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Review

Advances in efficient electrocatalysts based on layered double hydroxides and their derivatives

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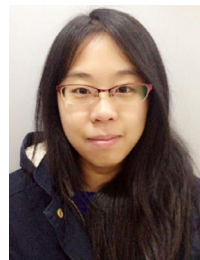
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ABSTRACT

The explore and development of electrocatalysts have gained significant attention due to their indispensable status in energy storage and conversion systems, such as fuel cells, metal–air batteries and solar water splitting cells. Layered double hydroxides (LDHs) and their derivatives (e.g., transition metal alloys, oxides, sulfides, nitrides and phosphides) have been adopted as catalysts for various electrochemical reactions, such as oxygen reduction, oxygen evolution, hydrogen evolution, and CO₂ reduction, which show excellent activity and remarkable durability in electrocatalytic process. In this review, the synthesis strategies, structural characters and electrochemical performances for the LDHs and their derivatives are described. In addition, we also discussed the effect of electronic and geometry structures to their electrocatalytic activity. The further development of high-performance electrocatalysts based on LDHs and their derivatives is covered by both a short summary and future outlook from the viewpoint of the material design and practical application.

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Lei Zhou received her Bachelor's degree in 2013 from Beijing University of Chemical Technology. She joined Professor Xue Duan's group as a Ph.D. candidate at Beijing University of Chemical Technology in 2013. Her research interests currently focus on the design and fabrication of structured transition metal sulfides/phosphides, which serve as electrocatalyst for small molecules catalytic reactions.



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1. Introduction

The ever increasing demand and environmental concerns arise from fossil-fuel combustion have attracted tremendous attention for exploring sustainable and renewable alternative

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energy sources [1–3]. Electrocatalysis with high efficiency and selectivity plays an indispensable role in future clean energy storage and conversion systems, such as fuel cells [4–8], metal–air batteries [9–15] and solar water splitting cells [16–21]. Recently, the rapid development for the design and controlled synthesis of electrocatalyst has greatly promoted the advances and outspread applications of this electrochemical and catalytic technology. Various electrocatalysts for different molecular reactions (e.g., water splitting [22–28], O₂ and CO₂ reduction reactions [29–38]) have been constructed with fine-tuned nanostructures (ultrathin nanosheets [39–42], nanorods [43–46] and hierarchical microspheres [47–51]), giving rise to largely enhanced performances. However, electrocatalysts meeting the requirements for commercial consideration with excellent activity, high selectivity and stability are still far from satisfaction.

Noble metal based electrocatalysts (such as Pt, Pd, and Ru) [52–59] have been demonstrated with superior behavior in various electrocatalytic reactions; however, they cannot afford wide range commercial application due to their scarcity and exorbitant price. To overcome this issue, tremendous endeavor has been devoted to non-precious candidates with low cost and high activity. The first row transition metal (Fe, Co, and Ni, etc.) based electrode materials, including transition metals [60–65], hydroxides [66–72], oxides [73–79], sulfides [79–86], nitrides [87–93], phosphides [94–101], have attracted extensive attention in energy conversion applications owing to their satisfactory catalytic performance in electrochemical reactions. Consequently, great efforts have been focused on the controlled synthesis of these non-precious catalysts with desired structure and promising performances. Exploring a facile and efficient strategy to achieve a high dispersion of active sites for the maximum utilization of material resources is an important part for their practical applications. On the other hand, the tunability of geometric and electronic state of active sites at the atomic scale is a fundamental factor for delivering a high performance in specific electrochemical reactions [102–104]. Thus, the development of green and facile synthetic routes of well-designed non-precious nanocatalysts with tunable composition, structure and morphology toward electrocatalytic energy production still remains highly desirable and challenging.

Two-dimensional (2D) nanomaterials afford great opportunities to create advanced electrocatalysts with high performance and stability due to their unique physicochemical properties [105–109]. Layered double hydroxides (LDHs) are a typical family of 2D clay materials, whose structure is based on well-defined intralayer multi-metal cations and interlayer anions [110–116]. By virtue of their versatility in chemical composition and architectural structure, LDHs materials have been widely explored as efficient electrocatalysts for the energy storage and conversion [117–123]. Moreover, various transition metal nanosheets can be fabricated via a topotactic transformation of LDHs precursors, which takes advantage of highly-uniform and ordered dispersion of active sites [61,62]. In addition, the two-dimensional nanostructure of LDHs provide the confined space for directed growth of other functional materials, also giving rise to a chance to design efficient electrocatalysts [124–126].

Up to now, several important reviews have been reported to interpret the development of LDHs-based materials for electrochemical energy storage and conversions [127–131]. However, a summary on LDHs derived electrocatalysts has not been reported until now, which we believe is of great significance to design sustainable energy materials. In this review, a brief highlight about the new ideas in the exploring of LDHs-based electrocatalysts is presented. To demonstrate more possibilities of using LDHs to fabricate efficient electrocatalysts, we will focus on the first row transition metal based catalysts derived from corresponding LDH precursors, including transition metal alloys, oxides, sulfides,

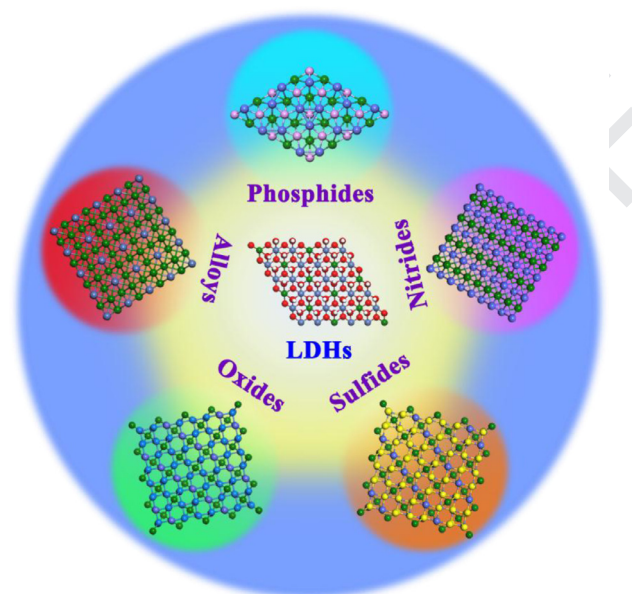


Fig. 1. Schematic presentation of transition metal alloys, oxides, sulfides, nitrides and phosphides derived from LDHs precursors.

nitrides and phosphides. Their synthesis strategies, structural characters and electrochemical performances will be introduced in every section. Current challenges and future outlook are also proposed from the viewpoint of the material design and practical application.

2. LDHs-based electrocatalysts

Transition metal containing LDHs have demonstrated large potential as efficient electrocatalysts, especially for oxygen evolution reaction (OER) [132–137]. Since the pioneer work reported by Dai et al. in 2013 [138], various LDHs and their hybrids have been investigated for OER or photoelectrochemical (PEC) water splitting [139–143]. Dai et al. also summarized the advances for the NiFe-based materials (including NiFe alloy, oxides and hydroxides) as highly active OER electrocatalysts in 2015 [130]. To overcome the intrinsic drawbacks of LDHs in conductivity as well as to modify the active sites, coupling LDHs with conducting materials, especially carbon materials, have attracted much interest [67,117,136]. According this new research upsurge, Zhang et al. gave an excellent summary and perspective in the rational integration of LDHs and nanocarbon [127].

In this work, we try to shed light on some new strategies in the design of LDHs-based electrocatalysts. It is reported that the electronic structures of LDHs determines their electrocatalytic activity by adjusting the ability of adsorption and desorption of intermediates [119,144]. For instance, the moderate guest-metal substitution into the host-oxyhydroxide framework (Fe into Ni or Ni into Fe) substantially enhanced the oxygen evolution activity due to the different metal–oxygen bond lengths and adsorption energies of the intermediates [144,145]. Very recently, Zhang et al. further propose an interesting concept of anionic regulation strategy to modify the electronic structure of CoFe-LDH, which significantly improves the OER performance by facilitating the adsorption, electron transfer, and desorption process simultaneously [146]. As shown in Fig. 2, although the cations in LDHs can provide strong positive electric field, the practical adsorption of hydroxyl is deficient when the easy polarization of anions adjacent to the central cations, which makes the reactants cannot feel enough positive electric field. To solve this problem, the OH[−] in the solid

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