UDC 622

CONTROL AND MECHANISM STUDY OF TWO DIMENSIONAL SUBNANO LAYERED SPACE OF NON PT CARBON BASED OXYGEN REDUCTION CATALYSTS

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Carbon materials have advantages such as rich pore structure, large specific surface area, easy control of surface chemical properties, favorable reduction of metal phases, and low cost.[1, 2].Carbon based carriers not only introduce heteroatom anchoring points to fix metal single atoms, but also have a positive effect on the electrocatalytic process of ORR by doping heteroatoms with carbon[3,4]. The "spatial confinement" strategy is an effective way to solve the problem of low metal loading and aggregation of metal particles or clusters. On the one side, molecular pores in materials such as molecular sieves[2], metal organic framework materials(MOFs)[5,6,7], covalent organic framework materials (COFs)[8] serve as "confinement cages" to restrict the migration of metal atoms. On the other side, by using two-dimensional layered materials (such as layered compounds) with interlayer spatial confinement, the interactions between molecules within the layers and between molecules and laminates can be effectively regulated[9], enhancing the coordination between metals and nitrogen, and thereby achieving molecular configuration and coordination control under two-dimensional confinement[10]. The multi-level pore structure can maximize the utilization of active sites, enhance material mass transfer ability, and enhance ORR catalytic activity. Porous carbon materials with different morphologies and pore sizes were prepared using methods such as template method[11,12], metal organic framework materials[5, 6] and so on. Among them, template method is particularly convenient and practical for precisely customizing the pore structure in carbon materials[13,14,15]. It can construct defective porous microstructures in target products, including N-doped carbon materials, which is conducive to improving the density of active sites in M-Nx and other C-N configurations[16,17,18,19].

In response to the shortcomings of low exposure of active sites, low utilization of metal atoms, and slow mass transfer rate in electrocatalytic ORR process, starting from the construction and optimization of nanospace confined structures and molecular assembly processes, the guest (short chain quaternary ammonium salts, etc.) and the host (two-dimensional layered materials, etc.) are first subjected to ion exchange, and the interlayer spacing of the two-dimensional confined space is modified and optimized, Forming a two-dimensional layered bulk structure with intercalation reaction and complexation ability, and then synchronously introducing small molecule amine compounds to achieve lateral ordered self-assembly, controllable formation of stable layered metal based complexes, and based on block polymer molecules as self sacrificial and easily thermally removed "pore regulators" to achieve ordered regulation and control of pores under high-temperature calcination conditions, thereby achieving localized nitrogen doping and bimetallic atom loading, Targeted construction of rich bimetallic active sites porous structure coupling sites, ultimately achieving in-situ preparation and optimization of high-performance porous ORR catalysts doped with bimetallic single atom active sites. Further coupling theoretical calculations, physical and chemical characterization, in situ experiments, and battery testing analysis, to construct the interrelationships between the surface interface structure, components, electronic distribution state of the catalyst and the ORR catalytic activity and stability, to identify the actual

role of metal atoms in nitrogen-containing active sites, and to reveal the basic laws and dynamic mechanisms of the catalyst's active sites and electrochemical catalytic ORR, Explore the degradation process and mechanism of active catalytic sites during actual stability testing. The development of this study is expected to obtain efficient and inexpensive porous bimetallic single atom ORR catalysts, which will help deepen the understanding of the active site structure and catalytic laws of catalysts, and provide 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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