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Recent advances in surface and interface engineering for electrocatalysis

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ABSTRACT

Electrocatalysis has attracted extensive attention for its promise in converting chemical energy of fuels and oxidants into electrical energy. In this review, we use our recent progress in electrocatalysis as examples to demonstrate how to rationally design and fabricate noble metal-based nanostructures. This information will enable the optimization of nanocatalysts, in terms of both cost and performance, from the viewpoint of surface and interface engineering. We first outline the key features related to surface and interface that may significantly impact on electrocatalytic performance. We then summarize various approaches to surface and interface modulations by highlighting materials synthesis, design and electrocatalytic performance for specific cases. Finally, we propose the challenges and opportunities to perform materials design for electrocatalysis from the aspect of surface and interface engineering.

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1. Introduction

Grubb [1] first put forward the concept of electrocatalysis in 1963, which he defined as the enhancement of the rate of an electrochemical reaction by a substance. Electrocatalysis has experienced great progress and gradually developed into an interdisciplinary science and comprehensive technology that involves electrochemistry, materials science, surface science, catalytic science and many other branches of science. Electrocatalysis plays an indispensable role in industrial and agricultural production, national defense, economic construction, energy development and environmental protection [2–10]. At present, electrocatalysis, as an essential and effective means for an electrochemical and catalytic united-technology, is widespread, and has been applied to the controllable synthesis and functional assembly of nanomaterials [11–14], micro–nano fabrications and electronic devices [15–20], metal processing, forming, plating and finishing [21–25], organic chemical reactions [26–28], nitrogen cycle and chlor-alkali industry [29–31], electrochemical luminescence and photosensitive measurement [32–34], electrochemical sensors and biosensors [35–38], electrochemical engineering for cell biology and medical therapy [39–43], electrochemical process of detection and analysis [44–47], and organic waste degradation and wastewater treatment [48–51]. In particular, to alleviate and address the increasingly severe energy and environmental crisis, efforts have been intensively focused on electrocatalysis for energy development, conversion and storage, such as chemical batter-

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ies or metal-air batteries [52–55], super-capacitors [56–59], solar water splitting cells [60–63], and low-temperature fuel cells [64–69].

The great advances and broad applications of electrocatalysis are greatly facilitated by the design and controlled synthesis of metal nanocatalysts [52–69]. The metal nanocatalysts (especially transition metal and noble metal nanocatalysts) have been widely used as electrocatalysts, mainly because the transition metals and noble metals may provide *d*-orbital lone-pair electrons as nucleophilic reagents or provide empty *d*-orbitals as electrophilic reagents in chemical reactions. The metals may coordinate with alien species to form intermediates to reduce the reaction activation energy, eventually promoting the catalytic reaction. Meanwhile, they may also provide adjustable *d*-band locations and electronic structures by tuning their composition and structural features, which alters the adsorption and stripping abilities of reactants/products on their surface/interface and further improves the activity, selectivity and stability of nanocatalysts [70–78]. Thus far, various unique transition/noble metal nanocrystals and their hybrid nanostructures have been synthesized through a variety of gas-phase, solid-phase and solution-phase synthetic methods as well as loading and assembly techniques. In particular, the solution-phase synthesis provides a versatile platform for controlled synthesis by manipulating the nucleation and growth process that involves atom generation, nucleation, seed formation, seeding growth and/or epitaxial growth. The experimental parameters that can be tuned in a reaction system include precursors, reducing agents, capping agents, etching agents, solvents, temperature and many others. Furthermore, combined with post-synthesis techniques, such as etching, loading and assembly, the synthesis can yield metal nanocrystals and their hybrid nanostructures with well-defined sizes, shapes and structures [72–78].

Noble metal catalysts (mainly Pt, Pd, Ru and Rh) with high activity, selectivity and stability are prevalent in catalysis and still cannot be completely replaced by other materials [79–82]. The major bottleneck limiting the catalytic applications of noble metals is their high cost and unsustainability owing to their relatively low abundance in nature. For this reason, an urgent challenge facing the human race is how to design, synthesize and assemble high-performance and low-cost metal nanocatalysts. In the past decade, great efforts have been made towards this goal, leading to numerous gratifying and encouraging achievements [72–82]. The catalytic properties of metal nanocatalysts are closely related to their shapes, structures, sizes and compositions that are essentially correlated with atomic arrangements, molecular adsorption and activation, electronic structures, charge transport, and other surface and interface features. Inspired by previous researches, we can simply define electrocatalysis as the heterogeneous catalytic reaction that occurs on the surface and interface of electrode materials and electrolytes [2–10,83]. Thus, surface and interface engineering is vital to the design, synthesis, construction and optimization of metal nanostructures and their hybrid nanostructures.

Fuel cells (FCs) are a class of electrochemical power-generation devices which can directly and continuously convert the chemical energy of fuels and oxidants into electrical energy. Compared with other traditional power-generation devices, fuel cells have the merits of low noise, fast response, good modularization, high operation quality and high energy conversion efficiency and can adapt well to different load power requirements, waste-heat utilization and catalyst loading/manufacturing solutions [84–88]. Fuel cells can be classified as molten carbonate fuel cells (MCFCs), solid oxide fuel cells (SOFCs), alkaline fuel cells (AFCs), phosphoric acid fuel cells (PAFCs), and proton exchange membrane fuel cells (or polymer electrolyte membrane fuel cells, PEMFCs) according to the varied electrolytes used in the cells. Despite the difference in electrolytes, reactions at the anode and the cathode in all FCs are separated, in which ions move through the electrolyte while electrons transport along an external circuit. Among the five

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