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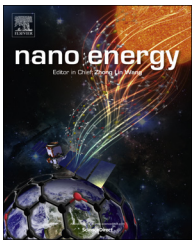

RAPID COMMUNICATION

Chemical adsorption: another way to anchor polysulfides

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Abstract
Lithium-sulfur (Li-S) batteries are known for their high energy density but suffer from poor cycling performance due to severe dissolution of the polysulfides (PSs) in liquid electrolyte and their shuttle effect. Effectively anchor the PS should combine the physical and chemical adsorption. However, most of the current studies are focused on confining the PS mobility by physical adsorption alone. Herein, chemical adsorption of the PS is emphasized in a special cell with functionalized graphene adsorbers. Both the experimental and calculated results show that functional groups containing C, N, O and P atoms are beneficial for immobilizing the PSs via chemical bonding, such as C-S, O-S, P-S. Such cells deliver a reversible capacity as high as 1218 mA h g^{-1} and cycle stably by solely applying the chemical adsorption, comparable to the current reports mostly based on physical adsorption. Our findings enrich the fundamental understanding of the chemical adsorption of the PS and shed light on the design of sulfur electrodes.

Introduction

Lithium-sulfur (Li-S) batteries are expected to find applications in various energy storage systems. However, high dissolution of polysulfides (PSs) in liquid electrolyte and their shuttle effect deteriorate the cycling performance of the Li-S battery [1–4]. Therefore, effectively anchoring the PSs is critical to improving the electrochemical performances of these batteries.

Various physical effects have been applied to confine the PSs in some substrates. Porous conducting matrices such as porous carbons [5,6], conducting polymers [7,8] and oxides [9] have been used to immobilize the PSs by the physical capillary effect. Core-shell [10] nano-architectures were tried to

Abbreviations: PS, polysulfide; FG, functional groups; FGNS, functionalized graphene

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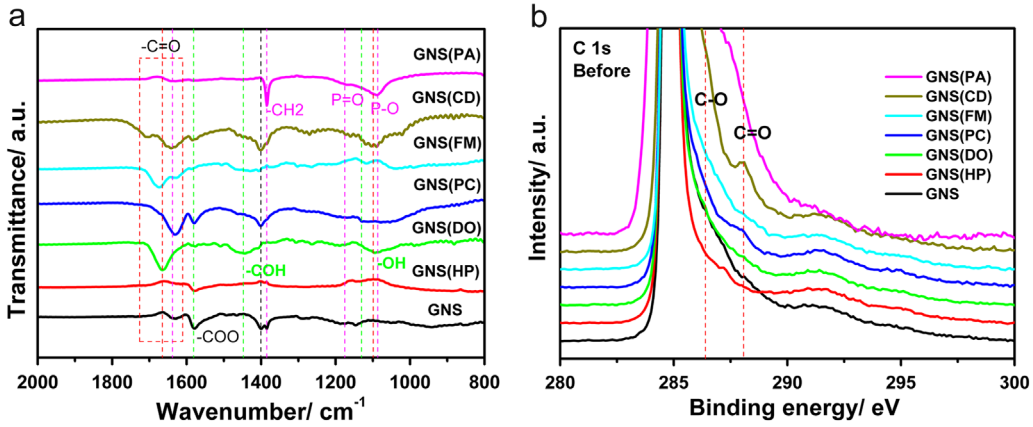

Fig. 1 FTIR (a) and XPS (b) spectra of different FGNS. The FTIR and XPS spectra show that FGs have been well grafted on the graphene.

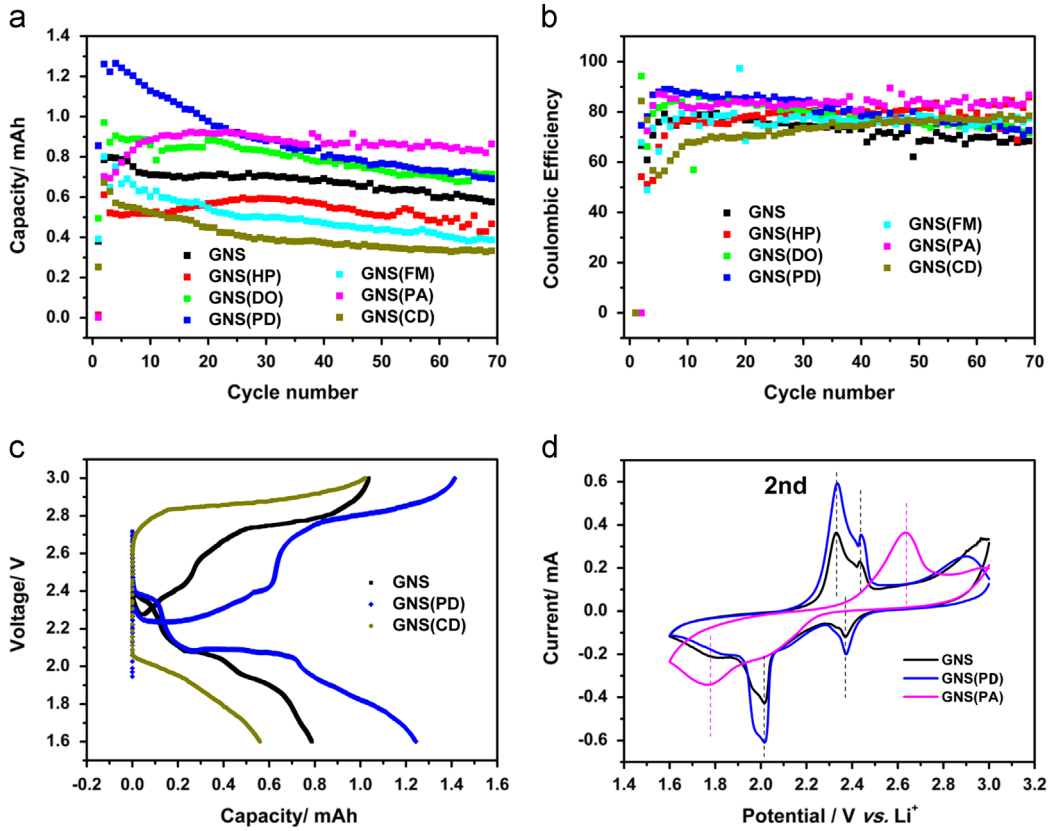

Fig. 2 Comparison of the cycling performance (a), Coulombic efficiency (b), potential profile of the second cycle (c) and CV curves (d) of the cells using different FGNS as electrode. Based on the capacity of the cells, the adsorption capability of different FGNS can be ranked as follows: GNS(PD) > GNS(PA) > GNS(DO) > GNS > GNS(HP) > GNS(FM) > GNS(CD) initially.

separate the active sulfur or PS from the electrolyte in two spaces while the yolk-shell structure [11] provides extra voids for the volume expansion of the active material during cycling. Sulfur electrodes with such configurations show improved cycling stability. However, large-scale synthesis of materials with well-defined morphology is complicated and expensive.

The strong interaction between the substrate and the active material in a chemical adsorption mode is expected to reinforce the trapping strength. However, immobilization of the PSs by chemical adsorption has been rarely explored and is lack of fundamental understanding. O-containing functional groups (FGs) on graphene oxide [12-15] are conducive to immobilize PSs. *Ab initio* calculations based on the adsorption of S [15] or S_3 [14] clusters indicated that both epoxy (C=O) and hydroxyl (-OH) groups can enhance the binding of S to the C-C bonds due to the induced charge transfer. However, changes of the FGs after cycling were ignored. The chemical adsorption of the PSs during cycling is difficult to be evidenced