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Coupled polaronic and ion transport in nanocrystalline metal oxide electrodes

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We report new computational methods and fundamental understanding in the dynamics of coupled charge and ion transport in nanoscale metal oxides. The methods attack the multi-scale problem of simulating the collective diffusivities of ions and charge compensating e-/h+ carriers in single crystal particles, across particle-particle grain boundaries, and through networks of grains for select systems. Methods include embedded quantum mechanical clusters at the DFT and MP2 levels of theory for atomic-scale polaronic and ion transport kinetics, classical DFT-based free energy calculations for grain-scale conductivity in the framework of the Poisson-Nernst-Planck formalism, and phase field simulation of charged particle diffusivity for conductivity at the grain network scale. This combination of approaches is one of a kind in terms of its multi-scale range, scaling, and computational efficiency. We are presently focused on coupled electron and Li+ ion transport in polymorphs of TiO2, and also in mixed valence spinel oxides, for electrode conductivity optimization and improving energy storage materials performance for Li+ batteries.