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An ab-initio approach to modeling high temperature thermodynamics of non stoichiometric ceria CHIRRANJEEVI BALAJI GOPAL, AXEL VAN DE WALLE, None — Ceria is a very promising material for fuel cell electrolytes because of its high oxygen ion diffusivity. Also, its ability to thermodynamically create oxygen vacancies in its structure at high temperatures and get oxidized at low temperatures has found use in thermochemical splitting of water to generate hydrogen. Although the experimental phase diagram for ceria is well established in the composition of interest, there have not been significant attempts at studying the oxygen vacancy thermodynamics at elevated temperatures from first principles. We performed GGA+U calculations ceria to study the electronic structure and ground state energies of various concentrations and configurations of oxygen vacancies in ceria. The energies are then fitted to a Cluster expansion Hamiltonian to efficiently model the interactions between the different species : Ce^{3+}, Ce^{4+} , O^{2-} and oxygen vacancies . Lattice Monte Carlo simulations are then performed to obtain the free energy as a function of temperature and oxygen chemical potential through thermodynamic integration. We are also investigating the effect of lattice vibrational contribution to the phase diagram by including a temperature dependent cluster expansion.

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