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Revisiting Demetallation of Single Atom Iron in Nitrogen-doped Carbon Electrocatalysts for Proton Exchange Membrane Fuel Cells

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Low temperature proton exchange membrane fuel cells (PEMFCs) powered by green hydrogen provide a means to sustainable energy production for stationary and transport applications. Their widespread commercialization is limited by the cost of the platinum catalyst at the cathode, where oxygen reduction occurs. Atomic Fe sites within nitrogen-doped carbon (Fe-N-C) offer a cheap and sustainable alternative, exhibiting the most promising non-precious metal activity for oxygen reduction. However, their stability is still below commercial realization owing to several degradation routes (Figure 1). Steps can be taken to minimize these pathways,^{1,2} however demetallation of FeN_x active sites is still suggested as a primary performance degradation mechanism in PEMFCs.³

To shed new light into Fe demetallation, in this work we focus on monitoring Fe dissolution of a highly porous and >50% FeN_x utilization Fe-N-C catalyst.⁴ We compare *operando* Fe dissolution under O₂ (active) and Ar (inert) conditions in both flow cell and gas diffusion electrode (GDE) setups coupled with inductively-coupled plasma mass spectrometry. During GDE measurement we observed very little Fe dissolution under O₂, but extensive detection under Ar. Post-mortem transmission electron microscopy and high angle annular dark field scanning transmission electron microscopy shows formation of Fe_xO_y nanoparticles under O₂, but not under Ar, in GDE setups. Meanwhile Fe was detected under both atmospheres in flow cell configuration, without Fe_xO_y formation in either atmosphere. We propose under device relevant O₂ reduction GDE conditions a degradation mechanism of Fe dissolution from FeN_x sites and subsequent reprecipitation into Fe_xO_y due to a proposed local pH increase. We also report the fate of Fe being dependent on temperature and material properties.

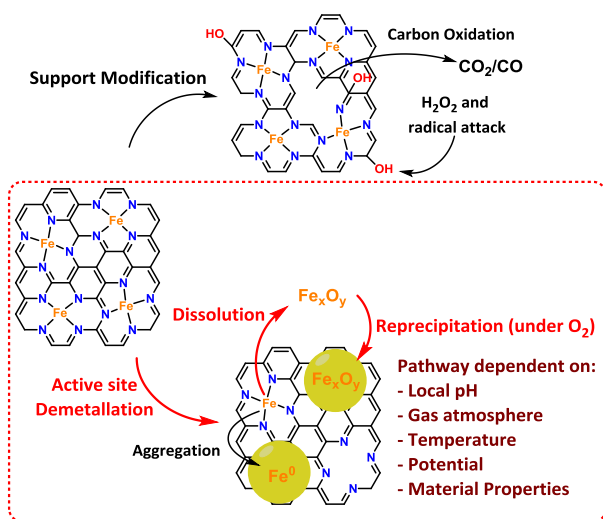


Figure 1. Fe-N-C degradation pathways.

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