Constructing an Improved Ce Pseudopotential in OPIUM Code

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We construct a Ce pseudopotential in OPIUM that demonstrates excellent agreement with both experimentally and computationally determined physical properties of bulk ceria (Ce_2O_3 and CeO_2) in density functional theory calculations. Our pseudopotential contains a mixture of semicore and partial core corrections, both of which are necessary to predict the correct energy level of *4f* states. An affectedly low energy for *4f* results in unphysical interactions between Ce-*4f* and O-*2p* states in bulk Ce oxides. Our Ce pseudopotential reproduces the reaction enthalpy for the reduction of CeO_2 to Ce_2O_3 with good agreement and similarly reproduces the activation barrier for bulk oxygen transport in pure CeO_2. With the new pseudopotential, we investigate the energetics and kinetics of Pd-doped $BaCeO_3$, a potential candidate of self-regenerative catalysts.

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