Thermodynamic Margin in Carbon Network Modulated Activity Control of Oxygen Reduction Reaction Iron Catalyst

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We investigate how structural changes in the carbon network around the active site are correlated with the oxygen reduction reaction activity of the FeNC catalyst by performing a series of first-principles simulations. We find that the defective carbon networks tend to negatively affect the activity, while the FeNC catalyst with a perfect graphene network yields a near-peak activity. Descriptor analysis demonstrates that there is a limited thermodynamic margin in improving the activity by changing the carbon network structures. Such a limited thermodynamic margin proves to be associated with the tight linear scaling relationships between certain reaction intermediates. Subsequent activity prediction based on the oxygen reduction reaction potential landscape suggests the synthesis of a novel motif capable of exclusively stabilizing the hydroperoxyl radical binding as a practical route to the development of highly active FeNC catalysts.