a large surface area to volume ratio, which results in small Li-ion diffusion distances and therefore enhanced power and rate capabilities [5,22]. Other promising structures include thin-film silicon anodes [4,23–25], which feature small charge transport lengths and a relatively low state of stress upon lithiation. However, a trade-off is required between the very small film thickness required for mechanical robustness, and sufficient active material necessary to achieve the desired electrochemical performance of the cell. Multiphase composite structures [9] are also of interest, where the active material is dispersed within a composite matrix. This host matrix buffers the large volume change upon lithiation and allows for the efficient transport of both electrons and Li-ions.

These silicon anode designs, crafted by experimentalists, are primarily based on design intuition and historical testing results. Despite showing some very promising results, the anode structures are not necessarily optimal from the outset. As such, there exists huge potential to use structural optimization methods to produce high performance silicon anode designs. These optimal designs may verify the experimentalists' design choices, or may provide new structural designs that could then be manufactured, tested and further refined.

We therefore propose the use of topology optimization methods to address this silicon anode design problem. Topology optimization is the most generalized structural optimization method, and is used to determine the optimal material distribution within a design domain for a given set of loading and boundary conditions. Unlike sizing and shape optimization, the optimum design is not based on a predefined structural configuration, therefore the solution obtained by topology optimization is regarded as the true optimum in a design space for a specific problem [26]. Topology optimization methods are implemented through the use of finite element and mathematical programming methods, and typical design objectives include minimizing compliance, displacement, or stress. This method is an extremely useful tool for conceptual design stages, or for problems where there is limited physical intuition of the optimal structural design, as is the case for designing multifunctional silicon anode structures.

To date, there have been no instances of topology optimization methods being applied to the silicon anode design problem. In fact, topology optimization methods have been completely underutilized for battery systems in general. Several authors have applied these structural optimization methods to solid oxide fuel cells (SOFCs) [27–30], an entirely different electrochemical energy storage system. These works focused on the shape optimization, rather than topology optimization, of cathodes or gas channels in SOFCs, typically performing only two-dimensional analysis. Their results illustrate the potential of topology optimization methods to provide a useful contribution to the design and development of energy storage systems.

In this paper, we use a density-based multiobjective topology optimization method for the three-dimensional design of multifunctional silicon anode structures for Li-ion battery applications. We address the design challenges associated with the large volume expansion upon lithiation, and also the low intrinsic electronic conductivity of the anode material. The underlying design is chosen to be a porous nanoscale structure in order to capitalize on the performance advantages of size-induced ductility, small Li-ion diffusion distances, and the porosity of the structure helping to accommodate the volume change upon lithiation. The structural and conduction design requirements are first optimized individually by considering a minimum compliance objective subject to design dependent volumetric expansion, and a maximum electrical conduction objective, respectively. In order to produce multifunctional designs, the design criteria are subsequently addressed simultaneously using a bi-objective formulation. The optimal structures provide a solid foundation for the informed design and development of silicon anode structures, and may subsequently be used by experimentalists for testing, or be incorporated into numerical models of Li-ion battery systems.

The paper is structured as follows. In Section 2, we present the topology optimization methodology and bi-objective formulation that simultaneously addresses the structural and conduction design criteria. Section 3 details the problem set up, including the loading, boundary conditions and material properties used in the analysis. In Section 4, the results for each design objective are first presented and discussed individually, followed by the results and discussion for the multiobjective topology optimization problem. Finally, concluding remarks are provided in Section 5 with an outlook on future work given in Section 6.

2. Methodology

In this section we outline the multiobjective topology optimization methodology used to obtain the optimal multifunctional silicon anode structures. We first briefly summarize density methods and the primary techniques used in this analysis, followed by an introduction to Pareto optimality and the multiobjective formulation used for the silicon anode design problem.