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Asymmetric 3d Electronic Structure for Enhanced Oxygen Evolution Catalysis

Abstract
With the deterioration of the environment, human concern with clean energy has increased considerably. Hydrogen is a relatively cleaner energy, and hence has a wide range of applications in various fields. Hydrogen can be obtained through many ways, such as electrolysis of water, photo-catalytic water splitting, etc. The process of water splitting involves the production of hydrogen and oxygen, where the evolution of these gases is interactional.[1] The theoretical voltage of hydrogen and oxygen evolution from water splitting are 0 and 1.23 V, respectively.[2] The Pt/C and IrO_2 catalysts have been reported as the best hydrogen and oxygen evolution catalysts with overpotentials of 0 and 270 mV, respectively.[3,4] Obviously, the oxygen evolution reaction (OER) is the biggest obstacle in water splitting. The oxygen evolution reaction (OER) is a four electron transfer process in acid or alkaline medium, where OH^- loses electrons to become O_2. Owing to the fact that the transition metals (such as Fe, Co, Ni, etc.) possess unpaired d orbital electrons, they can be used to open the O−H bond. But due to high degree of localization and overspreading of the d orbital, intermediate products (such as Oad, OOH_{ad}) and OHad, OOH_{ad} could be resulted, which would be difficult to be desorbed and dissociated, respectively. In this work, FeS, Ni_3S_2, $Fe_5Ni_4S_8$, and N, O, S-doped meshy carbon base were successfully synthesized. The sample containing $Fe_5Ni_4S_8$ exhibited excellent OER performance. The density functional theory calculations indicate that the partial density of states for 3d electrons (3d-PDOS) of Fe and Ni atoms are changed from monometallic sulfide to bimetallic sulfide at the sulfur vacancy. The asymmetric 3d electronic structure optimizes the 3d-PDOS of Fe and Ni atoms, and leads to an enhanced OER activity. This work provides a new strategy to prepare a low-cost electrocatalyst for oxygen evolution with high-efficiency.

References

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[4] Yu, X.; Zhang, S.; Li, C.; Zhu, C.; Chen, Y.; Gao, P.; Qi, L.; Zhang, X., Nanoscale, , 8 (2016) 10902−10907.

Acknowledgment

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Figures

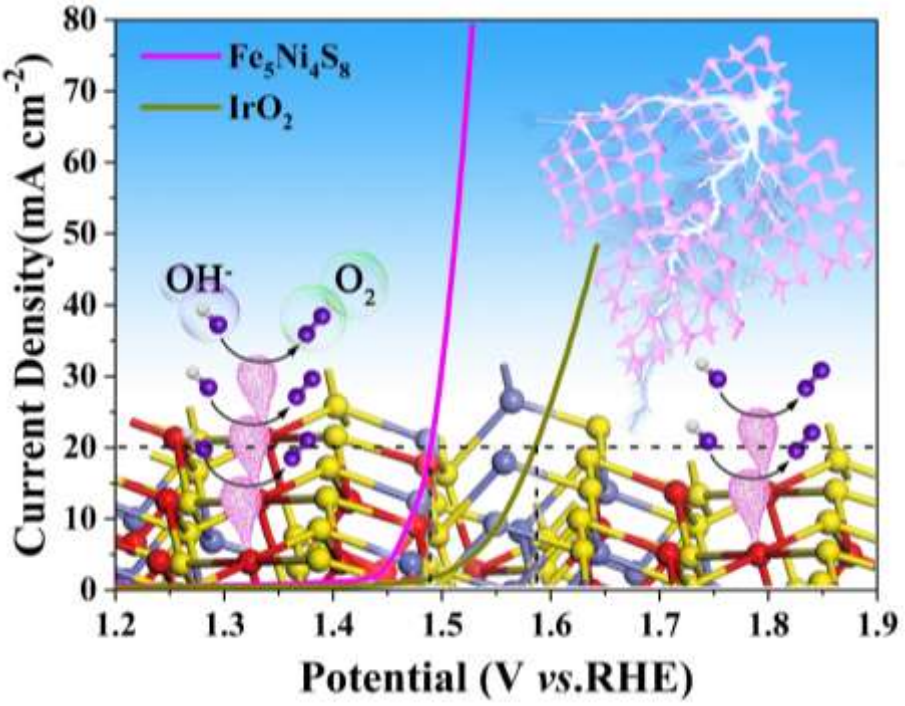

Figure 1: The cover of this paper.

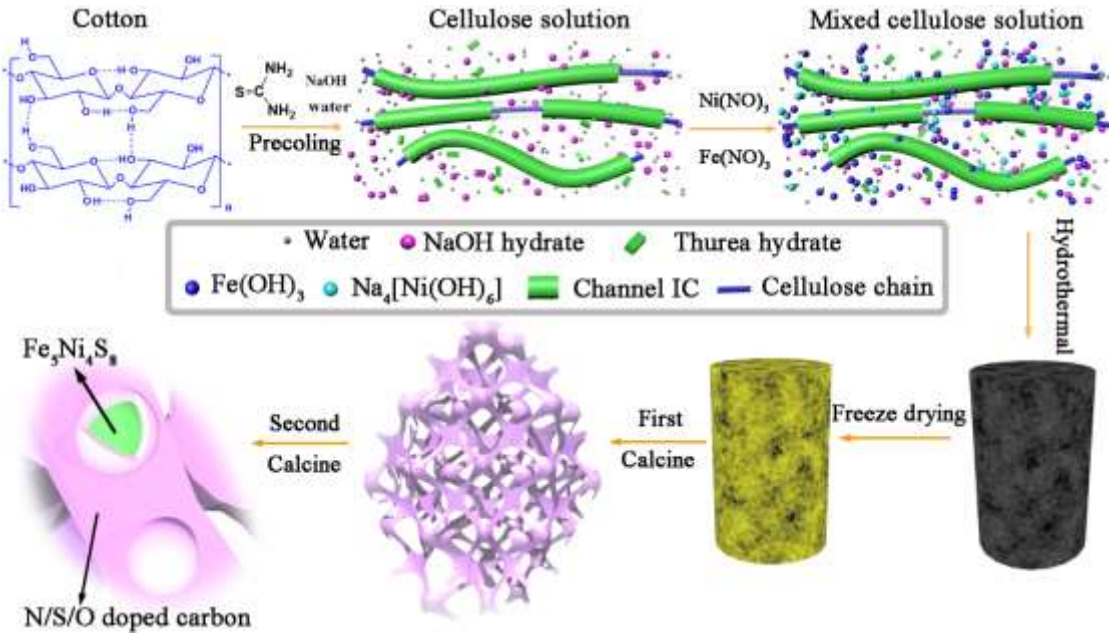

Figure 2. Synthesis procedures for meshy carbon materials loaded by carbon-encapsulated Fe5Ni4S8 nanoparticle.