Plane wave Density Functional Theory Investigation of Oxygen Migration Energetics and Vacancy Structure Combined With Spectroscopic Study of Barium/Strontium Ferrate/Cobaltate (BSCF) as a Promising Material For Solid Oxide Fuel Cell (SOFC)

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ABSTRACT

Mixed oxides of the general formula ABO_3 which crystallize in perovskite structures often have large mobility of the oxygen vacancies and exhibit strong ionic conductivity and used for several practical applications, including Solid Oxide Fuel Cells (SOFC). Barium/Strontium Ferrate/Cobaltate (BSCF) was recently identified as a promising candidate for cathode material in intermediate temperature SOFCs. We apply multiscale technique to determine its vacancy diffusion coefficient. At the small (atomic) scale Density Functional theory (DFT) is used to calculate activation energy barriers for oxygen migration in different local cation distribution. Activation barriers are used in Arrhenius equation to predict the rates for elementary steps in diffusion processes. These rates are then input into Kinetic Monte Carlo (KMC) at large (meso) scale simulations to obtain long time oxygen diffusivities and apparent activation energies. Since KMC method does not need energy evaluations, it is computationally inexpensive and allows treating millions of atoms explicitly. KMC approach can readily describe the macroscopic properties as a function of material morphology. In this contribution we report atomic scale study of BSCF electronic structure using plane wave pseudopotential DFT implemented in Quantum-ESPRESSO. We report cations are completely disordered, while oxygen vacancies exhibit a strong trend to from L-shaped trimers and tetramers, Löwdin population analysis of the spin density indicates that the cobalt cations shows Jahn-Teller distortion of octahedral coordination around Cobalt cations, both theoretically and experimentally and confirmed its intermediate spin state. We compute oxygen vacancy migration activation energy and find it to be in good agreement with experimental data.

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