Understanding and tailoring spinel and perovskite surfaces for energy conversion applications

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Transition metal oxides play an important role as anode materials in the (photo-) electrocatalytic water splitting. Based on density functional theory (DFT) calculations with a Hubbard *U* term, we focus on the oxygen evolution reaction (OER) activity of iron, cobalt and nickel containing spinels [1-3] vs. perovskites [4-5]. This allows us to disentangle the role of structural motifs, crystallographic orientation and termination. Analysis of the underlying electronic and magnetic properties reveals dynamic variation of oxidation state during OER with marked differences in the degree of localization spinels and perovskites. Octahedral Co at the (001) surface emerges as a potential active site with the lowest overpotentials. Last but not least, we elucidate the effect of a surface transformation into an oxyhydroxide layer at the perovskite surface and identify important distinctions for different surface facets [5].

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