Investigating How Oxidation Impacts Grain Boundary Structure, Energetics, and Activity in Electrocatalysis via Simulation

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Electrocatalyst design seeks to minimize key bond-breaking event reaction energies, such as O-O dissociation in the oxygen reduction reaction. Grain boundaries (GBs) introduce structural disorder to minimize such energies, improving reaction activity, selectivity, and yields. Such catalytic reactions occur over sequential mechanisms, which are complicated by variable reactant stabilities within heterogeneous interfacial environments. Changes in favorable mechanisms render whole reaction process characterization difficult, as seen in atypical charge vs. potential trends and anti-thermal kinetics over varied reaction conditions. Previous work has investigated the link between elastic energetics and metal GB dislocations, and how catalytic activity is improved by deriving Cu GBs from oxides. Current research seeks to reconcile Cu GB oxidation energetics, structure, and catalytic activity, determining the atomistic origins of discontinuities in electrocatalytic reaction rates over multi-step mechanistic processes. This is accomplished via simulation methods including density functional theory, deep neural network force field development, and both molecular mechanics and dynamics.