

Influence of Graphite on Lithiation-Induced Strain in Crystalline Silicon Anodes

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Influence of Graphite on Lithiation-Induced Strain in Crystalline Silicon Anodes

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Abstract

Silicon has attracted considerable attention as a next-generation anode material for lithium-ion batteries because of its high theoretical capacity. However, severe volume expansion and structural degradation during lithiation remain major barriers to practical application. In this study, we investigated the effect of graphite presence on the suppression of lithiation-induced strain in crystalline silicon using molecular dynamics (MD) simulations. A graphite-containing Si-C-Li composite system (Group A) and a graphite-free Si-Li system (Group B) were compared under identical conditions using a machine-learning potential based on MACE. The structural evolution of the two systems was examined using spatial number-density profiles, coordination number, radial distribution function, and the tetrahedral order parameter. Analysis of local Green-Lagrangian strain showed that graphite-containing system maintained lower strain, approximately 5.8 times lower than system without graphite, whereas the in-plane components differed by only around 1.7 times. These results suggest that the presence of graphite may mitigate the spatial localization of lithiation-induced structural and mechanical deformation.

Keywords : Silicon anode; Lithiation; Graphite; Strain localization; Molecular dynamics

1. Introduction

As demand for lithium-ion batteries (LIBs) continues to increase, silicon (Si) has attracted considerable attention as a high-capacity anode material capable of overcoming the energy-density limitations of conventional graphite anodes, which have a theoretical capacity of

approximately 372 mAh/g [1, 2]. Silicon offers an exceptionally high theoretical capacity of up to approximately 4200 mAh/g through alloying with lithium, but it suffers from severe volume expansion of more than 300% during lithiation. This extreme volume change generates substantial mechanical stress within anode particles, resulting in particle cracking, pulverization, and repeated failure of the solid electrolyte interphase (SEI) [3]. In particular, volume expansion during the lithiation of crystalline silicon does not proceed isotropically but exhibits a high degree of anisotropy along specific crystallographic directions, such as the $\langle 110 \rangle$ direction [4]. This heterogeneous expansion mechanism intensifies local stress concentration within the structure and is considered a key factor that accelerates mechanical failure of the electrode [5].

To address these mechanical degradation issues, extensive efforts have focused on nanostructuring silicon or designing Si@C composites coated with carbon-based materials to buffer volume expansion [6-7]. More recently, machine-learning potentials (MLPs), which combine density functional theory (DFT)-level accuracy with the computational efficiency of molecular dynamics (MD), have enabled large-scale atomistic simulations [8-9]. For example, previous studies have used active-learning-based neural network potentials to simulate Li diffusion at silicon-graphite interfaces and to analyze how the presence of carbon layers affects the chemical diffusion pathways and structural evolution of Li [10].

However, previous studies have focused mainly on interfacial chemical reactions and Li diffusion, whereas atomistic comparisons of how lithiation-induced local structural rearrangement and anisotropic deformation are spatially distributed remain relatively limited. In this study, we correlated the effects of graphite presence or absence on Li distribution, local coordination environment, structural order, and Green-Lagrangian strain [11]. In particular, models trained primarily on relaxation trajectories, such as those based on MPtrj, may systematically underestimate energies and forces in high-energy, non-equilibrium states involving bond breaking or severe structural distortion [12].

To overcome this limitation, we adopted the MACE (Higher Order Equivariant Message

Passing Neural Network) architecture based on the OMat24 (Open Materials 2024) dataset, which contains more than 110 million diverse non-equilibrium atomic configurations [13]. This approach was selected to provide broader coverage of non-equilibrium configurations and large local distortions encountered during lithiation. Beyond the conventional perspective of chemical diffusion, this study focuses on the spatial distribution of lithiation-induced local structural rearrangement and anisotropic deformation.

By comparing a graphite-containing Si-C-Li composite system with a graphite-free Si-Li system under identical molecular dynamics simulation conditions, we aim to determine how the graphite layer affects the spatial distribution of lithiation-induced structural rearrangement and strain.

2. Materials and Methods

Two systems were constructed such that the Si slab and Li reservoir were kept identical where possible, while the interfacial region was either graphite separated or direct Li-Si contact. A graphite-containing Si-C-Li system (Group A, 1424 atoms) and a graphite-free Si-Li system (Group B, 752 atoms) were compared under identical conditions. Both systems were modeled as quasi-two-dimensional slab structures with free surfaces along the x direction and periodic boundary conditions along the y and z directions (Fig. 1). In Group A, Si-C-Li were sequentially arranged along the x direction to represent a pathway in which Li penetrates through the graphite layer into the Si core with the (100) crystal plane. Group B was constructed as a control structure by removing only the graphite region from Group A so that Li and Si were in direct contact. Interatomic interactions were described using a potential based on the MACE architecture [14]. The MACE-OMAT model trained on the OMat24 dataset was selected because of its broad coverage of non-equilibrium atomic configuration relevant to the large local distortions and amorphization occurring during lithiation [13].

All simulations were performed using LAMMPS [15] with the metal unit system, and f p

p boundary conditions were applied. The timestep was set to 1 fs. Following energy minimization, the systems were heated from 300 K to 600 K under the NPT ensemble for 30 ps, followed by a 1 ns production run at 600 K under the NPT ensemble. Structural changes were analyzed using five metrics: spatial number-density profiles of Si and Li (density profile), changes in Si-Si and Si-Li coordination numbers, local coordination structure through radial distribution function (RDF), tetrahedral order parameter, and atomistic local strain (Green-Lagrangian strain). Rather than presenting every frame in the figures describing structural evolution, representative time points corresponding to the early, middle, and late stages of lithiation were selected for visual clarity. Further details on the simulations are included in Supplementary Information.

3. Results and Discussion

We compared the differences in atomic structure and strain that developed during lithiation in Group A and Group B. First, the spatial distribution of Li and the local structural changes in Si were analyzed, and these results were then correlated with Green-Lagrangian strain to evaluate the role of graphite.

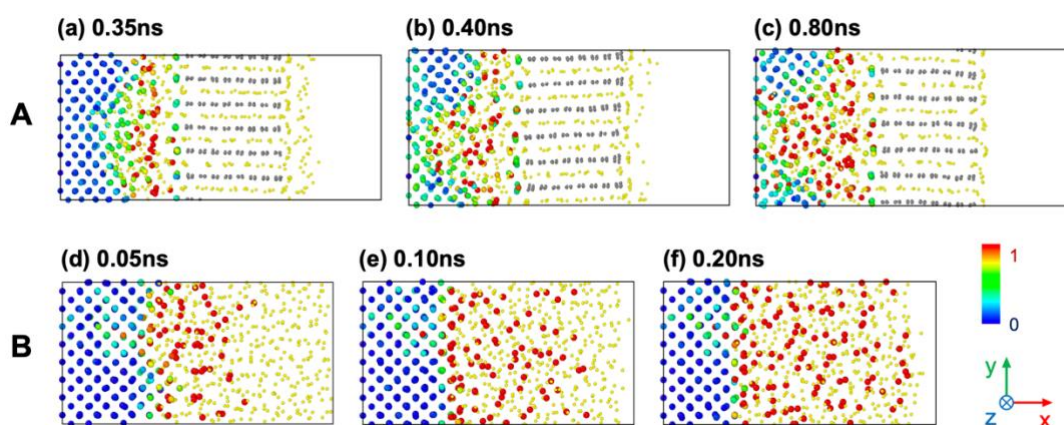


Fig. 1 Time-resolved atomic configurations during lithiation of (a) Group A (Si-C-Li) and (b) Group B (Si-Li). Silicon atoms are colored according to local shear strain; graphite carbon and Li atoms are shown in gray and yellow, respectively.

Fig. 1 shows representative atomic configurations of Group A and Group B during

lithiation. Both systems initially maintain relatively ordered crystalline Si structures, but as lithiation proceeds, the influx of Li into the Si region changes the arrangement and local structure of Si atoms. To quantitatively compare these structural differences, the spatial number-density profiles of Li and Si along the x direction were analyzed (Fig. 2). The Li distribution patterns were markedly different between Group A and Group B. In Group A, which contains graphite, the reaction between Li and Si occurred over a relatively broad spatial region rather than being confined to a specific surface region. In contrast, in Group B, where Li and Si were in direct contact, the presence of Li and the associated structural changes were concentrated near the Si-Li surface [16]. This spatial difference was also consistently observed in the subsequent spatial distributions of Si-Li and Si-Si coordination-number changes.

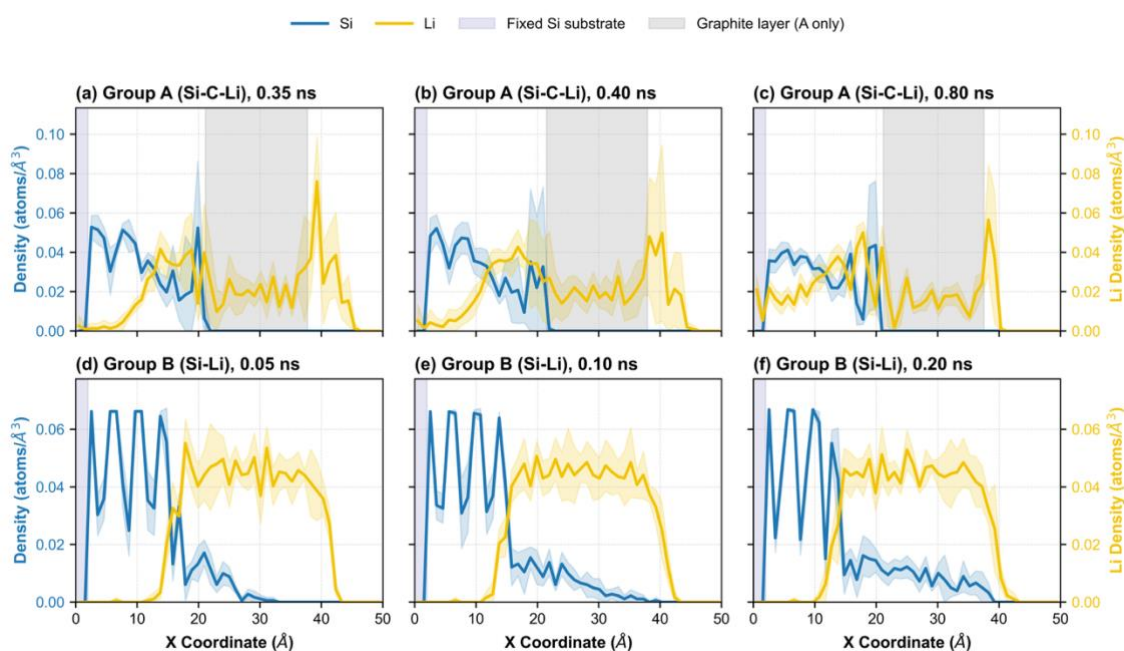


Fig. 2 Spatial number-density profiles of Si and Li along the x direction during lithiation (a)-(c) Group A and (d)-(f) Group B. The fixed Si substrate and graphite layer are indicated by blue and gray shaded regions, respectively.

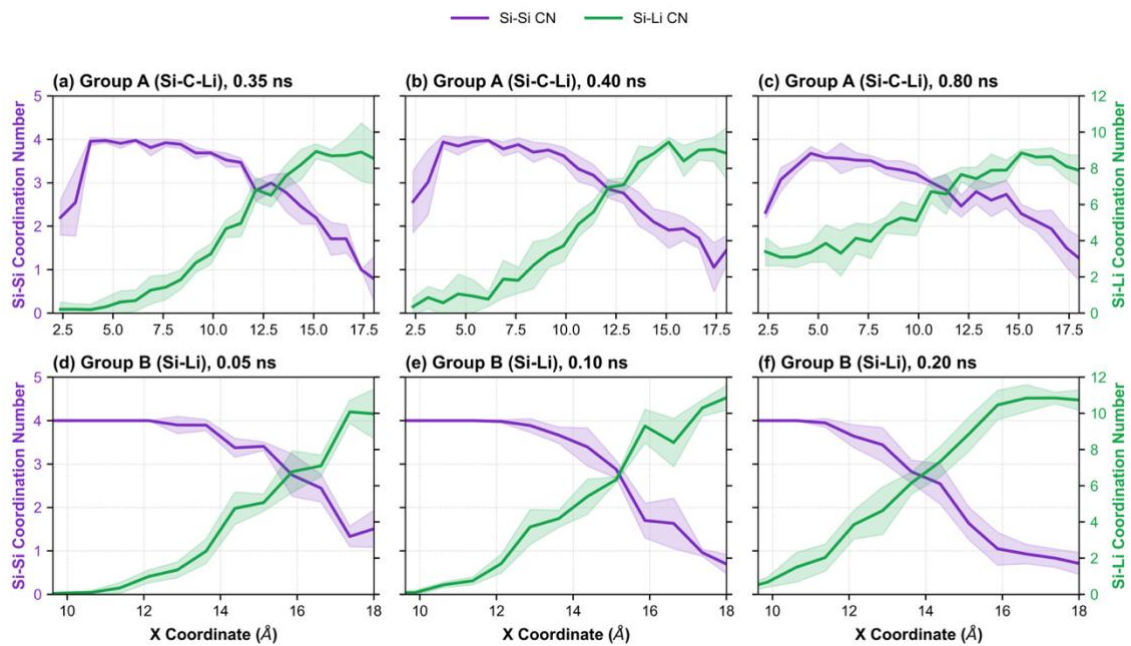


Fig. 3 Spatial profiles of Si-Si coordination number (CN, purple) and Si-Li coordination number (CN, green) along the x-axis during lithiation for Group A and Group B.

To examine the changes in interatomic coordination, Si-Si and Si-Li coordination numbers (CNs) were analyzed (Fig. 3). The spatial evolution of the Si-Li interfacial region differed between the two systems. In Group A, the increase in Si-Li CN progressively extended toward the interior of the Si region with increasing lithiation time, whereas in Group B, the Si-Li interfacial region shifted from approximately 16 Å toward 14 Å. The cutoff used to calculate the Si-Li CN was 3.75 Å, corresponding to the first Si-Li coordination shell identified from the Si-Li radial distribution function. An increase in Si-Li CN indicates that Li enters the vicinity of Si and forms new nearest-neighbor coordination environments, while a decrease in Si-Si CN indicates changes in the bonding environment of crystalline Si [17]. These spatial differences became more apparent in the magnitude and extent of the coordination-number changes. In Group A, with increasing lithiation time, the increase in Si-Li CN and decrease in Si-Si CN progressively extended over a broader Si region. In particular, at 0.80 ns, these coordination changes occurred over the range of $x = 2\text{-}18$ Å, with a greater magnitude closer to the Li-facing surface. In contrast, in Group B, high Si-Li CN and low Si-Si CN were concentrated mainly in the $x = 10\text{-}18$ Å region

in contact with Li. In this region, Si-Li CN increased to approximately 10 or higher, while Si-Si CN decreased to approximately 1 or lower. In contrast, relatively high Si-Si CN was maintained in the interior region. Thus, the coordination rearrangement in Group A progressively extended across the Si region, whereas in Group B it remained localized near the Li-facing surface.

To examine how these changes in coordination relate to the loss of order in the crystalline structure, the tetrahedral order parameter, q , was analyzed (Fig. 4). The parameter q measures how closely the local bonding angles of Si atoms resemble an ideal tetrahedral structure. A high q indicates preservation of the tetrahedral structure of crystalline Si, whereas a decrease in q indicates disruption of bond-angle order and progression toward amorphization [18-19]. In addition, the Si region was divided into three regions—inner, middle, and surface—according to their distance from the Li reservoir, and the temporal evolution of structural order was compared. In the surface region, q decreased in both systems, but the rates differed substantially. In Group B, q decreased to 0.2 or below within approximately 0.1 ns, whereas in Group A it reached approximately 0.2 only after about 0.5 ns. This is consistent with the much faster increase in Si-Li CN and decrease in Si-Si CN observed in the surface region of Group B. Thus, in Group B, where Li is in direct contact with Si, the rapid formation of Si-Li coordination and rearrangement of the Si-Si coordination environment at the early stage leads to rapid loss of tetrahedral order in the surface Si.

In the middle region, q decreased with time in both systems. In Group A, q decreased over approximately 0.3-0.6 ns and then remained at approximately 0.5, whereas in Group B it continued to decrease to approximately 0.4 at 1.0 ns.

In the inner region, however, the behavior of the two systems again differed clearly. In Group A, q gradually decreased after approximately 0.3 ns and reached approximately 0.6 at 1.0 ns, whereas Group B maintained a relatively high q value through 1.0 ns. Thus, the q analysis shows that the difference between Group A and Group B is not simply a difference in the overall magnitude of structural order, but rather a difference in how structural amorphization is spatially

distributed. In Group A, the reduction in structural order is distributed over a relatively broad region, whereas in Group B, rapid loss of order occurs in the surface region while relatively high structural order is retained in the interior.

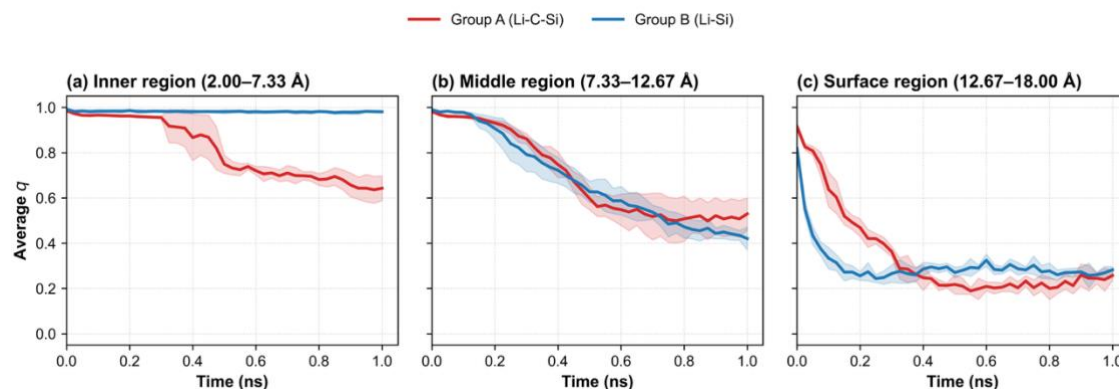


Fig. 4 Time evolution of the tetrahedral order parameter q for Inner region (a, farthest from the Li reservoir), Middle region (b, middle), and Surface region (c, adjacent to the Li reservoir).

The preceding CN and q analyses show changes in the local coordination environment and bond-angle order of Si. To further determine how these changes affect the distribution of interatomic distances, Si-Si and Si-Li radial distribution functions (RDFs) were analyzed (Fig. 5) [17,20]. At the initial state of 0.0 ns, both systems exhibited a distinct first Si-Si peak near 2.4 Å, followed by additional peaks near 3.8 Å and 4.6 Å. This indicates that both systems maintained the ordered atomic arrangement characteristic of crystalline Si in the initial state.

As lithiation proceeded, the second and third peaks in the Si-Si RDF of both systems gradually decreased in intensity and broadened. This indicates that, rather than a substantial change in the distance of the nearest-neighbor Si-Si bond itself, the regularity of the atomic arrangement maintained at longer distances progressively broke down. In particular, in Group B, the second and third peaks weakened and broadened rapidly from approximately 0.25 ns onward, whereas the same changes occurred in Group A but progressed more gradually. In contrast, the position of the first Si-Si peak showed no substantial change in either system. Therefore, the structural changes occurring during lithiation can be interpreted as a process in which structural

order of the crystalline atomic arrangement breaks down before a substantial change in the nearest-neighbor Si-Si bond distance. This is consistent with the decrease in Si-Si coordination and tetrahedral order observed in the CN analysis.

In the Si-Li RDF, no distinct peak was observed initially, but a new first peak formed near 2.7 Å as lithiation proceeded. This indicates the formation of a new nearest-neighbor Si-Li coordination environment as Li entered the vicinity of Si. In particular, the Si-Li RDF peak began to appear after approximately 0.25 ns in Group B, whereas it became more pronounced after approximately 0.5 ns in Group A. This temporal difference indicates that the initial formation of Si-Li coordination proceeds rapidly in Group B because of direct Li-Si contact, whereas Si-Li-related structural changes proceed more gradually in the graphite-containing Group A.

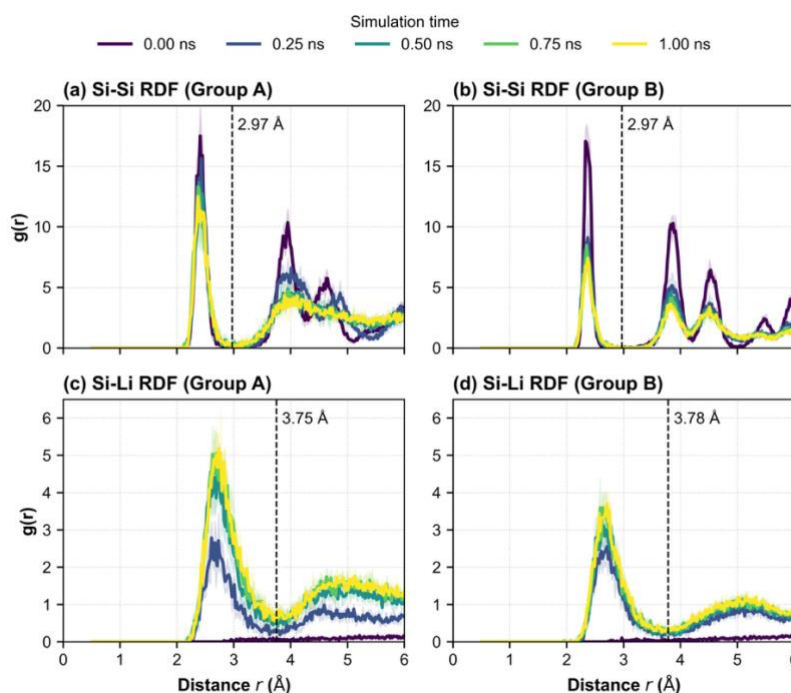


Fig. 5 Time evolution of Si-Si and Si-Li RDF for Group A and Group B ($t = 0-1.0$ ns). (a) Si-Si RDF, Group A; (b) Si-Si RDF, Group B; (c) Si-Li RDF, Group A; (d) Si-Li RDF, Group B. Dashed lines and annotated values indicate the coordination cutoff radius (first minimum after the nearest-neighbor peak) used for coordination number calculations.

The spatial differences in these structural changes were ultimately reflected clearly in the atomistic strain behavior. Fig. 6 compares the evolution of the E_{xx} , E_{yy} and E_{zz} components of the local Green-Lagrangian strain tensor in Group A and Group B during lithiation [21]. Because the x direction corresponds to the direction of Li penetration into Si, the component can reflect the spatial characteristics of Si-Li coordination formation and structural breakdown identified above.

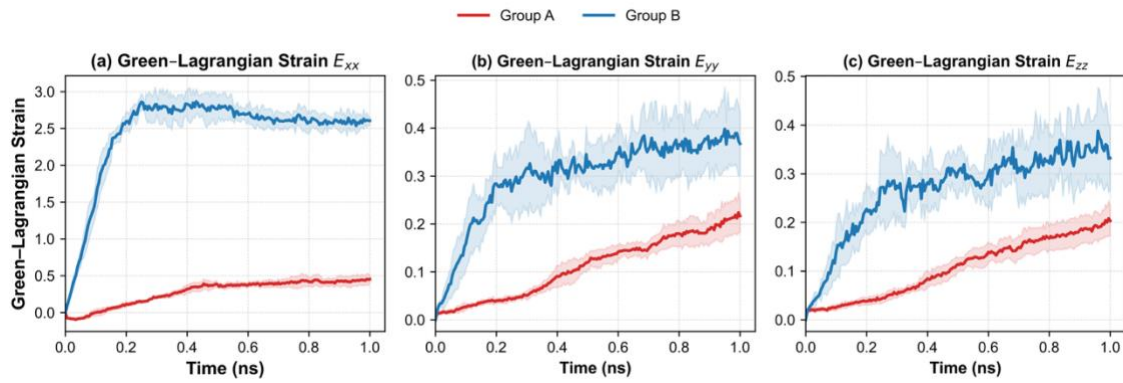


Fig. 6 Time evolution of the E_{xx} , E_{yy} and E_{zz} components of the local Green-Lagrangian strain tensor for Group A and Group B. The x-direction corresponds to the Li insertion direction.

Table 1. Strain values represent the ensemble-averaged E_{xx} , E_{yy} and E_{zz} components of the dimensionless local Green-Lagrangian strain tensor.

Direction	Group A (1.0 ns)	Group B (1.0 ns)	B/A ratio
XX (Li insertion direction)	0.45	2.6	5.8
YY (in-plane)	0.22	0.37	1.7
ZZ (in-plane)	0.21	0.33	1.6

At 1.0 ns, the average E_{xx} was approximately 0.45 in Group A and approximately 2.60 in Group B, indicating that Group B exhibited a value approximately 5.8 times higher (Table 1). In particular, E_{xx} in Group B increased rapidly during the initial approximately 0.2 ns and then remained at a high level. This temporal behavior corresponds to the rapid formation of Si-Li coordination, breakdown of Si-Si coordination, and decrease in tetrahedral order observed in the surface region of Group B at the early stage. Thus, the large Green-Lagrangian strain along the x

axis, the Li insertion direction, in Group B is consistent with rapid structural rearrangement occurring within a narrow surface region in direct contact with Li.

In contrast, the increase in E_{xx} was relatively gradual in Group A. As described above, this can be interpreted as resulting from the fact that Li-related structural changes and rearrangement of Si-Si coordination were not confined to a specific surface region but were distributed over a broader Si region in Group A. Accordingly, Group A exhibited structural rearrangement over a wider region rather than abrupt local deformation concentrated in a specific region, leading to mitigation of strain localization along the x direction.

The strain in the y and z directions was also higher in Group B than in Group A. However, the ratios of the strains obtained from Group B to those for Group A (B/A ratios) at 1.0 ns were approximately 1.7 and 1.6, respectively, which were smaller than the value of approximately 5.8 in the XX direction. This indicates that the difference between the two systems is not limited to the magnitude of the overall strain but also involves a directional difference in the degree to which strain becomes localized along the Li insertion direction. In Group B, in particular, the E_{xx} was much larger than E_{yy} and E_{zz} suggesting that the surface-localized lithiation and the associated structural rearrangement are closely related to strain localization along the x axis, the Li insertion direction. In contrast, this directional localization was relatively mitigated in Group A, resulting in a more spatially distributed strain state. Taken together, these results suggest that the presence of graphite does not simply reduce Si strain uniformly, but rather changes the spatial extent of lithiation and the manner in which the associated structural rearrangement proceeds.

In graphite-containing Group A, Li was distributed over a broad region including the graphite and Si-graphite interface, and the increase in Si-Li coordination and decrease in Si-Si coordination consequently progressed toward the interior of the Si region. At the same time, the reduction in tetrahedral order extended over a broader Si region, while the Si-Si RDF showed a more gradual loss of structural order in Group A at the system level. The resulting strain was distributed relatively broadly rather than becoming concentrated in a specific surface region.

In contrast, in graphite-free Group B, direct Li-Si contact led to rapid formation of Si-Li coordination near the surface, accompanied by rapid decreases in Si-Si coordination and tetrahedral order. These changes occurred together with the rapid attenuation of the higher-order peaks in the Si-Si RDF, indicating accelerated loss of structural order in Group B. Combined with the spatially resolved CN and q analyses, these results indicate that structural disorder developed more rapidly near the Li-facing surface than in the interior [22]. However, relatively high Si-Si coordination and tetrahedral order were retained in the interior, limiting the loss of structural order compared with the surface. As a result, a pronounced spatial heterogeneity developed between the rapidly changing surface structure and the interior crystalline structure, and this heterogeneous response is considered to have manifested as the large Green-Lagrangian strain particularly along the Li insertion direction.

The primary difference observed between the two systems is the spatiotemporal mode in which lithiation and structural rearrangement proceed. In Group B, direct Li-Si contact leads to a rapid structural rearrangement at the surface, which is associated with localized Si bond breakdown and large x-direction strain. In contrast, in Group A, the reaction between Li and Si is distributed over a broader region in the presence of graphite, and the associated structural changes and strain are likewise distributed over a broader Si region. Therefore, the role of graphite in this study can be interpreted not simply as a physical barrier that suppresses volume expansion, but as a factor that may mitigate the spatial localization of the structural and mechanical responses accompanying lithiation.

4. Conclusions

In this study, we compared a graphite-containing Si-C-Li system (Group A) and a graphite-free Si-Li system (Group B) to analyze the effect of the graphite layer on lithiation-induced structural changes in Si in terms of strain, local coordination environment, and the spatial distribution of lithiation. At 1.0 ns, Group B exhibited substantially larger strain than Group A, with average

strain values of 2.6 and 0.45, respectively, along the Li insertion direction, corresponding to a 5.8-fold difference. In contrast, the differences between the two groups in the in-plane directions were relatively small, with B/A strain ratios of 1.7 and 1.6 in the YY and ZZ directions, respectively. These results show that the strain difference associated with the presence of the graphite layer is particularly pronounced along the Li insertion direction.

Therefore, the difference observed between the two systems cannot be explained simply as a temporal delay in lithiation-induced structural evolution, but instead reflects differences in the spatial pathway of structural rearrangement and deformation. In Group A, Li-related structural changes in the Si-Si coordination environment were distributed over a relatively broad Si region, and the resulting strain was likewise relatively distributed. In contrast, in Group B, direct contact between Li and Si induced rapid structural rearrangement near the surface, producing strong structural heterogeneity relative to the interior and consequently large E_{xx} Green-Lagrangian strain along the Li insertion direction.

Thus, the role of graphite can be interpreted as mitigating the spatial localization of the structural and mechanical changes accompanying lithiation rather than simply reducing the total strain of Si. These results suggest that graphite may serve as a structural component for regulating lithiation pathways and strain distributions in Si-based anodes.

Nevertheless, this study has several limitations. A simplified quasi-two-dimensional model was used, and only a single lithiation process was considered over a limited simulation time. In addition, because only one Li species and a single lithiation cycle were considered, further studies incorporating three-dimensional structures, longer simulation times, repeated lithiation/delithiation cycles, and various carbon-layer structures are required to determine whether the same trends persist under more realistic conditions.

Data Availability

The data that support the findings of this study are available from the corresponding author upon reasonable request.

REFERENCES

1. J. Sakabe, N. Ohta, T. Ohnishi, K. Mitsuishi, K. Takada, Porous amorphous silicon film anodes for high-capacity and stable all-solid-state lithium batteries. *Commun. Chem.* 1, 24 (2018). <https://doi.org/10.1038/s42004-018-0026-y>
2. H. Ahmed, G. Simoes dos Reis, P. Molaiyan, A. Lähde, U. Lassi, Silicon/carbon composite anode materials for lithium-ion batteries: materials design and synthesis, current state, progress, and future perspectives. *Prog. Energy* 7, 022003 (2025). <https://doi.org/10.1088/2516-1083/ada90f>
3. Y. Zhang, N. Du, D. Yang, Designing superior solid electrolyte interfaces on silicon anodes for high-performance lithium-ion batteries. *Nanoscale* 11, 19086-19104 (2019). <https://doi.org/10.1039/C9NR05748J>
4. S.W. Lee, M.T. McDowell, J.W. Choi, Y. Cui, Anomalous shape changes of silicon nanopillars by electrochemical lithiation. *Nano Lett.* 11, 3034-3039 (2011). <https://doi.org/10.1021/nl201787r>
5. M.T. McDowell, S.W. Lee, W.D. Nix, Y. Cui, 25th Anniversary Article: Understanding the lithiation of silicon and other alloying anodes for lithium-ion batteries. *Adv. Mater.* 25, 4966-4985 (2013). <https://doi.org/10.1002/adma.201301795>
6. X. Zhang, J. Weng, C. Ye, M. Liu, C. Wang, S. Wu, Q. Tong, M. Zhu, F. Gao, Strategies for controlling or releasing the influence due to the volume expansion of silicon inside Si-C composite anode for high-performance lithium-ion batteries. *Materials* 15, 4264 (2022). <https://doi.org/10.3390/ma15124264>
7. J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries. *Carbon Energy* 1, 57-76 (2019). <https://doi.org/10.1002/cey2.2>
8. J. Behler, Perspective: Machine learning potentials for atomistic simulations. *J. Chem. Phys.* 145, 170901 (2016). <https://doi.org/10.1063/1.49666192>
9. V.L. Deringer, M.A. Caro, G. Csányi, Machine learning interatomic potentials as emerging tools for materials science. *Adv. Mater.* 31, 1902765 (2019).

<https://doi.org/10.1002/adma.201902765>

10. M. Babar, H.L. Parks, G. Houchins, V. Viswanathan, An accurate machine learning calculator for the lithium-graphite system. *J. Phys. Energy* 3, 014005 (2021). <https://doi.org/10.1088/2515-7655/abc96f>

11. Y. Liao, P. Suo, C. Wang, J. Zhang, Y. Li, Advanced machine learning interatomic potential for accelerated atomistic simulations of lithiation dynamics in large-scale Si@C core-shell anodes. *ACS Appl. Mater. Interfaces* 18, 2996-3006 (2026). <https://doi.org/10.1021/acsami.5c18142>

12. J.D. Morrow, J.L.A. Gardner, V.L. Deringer, How to validate machine-learned interatomic potentials. *J. Chem. Phys.* 158, 121501 (2023). <https://doi.org/10.1063/5.0139611>

13. L. Barroso-Luque, M. Shuaibi, X. Fu, B.M. Wood, M. Dzamba, M. Gao, A. Rizvi, C.L. Zitnick, Z.W. Ulissi, The Open Materials 2024 (OMat24) inorganic materials dataset and models. *Nat. Comput. Sci.* 6, 642-652 (2026). <https://doi.org/10.1038/s43588-026-00996-w>

14. I. Batatia, D.P. Kovács, G.N.C. Simm, C. Ortner, G. Csányi, MACE: Higher order equivariant message passing neural networks for fast and accurate force fields, in *Advances in Neural Information Processing Systems*, vol. 35 (Curran Associates, Red Hook, 2022), pp. 11423-11436

15. S. Plimpton, Fast parallel algorithms for short-range molecular dynamics. *J. Comput. Phys.* 117, 1-19 (1995). <https://doi.org/10.1006/jcph.1995.1039>

16. B. Zhang, T. Ma, N. He, C. Wang, H. Tan, J. Han, Y. Liu, Machine learning driven insights into lithiation mechanisms at the silicon-graphite interface within composite electrode. *Acta Mater.* 292, 121072 (2025). <https://doi.org/10.1016/j.actamat.2025.121072>

17. S.-P. Kim, D. Datta, V.B. Shenoy, Atomistic mechanisms of phase boundary evolution during initial lithiation of crystalline silicon. *J. Phys. Chem. C* 118, 17247-17253 (2014). <https://doi.org/10.1021/jp502523t>

18. S. Kohara, M. Shiga, Y. Onodera, H. Masai, A. Hirata, M. Murakami, T. Morishita, K. Kimura, K. Hayashi, Relationship between diffraction peak, network topology, and

amorphous-forming ability in silicon and silica. *Sci. Rep.* 11, 22180 (2021).
<https://doi.org/10.1038/s41598-021-00965-5>

19. S.C. Jung, Y.-K. Han, Ab initio molecular dynamics simulation of lithiation-induced phase-transition of crystalline silicon. *Electrochim. Acta* 62, 73-76 (2012).
<https://doi.org/10.1016/j.electacta.2011.11.093>

20. S. Chen, A. Du, C. Yan, Molecular dynamic investigation of the structure and stress in crystalline and amorphous silicon during lithiation. *Comput. Mater. Sci.* 183, 109811 (2020). <https://doi.org/10.1016/j.commatsci.2020.109811>

21. J. Griebner, L. Frérot, J.A. Oldenstaedt, M.H. Müser, L. Pastewka, Analytic elastic coefficients in molecular calculations: Finite strain, nonaffine displacements, and many-body interatomic potentials. *Phys. Rev. Mater.* 7, 073603 (2023).
<https://doi.org/10.1103/PhysRevMaterials.7.073603>

22. M.J. Chon, V.A. Sethuraman, A. McCormick, V. Srinivasan, P.R. Guduru, Real-time measurement of stress and damage evolution during initial lithiation of crystalline silicon. *Phys. Rev. Lett.* 107, 045503 (2011).
<https://doi.org/10.1103/PhysRevLett.107.045503>

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397 Conceptualization, Writing-review & editing, Funding acquisition

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400

401 **Ethics Declarations**

402 **Competing Interests**

403 The authors declare that they have no known competing financial interests or personal
404 relationships that could have appeared to influence the work reported in this paper.

405

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