UDC 66

PREPARATION AND PROPERTIES OF POROUS DOPED CARBON BASED CATALYSTS WITH BIMETALLIC ACTIVE SITES

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As an inevitable path to achieving carbon peak and carbon neutrality, new and renewable energy is bound to accelerate its entry into the mainstream of the energy system, with fuel cells as the key core technology of the new energy industry. However, the development of electrocatalysts for oxygen reduction reactions (ORR) on air electrodes determines the energy conversion efficiency, application cost, durability, and lifetime of the battery in fuel cells[1]. Due to the high price and resource scarcity of platinum catalysts, their large-scale production and commercial application are limited[2-]34]] . Therefore, the development of oxygen reduction catalysts with low cost, good catalytic activity, and high stability has become the focus of current research in this field.

Monometallic atomic catalysts have attracted increasing attention due to their excellent catalytic activity and high atomic utilization. However, SACs have two inherent defects: a single catalytic structure and a lack of coordination of multiple active sites. Therefore, its application in multi molecular catalytic reactions will be greatly limited. Introducing additional metal atoms (i.e., dual metal site catalysts, DMSCs) near the active site of a single metal atom can effectively solve this problem. DMSCs not only maintain a high atomic utilization rate similar to that of single metals, but also have strong synergistic effects between adjacent active sites [5-]6]]]]]. The synergistic effects between bimetallic components can be divided into three types.[7-8] Geometric (or strain) effect: The addition of a second metal causes lattice constant mismatch (lattice distortion), resulting in changes in atomic spacing or average metal metal bond length, changing the geometric configuration of the catalyst, and improving catalytic activity. Electronic (or ligand) effect: The addition of a second metal changes the electronic configuration of active metal sites by changing the electronic environment on the metal surface or promoting electron transfer between metals. Stabilization effect: The addition of a second metal improves the stability of the catalytically active metal. For example, stability is maintained by adding a second metal element that is not toxic. The geometric effects of bimetallic catalysts are usually accompanied by electronic effects. In situ X-ray absorption spectroscopy (XAS), including X-ray absorption near edge structures (XANES) and extended X-ray absorption fine structures (EXAFS), has a strong correlation with DFT and can detect the electronic and geometric states of atoms in materials during catalytic reactions[6]. At present, the commonly used methods to synthesize bimetallic catalysts are: co reduction method, continuous

reduction method, microemulsion method, electrodeposition method, microwave heating method, etc., but these methods are difficult to accurately control the nano array structure of bimetallic components, so it is difficult to effectively control and adjust the space spacing of different metal components, making the research on the relationship between microstructure and catalytic performance a difficult problem. Therefore, it is urgent to develop a simple and efficient method to prepare porous catalysts with porous, high-density active sites, and atomic doping efficiency.

In this study, we propose to modify and doped bimetallic M (M=Ag, Co, Mn, Fe, etc.) by in-situ synthesis of MOFs (MET-6, ZIF-8, etc.) materials in order to address the shortcomings of catalysts in the electrocatalytic ORR process, such as low exposure of active sites, low metal atom utilization, and slow mass transfer rate. It is proposed to achieve porous exposure to more active sites through high-temperature pyrolysis or pore channel modifiers, and directionally construct rich bimetallic active site-porous structure coupling sites, ultimately achieving in situ preparation and optimization of high-performance porous ORR catalysts doped with bimetallic monatomic active sites. Through advanced structural characterization and electrochemical testing techniques, the intrinsic relationship between the active site structure, porous structure, and ORR catalytic properties of different carbon catalysts was studied. By using theoretical calculations and modern in situ characterization techniques, the mechanism of factors such as the single atom and its coordination environment in the catalyst, the doping of bimetallic single atom M-Nx sites, and the coupling site of porous structure on the ORR catalytic behavior and process was revealed. The implementation of this project is expected to obtain highly efficient and inexpensive porous bimetallic monoatomic ORR catalysts, which will help deepen the understanding of the active site structure and catalytic laws of the catalysts, and provide a theoretical and practical basis for the development of new high-performance catalysts and the commercial application of green environmental protection energy systems such as fuel cells and metal air batteries.

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