

Atomic Fe in N-doped Carbon for O₂ reduction: How to Achieve High Fe Loading?

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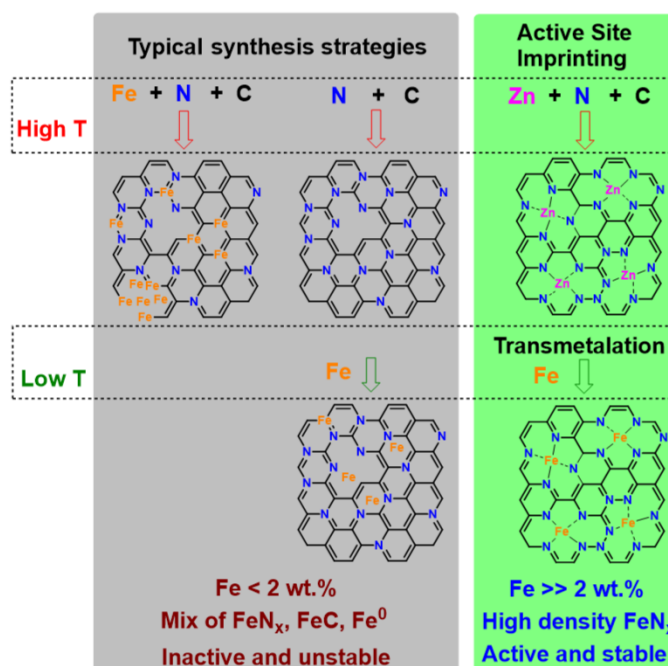
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Low temperature proton exchange membrane fuel cells powered by green hydrogen provide a means to sustainable energy production for stationary and transport applications, such as back-up power and fuel cell vehicles, respectively. Their widespread commercialisation is limited by the cost of the platinum catalyst at the cathode, where oxygen reduction occurs. Atomic FeN_x sites within carbon offer a cheap and sustainable alternative, exhibiting the most promising non-precious metal activity for oxygen reduction. However, atomic Fe loading typically cannot exceed >2 wt.% without unstable and inactive FeC and Fe⁰ formation due to high pyrolysis temperatures (700-1000°C) required during synthesis of the conductive catalyst support. Recent progress has identified the successful use of a decoupled two-step procedure whereby Fe is incorporated at low temperature, following the high temperature pyrolysis, which has enabled >2 wt.% Fe.^[1-3] In our work, we adapt a Zn active site imprinting and subsequent low temperature

(170°C) Fe (trans-)metalation process,^[1] instead using a zeolitic imidazolate framework-8 precursor to yield Fe >5 wt.% (ICP-MS) with oxygen reduction active FeN_x sites. The ex-situ atomic nature of the FeN_x active site is elucidated by aberration corrected high-angle annular dark field scanning transmission electron microscopy, x-ray absorption spectroscopy, and electron paramagnetic resonance. The high Fe loading also enables novel characterisation by time-of-flight secondary ion mass spectrometry.

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