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Review

Two-dimensional nanosheets as building blocks to construct three-dimensional structures for lithium storage

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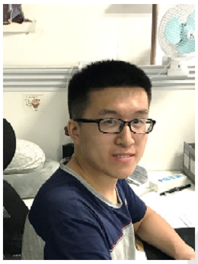
ABSTRACT

2D nanosheets such as graphene, silicene, phosphorene, metal dichalcogenides and MXenes are emerging and promising for lithium storage due to their ultrathin nature and corresponding chemical/physical properties. However, the serious restacking and aggregation of the 2D nanosheets are still hampering their applications. To circumvent the issues of 2D nanosheets, one efficient strategy is to construct 3D structures with hierarchical porous structures, good chemical/mechanical stabilities and tunable electrical conductivities. In this review, we firstly focus on the available synthetic approaches of 3D structures from 2D nanosheets, and then summarize the relationships between the microstructures of 3D structures built from 2D nanosheets and their electrochemical behaviors for lithium storage. On the basis of above results, some challenges are briefly discussed in the perspective of the development of various functional 3D structures.



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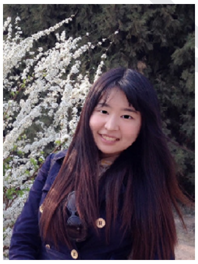

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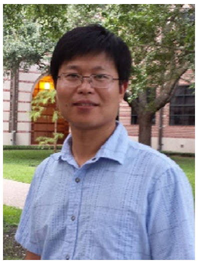

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1. Introduction

Lithium-ion batteries (LIBs) have been widely utilized as power sources for various portable electronic devices due to their high energy density and environmental friendliness [1–3]. However, the power density and cycle life are still needed to be further improved when LIBs are employed in electric-vehicles (EV), station-

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ary electricity storage and other large-scale applications. To improve the electrochemical performances of LIBs, three-dimensional (3D) electrode materials are highly desired as they are easy to access the electrolyte and have short ion diffusion lengths. According to the equation of $\tau = L^2{}_{ion}/D_{ion}$ [4,5], where τ is ionic diffusion time in the host material, L_{ion} is the ionic diffusion length, and D_{ion} is the ionic diffusion coefficient, it is known that lithium-ion diffusion time is in direct proportion to the square of ionic diffusion length. Thus, designing and fabricating 3D structured electrode materials with short ionic diffusion lengths are becoming an efficient strategy to dramatically reduce the diffusion time of lithium ions and meanwhile enhance their electrochemical performances especially at high current densities. Although 3D nanostructures can be realized via various approaches, those built from 2D nanosheets have recently attracted much attention owing to their high surface-to-volume ratio, good mechanical properties, tunable active materials and fully exposed active surfaces based on the large family of emerging 2D nanosheets or atomic layers.

To date, various 2D nanosheets including graphene and graphene analogies (hexagonal boron nitride (h-BN) [6,7], graphitic carbon nitride (g-C_3N_4) [8,9], transition metal dichalcogenides (TMDs) [10,11], black phosphorus (BP) [12,13] and MXenes [14–17]) have been widely explored via some manners such as mechanical exfoliation, liquid-phase exfoliation, chemical vapor deposition and hydrothermal approaches. These free-standing 2D nanosheets can be further employed as building blocks to construct various 3D structures with hierarchical porous structure and tunable components. In this review, we focus on recent advances in the controlled synthesis of 3D structures by ultrathin 2D nanosheets and their potential for lithium storage. This review is divided into three major categories based on the types of employed 2D nanosheets: 3D structures built from graphene nanosheets, 3D structures built from graphene and graphene analogies, and 3D structures built from graphene analogies. We firstly highlight their available synthetic methods, and then summarize the relationships between the microstructures of 3D structures built from 2D nanosheets and their electrochemical behaviors for lithium storage. On the basis of above results, we give potential directions to produce 3D structures and identify some of the remaining challenges.

2. Synthetic approaches of 3D structures based on 2D nanosheets

2.1. Solvothermal/hydrothermal approaches

Generally, free-standing 2D nanosheets such as graphene, graphene oxide (GO), reduced graphene oxide (rGO), MoS_2 and MXenes can be facilely dispersed in some solvents such as water, isopropanol (IPA), *N*-methyl pyrrolidone (NMP) and/or *N*,*N*-dimethylformamide (DMF) [4,18–23]. The homogeneous dispersions usually keep a stabilized state in equilibrium due to the balance between attractive and repulsive forces [24]. For instance, in

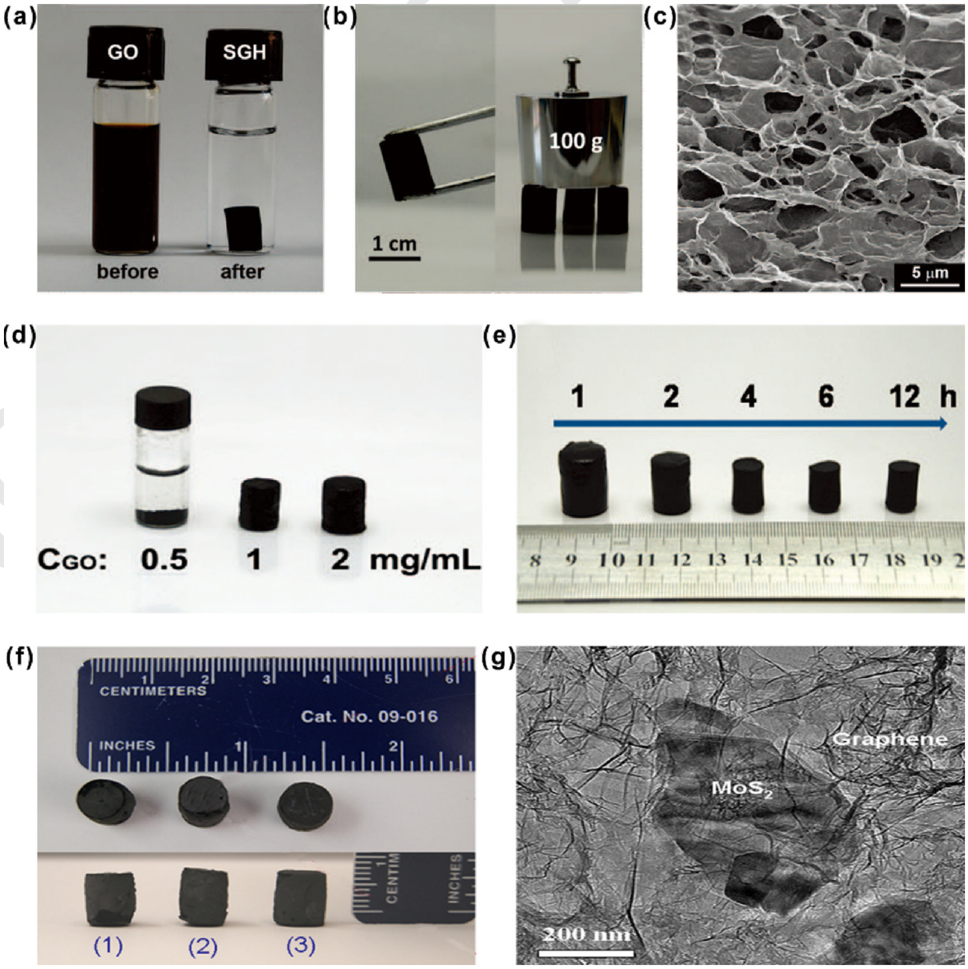

Fig. 1. (a) Photographs of a 2 mg/mL homogeneous GO aqueous dispersion before and after hydrothermal reduction at 180 °C for 12 h; (b and c) Photographs and SEM image of the SGH interior microstructures; (d and e) Photographs of the products prepared by hydrothermal reduction of GO dispersions under different conditions; (f and g) Photographs and HRTEM image of MoS_2-graphene architectures. Reprinted from Refs. [25,31] with permission of American Chemical Society and Wiley.

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