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Article

Modeling Short-Range and Three-Membered Ring Structures in Lithium Borosilicate Glasses Using a Machine-Learning Potential

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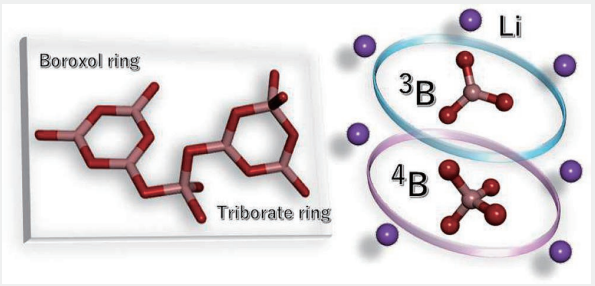
ABSTRACT: Lithium borosilicate (LBS) glass is a prototypical lithium-ion conducting oxide glass available for an all-solid-state buttery. Nevertheless, the atomistic modeling of LBS glass using *ab initio* (AIMD) and classical molecular dynamics (CMD) simulations has critical limitations due to computational cost and inaccuracy in reproducing the glass microstructures, respectively. To overcome these difficulties, a machine-learning potential (MLP) was examined in this work for modeling LBS glasses using DeepMD. The glass structures obtained by this MLP possessed 4-fold coordinated boron (^{4}B) confirmed well with the experimental data and abundance of three-membered rings. The models were energetically more stable compared with those constructed with a functional force field even though both the models included reasonable ^{4}B. The results confirmed MLP to be superior to model the boron-containing glasses and address the inherent shortcomings of the AIMD and CMD. This study also discusses some limitations of MLP for modeling glasses.



1. INTRODUCTION

Boron-containing silicate glasses are widely used for a variety of products such as tableware,[1] optical fibers,[2] display glass,[3] sealing glass,[4] bioactive glass,[5,6] and nuclear waste glass[7] because of their specific features such as high chemical durability, low thermal expansion coefficient, and high thermal shock resistance.[8] Among them, lithium-containing borosilicate glasses are expected to play an important role in all-solid-state battery technologies[9−12] because of their flexibility, which enables a smooth connection between the electrolyte and electrode to suppress the interfacial resistance.[13−15]

An extensive understanding of the atomistic view of the microstructure is indispensable to precisely preserve glass properties such as ion conductivity, mechanical response, and crack resistance. Quantum calculation with density functional theory (DFT) is a promising method to accurately obtain material structures in the nanoscale because the computational cost of DFT calculations is relatively low compared with that of more precise approximations such as the Møller−Plesset perturbation, coupled cluster calculations, and configuration interaction method. Nevertheless, DFT calculation is still an expensive tool for modeling amorphous oxide glasses, mundanely because many atoms should be considered to represent the variations of the amorphous structure while avoiding the artifact of the periodic boundary condition, which is often assumed to represent an infinite material structure in computational modelings. Besides, a significantly long cooling time is usually required to obtain well-equilibrated glass structures. Therefore, even the DFT calculations often only qualitatively reproduce the glass microstructures; i.e., the numbers of tetrahedral BO_4 units and planar three-membered rings, such as a boroxol ring, are underestimated.[16,17]

Although a possible alternative method to obtain adequately large and well-equilibrated glass structures is the classical molecular dynamics (CMD) simulation, CMD simulations often fail to reproduce borosilicate glass structures. Contrary to silicate glasses, borosilicate glasses including modifiers are difficult to be modeled accurately with simple functional force fields because boron atoms flexibly vary the oxygen coordination number, 3-fold (^{3}B) or 4-fold coordinated (^{4}B).[18−20] To accurately model sodium borosilicate glasses using CMD simulations, several force fields were extended by employing composition-dependent parameters.[21−24] Analogously, a composition-dependent force field for lithium borosilicate glass was also proposed by modifying the Buckingham-type force field,[25−27] whose parameters were optimized to reproduce force and energy evaluated by DFT calculations.[28] The empirical force field for LBS glasses is hereafter abbreviated as FMP-LBS.[28] These extended empirical force fields can reproduce ^{3}B/^{4}B ratios in alkaline borosilicate glasses; however, it is known

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