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Application of Carbon Nanotube-Based Materials as Interlayers in High-Performance Lithium-Sulfur Batteries: A Review

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With the ever-increasing demands of electrochemical energy storage, lithium–sulfur (Li–S) batteries have drawn more attention because of their superior theoretical energy density and high specific capacity. However, practical applications of Li–S batteries suffer from problems such as low conductivity of sulfur and discharged products, severe polysulfide shuttling effect, and large volume change of sulfur during cycling, resulting in sluggish rate performance, and unsatisfactory cycle life. Various nanostructured carbon materials have been served as barrier layers to overcome these problems. In particular, carbon nanotubes (CNTs) with unique 1D nanostructure, have been introduced to Li–S batteries as the intermediate layers because of its superior flexibility, excellent electrical conductivity, and good chemical stability. Moreover, CNTs and CNTs-based barrier layers could also curb lithium polysulfides shuttling. In the minireview, we summarize recent works of CNTs-based materials as modifying interlayers for Li–S batteries. In addition, the strategies to enhance electrochemical performances of the batteries are summarized and discussed. Finally, the challenges and prospects for future research of CNTs-based materials as interlayer are proposed. We hope this review will be useful for designing and fabricating high-performance Li–S batteries and boost their practical applications.

Keywords: lithium–sulfur batteries, carbon nanotubes, CNTs-based materials, interlayer, lithium polysulfides

INTRODUCTION

Nowadays, electric energy storage technology and equipment are gradually becoming a critical issue with the development of society (Wang F. et al., 2018; Wang R. et al., 2020; Liu et al., 2019a,b; Wu et al., 2019a; Zhao X. et al., 2019; Zhao et al., 2020; Li et al., 2020f; Ma et al., 2020b; Pan K. et al., 2020). In recent years, various electrochemical energy storage systems have been extensively studied (Ma et al., 2013, 2017, 2019; Luo et al., 2017; Chen H. et al., 2018; Guo et al., 2019, 2020; Liu et al., 2019c, 2020; Song C. et al., 2020; Wang F. et al., 2020; Yu et al., 2020; Zou P. et al., 2020). Among them, Li–S batteries have become one of the most promising candidates for next-generation electrochemical energy storage owing to their high theoretical specific capacity (1675 mAh g^{-1}) and superior theoretical specific

energy density (2600 Wh Kg^{-1}), together with non-toxicity, low cost and environmental friendliness of sulfur (Yu et al., 2018; Gao et al., 2020).

However, there are some challenges for Li–S batteries to overcome, including the electrical insulating characteristic of sulfur and the discharge product (Li_2S_2/Li_2S), the large volumetric change of sulfur during the charge/discharge process (approximately 80%), as well as the notorious shuttle effect caused by dissolved lithium polysulfides (LiPSs) (Huang et al., 2015; Cao et al., 2019; Chen Y.T. et al., 2019).

To address these problems, researchers have made considerable effort with regard to cathode and separator modification (Wang X. et al., 2019; Wei et al., 2019; Li et al., 2020a), optimization of electrolytes (Ding et al., 2016), and lithium metal anode protection and stabilization (Paolella et al., 2019; Pei et al., 2019). Among them, the interlayers could selectively control the shuttle effect of polysulfides, while not disturbing the Li^+ transfer. As a superior type of carbon material, carbon nanotubes (CNTs) have been intensively investigated as intermediate layers due to their excellent mechanical durability and high electrical conductivity as well as stable chemical properties. Together with other materials, CNTs-based interlayers are receiving considerable attentions for Li–S batteries in recent years (Wang et al., 2017; Jiang et al., 2018; Li et al., 2018; Son et al., 2019). For example, Yao M. et al. (2018) reported the multifunctional layer consisting of MnO_2 and multi-walled CNTs modified polypropylene (PP) separator could tightly attract polysulfides and promote the transfer of electrons and ions, which could enhance the electrochemical performance of Li–S batteries. Li et al. (2018) designed and constructed a MoS_2/CNTs interlayer to insert between separator and cathode, exhibited an optimized cycle performance and excellent rate capability. Some reviews also discussed recent developments on the interlayer materials for Li–S batteries. For instance, Chen et al. reviewed on interlayer design of Li–S batteries by coating and inserting different types of carbon-based materials (Chen L. et al., 2020). Furthermore, Jiao et al. reviewed recent articles regarding to the interlayers for Li–S batteries including carbon-based interlayers (Jiao et al., 2019); Pang and co-workers reviewed on recent research work on CNT-based materials for Li–S batteries (Zheng et al., 2019). However, there is still a lack of review to exclusively cover the state-of-the-art developments of CNTs-based interlayer materials for Li–S batteries.

Herein, we provide an overview on carbon nanotubes-based materials as inter-layers of Li–S batteries. Their nano/microstructure and electrochemical performances are summarized. In addition, some reasonable suggestion on their design and development are proposed to facilitate breakthroughs. We hope this review could attract more attentions to CNTs-based interlayer materials for Li–S batteries and boost their practical applications.

MWCNTs

CNTs can be regarded as the curled graphene sheets, and they could be classified into two catagories, including multi-walled CNTs (MWCNTs) and single-walled CNTs (SWCNTs) (Kumar S. et al., 2018; Ali et al., 2019). MWCNTs could be seen as the collection of multiple curled graphene sheets. Compared to SWCNTs, they are cheaper and easier to fabricate. Moreover, they remains the merits of high conductivity, low thermal expansion coefficient and stable structure (Zheng et al., 2019). For interlayers for Li–S batteries, MWCNTs are used mainly in two ways, (1) as the coating layer on separator (Cheng et al., 2016; Wang J. et al., 2018); (2) as the self-supporting film (Kim H.M. et al., 2016; Wang X. et al., 2020). The electrochemical performances of Li–S batteries with MWCNTs-based interlayers are summarized in **Table 1**.

Separator-Based Interlayer

As an essential part of Li–S batteries, separators could separate the cathode and anode and prevent the internal short circuit. However, the pore size (∼100 nm) of commercial separators is much larger than the average size of long-chained polysulfides, which may cause the soluble polysulfides to easily migrate to the anodes, leading to notorious shuttle effect (Li et al., 2020d). To prevent the shuttling of dissolved polysulfides, various MWCNTs-based materials have been developed to modify separators to provide physical entrapment and chemical adsorption for LiPSs, enhancing the electrochemical performances of Li–S batteries (Chang et al., 2015; Tan et al., 2018; Li et al., 2019e; Xiang et al., 2019).

Pure MWCNTs

As the most common CNT materials, MWCNTs have been intensively applied as interlayer coated on separator for Li–S batteries. For example, Chung and Manthiram (2014) developed a MWCNT-coated separator as a barrier to anchor dissolved LiPSs, as shown in **Figure 1A**. The MWCNTs layer not only act as an upper current collector for rapidly electron transport and high sulfur utilization but also serve as a filter to trap and adsorb LiPSs. Meanwhile, the porous network of MWCNTs layer is conducive to electrolyte infiltration and electron/ion diffusion. Hence, the cell with MWCNTs-coated separator exhibited a superior long-term cycling performance. After 150 cycles, the cell with MWCNT layer delivered a specific capacity of 881, 809, and 798 mAh g^{-1} at 0.2, 0.5, and 1 C, respectively. After that, researchers have paid more attention on carbon nanotubes modified separator to improve the performance of Li–S batteries. For instance, Ponraj et al. (2017) synthesized a hydroxyl-functionalized carbon nanotube (CNTOH) coated on the separator, which could address the poor electronic conductivity of active materials and mitigate the diffusion and migration of LiPSs in Li–S batteries owing to the good conductivity and hydroxyl groups of CNTOH.

MWCNTs-Based Composites

Nano-composite materials normally have a better performance than single-component materials due to the synergistic effect of different components, which have attracted extensive attention from researchers in recent years (Luo Y. et al., 2018; Guan B. et al., 2019; Kim et al., 2020; Ma et al., 2020a; Wang W. et al., 2020). Many types of MWCNTs-based composites,

TABLE 1 | Electrochemical performance of Li–S batteries employing CNT-based materials as interlayer.

Materials	Mass load (mg cm^{-2})	Sulfur contents (wt%)/loading (mg cm^{-2})	Current density	Cycle number	Initial capacity (mAh g^{-1})	Post-cycle capacity (mAh g^{-1})	References
Modify separator							
MWCNT	0.17	70/2	C/5	150	1324	881	Chung and Manthiram, 2014
DWCNT	0.00477	60/1.5–2	0.1 C	100	598	508	Sun et al., 2020
o-MWCNT	0.4	90/5	0.1 A g^{-1}	50	1105	925	Cheng et al., 2016
PAN/SiO_2-MWCNT	3.44	70/1–1.2	0.2 C	100	1182	741	Zhu et al., 2016
MWCNT/GO/MWCNT	0.23	70/7	0.2 C	200	1290	620	Chang et al., 2017
CNT	–	–/–	1 C	500	684	361	Liu et al., 2017
aCNT	∼0.15	67/1.5	0.5 C	500	1306	621	Huang et al., 2018
CNTOH	1.34	70/3	0.5 C	400	1056	∼591	Ponraj et al., 2017
OCNT	–	-/1.5	0.5 C	400	1034.9	∼732	Kim et al., 2018
CNT	(5	80/1.3	1 C	200	968	∼670	Manoj et al., 2018a
AB/MWCNT	–	70/3.2	1.6 A g^{-1}	200	1500	662	Tian et al., 2018
CNT/PVP	0.4	70/1–1.2	0.1 C	100	1226	887	Li et al., 2019c
Ni@NG-CNTs	0.6	70/1–1.2	1 C	800	1144.2	655.5	Zuo et al., 2019
Co-NCNTs	0.23	70/1.8	0.2 C	200	1251.4(20^{th})	857.7	Wei et al., 2020
SCL	0.25	70/1.35–1.55	1 C	400	1073	700.4	Geng et al., 2020
MoS_2@CNT	–	77.5/–	1 C	500	∼690	∼670	Jeong et al., 2017
Sb_2S_3/CNT	0.4	65/1.0	1 C	1000	∼720	373	Yao S. et al., 2018
SnS_2/CNT	0.4	78/1.4	2 C	800	∼782	555	Jiang et al., 2019
NSCNTs/MoS2	0.5	88.3/2.2	1 C	1000	1024	814	Xiang et al., 2019
SWCNT	0.13	80/3.0	0.2 C	100	953	713	Chang et al., 2016
PANiNF/MWCNT	0.01	60/1.4	0.2 C	100	1020	709	Chang et al., 2015
MCNT@PEG	0.26	60/1.6	0.5 C	200	1283	727	Wang et al., 2015
MWCNT/PEG	0.12	78/6.5	0.2 C	300	1206	630	Luo et al., 2016
PEI/MWCNT)	–	75/1.1	2 C	300	819	∼590	Lee Y.-H. et al., 2018
PEDOT: PSS-CNT	–	80/–	0.2 C	–	950	–	Manoj et al., 2018b
PAMAM-CNTs	0.34	60/0.6–0.8	2 C	1200	1050	573	Li et al., 2020c
MWCNT/SPANI	0.4	72/5	0.1 A g^{-1}	100	∼1127	913	Shi et al., 2018
PDAAQ/K-FGF/MWCNT/CTAB	–	70/1.8	1 C	200	1361	968	Kiai and Kizil, 2019
CNT/AC	–	70/–	0.2 C	200	1495.6	742	Guo et al., 2017
G@CNT	0.33	60/1.4	0.2 C	200	1231.3	935.1	Wu et al., 2017
CNT/CH	0.35	75/1.2	0.5 C	200	894	826	Jiang et al., 2018
MWCNT/NCQD	0.15	60/1.3–1.5	0.5 C	1000	1330.8	507.9	Pang et al., 2018
PC/MWCNT	0.51	70/1.6–1.7	0.5 C	200	911	659	Tan et al., 2018
G/CNT	0.17	75/1.7–2	0.5 C	100	∼913.5	813	Gao et al., 2019b
GO/CNT	–	60/1.1	0.2 C	50	1591.56	1003	Lee et al., 2019
EB	0.2	70/1.8	0.2 C	100	950 (25^{th})	980	Lu et al., 2019
Co/NCNS/CNT	0.2	80/2	2 C	1000	972.4	∼486	Song C.L. et al., 2020
CNT/Al_2O_3	–	60/1	0.2 C	100	1282	807.8	Xu Q. et al., 2015
Al_2O_3/CNT	–	70/1.0–1.2	0.2 C	100	1096	760.4	Chen X. et al., 2020
Fe_3O_4@C/CNTO	0.288	65/1.5	2 C	1000	∼833	335	Du et al., 2020
CNTs@FeOOH	–	55/0.8	3.2 A g^{-1}	350	1121.9	556	Li et al., 2020e
$MgBO_2$(OH)/CNT	0.3	70/1.1	0.5 C	200	924	785	Kong L. et al., 2017
CNT@TiO_2	0.7	66.4/1.7	0.1 C	200	1351	803	Yang et al., 2017
TiO_2/CNTs	0.3	70/1.5	1 C	250	1036.9	763	Guan Y. et al., 2019
MWCNTs @TiO_2	–	60/0.8–1.0	838 mA g^{-1}	600	1083	610	Ding et al., 2018
Ni-MOF/MWCNTs	0.36	60/1.58	0.5 C	300	1358	1183	Lee D.H. et al., 2018
CNT/ZrO_2	–	50/1.5	0.1 C	120	1207	685	Liu et al., 2018
MoO_3@CNT	0.577	60/1	0.3 C	200	1251	755	Luo L. et al., 2018
CNT/MoP_2	0.34	50/1.2	0.2 C	100	1223	905	Luo Y. et al., 2018

(Continued)

TABLE 1 | Continued

Materials	Mass load (mg cm^{-2})	Sulfur contents (wt%)/loading (mg cm^{-2})	Current density	Cycle number	Initial capacity (mAh g^{-1})	Post-cycle capacity (mAh g^{-1})	References
CNT@ZIF	0.9	70.2/1.2	0.2 C	100	1588.4	870.3	Wu et al., 2018
COF-CNT	–	80/2	1 A g^{-1}	500	1068	621	Wang J. et al., 2020
MoS_2/CNT	0.25	50/1.4	0.5 C	500	1237	648	Yan et al., 2018
MWCNTs/MnO_2	0.20–0.48	60/1,4	1 C	500	∼880	∼610	Yao M. et al., 2018
TiO_2/CNT	–	73/2	1 C	200	936	557	Chen A. et al., 2019
TiO_2/CNT	–	70/0.86	0.2 C	100	1247	627	Chen P. et al., 2020
TiS_2/CNT	0.6	69.7/2	0.5 C	100	1012	848	Pan S. et al., 2020
MWCNTs/CeO_2	0.15	60/1.8–2.0	0.2 C	300	898.3	520.7	Zhu et al., 2020
Sc_2O_3@CNT	0.17	65/1.5	1 C	500	1037	788	Xu et al., 2020
Co_2B@CNT	–	72.45/3.6	0.2 C	200	1430	1283	Guan B. et al., 2019
PCCNT/Ni_2P	0.8	78.8/1.1–1.2	1 C	500	1067	654.2	Guang et al., 2019
Ce-MOF/CNT	0.4	80/2.5	1 C	800	1021.8	838.8	Hong et al., 2019
CNTs/MXene	0.16	70/0.8–2.5	1 C	600	987	614	Li et al., 2019a
$Ti_3C_2T_x$/CNTs	0.016	70/1.2	1 C	200	760	640	Li et al., 2020b
TiO/MWCNT	0.7	60/1.4–1.6	0.5 C	200	1527.2	1033.8	Li et al., 2019e
Ta_2O_5/CNT-O	–	80/1.5	0.2 C	100	1230.7	932.6	Li et al., 2019f
CNTs/Fe_3O_4	0.38	72/1.4	1 C	1000	∼750	651	Sun et al., 2019b
MnO_2/CNT	0.35	70/0.8	1 C	500	843.7	∼574	Wang Y. et al., 2019
Co/NCNS/CNT	0.2	80/2	2 C	1000	972.4	434	Song C.L. et al., 2020
B-CNT	0.06	70/2.4–2.5	0.2 C	500	∼675	509	Chung et al., 2016
Gr-CNT-Ni	0.2	64/7.68	0.2 C	100	849	549	Kumar G.G. et al., 2018
W-V_2O_5-G/CNT	0.11	75/1.7–2	0.5 C	200	∼910	815	Gao et al., 2019a
HfO_2/CNT	0.087	75/1.80–2.22	1 C	500	∼1060	721	Kong et al., 2018
ACNTs	–	90/2.2–2.5	0.5 C	1400	897	713	Chen M. et al., 2018
Freestanding interlayer							
MWCNT/RGO	1	70/1.5–1.8	1 C	350	908	611	Sun et al., 2018
MWCNT	0.83–1.1	70/–	0.2 C	50	∼1420	962	Su and Manthiram, 2012
GF/CNT	0.54	70/1.6	0.2 C	230	1111.7	802.8	Lee and Kim, 2015
DF/PCW	–	56/1.8–2.7	1 C	100	1113	1040.6	Hwang et al., 2016
MWCNT	1.95	80/3	0.5 C	100	851	∼664	Kim H.M. et al., 2016
CNT	∼0.256	60/–	0.5 C	500	872	460	Sun et al., 2016
CNTs	1.0	70/0.974	0.25 C	100	1658	556	Li et al., 2017
CNT	0.45	45/4	0.5 C	80	∼1160	981	Xie et al., 2017
CNTs	1	-/3	1 C	200	943	802	Peng et al., 2019
SMAP	–	70/–	1 C	200	849	806	Rui et al., 2019
C-C-N-Co	0.45	70/1	0.5 C	100	∼1060	787	Li et al., 2020g
UPHC	–	60/1.5–2.0	0.5 C	300	704.5	608.8	Wang X. et al., 2020
SWCNT	–	70/–	0.1 C	50	1674	1052	Kaiser et al., 2015
PAA-SWNT	0.82	65/2.7	1 C	200	∼770	573	Kim J.H. et al., 2016
SWCNT	1	50/1.0–1.2	0.8 A g^{-1}	1500	1034	408	Jin et al., 2018
SWCNT/rGO	0.1	60/1	1 A g^{-1}	80	773	∼605	Hao J. et al., 2019
PEDOT:PSS-CNT	0.7	60/2	0.5 C	50	793	690	Wang et al., 2017
GO/CNT	1.1	70/1	0.2 C	300	1370	671	Huang et al., 2016
CNTs/RGO	0.34	70/2	4 C	600	∼765	597	Liu et al., 2016
MWCNTs/RGO	1	70/1.5–1.8	0.2 C	100	1224	854	Sun et al., 2018
CNTF	5–7.6	80/1.3	1 C	200	542	384	Manoj et al., 2019
G/CNT	0.2	50/2.46	2 C	500	∼460	445	Shi et al., 2019
CNTP/TiO_2	–	70/0.98	0.5 C	250	∼1575	575.8	Xu G. et al., 2015
G/M@CNT	0.104	60–80/1.11–1.37	1 C	2500	∼1065	293	Kong W. et al., 2017
MoS_2/CNTP	0.04	70/3.4	0.5 C	100	1233	850	Li et al., 2018
CNF@VS_2/CNT@GN	1.9	60/1	1 C	1145	1138.4	605	Wang L. et al., 2018

(Continued)

TABLE 1 | Continued

Materials	Mass load (mg cm^{-2})	Sulfur contents (wt%)/loading (mg cm^{-2})	Current density	Cycle number	Initial capacity (mAh g^{-1})	Post-cycle capacity (mAh g^{-1})	References
Fe_3C-C-CNTs	–	–/1.5	0.2 C	100	1243	870.2	Wang S. et al., 2020
CeF_3/CNTs	0.8–2	90/1.2–1.4	0.2 C	100	1015	951	Zou K. et al., 2020
Pd_3Co/MWCNTs	–	60/1	2 C	300	953	752	Cho et al., 2019
Pt@CNT	–	60/0.7	0.5 C	400	1120	660	Ding et al., 2019
MWCNTs-OH	–	70/2–2.3	1 C	500	601	516	Huang et al., 2019
MTO-CNT	0.06	65/1.1–3	0.5 C	500	931.7	634.6	Li et al., 2019b
SiO_2/AP	–	87/–	1 C	200	∼1087	1039	Li et al., 2019d
BTO/C	–	60/0.94	0.2 C	200	∼1201	908	Son et al., 2019
ZnO/CNT/RGO	0.85	–/1.7	0.2 C	150	1061	768	Sun et al., 2019a

o-MWCNT, oxidized multiwall carbon nanotube; PAN/SiO_2-MWCNT, poly-acrylonitrile/silica-MWCNT; aCNT, activated carbon nanotube; AB/MWCNT, acetylene black/MWCNT; SCL, sandwich-structure composite carbon layer (a N-doped mesoporous carbon sandwiched between two carbon nanotube); NSCNTs/MoS_2, nitrogen and sulfur codoped carbon nanotubes/MoS_2; SWCNT, single-walled carbon nanotube; MWCNT/PEG, MWCNT/polyethylene glycol; MWCNT/SPANI, MWCNT/sulfonated polyaniline; SMAP, nano-SnO_2/MWCNTs/aramid paper; PDAAQ/K-FGF/MWCNT/CTAB, poly 1,5-diaminoanthraquinone/potassium functionalized graphene/MWCNT/cetyltrimethyl-ammonium-bromide; CNT/AC, CNT/active carbon; PAMAM-CNTs, poly(amidoamine)-modified multiwalled carbon nanotubes; COF-CNT, Covalent organic frameworks-CNT; CNT/CH, CNT/chitosan; MWCNT/NCQD, MWCNT/N-doped carbon quantum dot; PC/MWCNT, porous carbon/MWCNT; EB, the mixture of H_xMnO_{2+x} nanosheets, graphene and CNTs; PCCNT/Ni_2P, P-doped carbon (PC) wrapped carbon nanotubes/Ni_2P; Ta_2O_5/CNT-O, Ta_2O_5/oxidized multi-walled carbon nanotubes; MTO-CNTs, mesoporous TiO_2-carbon nanotubes; Gr-CNT-Ni, graphene-CNT-nickel; C-C-N-Co, single-atom cobalt-anchored nitrogen-doped carbon nanosheets and dual network of carbon nanotube-cellulose nanofiber hybrid; UPHC, ultraviolet polymerized PVDF-HFP with the addition of MWCNTs; ACNTs, N, B, S tri-doped active carbon nanotubes; PEDOT:PSS-CNT, poly(3,4-ethylenedioxythiophene):polystyrene sulfonate–carbon nanotube; PAA-SWNT, poly(acrylic acid) coated single-walled carbon nanotube; CNTF, acid-functionalized carbon nanotubes; CNTP/TiO_2, carbon-nanotube paper/TiO_2; SiO_2/AP, nano-SiO_2 and multi-walled carbon nanotubes and aramid paper; BTO/C, $BaTiO_3$/CNT; NMC/CNT, N-doped mesoporous carbon/CNT; Co/NCNS/CNT, Co/nitrogen-doped carbon nanosheets/CNT. The symbol "-" in the table means that there is no relevant information in the reference.

such as MWCNTs/carbonaceous material composites, MWCNTs/polymer composites and MWCNTs/metal-based compound composites have been applied as interlayer coated on separator for Li–S batteries. For example, Pang et al. (2018) fabricated a MWCNT/N-doped carbon quantum dots (NCQDs) composites as coating layer onto the separator for Li–S batteries (**Figure 1B**). The NCQDs with large surface area and rich oxygenated functional groups could be regarded as an efficient adsorbent for LiPSs, meanwhile, MWCNTs are beneficial to enhance the conductivity of composites. Therefore, the cell with MWCNT/NCQDs-coated separators displayed an excellent electrochemical performance, as well as an average self-discharge value of ∼11% after resting for 48 h, which is lower than that with the pristine MWCNTs-modified separator (**Figure 1B**).

MWCNTs/Polymer composites have also been fabricated to trap LiPSs through physical obstructing and chemical bonding in Li–S batteries (Chang et al., 2015; Luo et al., 2016; Lee Y.-H. et al., 2018). For Lee Y.-H. et al. (2018) instance, developed a spiderweb separator consisting of three functional nanomats with good mechanical properties and excellent electrical conductivity. In this sandwich-type separator, MWCNT-wrapped polyetherimide (PEI/MWCNT) nanomats were used as the top and bottom layer, and the PVIm[TFSI]/poly(vinylidene fluoride-co-hexafluoropropylene) (PVDF-HFP) with a polyethylene (PE) nanomat was the middle layer (**Figure 1C**). When tested in Li–S batteries, the cell with this composites separator exhibited higher discharge capacity and more stable cycling performance than that with pristine PE separator (Lee Y.-H. et al., 2018).

Metal-based compounds are mainly including metal oxides (Ding et al., 2018; Luo L. et al., 2018), metal sulfides (Jeong et al., 2017; Xiang et al., 2019), metal phosphides (Luo Y. et al., 2018), metal borides (Guan B. et al., 2019), metal hydroxides (Kong L. et al., 2017) and metal-organic frameworks (MOFs) (Lee D.H. et al., 2018; Wu et al., 2018), which could offer a polar chemical interaction with LiPSs and curb polysulfides shuttling in Li–S batteries. For instance, Hong et al. (2019) fabricated cerium-MOFs/MWCNTs composites as coating material on separators for Li–S batteries at a high sulfur loading of 6 mg cm^{-2}, the cell with the modified separators could deliver an initial specific capacity of 993.5 mAh g^{-1} at 0.1 C, and the specific capacity of 886.4 mAh g^{-1} was retained after 200 cycles (**Figure 1D**). The superior electrochemical performance is ascribed that Ce-MOFs/MWCNTs can mitigate the transmission and diffusion of polysulfides and boost the efficient catalytic conversion of LiPSs, owing to their uniformly dispersed catalytic active sites and large specific surface area as well as good electronic conductivity. Yao S. et al. (2018) successfully exfoliated a 2D antimony sulfides (Sb_2S_3) sheets and combined them with MWCNTs (Sb_2S_3/MWCNTs) to modify the Celgard PP separator (**Figure 1E**). With Sb_2S_3/MWCNTs modified separator, the Li–S cell exhibited a low average capacity decay rate (∼0.049% per cycle after cycling 1000 cycles at 1 C), indicated that the Sb_2S_3/MWCNTs has a strong interaction with polysulfides which was confirmed by the first-principle calculation.

Free-Standing Interlayer

In addition to coating MWCNTs-based materials onto the separator to curb the shuttling of lithium polysulfides, another alternative strategy is to insert a self-supporting interlayer between the cathode and separator (Wang et al., 2017; Sun et al., 2018). The self-supported interlayers are usually much thicker than the modified-separators interlayers, and they are supposed

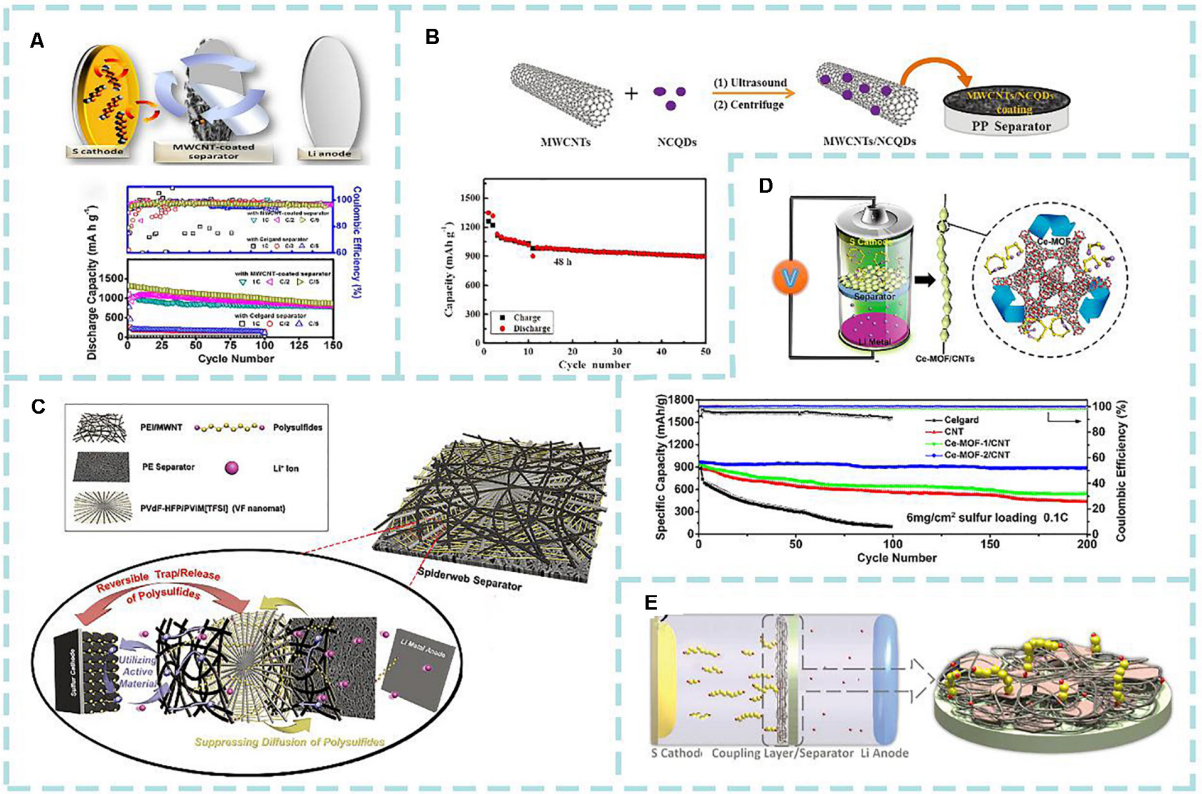

FIGURE 1 | (A) Mechanism of multi-walled carbon nanotubes (MWCNTs)-coated separator for Li–S batteries and the corresponding cycling performance; **(B)** Schematic diagram of MWCNTs/N-doped carbon quantum dots (MWCNTs/NCQDs)-coated separator preparation process and the cycle performance of the Li–S battery with MWCNTs/NCQDs-coated separator; **(C)** Schematic illustration of spiderweb separator and its application in Li–S batteries; **(D)** Scheme of Ce-MOFs/CNTs as coating materials in Li–S battery and the corresponding cyclic performance; **(E)** Schematic configuration of the Li–S cell with a Sb_2S_3/CNTs coated membrane. **(A)** Reproduced with permission. Chung and Manthiram (2014) Copyright 2014, American Chemical Society. **(B)** Reproduced with permission. Pang et al. (2018) Copyright 2018, Wiley-VCH. **(C)** Reproduced with permission. Lee Y.-H. et al. (2018) Copyright 2018, Wiley-VCH. **(D)** Reproduced with permission. Hong et al. (2019) Copyright 2019, American Chemical Society. **(E)** Reproduced with permission. Yao S. et al. (2018) Copyright 2018, Wiley-VCH.

to have the characteristics of strong chemical and/or physical adsorption toward polysulfides, good container for polysulfides and good electronic conductor. Also, the interlayer should allow smooth Li-ion diffusion. Therefore, with the insertion of interlayers, the LiPSs could be blocked at the cathode side, and the Li^+ diffusion and electron tranfer could also be promoted (Huang et al., 2016).

Pure MWCNTs

Because of intrinsic interweaving properties of CNTs, it is easy to prepare freestanding CNT interlayers with good flexibility and high porosity. Manthiram (Su and Manthiram, 2012) firstly fabricated a freestanding MWCNTs membrane by ultrasonic dispersion of synthesized MWCNTs, followed by a facile vacuum filtration (**Supplementary Figure S1A**). With the insertion of this freestanding interlayer, interfacial resistance in cathode was greatly reduced, and the intermediate polysulfides was efficiently localized at cathode side. The enhanced cycling performance of Li–S battery with MWCNTs interlayers could be ascribed that MWCNTs can prevent the undesirable migration and diffusion of polysulfides (Su and Manthiram, 2012). After that, many other interlayers based on MWCNTs have been explored for Li–S batteries (Lee and Kim, 2015; Sun et al., 2016). For instance, Kim H.M. et al. (2016) prepared a self-assembled MWCNTs membrane by mixing MWCNTs and electrolyte, followed by being applied to the top of sulfur electrode (**Supplementary Figure S1B**). The self-assembled MWCNTs membrane has more tightly interwoven structure and denser than bare MWCNTs interlayer. When tested in Li–S batteries, after 100 cycles at 0.5 C, the surface of the self-assembled MWCNTs interlayer only has tiny pores and more densely packed shape than that of bare MWCNTs interlayer. The enhanced electrochemical performance of the cell with self-assembled MWCNTs interlayer could be attributed to efficiently hinder polysulfide diffusion and enhance sulfur utilization through supplying polysulfide capturing sites (Kim H.M. et al., 2016).

MWCNTs-Based Composites

Owing to the abundance of functional groups, large specific surface area, and excellent electrical conductivity, graphene has attracted more interest in energy storage materials fields

(Wu et al., 2019b; Ma et al., 2020c; Sui et al., 2020). In Li–S batteries, many researchers combined graphene with MWCNTs as barrier materials to inhibit polysulfides migration (Sun et al., 2018; Shi et al., 2019). For instance, Huang et al. synthesized a freestanding graphene oxide/MWCNTs (GO/MWCNTs) film through combining MWCNTs with GO sheets (Huang et al., 2016), which exhibited high electronic conductivity and strong ability to immobilize LiPSs in Li–S batteries, resulting in improved electrochemical performance. The cell with GO/MWCNTs depicted an outstanding initial specific capacity of 1370 mAh g^{-1} at 0.1 C. After 300 cycles at 0.2 C, it still maintained a discharge capacity of 671 mAh g^{-1}, and the corresponding capacity decay rate was about 0.043% per cycle. More recently, Shi et al. (2019) reported a three-dimensional graphene/multi-walled carbon nanotube aerogels (G/MWCNTs) as self-supporting interlayer to improve the performance of Li–S cells. As displayed in **Supplementary Figure S1D**, this 3D G/MWCNTs aerogels with a 3D interconnected porous network were fabricated by the rapid reduction of graphene oxide/MWCNTs aerogel via self-propagating combustion. Cross-linked MWCNTs with graphene can give the membrane good mechanical flexibility and abundant pores, so that it can prevent the damage of interlayer during battery assembly and has more active sites to anchor polysulfides by chemical interaction and physical confinement.

In addition to combining with graphene, some researchers also combined MWCNTs with polar materials such as metal oxides (Xu G. et al., 2015; Kong W. et al., 2017; Kong et al., 2018; Li et al., 2019b), metal sulfides (Li et al., 2018; Wang L. et al., 2018), and metal catalyst (Cho et al., 2019; Ding et al., 2019), which could have produced strong chemical interactions with polysulfides. For instance, Wang L. et al. (2018) integrated a multicomponent sandwich structure interlayer as freestanding interlayer inserting between the sulfur cathode and separator, and the Li–S cell has been displayed outstanding electrochemical performance. In the structure, CNF is used as a scaffold to support CNT and vanadium disulfide compounds (VS_2/CNT), and covered a graphene layer onto it in the end. As shown in **Supplementary Figure S1C**, the CNT containing VS_2 had stronger adsorption ability toward polysulfides than pure CNT, suggesting that the polar V–S bonds in VS_2 promote the adsorption toward polysulfide. With the introduction of VS_2/CNT, the interlayer exhibited excellent synergetic effects to trap LiPSs and suppress the self-discharge effect (Wang L. et al., 2018).

SWCNTs

SWCNTs can be regarded as an ideal scaffold to anchor polysulfides in Li–S batteries due to their relatively uniform diameters and excellent mechanical and electrical properties, which could allow fast electron transfer (Zhao et al., 2012; Fang et al., 2017). Many efforts have been devoted to SWCNTs-based materials as interlayer for Li–S batteries (Kaiser et al., 2015; Chang et al., 2016; Hao J. et al., 2019). For example, Kaiser et al. (2015) synthesized a refined and interwoven structured SWCNTs freestanding film as the barrier layer, and inserted it between sulfur cathode and separator, which could effectively restrain polysulfides, enhancing the electrochemical performance of Li–S batteries.

Later, Kim J.H. et al. (2016) fabricated a polymer-coated single-wall carbon nanotube membrane (PAA-SWCNTs) as functional interlayer, which could efficiently restrain the diffusion of polysulfides via physical blocking of SWCNTs scaffold and hydrogen-bonding interaction of PAA. Moreover, SWCNTs films can act as second current collector to smooth the transfer of electron and Li^+. As a result, due to the synergic effect of PAA-SWCNT interlayer, the assembled cell exhibited an initial capacity of 770 mAh g^{-1}, it still retained a capacity of 573 mAh g^{-1} after 200 cycles at 1 C.

Apart from combined polymer with SWCNT, there are also researchers combined rGO with SWCNT as intermediate layer to anchor polysulfides (Hao J. et al., 2019). The composite membrane not only improve the internal electronic conductivity but also efficiently anchor polysulfides, resulting in an improved performance in Li–S batteries.

CONCLUSION AND OUTLOOK

In summary, we briefly review the recent development of CNT-based materials, including heteroatom-doped CNTs, carbonaceous materials/CNT composites, and metal-based materials/

CNT composites, as interlayers for Li–S batteries. Their nano/microstructure and electrochemical performances are intensively discussed. As we can see, even though the electrochemical properties of Li–S batteries with CNT-based interlayer have been significantly improved, several problems still exist and need to be addressed before practical application of Li–S batteries.

1. As mentioned above, CNTs-based materials are beneficial to immobilize the LiPSs when it is designed as an intercept layer, however, it should be noted that these materials could just relieve the shuttle effect to a certain degree physically or chemically, and the active materials still be possible to shuttle to the anode during the cycling. Moreover, the sulfur loading is normally about 1–2 mg cm^{-2} in most of above-mentioned work, and the highest loading does not exceed 10 mg cm^{-2}. With gradually increased sulfur loading, despite the existence of intercept layers, the issues of active material loss and polysulfide shuttling are severe. Hence, it is imperative to develop new materials which have strong binding energy and interactions with polysulfides.
2. The mechanism of CNTs-based materials for immobilization and catalytic conversion of LiPSs and their phase transformation during cycling are not clear enough, and still need to be explored. In-operando techniques, such as *in situ* electron microscopy, and *in situ* X-ray

technique, as well as synchron X-ray based techniques, are very helpful to obtain time dependent information. So, more research using in-operando techniques need to conduct to get insight understanding of the CNTs-based materials as interlayers for Li–S batteries.

3. Another issue to be tackled for practical application is lithium metal anode for lithium–sulfur batteries. There is always a problem in battery systems in which lithium metal exists: the formation and growth of lithium dendrites and the resulting low Coulombic efficiency and volume changes (Wang Y. et al., 2018; Wang G. et al., 2020; Zhao Q. et al., 2019). Therefore, in order to obtain a stable and safe lithium anode, a suitable electrolyte additive can be used. At present, the research on electrolyte additives has been highly advanced. Further, it is also possible to insert an artificial layer on the side of the lithium anode (Hao X. et al., 2019; Wang G. et al., 2020). However, there is little research on application of interlayers of CNTs-based composites in anodes, and this area requires further effort.
4. The reported literatures on CNTs-based materials coating layer/freestanding interlayers have developed many high-loading sulfur cathodes with excellent performances. However, the introduction of a coating/freestanding interlayer will increase the inactive weight ratio of the whole battery. Therefore, the direction to balance the relationship between the introduced CNTs-based interlayers and the total energy densities of batteries will be very important in the development of high-energy Li–S batteries.

AUTHOR CONTRIBUTIONS

YL and FR conceived the idea. HW and YL wrote the draft. All authors contributed to the writing, discussion, and revision of the final version of the manuscript.

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SUPPLEMENTARY MATERIAL

The Supplementary Material for this article can be found online at: https://www.frontiersin.org/articles/10.3389/fenrg.2020.585795/full#supplementary-material

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