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Rational Design of Hollow N/Co-Doped Carbon Spheres from Bimetal-ZIFs for High-Efficiency Electrocatalysis

Xiaodong Chen, Kui Shen,* Junying Chen, Binbin Huang, Danni Ding, Lei Zhang and Yingwei Li*

State Key Laboratory of Pulp and Paper Engineering, School of Chemistry and Chemical Engineering, South China University of Technology, Guangzhou 510640, People's Republic of China.

* Corresponding author. Email: cekshen@scut.edu.cn (K. S.), liyw@scut.edu.cn (Y. L.)

Abstract

To explore efficient non-noble metal-based electrocatalysts for oxygen reduction reaction (ORR), herein we developed a facile bottom-up approach for the fabrication of a hollow porous carbon sphere codoped with ultra-small Co nanoparticles and uniform nitrogen distribution (Co-HNCS) *via* one-step pyrolysis of a core-shell type precursor composing of polystyrene (PS) core and bimetallic ZIF (zeolite imidazolate framework) shell. The bimetallic Co-Zn-ZIFs (BMZIFs) was selected as the sacrifice template due to not only its high nitrogen content and regular porosity but also the superiority that Zn species in BMZIFs can both spatially separate Co species to suppress the aggregation of ultra-small Co NPs and be evaporated to afford extra pores during high-temperature pyrolysis. As expected, by adjusting the starting molar ratio of Zn to Co, we were able to prepare Co-HNCS-*x* (*x* represent the molar ratio of Co to total starting metal feeding) that exhibited unique hollow structure with large surface areas, enhanced mass transport, high porosities, tunable particle sizes and graphitization degrees, abundant highly active CoN_x sites, and thus significantly improved ORR performance. Particularly, the optimal Co-HNCS-0.2 exhibited the remarkable ORR activity (the onset and half-wave potentials were 0.94 and 0.82 V vs. RHE, respectively) *via* an efficient four-electron-