Towards real-time exploration of grain boundary effects on diffusion in solid electrolytes

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Abstract

Major progress in battery technology is a key to the decarbonization of industry and everyday life. In All-Solid State Lithium Ion Batteries (ASSLIBs) the liquid electrolyte of conventional Lithium ion batteries is replaced by a polycrystalline solid-state electrolyte, promising increased capacity, operational safety, and charging performance. Therefore, an in-depth understanding of the multiphysical multiscale effects within the solid-state electrolyte is the subject of research across various disciplines.

The diffusion behavior of Li ions in the solid-state electrolyte is crucial for the battery's overall performance. Studies on the atomistic scale indicate that grain boundary effects and grain size significantly impact overall diffusivity. Recent advances in active learning methods have enabled large-scale, long-time, and high-accuracy simulations of complex atomic structures using ab initio-based machine-learning interatomic potentials. Despite their promising insights, these approaches still go along with massive computational demands, thereby leaving a significant gap between atomistic and application length scales of interest. In simulations on a larger scale, however, the details of the polycrystalline structure are often neglected or only considered under strong assumptions such as isotropy.

Our approach considers the fully resolved crystalline structure with a parametrization that aligns with the atomistic perspective to find a microscopic description that accounts for diffusion along as well as across grain boundaries. This is embedded into a finite element simulation by introducing a novel collapsed interface element. The results are governed by different and potentially anisotropic diffusion coefficients in bulk and grain boundary domains gained from calculations on the atomistic scale. By means of homogenization, an effective model on the mesoscale is derived based on a periodic reference volume element (RVE). Using the hybrid bulk-interface diffusion model, the contribution of the grain boundary to the overall diffusivity can be studied quantitatively.

Furthermore, the affine structure of the problem is exploited to derive a reduced-order model that offers massive performance improvements at little sacrifice in accuracy: The availability of just a few full simulations enables highly accurate real-time estimates for the effective diffusion coefficients across the parameter space. Moreover, the reduced-order model allows for an in-depth analysis of the material and structural features that govern the effective diffusion behavior. For instance, it enables the investigation of preferred diffusion paths. Interactive parameter space exploration supports bridging the gap between the concepts on different scales as well as computational and experimental results. This will facilitate in silico exploration of microstructures and their implications on larger scales, which, at a later stage, will be used to design improved materials and batteries.

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