

Abstract Submitted
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First Principles Modeling of BaCeO₃: Stabilization of O Vacancies JOSEPH BENNETT, ANDREW RAPPE, University of Pennsylvania — We use first-principles density functional theory (DFT) calculations to investigate the ground state structures of both BaCeO₃ (BC) and Pd-doped BC (BCP) perovskites of general formula ABO₃. The relaxed structures match closely with recent experimental scattering studies, and also provide a local picture of how the BC perovskite lattice accommodates Pd. Both stoichiometric and oxygen-deficient materials are considered, and structures with an O vacancy adjacent to each Pd are predicted to be favored. The oxidation state of Pd in each doped structure is investigated through a structural analysis, the results of which are supported by an orbital-resolved projected density of states. The local bonding environments around Pd as well as the electron fillings of atomic orbitals on Pd and its neighbors are examined. A particular focus is the character of the highest occupied molecular orbital (HOMO) and the few lowest unoccupied molecular orbitals (LUMO). These electronic states are compared with expectations based on crystal field splittings in the computed atomic geometries. The vacancy stabilization by Pd in BCP is explained through redox chemistry and lattice strain relief.

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