

Exploring Surface Magnetism in Transition Metal Oxides for Oxygen Electrolysis

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Transition metal oxides (TMOs) are abundant and cost-effective materials with significant potential for oxygen electrocatalysis. However, the influence of their intrinsic magnetic properties on electrocatalytic activity remains unclear. This study investigates the correlation between magnetic ordering and catalytic performance on TMO surfaces, specifically Mn, Fe, Cr, and Co, by analyzing the Gibbs free energies of oxygen reduction reaction (ORR) intermediates (OO*, O*, HOO*, HO*).

Our findings reveal that magnetic order strongly influences ORR intermediate adsorption, with antiferromagnetic (AFM) surfaces exhibiting weaker binding energies, making them more inert compared to ferromagnetic (FM) and ferrimagnetic (FERM) surfaces. The higher reactivity of FM and FERM configurations is attributed to a broader d-band, charge delocalization, and enhanced surface magnetization relative to AFM configurations. Additionally, FM surfaces facilitate greater charge transfer to ORR intermediates, further enhancing their catalytic activity.

We also examine the impact of biaxial strain on adsorption energies, showing that compressive strain strengthens adsorption across all magnetic surfaces. Notably, FM surfaces gain additional stability under compressive strain compared to AFM surfaces. These results underscore the crucial role of magnetic order in governing the adsorption and catalytic behavior of TMO surfaces, offering valuable insights for the design of more efficient electrocatalysts.