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

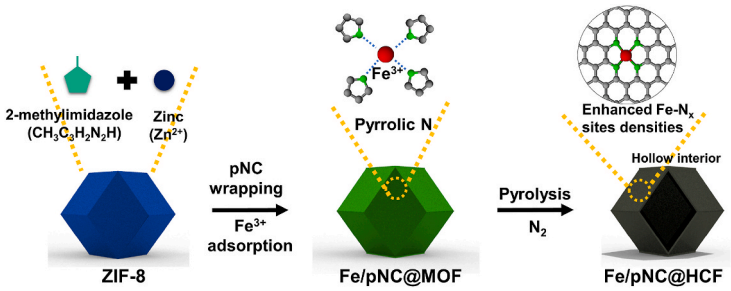
Gil-Seong Kang [a,b], Jue-Hyuk Jang [c], Su-Young Son [d], Youn-Ki Lee [a], Doh C. Lee [b], Sung Jong Yoo [c,***], Sungho Lee [a,e,**], Han-Ik Joh [d,*]

[a] Carbon Composite Materials Research Center, Korea Institute of Science and Technology (KIST), 92 Chudong-ro, Bongdong-eup, Wanju-gun, Jeollabuk-do, 55324, South Korea
[b] Department of Chemical and Biomolecular Engineering, KAIST Institute for the Nanocentury, Korea Advanced Institute of Science and Technology (KAIST), 291 Daehak-ro, Yuseong-gu, Daejeon, 34141, South Korea
[c] Center for Hydrogen Fuel Cell Research, Korea Institute of Science and Technology (KIST), Seoul, 02792, South Korea
[d] Department of Energy Engineering, Konkuk University, 120 Neungdong-ro, Gwangjin-gu, Seoul, 05029, South Korea
[e] Department of Quantum System Engineering, Jeonbuk National University, 567 Baekje-daero, Deokjin-gu, Jeonju, Jeollabuk-do, South Korea

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

* Corresponding author.
** Corresponding author. Carbon Composite Materials Research Center, Korea Institute of Science and Technology (KIST), 92 Chudong-ro, Bongdong-eup, Wanju-gun, Jeollabuk-do 55324, South Korea.
*** Corresponding author.
E-mail addresses: ysj@kist.re.kr (S.J. Yoo), sunghol@kist.re.kr (S. Lee), hijoh@konkuk.ac.kr (H.-I. Joh).

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