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

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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.

Keywords: Topology optimization; multiobjective; conduction; compliance; design dependent loads

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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 mAh/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 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, 12, 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, 15, 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, 20, 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 [23, 24, 4, 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].

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6 Topology optimization methods are implemented through the use of finite element and mathe-
7 matical programming methods, and typical design objectives include minimizing compliance,
8 displacement, or stress. This method is an extremely useful tool for conceptual design stages, or
9 for problems where there is limited physical intuition of the optimal structural design, as is the
10 case for designing multifunctional silicon anode structures.
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14 To date, there have been no instances of topology optimization methods being applied to
15 the silicon anode design problem. In fact, topology optimization methods have been completely
16 underutilized for battery systems in general. Several authors have applied these structural opti-
17 mization methods to solid oxide fuel cells (SOFCs) [27, 28, 29, 30], an entirely different elec-
18 trochemical energy storage system. These works focused on the shape optimization, rather than
19 topology optimization, of cathodes or gas channels in SOFCs, typically performing only two-
20 dimensional analysis. Their results illustrate the potential of topology optimization methods to
21 provide a useful contribution to the design and development of energy storage systems.
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25 In this paper, we use a density-based multiobjective topology optimization method for the
26 three-dimensional design of multifunctional silicon anode structures for Li-ion battery appli-
27 cations. We address the design challenges associated with the large volume expansion upon
28 lithiation, and also the low intrinsic electronic conductivity of the anode material. The underly-
29 ing design is chosen to be a porous nanoscale structure in order to capitalize on the performance
30 advantages of size-induced ductility, small Li-ion diffusion distances, and the porosity of the
31 structure helping to accommodate the volume change upon lithiation. The structural and conduc-
32 tion design requirements are first optimized individually by considering a minimum compliance
33 objective subject to design dependent volumetric expansion, and a maximum electrical conduc-
34 tion objective, respectively. In order to produce multifunctional designs, the design criteria are
35 subsequently addressed simultaneously using a bi-objective formulation. The optimal structures
36 provide a solid foundation for the informed design and development of silicon anode structures,
37 and may subsequently be used by experimentalists for testing, or be incorporated into numerical
38 models of Li-ion battery systems.
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42 The paper is structured as follows. In Section 2, we present the topology optimization
43 methodology and bi-objective formulation that simultaneously addresses the structural and con-
44 duction design criteria. Section 3 details the problem set up, including the loading, boundary
45 conditions and material properties used in the analysis. In Section 4, the results for each design
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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.

2.1. Density methods

The general aim of topology optimization is to determine the material distribution and therefore structural design that optimizes an objective function, such as compliance, mass or displacement, in conjunction with imposed constraints, for example a limit on the total material volume of the structure. Density-based topology optimization methods utilize continuous design variables and finite element techniques. The design domain is discretized into finite elements, where each finite element is typically comprised of either material or void, and is assigned a design variable x_e that represents the density of the material in that element. To ensure the problem is well-posed, the values of the element density are allowed to vary continuously between 0 and 1, representing void and solid material, respectively. Therefore, material properties such as stiffness or conductivity are a function of these continuous design variables. In order to obtain a discrete or solid-void final design, the intermediate densities are penalised through the use of an interpolation scheme. This work uses both the Solid Isotropic Material with Penalization (SIMP) [32, 33], and the Rational Approximation of Material Properties (RAMP) [34] interpolation schemes.

In addition, regularization techniques such as density filtering are utilized to prevent numerical difficulties, and a continuation scheme is implemented to assist with convergence to a globally optimal solution. Furthermore, mathematical programming methods are used to obtain the updated design variables. For the silicon anode design problem, we utilize the globally convergent method of moving asymptotes (GCMMA) by Svanberg [35].

2.2. Multiobjective formulation

In this study, we perform multiobjective topology optimization to produce anode structures that simultaneously address the mechanical degradation associated with the large volume expansion upon lithiation, and the poor electric conductivity of silicon. Due to the competing nature of these design objectives, there exists no single global solution that would simultaneously give an optimum for both objectives. Therefore, a trade-off between these design criteria is required, and the concept of Pareto optimality [36] is used to determine the series of solutions that make up the optimum. A solution is Pareto-optimal if an improvement in one objective is accompanied by the deterioration of at least one other objective. In this analysis we utilize the weighted sum method [37] to obtain the Pareto optimal solutions. The discrete multiobjective problem formulation is therefore given by

$$\min_x : f(\tilde{\mathbf{x}}) = w_s f_s(\tilde{\mathbf{x}}) + w_c f_c(\tilde{\mathbf{x}}) \quad (1)$$

$$= w_s \frac{\mathbf{U}(\tilde{\mathbf{x}})^T \mathbf{K}(\tilde{\mathbf{x}}) \mathbf{U}(\tilde{\mathbf{x}})}{f_s^*} + w_c \frac{\mathbf{\Phi}(\tilde{\mathbf{x}})^T \mathbf{C}(\tilde{\mathbf{x}}) \mathbf{\Phi}(\tilde{\mathbf{x}})}{f_c^*}$$

$$\text{subject to : } \mathbf{K}(\tilde{\mathbf{x}}) \mathbf{U}(\tilde{\mathbf{x}}) = \mathbf{F}_s(\tilde{\mathbf{x}}), \quad (2)$$

$$\mathbf{C}(\tilde{\mathbf{x}}) \mathbf{\Phi}(\tilde{\mathbf{x}}) = \mathbf{F}_c, \quad (3)$$

$$V_f^{\min} \leq \frac{V(\tilde{\mathbf{x}})}{V_0} \leq V_f^{\max}, \quad (4)$$

$$\mathbf{0} \leq \mathbf{x} \leq \mathbf{1}. \quad (5)$$

Here, $f(\tilde{\mathbf{x}})$ is the objective function, which is a function of the vector of physical element densities $\tilde{\mathbf{x}}$. $f(\tilde{\mathbf{x}})$ is given by the sum of contributions from the structural and conduction design objectives f_s and f_c , respectively. w_s and w_c are the associated weighting factors, where $0 \leq w_s \leq 1$, and $w_s + w_c = 1$. This means when $w_s = 1$, only the structural objective is considered, and when $w_s = 0$ only the conduction objective is considered. The weights may therefore be continuously altered to form the complete set of Pareto-optimal designs, which are visualised using a Pareto front.

The structural objective function f_s is equal to the compliance, which may be defined as the virtual work of the loads on the displacements they generate [38]. Minimizing this objective function effectively minimizes a global measure of deformation, and an average measure of stress. Here, $\mathbf{U}(\tilde{\mathbf{x}})$ is the global displacement vector, $\mathbf{K}(\tilde{\mathbf{x}})$ is the global stiffness matrix, and f_s^* is a normalization factor.

The second design objective is to maximize electric conduction through the anode structure by considering the scalar version of the minimum structural compliance formulation [39]. This objective function f_c is therefore termed the electrical compliance, where $\Phi(\tilde{\mathbf{x}})$ is the global voltage vector, $\mathbf{C}(\tilde{\mathbf{x}})$ is the global electrical conductance matrix, and f_c^* is a normalization factor.

The optimization problem is subject to the structural and conduction equilibrium equations, Equations 2 and 3, respectively. Here, $\mathbf{F}_s(\tilde{\mathbf{x}})$ is the global force vector for the structural objective, which is due to the design dependent volumetric expansion upon lithiation. This load vector is design dependent in the sense that it is a function of element density $\tilde{\mathbf{x}}$ and therefore changes upon each iteration. $\mathbf{F}_c$ is the global force vector for the conduction objective, which is due to a boundary current for the silicon anode design problem and is design independent.

This optimization problem is also subject to a constraint on material volume given by Equation 4, where $V(\tilde{\mathbf{x}})$ is the material volume, V_0 is the design domain volume, and V_f^{min} and V_f^{max} are the minimum and maximum allowable volume fractions, respectively. Both an upper and lower bound on material volume are used to prevent singular behavior resulting from the design dependent nature of the volume expansion. Finally, the vector of design variables $\mathbf{x}$ must attain a value between 0 and 1.

3. Optimization problem parameters

This section details the silicon anode design problem parameters, such as the loading, boundary conditions, and material properties. The design domain representing the anode structure and the applied loading and boundary conditions are depicted in Figure 1.

Figure 1a shows the overall anode design domain, where the bottom face of the anode is rigidly fixed to the substrate, while the sides are constrained by battery packaging and reinforcement. The design domain is comprised of two material phases, the silicon anode structure and the electrolyte. We consider the anode structure to be composed entirely of silicon active material. In this way we directly optimize the underlying silicon anode structure for the maximum electrical conductivity, before the addition of conductive additives or coatings. It is expected that a conductive copper coating or similar may be incorporated during manufacturing to further enhance electrical conductivity and improve cyclability. This design assumption of modelling only the active material and no conductive coatings also greatly improves computational efficiency

Figure 1: Schematics of the design domain, loading, and boundary conditions for the silicon anode design problem. (a) Anode design domain. (b) Volume expansion problem applied to the periodic base cell. (c) Electric conduction problem applied to the periodic base cell.

and results in a feasible required memory and computation time. The desired volume fraction of the silicon anode material is set to $V_f = 0.25 - 0.30$, with the initial element density set to the median value between these two bounds. This volume fraction was chosen to ensure there was adequate porosity of the overall anode/electrolyte structure, whilst also maintaining sufficient active material to achieve the required electrochemical performance of the battery.

We consider a $5 \mu\text{m} \times 5 \mu\text{m} \times 5 \mu\text{m}$ cubic base cell which is periodic in the x - z plane and discretized into $30 \times 30 \times 30$ or 27,000 finite elements. This base cell size was chosen to ensure consistency with the dimensions used by experimentalists who work with silicon anode structures [40, 24, 41]. Figures 1b and 1c depict schematics of the loading and boundary conditions applied to the periodic base cell for each of the structural and conduction design problems, respectively.

For the minimum compliance objective we prescribe a fixed bottom face, a free top face, and periodic boundary conditions on the side faces (Figure 1b). We also apply a design dependent volumetric strain to the structure to simulate the 300% change in volume of the silicon upon lithium insertion. The anode is assigned the material properties of $\alpha\text{-Li}_{15}\text{Si}_4$: a Young's modulus of $E_{\text{anode}} = 35$ GPa and a Poisson's ratio of $\nu = 0.23$ [42]. The other phase representing the electrolyte is modelled as void, which is a common practice in many topology optimization problems. As such, the Young's modulus of the electrolyte is set to a very small number not equal to zero in order to prevent singularities, i.e., $E_{\text{electrolyte}} = 1 \times 10^{-3}$ GPa.

For the maximum conduction objective, we model a silicon anode undergoing galvanostatic charging, where the current remains at a constant value (Figure 1c). This is implemented as a prescribed voltage of $\Phi = 0.0$ V on the top surface of the domain, and a current into the electrode from the bottom surface of the domain. We consider a maximum battery capacity of 4200 mAh/g, and a charging or C rate of $C/5$, which indicates that for the given current it will take 5 hours to fully lithiate or delithiate under ideal conditions [43]. Assuming 30% of the domain is occupied by active silicon material with a density of 2328 kg m^{-3} , we obtain a current influx of 7.33×10^{-11} A. For this problem we assume no flux on the remaining boundary faces. The silicon is prescribed an isotropic electrical conductivity of $d_{\text{anode}} = 0.067 \text{ Sm}^{-1}$ [44], while the electrolyte is an electrical insulator and is prescribed a very small conductivity value in order to avoid numerical difficulties.

Interpolation schemes are used for the design dependent material parameters, namely the Young's modulus $E(\tilde{x}_e)$, the volume expansion coefficient $\gamma(\tilde{x}_e)$, and the electrical conductivity $d(\tilde{x}_e)$. The RAMP scheme is used for the first two parameters with a maximum penalty value of $p = 8$, while the SIMP scheme is used for the electrical conductivity with a maximum penalty value of $p = 3$. Additionally, we apply density filtering and the GCMMA optimization algorithm.

4. Results and discussion

This section details the optimization results for both the individual structural and conduction objectives, and the multiobjective formulation.

4.1. Structural design objective

The optimal base cell designs for the structural objective are shown in Figure 2 for three different filter radii, $r = 2 \mu\text{m}$, $r = 1 \mu\text{m}$ and $r = 0.5 \mu\text{m}$. The filter radius is a parameter used

in a smoothing function for regularization purposes. The minimum length scale, or minimum size of structural features, is dependent on the comparative values of the domain size, number of elements and the filter radius. If the domain size and number of elements are kept constant, this filter radius variable becomes the dominant length scale parameter, controlling the size of structural features and therefore the complexity of the design. A larger filter radius results in larger structural features and therefore a more simple structure. This filter radius therefore serves the dual purpose of regularization and ensuring manufacturability of the optimal structures.

Figure 2: Optimized base cells for the structural design objective for a volume fraction of 30% and three different filter radii, (a) $r = 2 \mu\text{m}$, (b) $r = 1 \mu\text{m}$, (c) $r = 0.5 \mu\text{m}$. The top row depicts the original base cell, while the bottom row shows the shifted base cells.

Figure 2a depicts the optimal base cell for a large filter radius of $r = 2 \mu\text{m}$. For clarity, two equivalent base cells are shown. The top image depicts the original base cell, while the image beneath depicts a base cell that has been shifted by half a base cell in the in-plane directions. When repeated periodically in the x - z plane, both base cells would result in the same overall anode structure, which resembles a series of cubes that have been hollowed out by spherical shapes. Upon lithiation the struts would swell to fill the central hollow, while the entire structure would expand vertically in the unconstrained direction. This design provides the structural rigidity and porosity required to accommodate the large volume expansion due to lithiation. This

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6 simple, connected structure would be suitable for application to batteries involving both liquid
7 and polymer electrolytes.
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9 A reduction in filter radius to $r = 1 \mu\text{m}$ results in a structure with finer structural features (Fig-
10 ure 2b). In comparison to the $r = 2 \mu\text{m}$ design, the major changes include four small corner holes
11 on each face rather than one large central hole, which results in the vertical struts spanning the
12 entire face of the base cells, and the appearance of an internal floating structure that resembles a
13 wire-frame cube. During lithiation the walls of the cube would expand inwards to fill the internal
14 space, while the central floating structure would also expand. Furthermore, vertical expansion
15 would occur due to the unconstrained top face of the base cell. In terms of the objective func-
16 tion, we calculate a small 1.2% reduction in compliance relative to the $r = 2 \mu\text{m}$ design, which
17 indicates a slightly better performance possibly due to the internal structure being free to expand
18 in all directions within the central void space, thereby reducing the average stress. However the
19 free-floating nature of the internal structure introduces manufacturability challenges, particularly
20 when considering a liquid electrolyte.
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28 A further reduction of the filter radius to $r = 0.5 \mu\text{m}$ results in a very fragmented structure.
29 The side faces have now become disconnected vertical plates, and the internal structure features
30 a series of floating plates and rods. This design has severe manufacturing limitations, and also
31 features a compliance value 2.9% greater than the $r = 2 \mu\text{m}$ structure, indicating a worse perfor-
32 mance which is likely due to the fragmented nature of the design.
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36 Due to the very small difference in objective function values, the optimal design for the
37 structural objective is dictated by the simplicity and therefore manufacturability of the design.
38 The authors therefore recommend the designs obtained using the larger filter radii (Figures 2a or
39 2b) for the application of a silicon anode structure in a lithium-ion battery.
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42 We also investigate the influence of volume fraction on the optimal designs. Figure 3 plots
43 the compliance as a function of prescribed volume fraction for a filter radius of $r = 2 \mu\text{m}$. A
44 reduction in volume fraction results in thinning of the struts, thereby increasing the size of the
45 center spherical hollow. Furthermore, the compliance decreases with decreasing volume fraction,
46 indicating an improved structural performance. This is because less material in the structure
47 means there is less material to undergo lithiation so the overall volume expansion is smaller.
48 Additionally, the larger spherical hollow can better accommodate the volume expansion of the
49 silicon, therefore resulting in a lower state of stress. However, a trade-off is required as the
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anode must have sufficient active material to achieve the desired electrochemical performance, therefore, a volume fraction of at least $V_f = 0.3$ is recommended.

Figure 3: A plot of the structural objective value as a function of the prescribed volume fraction for $r = 2 \mu\text{m}$. This plot shows that a lower volume fraction design will result in improved structural performance.

4.2. Electric conduction design objective

The optimal base cell structures for the electric conduction objective are shown in Figure 4. Regardless of filter radius size, the optimal designs resemble rod-like structures that feature direct electronic pathways through the thickness of the electrode, similar to the silicon nanowire structures used experimentally. Upon lithiation the rods would expand in both diameter and length whilst remaining rigidly fixed to the current collector, or bottom face of the design domain. A reduction in filter radius allows for a slightly more complex structure, where the anode fans out towards the bottom of the domain. This feature results in a greater surface area in contact with the current collector, allowing more current to enter and flow through the structure. The smaller filter radii designs are accompanied by a reduction in the value of electrical compliance, indicating that the tapered structures offer improved electric conduction performance.

In Figure 5 the electric conduction objective is plotted as a function of volume fraction. For this design objective, an increase in volume fraction results in a larger radius of the cylindrical

Figure 4: Optimized base cells for the electric conduction design objective for a volume fraction of 30% and three different filter radii, (a) $r = 2 \mu\text{m}$, (b) $r = 1 \mu\text{m}$, (c) $r = 0.5 \mu\text{m}$.

structure. Furthermore, for larger volume fractions the structure becomes increasingly tapered, allowing more current to be transported through the silicon structure from the base of the cell. For this problem the electrical compliance decreases with increasing volume fraction, which indicates that a structure with a greater volume of conductive material will be a better conductor of electricity. It is interesting to note that the structural and conduction design objectives favor the lowest possible and highest possible volume fractions, respectively. Therefore these objectives are competing in nature, and a trade-off is required to adequately satisfy both design criteria.

Figure 5: A plot of the conduction objective value as a function of the prescribed volume fraction for $r = 2 \mu\text{m}$. This plot shows that a higher volume fraction design will result in improved electric conduction performance.

4.3. Combined objective results

The results of the multiobjective topology optimization analysis are shown in Figure 6 for a volume fraction of 30% and a filter radius of $r = 1 \mu\text{m}$. The Pareto front, presented in Figure 6a, illustrates how the competing design objectives influence the resulting topologies as the weights are varied from the full structural design, $w_s = 1$, to the full conduction design, $w_s = 0$.

(a)

(b)

(c)

Figure 6: (a) The Pareto front and corresponding optimal topologies for $V_f = 0.30$ and a filter radius of $r = 1 \mu\text{m}$. (b) and (c) show 2×2 base cells and 2×2 shifted base cells of the intermediate frame structure, respectively. This structure provides the best design compromise in terms of the combined minimum structural compliance and maximum electrical conduction objectives for $V_f = 0.30$.

When the weight is $w_s = 1$, only the structural objective is considered and the resulting optimal structure is the same as that shown in Figure 2b. The value of the normalized structural objective is at a minimum, while the normalized conduction objective takes its maximum value of 1. When the weight is $w_s = 0$, only the electric conduction objective is considered and the resulting optimal structure is the equivalent shifted base cell of Figure 4b. The value of the normalized conduction objective is at a minimum, while the normalized structural objective takes its maximum value of 1.

As the Pareto weight decreases from $w_s = 1$, the conduction objective begins to have an effect on the optimized structure, causing the top and bottom faces to lose material and the side faces to gain material so that the structure is more oriented in the direction of current flow. By $w_s = 0.65$ the internal floating structure disappears, resulting in a intermediate rigid frame structure that spans the central section of the Pareto front. A further reduction in the Pareto weight causes the conduction objective to dominate. Therefore, material begins to accumulate at the corners of the structure, while the central sections of the side faces lose material until the full conduction structure is obtained at $w_s = 0$.

The intermediate frame structure offers a clear compromise between the structural and conduction design objectives. The 2 x 2 base cell and shifted base cell representations of this frame structure are depicted in Figures 6b and 6c, respectively. This design provides the structural rigidity required by the minimum structural compliance objective. Furthermore, by virtue of the volume fraction, there are distinct spaces for the anode structure to expand into during lithiation. **The anode structure will swell upon lithiation to occupy the internal spaces, and also expand in the vertical direction.** The electrical conduction objective is achieved by using direct conduction pathways from the top to the bottom of the anode structure. For a volume fraction of $V_f = 0.30$, this frame structure is recommended as the best silicon anode design in terms of combined minimum structural compliance and maximum electrical conduction.

Multiobjective topology optimization was also undertaken for a larger prescribed volume fraction of 60% and a filter radius of $r = 1 \mu\text{m}$, shown in Figure 7. In addition to the original base cell (denoted A.), the shifted base cell (denoted B.) is shown for the higher Pareto weights to assist with visualization. The progression of structures along the Pareto front appears similar to the results shown in Figure 6. Starting at the full structural objective, $w_s = 1$, a cubic structure with holes on the side faces is observed. Unlike the 30% full structural design, the internal

structure is connected to the side walls due to the larger volume of material.

(a)

(b)

(c)

Figure 7: (a) The Pareto front and corresponding optimal topologies for $V_f = 0.60$ and a filter radius of $r = 1 \mu\text{m}$. (b) and (c) show 2×2 base cells and 2×2 shifted base cells of the intermediate frame structure, respectively.

A slight decrease in the Pareto weight results in the conduction objective beginning to influence the solution and create a more vertically-oriented structure. The holes on the side faces close, while holes on the top face appear. At $w_s = 0.80$, the internal structure becomes a connected conduction pathway between the top and bottom faces. A further reduction in Pareto

weight causes the internal structure to begin to disappear, as shown by base cell B of $w_s = 0.60$, and material begins to accumulate near the sides of the domain. In the center section of the Pareto curve, an intermediate structure is produced which is similar to the frame structure obtained for a $V_f = 0.30$. However, a notable difference is that this intermediate structure does not line the side faces of the base cell. For clarity, Figures 7b and 7c show 2 x 2 base cells and 2 x 2 shifted base cells of this intermediate structure, respectively, which exhibits direct conduction pathways and has the interesting feature of the side faces being slanted rather than vertical. This allows material within the original base cell being able to expand in both inward and outward directions upon lithiation. A small amount of expansion would result in the structure resembling a high volume fraction frame-like structure, similar to that depicted in Figures 6b and 6c. As the weight is further decreased and the conduction objective begins to dominate, the sides of the intermediate structure recede and material gathers at the corners to eventually produce the tapered cylindrical structures for $w_s = 0$.

Figure 8 shows the Pareto curves for a filter radius of $r = 1 \mu\text{m}$ and the volume fractions $V_f = 0.30$, $V_f = 0.45$ and $V_f = 0.60$ on the same plot. The individual Pareto curves appear to be constituents of the full Pareto front, which clearly depicts the competing nature of the objective functions. Furthermore, this plot illustrates the range of attainable objective function values for the different volume fractions. As expected, the lowest structural objective value is obtained at the lowest volume fraction, while the lowest conduction objective value is obtained by the highest volume fraction. Therefore, in order to best resolve the primary design concern of lithiation-induced mechanical degradation, the recommended multifunctional silicon anode design is the rigid frame structure using the lowest practicable volume fraction.

5. Summary and conclusions

This study utilizes topology optimization methods to determine optimal multifunctional silicon anode structures for Li-ion battery applications. An improvement in battery performance may be achieved through the replacement of traditional graphite anodes with high capacity silicon, however, the use of silicon as an anode material has resulted in several design challenges. The silicon anode structures are subject to a 310% volume expansion upon lithiation, which causes severe damage, such as pulverization of the active particles and disconnected charge transport paths. Additionally, the low intrinsic electric conductivity of silicon results in poor

Figure 8: Pareto curves for $r = 1 \mu\text{m}$ and $V_f = 0.30$, $V_f = 0.45$ and $V_f = 0.60$.

rate performance due to slow electron transport through the material.

This study addresses these structural and conduction design challenges using topology optimization methods. Topology optimization is the most general form of structural optimization that is used to determine the optimal distribution of material within a design domain for a given set of loading and boundary conditions. The methodology employed in this study utilizes density methods, regularization techniques, continuation schemes and mathematical programming methods such as the globally convergent method of moving asymptotes. The design challenges associated with the lithiation-induced volume expansion and the low electrical conductivity of silicon are addressed using two design objectives, namely minimum compliance subject to design dependent loading, and maximum electrical conduction through the structure, respectively. The objectives are first considered individually, then subsequently addressed simultaneously in a multiobjective formulation.

For both design objectives, the concept of a minimum structural feature size is used to investigate the manufacturability of the optimal designs. It is found that specifying a larger minimum structural feature size results in a more simplistic, connected structure that is easier to manufac-

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6 ture. For the structural objective, the optimal design resembles a series of cube-like structures
7 with internal hollow spaces and holes on the side faces. This design provides the rigidity and
8 porosity required by the minimum compliance objective. A smaller volume fraction is found to
9 yield an improved structural performance due to less material undergoing lithiation and therefore
10 volume expansion, and also larger void spaces to better accommodate this expansion.
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14 For the electric conduction design objective, the analysis produces rod-like structures that
15 provide efficient conduction pathways through the thickness of the anode structure, similar to
16 the silicon nanowire structures used by experimentalists. A reduction in the minimum length
17 scale results in an increased tapering out from the base of the structure, allowing more current to
18 enter the anode. Furthermore, a greater volume fraction of silicon, by means of a larger cross-
19 sectional area of the rod structure, is found to yield a lower value of electrical compliance and
20 therefore improved electric conduction performance. These structural and conduction design
21 criteria favor opposing volume fractions, indicating that a trade-off is required to meet both
22 design requirements.
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26 A multiobjective topology optimization formulation is subsequently used to simultaneously
27 address the competing design criteria. The Pareto fronts for volume fractions of 30% and 60%
28 are derived using a weighted sum method. The optimal structures show a clear progression from
29 the full structural objective $w_s = 1$ to the full conduction objective $w_s = 0$, where the influence
30 of the conduction objective causes the anode structure to be increasingly aligned in the direction
31 of current flow. For both volume fractions, an intermediate frame structure is deemed to be an
32 excellent compromise between the competing design criteria, as it provided both the structural
33 rigidity and direct conduction pathways required by these design objectives. Furthermore, the
34 frame structure for the larger volume fraction features slanted sides, allowing expansion in both
35 internal and external directions.
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39 The Pareto fronts for the three volume fractions of 30%, 45%, and 60% are then combined to
40 form an overall Pareto front, which provides a key insight into the competing nature of the objec-
41 tive functions, and also the attainable range of objective function values for the different volume
42 fractions. As the mechanical degradation associated with the lithiation-induced expansion is the
43 primary design concern for this study, the recommended multifunctional silicon anode design is
44 the rigid frame structure using the lowest practicable volume fraction whilst ensuring sufficient
45 active material to yield the required electrochemical performance of the battery.
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6. Outlook

The developments and results presented in this **computational** study provide a solid foundation for the informed design and development of optimal multifunctional silicon anode structures for use in lithium-ion batteries. The next phase would be for these structures to be manufactured and tested by experimentalists. The recommended optimal silicon anode structures depicted in Figures 6b and 7b exhibit features similar to various silicon anode structures that have been manufactured experimentally, including the one dimensional electronic pathways of silicon nanowires [19, 20, 21], the novel honeycomb thin film structures by Baggetto et al. (2011) [40], and the one-dimensional porous silicon structures by Shin et al. (2012) [17]. It is therefore anticipated that similar methods of manufacture could be used to produce these optimal designs, namely deposition and electrochemical etching techniques.

Potential areas of experimental investigation include testing the storage performance, lithiation/delithiation behaviour, and cyclability of the optimal structures. These silicon anode designs have been optimized to minimize the state of stress in the structure and therefore, for a structure on this length scale with a copper coating, these designs are expected to achieve good performance in terms of surface cracking, particle fracture, and cyclability. Improved cycling performance may be obtained by further tailoring of the geometric dimensions of the base cell. For example, Liu et al. (2012) [45] reported greatly improved cyclability for particle sizes below 150 nm in silicon nano powder, therefore a reduction in length scale may yield improved performance.

It is anticipated that these designs may undergo other slight modifications and enhancements during the experimental testing phase. For example, in order to most effectively optimize the underlying silicon structure and meet computational demands, the optimal structures presented in this paper were designed using silicon active material only. Experimental studies have shown that the addition of a conductive coating, such as copper, yields improved performance in terms of electrical conductivity and cyclability [46, 47]. Therefore, the addition of a copper coating to these silicon anode structures may prove beneficial.

The results of this work may also be incorporated into numerical models to simulate the electrochemical performance of the entire battery system. The present study can also be extended to incorporate non-linear elasticity, direct stress-based topology optimization, or additional objective functions such as maximizing the diffusion of Li-ions into the anode structure or maximizing

the buckling load. Furthermore, the methodology developed in this study can be directly applied to other design problems, such as electrochemical systems, thermal insulators, propellants, or aerospace applications.

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