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Review article

Recent progress of advanced binders for Li-S batteries

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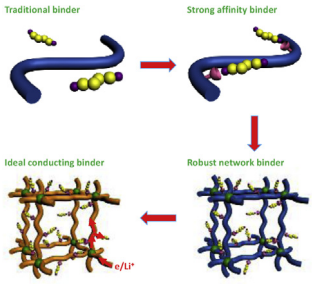
HIGHLIGHTS

- Literature on binders for Li-S batteries has been reviewed.
- Properties of binders for practical-loading sulfur cathodes have been discussed.
- A perspective on the future development of practical binders is given.

GRAPHICAL ABSTRACT



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ABSTRACT

Lithium-sulfur (Li-S) batteries are considered as one of the most promising energy storage systems. However, the commercial application of Li-S batteries with practical loading levels ($> 7\ mg\ cm^{-2}$) still remains several obstacles, including low sulfur utilization, short lifespan, and poor rate property. Exploiting advanced multifunctional binders is an effective and straightforward approach to improve electrochemical performance and this method has the inherent advantage of not introducing additional weight and volume, which will undermine maximizing the cell energy density. Traditional PVDF binder fails to withstand mechanical instability of high-loading electrode; moreover, the binder possesses poor affinity for polysulfides, making it unsuitable for high-loading Li-S battery. Hence, designing rationally advanced binder systems is urgently required to fabricate high-loading sulfur electrodes. Although much effort has been devoted in this regard, practical binders for high-loading sulfur electrodes are still absent. To accelerate the development of such binder systems, we review the recent progress on the advanced binders in high-performance Li-S batteries. We discuss their functional mechanisms, summarize their desirable properties, and then provide perspective on the future development of advanced binders for practical Li-S batteries.

1. Introduction

Owing to the ever-increasing demands of high-energy-density and long-life energy sources for electric vehicles (EVs), hybrid electric vehicles (HEVs), and smart electric grids, developing advanced energy storage systems is particularly urgent [1–3]. Lithium-sulfur (Li-S) batteries with a high theoretical gravimetric energy density of 2500 Wh kg^{-1} have been considered as one of the most promising candidates in this regard [4–8]. In a typical Li-S cell, Li metal is used as the negative electrode and is separated from the sulfur positive electrode by an ion-conducting liquid or solid electrolyte [8]. A widely accepted illustration of the reaction process of Li-S batteries is given [9]. The sulfur is reduced in a stepwise fashion as $S_8 \rightarrow Li_2S_8 \rightarrow Li_2S_6/Li_2S_4 \rightarrow Li_2S_2/Li_2S$. Additionally, some other reaction routes forming $S_3^{-\cdot}$ and other polysulfide intermediates via disproportionation/decomposition reactions have also been presented [9]. The overall redox reaction of a Li-S

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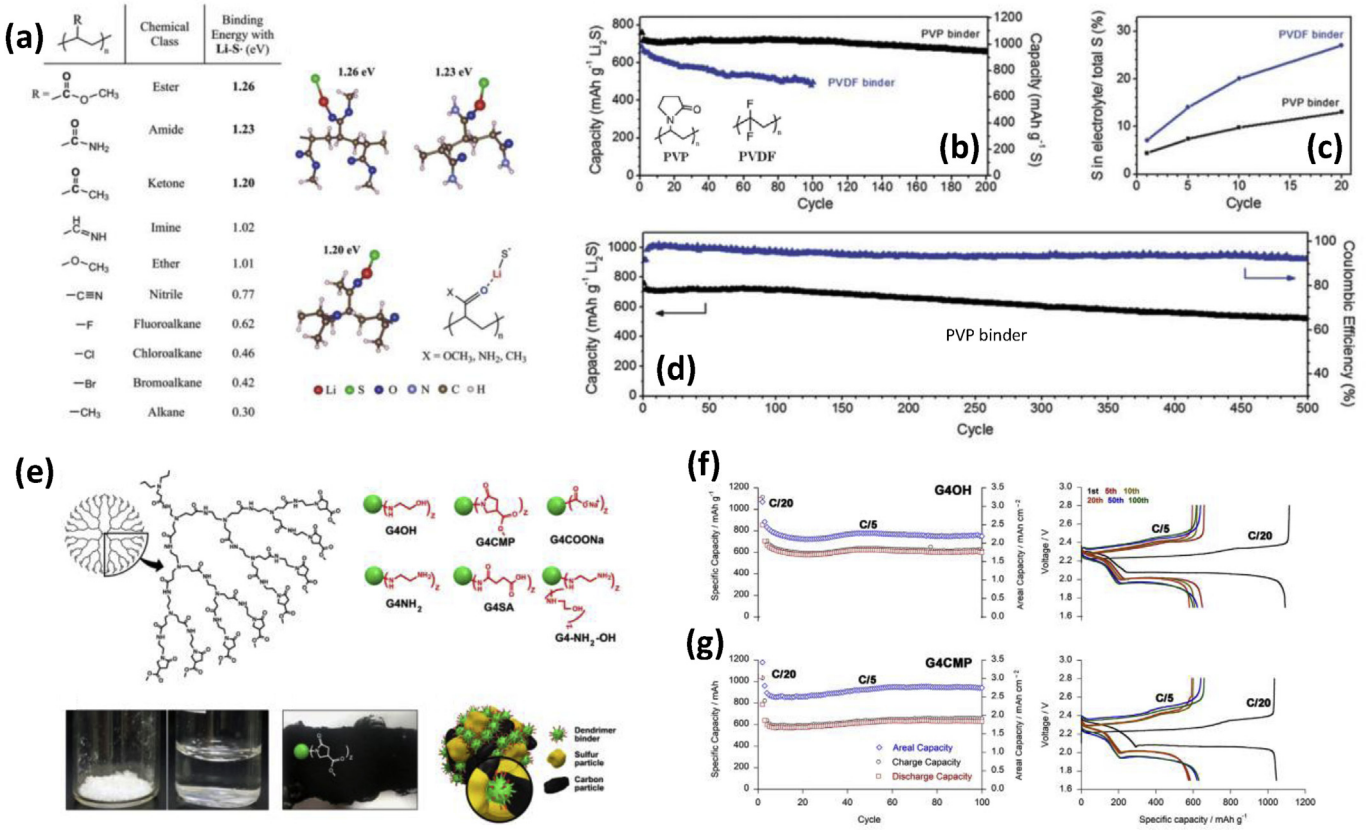

Fig. 1. (a) Calculated binding energies of Li–S• species with various functional groups and the most stable configuration of Li–S• with ester, amide, and ketone groups; (b) Specific capacity of Li_2S cathodes using a PVP binder at 0.2 C compared with the PVDF binder; (c) Percentage of sulfur in the electrolyte relative to the total sulfur mass on the electrode after cycling at 0.2 C using the PVP and PVDF binders; (d) Specific capacity and coulombic efficiency of a Li_2S cathode using a PVP binder upon prolonged cycling over 500 cycles at 0.2 C. Reprinted with permission from ref [64]. Copyright 2013 The Royal Society of Chemistry. (e) Schematic representation of a G3CMP dendrimer with a magnified image of its branch and image of electrode coated with a slurry having the water-soluble G4CMP binder; Electrochemical cycling and capacity–voltage profiles of (f) G4OH and (g) G4CMP dendrimer binder-based Li–S cells. Reprinted with permission from ref [71]. Copyright 2015 Elsevier.

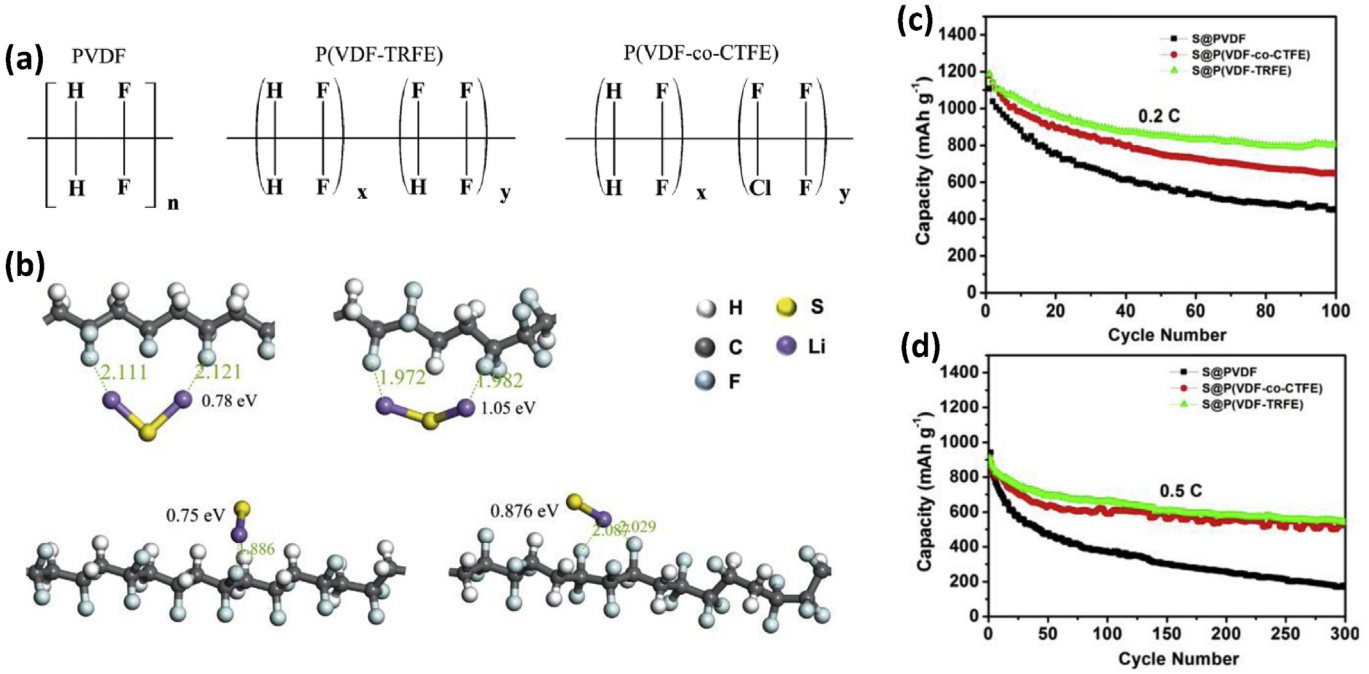

Fig. 2. (a) Chemical structures of PVDF, P(VDF-TRFE), and P(VDF-co-CTFE) binders; (b) *Ab initio* simulations showing the most stable binding configurations of Li_2S and Li–S• species with PVDF (left) and P(VDF-TRFE) (right) binders; (c) Cycling stability of sulfur electrodes with different binders at a current density of 0.2 C; (d) Long-term cycling performance at 0.5 C of sulfur electrodes with different binders. Reprinted with permission from ref [72]. Copyright 2016 Elsevier.