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Pyrrolic N wrapping strategy to maximize the number of single-atomic $Fe-N_x$ sites for oxygen reduction reaction

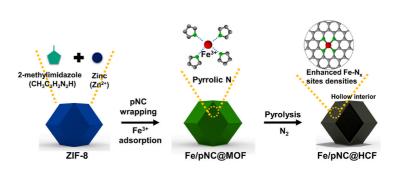
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HIGHLIGHTS

- Pyrrolic N wrapping strategy is devised to maximize the number of Fe-N_x sites.
- Pyrrolic N-rich carbon (pNC) are uniformly coated onto ZIF-8.
- pNC plays two critical roles: providing pinning sites and maintaining ZIF-8 structure.
- The increase of active sites significantly affects single-cell performance of AEMFC.

G R A P H I C A L A B S T R A C T



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ABSTRACT

Iron-nitrogen-carbon (Fe–N–C) catalysts with a representative single-atomic structure are promising platinum group metal-free catalysts for the oxygen reduction reaction (ORR) as they exhibit comparable activity to commercial catalysts. To enhance the ORR activity of Fe–N–C catalysts, the number of single Fe atoms coordinated N (Fe-N $_{\rm x}$) should be maximized. In this study, a strategy is devised to increase the number of Fe-N $_{\rm x}$ sites using electrostatic interactions between electronegative pyrrolic-N and electropositive Fe ions. Pyrrolic N-rich carbon (pNC) is dispersed on the surface of the metal-organic framework (MOF) to form composite supports (pNC@MOF). Owing to the well-dispersed pNC and electrostatic interactions, the number of Fe-N $_{\rm x}$ sites on the pNC@MOF-derived hollow carbon framework (Fe/pNC@HCF) increases dramatically compared to that on the pristine MOF (Fe/HCF). The original shape of the Fe-absorbed MOF is maintained by the conversion of pNC into carbon layer within the framework by pyrolysis at 1000 °C even though pure Fe-absorbed MOF collapses. An

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