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Ab initio molecular dynamics simulations of organic electrolytes, electrodes, and lithium ion transport for Li-ion batteries P.R.C. KENT, P. GANESH, Center for Nanophase Materials Sciences, Oak Ridge National Laboratory, Oak Ridge 37831, DE-EN JIANG, Chemical Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831, O. BORODIN, Electrochemical Branch, U.S. Army Research Laboratory, Adelphi MD 20783 — Optimizing the choice of electrolyte in lithium ion batteries and an understanding of the solid-electrolyte interphase (SEI) is required to optimize the balance between high-energy storage, high rate capability, and lifetime. We perform accurate ab initio molecular-dynamics simulations of common cyclic carbonates and LiPF6 to build solvation models which explain available Neutron and NMR spectroscopies. Our results corroborate why ethylene carbonate is a preferred choice for battery applications over propylene carbonate and how mixtures with dimethyl carbonate improve Li-ion diffusion. We study the role of functionalization of graphite-anode edges on the reducibility of the electrolyte and the ease of Li-ion intercalation at the initial stages of SEI formation. We find that oxygen terminated edges readily act as strong reductive sites, while hydrogen terminated edges are less reactive and allow faster Li diffusion. Orientational ordering of the solvent molecules precedes reduction at the interphase. Inorganic reductive components are seen to readily migrate to the anode edges, leading to increased surface passivation of the anode. We are currently quantifying Li-intercalation barriers across realistic SEI models, and progress along these lines will be presented.

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