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First-principles estimates of free energy barriers for Mg desolvation and intercalation at electrolyte/electrode interfaces[1] LIWEN WAN, DAVID PRENDERGAST, The Molecular Foundry, Lawrence Berkeley National Laboratory — There is a growing interest in developing multivalent ion batteries that could, in principle, double or triple the energy density compared to the monovalent Li-ion batteries. However, the strong electrostatic interaction caused by the extra charge also makes it very challenging to find appropriate intercalation compounds that allow for relatively fast and reversible ion transport. An established working multivalent battery is comprised of Mg(AlCl2BuEt)2 salts in THF solution as the electrolyte, and Mg metal and Mo6S8 Chevrel phase as the anode and cathode, respectively. Currently, we lack a clear understanding of the mechanism for Mg desolvation and intercalation at the interface between the electrolyte and Chevrel phase surfaces, which is critical in designing new advanced battery systems with improved ion diffusion rate. Here, we present a theoretical investigation of the dynamics and kinetics of the Mg desolvation/intercalation process. The surface properties of Mo6S8 are studied for the first time using density functional theory (DFT) and its interaction with the electrolyte is simulated via an ab initio molecular dynamics (AIMD) approach. The free energy barrier for Mg diffusing through the interface is then calculated by performing a set of biased AIMD simulations.

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