Single Atom Fe-N-C Electrocatalysts for Oxygen Reduction Reaction

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Electrocatalytic oxygen reduction reaction (ORR) is the vital process in the cathode of next-generation electrochemical storage and conversion technologies, such as, metal-air batteries and fuel cells. Single-atom Fe-N-C carbonous electrocatalysts have emerged as attractive alternatives to noble-metal platinum to catalyze the kinetically sluggish ORR due to the high electrical conductivity, large surface area, structural tunability at the atomic/morphological levels. However, the ORR activity of current Fe-N-C is seriously limited by the low density, inferior exposure of active Fe-N_x species and low intrinsic activity of the Fe-N_x sites.[1] Here, a novel zinc-mediated template synthesis strategy is demonstrated for constructing densely exposed Fe-N_x moieties on hierarchically porous carbon (SA-Fe-NHPC). As a result, the SA-Fe-NHPC electrocatalysts exhibit an unprecedentedly high ORR activity in a 0.1 M KOH aqueous solution, which outperforms those for Pt/C catalyst and state-of-the-art noble metal-free electrocatalysts.[2] Furthermore, we discovered an edge tensile strain effect strategy to efficiently weaken the interaction between the single Fe site and the O* intermediate through atomic engineering of FeN_4 active sites at the armchair-edge of nitrogen-doped hierarchically porous carbon (e-FeN_4(A)-NHPC), thereby boosting its intrinsic ORR activity in acidic solution.[3] Increasing the site density and enhancing the intrinsic activity of the Fe-N_x site pave a new avenue toward high-performance ORR electrocatalysts.

References

[1] G. Chen, H. Zhong, X. Feng. Chem. Sci., 12 (2021) 15802–15820.
[2] G. Chen, X. Feng et al, Adv. Mater., 32 (2020) 1907399.
[3] G. Chen, X. Feng et al, Under Revision in Energy & Environ. Sci.

Figures

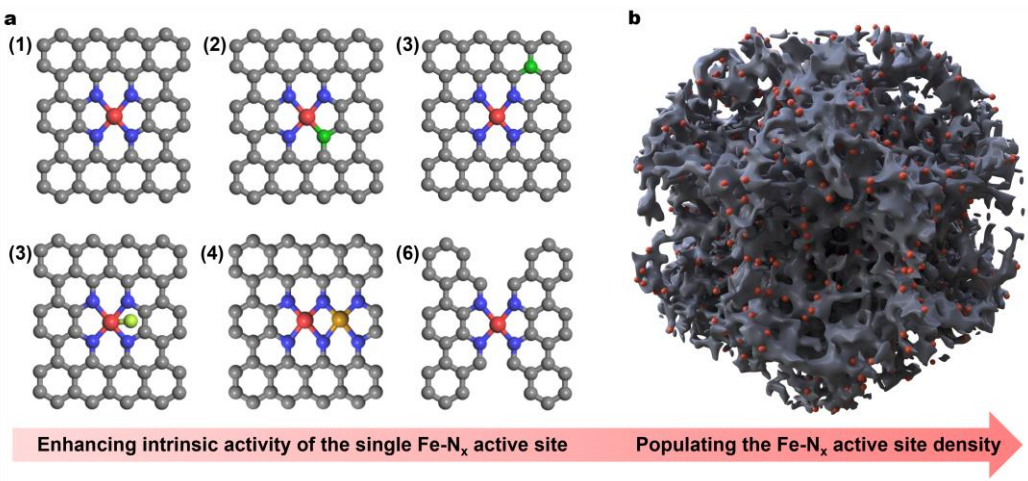

Figure 1: Promoting the ORR activity of Fe-N-C materials via (a) enhancing the intrinsic activity of single Fe-N_x site and (b) populating the Fe-N_x site density.