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Understanding ionic conductivity trends in polyborane solid electrolytes from ab initio molecular dynamics[1] JOEL VARLEY, KYOUNG KWEON, LLNL, PRATEEK MEHTA, University of Notre Dame, PATRICK SHEA, TAE WOOK HEO, LLNL, VITALIE STAVILA, Sandia National Laboratories, TERRENCE UDOVIC, National Institute of Standards and Technology, BRANDON WOOD, LLNL — Polyborane salts based on $B_{12}H_{12}^{2-}$, $B_{10}H_{10}^{2-}$, and their carboborane counterparts $CB_{11}H_{12}^{-}$ and $CB_9H_{10}^{-}$ demonstrate extraordinary Li and Na superionic conductivity that make them attractive as electrolytes in all-solid-state batteries. Their rich chemical and structural diversity creates a versatile design space that could be used to optimize materials with even higher conductivity at lower temperatures; however, many mechanistic details remain enigmatic, including reasons why certain modifications lead to improved performance. Here, we use extensive ab initio molecular dynamics simulations to broadly explore the dependence of ionic conductivity on cation/anion pair combinations for Li and Na polyborane salts. Further simulations based on $Li_2B_{12}H_{12}$ as a model system are used to probe the additional influence of local perturbations, including modifications to chemistry, stoichiometry, and composition. Carbon doping, anion alloying, and cation off-stoichiometry are found to be favorable because they introduce intrinsic disorder, which facilitates local deviations from the expected cation population. Anion reorientations are also discovered to be critical for conduction, with benefits associated with lattice expansion traceable to the facilitation of anion rotation at larger volumes.

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