21st International Conference on

Advanced Energy Materials and Research

July 11-12, 2019 | Zurich, Switzerland

Discrete Zn, Co bimetallic sites supported on N doped carbon for high performance oxygen reduction reaction catalysis

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A new design of discrete Zn, Co bimetallic sites supported on N-doped carbon was fabricated through a competitive complexation strategy. Aberration corrected atomic resolution high angle annular dark field scanning transmission electron microscopy (HAADF-STEM) measurements combined with X-ray absorption fine structure (XAFS) reveals the existence and the structure of the Zn-Co bimetallic sites. This Zn-Co dual atom catalysts exhibit significantly improved oxygen reduction catalytic activity compared to single atom catalysts in both acid and alkaline conditions. Density functional theory (DFT) calculations reveal that the enhanced catalytic activity can significantly be attributed to the elongated O-O bond length (from 1.23 Å to 1.42 Å), and thus facilitates the cleavage of O-O bond at the $ZnCoN_6(OH)$ sites, showing a theoretical over potential of 0.335 V during ORR process. In-situ XAS study demonstrates that Co serves as the active center during the catalysis. Furthermore, a highly active sulfur (S)-modified Zn, Co-Nx-C-Sy ORR catalyst is also developed. Besides the elongated O-O band length, the S doping can further modify the charges around Zn, Co active center and strengthen the interaction with oxygenated species by decreasing the free energy changes of $*O_2 + e^- + H_2O \rightarrow *OOH + OH^-$ step. The prepared catalysts show promising potential in practical applications in both fuel cell and Zn-air batteries. Particularly, the H_2/O_2 fuel cell tests based on the Zn-Co atomic pair presents a peak power density of 705 mW cm^{-2} along with excellent stability.

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