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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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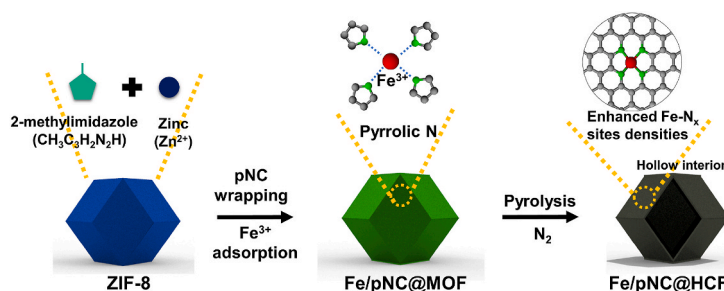
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HIGHLIGHTS

- 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.

GRAPHICAL ABSTRACT



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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_x) should be maximized. In this study, a strategy is devised to increase the number of Fe-N_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_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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