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Two-Dimensional Materials to Address the Lithium Battery Challenges

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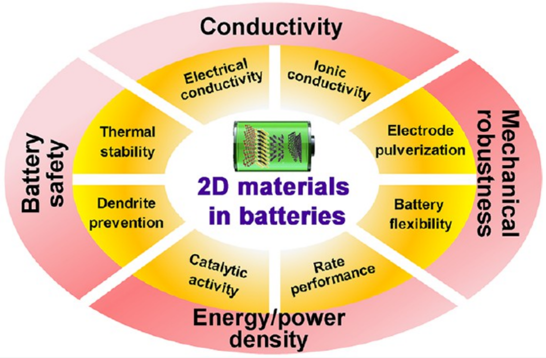

ABSTRACT: Despite the ever-growing demand in safe and high power/energy density of Li^+ ion and Li metal rechargeable batteries (LIBs), materials-related challenges are responsible for the majority of performance degradation in such batteries. These challenges include electrochemically induced phase transformations, repeated volume expansion and stress concentrations at interfaces, poor electrical and mechanical properties, low ionic conductivity, dendritic growth of Li, oxygen release and transition metal dissolution of cathodes, polysulfide shuttling in Li−sulfur batteries, and poor reversibility of lithium peroxide/superoxide products in Li−O_2 batteries. Owing to compelling physicochemical and structural properties, in recent years two-dimensional (2D) materials have emerged as promising candidates to address the challenges in LIBs. This Review highlights the cutting-edge advances of LIBs by using 2D materials as cathodes, anodes, separators, catalysts, current collectors, and electrolytes. It is shown that 2D materials can protect the electrode materials from pulverization, improve the synergy of Li^+ ion deposition, facilitate Li^+ ion flux through electrolyte and electrode/electrolyte interfaces, enhance thermal stability, block the lithium polysulfide species, and facilitate the formation/decomposition of Li−O_2 discharge products. This work facilitates the design of safe Li batteries with high energy and power density by using 2D materials.

KEYWORDS: *lithium batteries, two-dimensional materials, fast diffusion properties, energy storage, solid-state battery, rapid charging, thermal safety, dendrite prevention*

In light of expanding demands for energy storage devices to empower a wide range of applications from home-friendly appliances to transport systems and stationary storage grids, battery technology needs to set the pace to justify the supply. The key to achieving this goal lies in the fundamental understanding of a rechargeable battery electrochemistry. A typical Li battery (LIB) is composed of several stacked cells. Each consists of an electrolyte sandwiched between the two electrodes which undergoes a series of electrochemical reactions to introduce energy to the system. These redox reactions occur due to the difference of electrochemical potential of the cathode and anode.[1] Controlling the redox reactions plays a significant role to acquire high electrochemical performance without side reactions for long cycle life. Therefore, the choice of elements or compounds is very critical.

In search for more reliable materials to deliver higher energy storage capabilities, two-dimensional materials, known as 2D materials, have shown a growing interest in this field. The distinct structural properties and high specific surface area of this class of materials reduce the ion diffusion distance and promote fast surface redox reactions.[2,3]

The structural architectures of 2D materials, *e.g.*, planar, buckling, and puckered structures, could play an important role in introducing facile ion transport channels, leading to high ion conductivity.[4] This ability could be of a great interest since it potentially allows the support of high current densities without chemical and mechanical instabilities. Expectedly, 2D materials advocate directed diffusion properties and lower the ion mobility barriers for ultrafast ion transport due to their superionic conduction properties.[5] Moreover, these materials have shown outstanding mechanical properties, such as high Young's modulus and flexibility in comparison to their bulk counterparts.[6] The ability to endure high stress and strain

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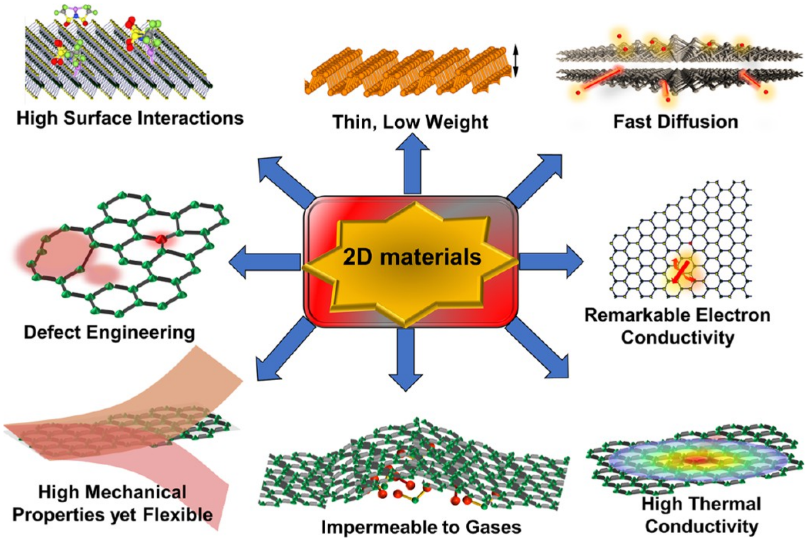

Figure 1. Some of the highlighted merits of 2D materials. 2D materials can introduce versatile properties in energy storage devices.

without structural collapse allows the application of 2D materials as mechanical supportive networks for high-capacity electrode materials.[7−9] Therefore, 2D material coatings on electrodes and separators can obstruct the Li metal dendrite formation and propagation very effectively.[10,11] Moreover, they have the advantage of band gap tunability and manipulating physical properties based on the applications.[12−14] This intrinsic advantage of 2D materials gives the opportunity to either enhance their electrical conductivity to be used as a cathode,[15] anode,[16,17] or current collector[18] or turning to the electrical insulative nature for separator[19,20] and electrolyte[21,22] applications. Studies on 2D materials are also expanded to their application as gas barrier membranes.[23−25] 2D materials such as graphene (G) and hexagonal boron nitride nanosheets (BNNs) have shown promising results for their use as impermeable gas coatings and films.[26,27] Consequently, these robust coatings potentially can address the environmental sustainability and safety issues in energy storage systems.[28,29] 2D materials can also offer great thermal management applications due to their intrinsic thermal conductivity.[30] Owing to the anisotropic thermal conductive framework, 2D materials such as G and BNNs have been used as efficient heat spreaders to mitigate the generated heat in LIBs.[31] Figure 1 depicts some of the abovementioned merits of 2D materials. On the other hand, this class of materials has shown promising corrosion resistance properties.[32,33] Hence, using 2D materials as corrosion protective coatings on electrodes or developing them as self-standing current collectors is increasing the focus of attention for LIBs.[34,35] These properties have led the energy storage community to perform intensive research on 2D materials to address the existing challenges of electrodes and electrolytes in rechargeable batteries. Table 1 summarizes the mostly used 2D materials in LIBs. These materials can be classified to graphene and graphene analogues, chalcogenides, transition metal oxides/hydrides, MXene materials, and elemental 2D materials.[36−39]

Table 1. Commonly Studied 2D Materials for Li Batteries

	category	2D material
1	graphene and graphene analogues	G,[a,40−42] FG,[b,43] rGO,[c,44−47] GO,[d,46,48,49] BNNS,[e,50] BCN[f51]
2	chalcogenides	GaS,[52] GaSe,[52] CoSe,[53] MoN,[54] NiS,[55] NiSe,[55] SnSe,[56] GeSe,[56] MoS_2,[57−59] WSe_2,[60] WS_2,[60,61] TiS_2,[62] SnS_2,[63] VS2,[64] $MoSe_2$,[65] $NbSe_2$,[66] Bi_2Se_3,[67] $MoTe_2$[68]
3	metal oxides/hydrides	MnO_2,[69,70] V_2O_5,[71−73] TiO_2,[74,75] Co_3O_4,[76] Bi_2O_3,[77] MoO_3,[78] VO_2,[79] WO_3,[80] SnO_2,[80] NiO,[81] $Ni(OH)_2$[82]
4	MXene	Ti_3C_2,[83] Ti_2C,[84] TaC_2,[85] Nb_2C,[86] V_2C,[87] Ti_3CN,[88] Ti_2N[89]
5	monoelemental	P,[90,91] Si,[92,93] Ge,[93] B,[94] Sb[95]

[a] Graphene. [b] Fluorinated-reduced graphene oxide. [c] Reduced graphene oxide. [d] Graphene oxide. [e] Hexagonal boron nitride nanosheets. [f] Hexagonal boron carbon nitride nanosheets.

In this Review, we address the current challenges of the Li battery materials and highlight the recent advances to overcome these limitations by using 2D materials. We discuss what types of 2D materials are appropriate for the LIB challenges and what progress has been made in that regard so far. The role of 2D materials on improving the performance of LIBs can be discussed from different viewpoints. 2D materials can enhance electrical conductivity and subsequently improve the electrochemical properties. These materials can be used as electrodes as well, such as cathode and anode with the goal to improve the cyclability and volumetric and gravimetric capacities. Other improvements can be seen in enhancing the mechanical stability and avoiding pulverization of high-capacity electrodes. Potential uses of 2D materials in trapping the polysulfides and tuning the discharge/charge products of Li−O_2 batteries are also discussed in this Review. 2D materials also can be engineered to achieve uniform Li^+ ion electrodeposition and prevent dendrite formation. Thermal safety of batteries is another important design consideration that is shown to be improved by the proper choice of 2D materials. In addition, 2D material contributions as current collectors are important to increase the energy density and to enhance the lifetime of LIBs. Moreover, the state-of-the-art of 2D materials as electrolyte additives and their role on boosting ionic conductivity and electrochemical stability of the electrolytes are comprehensively reviewed. In the last section, we summarize the current advances and discuss the future potentials of 2D materials in Li batteries.

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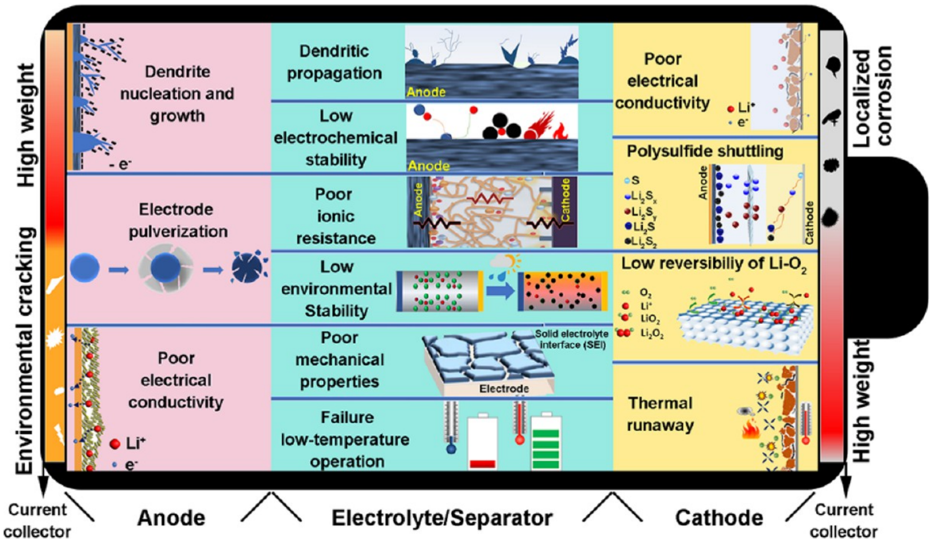

Figure 2. Common challenges in LIBs. Schematic showing the typical challenges for current collectors, cathodes, anodes, separators, and electrolytes of LIBs.

CHALLENGES OF BATTERY MATERIALS

A group of several electrochemical galvanic cells which can store and provide energy/power is defined as a battery. Typically, the battery components include electrodes (cathode, anode), separator, electrolyte, and current collector. Over the past few decades, a wide variety of primary (single use) and secondary (rechargeable) batteries have been developed. The LIB is an important class of secondary batteries which involve shuttling of Li$^+$ ions between the anode and cathode through the electrolyte.[96] The resulting electrons are transported through electrically conducting current collectors while the battery is in use.[97] Although there have been growing research studies on LIBs, major material-related issues have limited their utilization in high energy density and high-performance commercial applications. In this review, these material challenges are summarized based on material specifics to electrodes, electrolytes, separators, catalysts, and current collectors in LIBs. The conventional electrode materials suffer from poor electrical properties which leads to high polarization and low electrochemical performance. High-capacity electrode materials undergo major irreversible volume, structural, and compositional changes, leading to huge capacity loss and low electrochemical performance. Sulfur-based materials involve the formation and migration of insoluble and insulative polysulfides to the anode interface which results in the decaying performance in the course of few charge−discharge cycles.[98−101] In addition, cycling Li−O$_2$ batteries results in the deposition of electrically insulative products on a porous cathode which blocks the oxygen flow and leads to poor reversibility and high voltage polarization. On the other hand, formation of Li metal dendrites and nonuniform electrodeposition of Li$^+$ ions lead to fast capacity decay. Low cycle-life of the current LIBs is also a consequence of the limited ability of separators to block the dendrites and parasitic reaction products. In addition, the delamination of active materials from metal foil current collectors decreases the capacity of the batteries. These metal foil substrates increase the overall weight of the batteries which affects the production and operation expenses and capabilities. Due to chemical and physical instabilities of electrode and electrolyte materials, batteries often fail to operate at extreme environmental conditions such as elevated temperature and high moisture levels, which raise crucial safety concerns. Low ionic conductivity and poor diffusion properties of solid electrolytes are among other major impediments in commercializing all-solid-state batteries. Figure 2 summarizes the typical challenges in LIB components. These challenges are discussed in the following section.

Cathode materials are important initiative component of LIBs. Cathode materials' challenges such as limited specific capacity, electrical conductivity, mechanical properties, and electrochemical stability of such active materials play as pivotal impediments in the electrochemical performance of a cell.[102−104] Cathode materials such as nickel−manganese−cobalt (NMC), lithium−cobalt-oxide (LCO), lithium−iron-phosphate (LFP), and lithium−manganese spinel (LMO)[105,106] are among the mostly used cathodes in commercial applications. Conventional cathode materials often offer significantly lower specific capacities than anodes. For instance, graphite has a theoretical capacity of 372 mA h/g which is higher than that of the typical cathodes with a capacity of 150−200 mA h/g. Therefore, cathode materials pose a major limitation in the overall capacity of a cell.[107] Moreover, these materials have limited applications in developing robust energy storage devices and compatibility to extreme workload conditions.[107,108] Unfortunately, these developed electrodes often suffer from low thermal stability and fast capacity decay at high current densities or during deep cycling.[109] Therefore, cathode materials with higher specific capacity, improved safety, enhanced rate capability, and wide working voltage to improve the energy and power densities, reducing the size and cost of LIBs, are in the focus of attention.

The anode is another key component in LIBs, and even though significant efforts have been dedicated to develop high-capacity and stable anode materials, there are still significant issues in the utilization of these materials.[110,111] The Li metal anode with a high theoretical capacity of 3860 mA h/g and low potential of reduction of −3.04 V vs standard hydrogen electrode (SHE) is extremely reactive and is prone to dendrite formation and short circuit upon cycling.[112] Alternatively, graphite is the most used anode material with a moderate theoretical capacity of 372 mA h/g and low Li^+ ion intercalation potential of 0−0.3 V vs Li/Li^+.[113,114] Nevertheless, this limited capacity restricts improving the energy and power density of LIBs. On the other hand, emerging high-capacity alloying and conversion anodes pose several safety and electrochemical instability issues. Alloying anodes such as silicon (Si)- and germanium (Ge)-based materials, and conversion anodes like transition metal compounds, often have poor electrical conductivity and undergo *ca.* 200−500 % expansion and shrinkage during de/lithiation, leading to electrode pulverization and significant capacity loss.[115−119]

As far as the safety is concerned, separators play an important role to prevent the electrical short-circuit between the cathode and anode. Separators are often made of organic and inorganic materials, and due to their high controlled porosity can intake liquid electrolytes in their structure as ion conducting media.[120−123] These separators are fabricated from organic materials as well as inorganic materials. Polyolefin materials including polyethylene (PE) and polypropylene (PP) are among major commercial organic separators that have been used for LIBs.[124] However, poor wettability, low mechanical properties, and low thermal integrity are the major bottlenecks of such separators. The inorganic glass wool separator, on the other hand, can settle the above-mentioned challenges, but these separators often have high thickness and nonuniform porosity which can lead to premature cell performance fading.[125,126] The recent generation of these separators is also responsible for providing ion conducting networks and improving ionic conductivity, therefore improving the overall electrochemical performance.[127−129] Although there are good advancements in this regard, a promising electrolyte separator to endure extreme workload conditions and introduce superior electrochemical performance is still under investigation.

Current collectors are another important component of the LIBs. Conventionally, copper (Cu) and aluminum (Al) foils are used to support anode and cathode active materials, respectively,[130] though these metal foils are susceptible to localized corrosion and environmental cracking.[131,132] In addition, considering that these foils are inactive materials and increase the weight and cost of the battery production, further engineered techniques are required to manage this challenge.

On the other hand, developing compatible electrolytes to address high safety and prolonged cycle stability is inevitable. Conventionally, liquid electrolytes have been used in commercial batteries. However, the organic and aqueous liquid electrolytes often suffer safety and environmental issues and have a limited voltage stability window and low cationic transference capabilities. These electrolytes pose significant challenges in high-energy-density batteries due to flammability, lithium dendrite growth, and unfavorable side reactions.[48,133−135] Over the past two decades, a variety of solid-state electrolytes were introduced to fill this gap.[136] From the poly(ethylene oxide) (PEO)-based system,[137] poly(acrylonitrile) (PAN),[138−140] poly(vinylidene fluoride) (PVDF),[122,141,142] and poly(methyl methacrylate) (PMMA)[141,143] as polymer electrolytes to Li^+ ion conductive ceramic materials such as lithium phosphorus oxynitride (LiPON),[144,145] garnet-structured electrolytes,[146−148] and LISICON-type materials[149,150] have been increasingly studied for the development of all-solid-state LIBs. Tremendous efforts have been made in improving the ionic conduction and enhancing the electrochemical performance of the electrolytes by adjusting chemical composition,[151−153] evolving electrolytes with specific molecular and structural architectures,[154−156] and developing multilayered hybrid electrolytes containing ceramic−polymer−liquid materials.[157−159] Although there have been great improvements in developing organic and inorganic solid-state electrolytes, the slow electrochemical kinetics of such materials in the electrolyte and at the electrode/electrolyte interfaces limit their cyclability and commercial applications.[160−164]

Lithium−sulfur (Li−S)-based batteries are among the most studied post-LIBs for their capability to increase the specific energy in recent years. Owing to electrochemical interactions of Li and sulfur, soluble products of Li_2S_8, Li_2S_6, Li_2S_4, Li_2S_3, and insoluble Li_2S/Li_2S_2 lithium polysulfides form during cycling.[165−167] However, shuttling of dissolution products causes side reactions at the electrode surfaces and depletion of electrolytes, resulting in an irreversible capacity loss in these batteries.[98−101,168,169] Moreover, the low mechanical stability of the solid electrolyte interphase (SEI) layer at the anode downgrades the long-term cycling stability of Li−S batteries.[165]

Li−oxygen (Li−O_2) batteries are envisioned as the next-generation electrochemical energy devices owing to their superior theoretical energy density.[170−172] The Li−O_2 electrochemical system involves a series of redox reactions that leads to the formation of solid reaction products, dominantly the Li_2O_2 layer, on a porous-structured cathode.[173−175] However, Li_2O_2 is electrically insulative and blocks the effective Li^+ ion flux through the cathode pores, leading to fast capacity decay, low rate capability, high voltage polarization, and poor cycle stability.[176−178] Although considerable progress has been devoted in the development of Li−O_2 batteries, promising strategies to solve these ongoing challenges have not yet been realized.

2D MATERIALS TO BOOST THE ELECTRICAL CONDUCTIVITY OF THE ELECTRODES

High electrical conductivity of the electrode materials is of prime importance to achieve stable electrochemical properties. The majority of the electrode materials in LIBs have semiconductive properties and often suffer from low electrical conductivity. Poor electrical properties of cathode materials such as Li−Co−O (LCO), Li−Ni−Co−Mn−O (NCM), and Li−Fe−P−O (LFP) and anode materials such as Si- and Sn-based materials and $Li_4Ti_5O_{12}$ (LTO) lead to limited electrochemical stability, high polarization, and short-term cyclability.[179−181] In the search for electrode materials with better electrochemical properties, several 2D materials have been introduced to be great candidates as electrode or electrode additives.[38] 2D materials often have flexibility to tune the electrical properties and introduce higher gravimetric and volumetric capacities.[182] These materials can enhance the surface area of the electrodes and increase the availability of active material.[183] Graphene is the pioneering 2D material in this field due to its distinguished physical, chemical, and electrical properties.[184,185] High charge carrier mobility (200 000 $cm^2/(v\ s)$) and high electrical conductivity (ca. 10^6 S/cm) make it a great candidate as an anode active material.[186−188]

High electron transport in cathode materials plays an important role in the diffusion of Li^+ ions through electrodes, and it enhances the ion migration.[187] To this end, the family of carbon-based materials such as carbon black (CB), acetylene black (AB), carbon nanotube (CNT), graphite, and its 2D

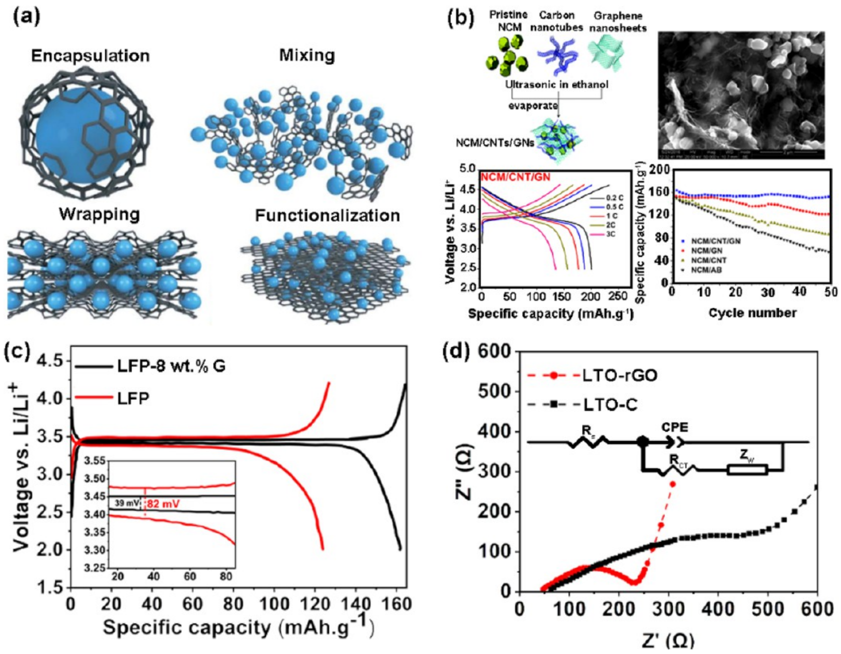

Figure 3. 2D material electrode additives to improve the electrical conductivity. (a) Schematic illustration of some of the typical structures of graphene-based composite electrode materials. Reprinted in part with permission from ref 189. Copyright 2017 Royal Society of Chemistry. (b) Schematic illustration of the NCM/CNT/GN hybrid cathode (top-left), and its corresponding SEM image (top-right) and voltage profile (bottom-left). The cycling performances of the NCM/CNT/GN, NCM/GN, NCM/CNT, and NCM/AB hybrids are compared at the 1 C rate. Reprinted in part with permission from ref 201. Copyright 2017 Royal Society of Chemistry. (c) Typical voltage profiles of LFPG-8 wt % G and LFP electrodes at a current density of 0.1 C. Reprinted in part with permission from ref 205. Copyright 2010 Elsevier Inc. (d) Nyquist plots for LTO-C and LTO-rGO samples. The equivalent circuit is shown in the inset. Reprinted in part with permission from ref 210. Copyright 2013 Elsevier Inc.

form of graphene have been widely used as electrode additives. Encapsulation, mixing, wrapping, and functionalizing the active materials with 2D materials is among the well-studied methods in reducing electrical resistance and improving Li^+ ion diffusion paths through the electroactive materials (Figure 3a).[189] It is worth noting that the addition of graphene-based frameworks has been shown to increase electrical contact and thus decrease the interfacial impedance of active electrode material particles and graphene additives.[190,191] A comparison study of LFP-5 wt % of various carbon materials showed that the graphene nanosheets tend to disperse more uniformly within the electrode matrix compared to AB and CNT. In addition, the LFP−graphene composite experienced a high diffusion coefficient of 5.9×10^{-9} cm^2/s and interface impedance value of ca. 130 Ω/cm^2. These values were shown to be roughly 150−300% times higher than those of the CNT and AB, respectively.[192] Although often 5−15 wt % of commercial carbon additives is required to enhance electrochemical properties of the target electrode material, it is shown that using only 0.2−5 wt % of graphene nanosheets could result in major improvements of electrochemical performance. This is related to the formation of long-range electron pathways and high electron conductivity of this type of 2D materials.[193−196]

Among the layered oxide cathodes, nickel-rich materials have been widely studied due to their high energy density and low cost.[197,198] However, this class of active material suffers from low electron transport properties and poor electrochemical stability.[199,200] To address these issues, Li *et al.*[201] studied the electrochemical performance of a developed hybrid NCM/carbon nanotube/graphene nanosheet (NCM/CNT/GN) as shown in Figure 3b. The NCM/CNT/GN hybrid material forms an electronic conducting network structure *via* the interconnection of one-dimensional CNT and two-dimensional GN. This three-dimensional hybrid cathode shows very low electrode polarization, enhanced rate capability with a high discharge capacity of 134 mA h/g at 3 C, and good capacity retention of ca. 93.8% compared to NCM/acetylene black (AB) composite electrodes. A similar study was performed by coating the NCM particles by SiO_2 and rGO single layers to improve structural stability, increasing Li^+ ion diffusion and electron conductivity.[202] $Li_3V_2(PO_4)_3$ (LVP) is another type of a cathode material that has large 3D channels for fast ionic conductivity and theoretically is expected to possess an excellent electrochemical performance at high current densities and long-cycle stability.[203] However, the low electrical conductivity of ca. 10^{-8} S/cm limits the usage of this cathode for high-power applications.[204] Jiang *et al.*[204] demonstrated that graphene-modified LVP composite not only improves the electrical conductivity but also can prevent the vanadium dissolution during Li^+ ion electrodeposition. It was reported that, upon addition of graphene-carbon to LVP, there would be about a 30% improvement in specific discharge capacity of *ca.* 110 mA h/g at the 5 C rate compared to conventional carbon (C) coating on these active material particles.[204] A similar electrochemical improvement has been reported for other electrode materials with low electrical performances upon adding graphene. Wang *et al.*[205] reported that adding 8 wt % of graphene nanosheets to LFP particles provides an effective conducting network that bridges Li^+ ions through the active materials. As shown in Figure 3c, the LFP-8 wt % G composite leads to lower voltage hysteresis values of 39 mV at a current density of 0.1 C whereas that of the bare LFP was 82 mV.[205] These graphene additives were found to significantly enhance the intergranular electrical conductivity

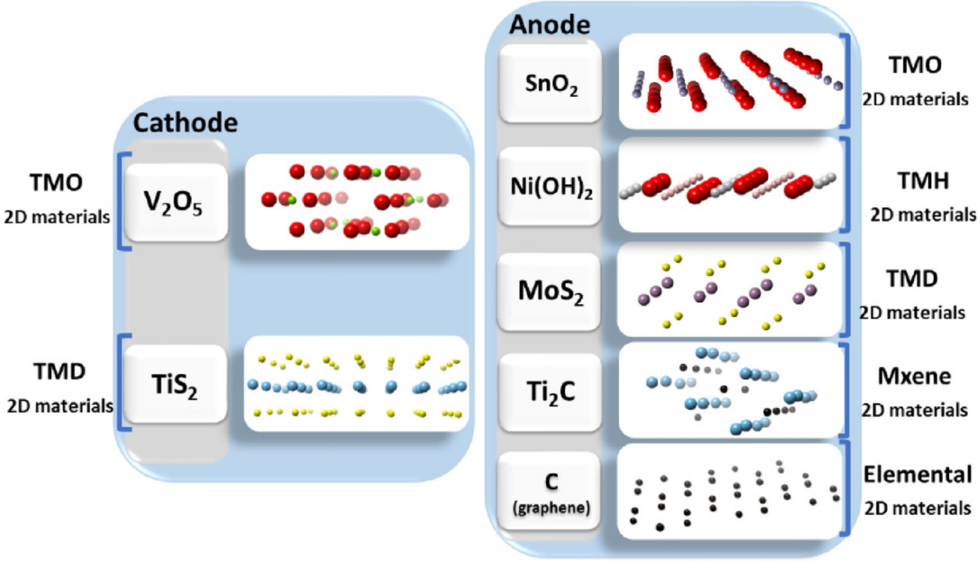

Figure 4. 2D materials as electrodes for Li batteries. Some examples of 2D materials are shown as cathodes or anodes, depending on their working potentials vs Li/Li$^+$.

and maintain the intact crystal structure of the orthorhombic LFP even at high current densities.[206]

Similarly, the improvement in electrical conductivity of anode materials can affect the electrochemical performance of the cell. For instance, due to low electrical conductivity of the spinel-LTO anode ($<10^{-13}$ S/cm at room temperature), this material has a poor rate capability, lithium storage capacity, and capacity retention.[207,208] There have been a number of studies for the composite LTO electrode material with two-dimensional reduced graphene oxide (rGO) nanosheets to address these issues. rGO additives shorten the Li$^+$ ion diffusion pathways and improve the rate performance and reversible capacity of LTO (124 mA h/g at a 20 C rate) by providing an electrically conducting network.[209,210] Figure 3d shows a comparison of the Nyquist plots for LTO-rGO and LTO-C composites. It has been shown that the value of charge transfer resistance (R_{ct}) is 450 and 230 Ω for LTO-C and LTO-rGO samples, respectively. The 50% reduction in R_{ct} of LTO-rGO indicates that there is more effective Li$^+$ ion and electron transportation through the electrode in charge–discharge processes by adding graphene. Other high-capacity anode materials such as Si-based and Sn-based suffer from poor electrical conductivity and electrical disconnection of the active materials due to pulverization of the particles.[211,212] Using graphene with such active materials offers efficient electrically conducting channels and improves their cyclability and large reversible capacity.[213,214] A similar strategy was also used to address the poor electrical conductivity of the other anode materials such as MnO_2,[215,216] MoS_2,[217,218] and phosphorus.[219,220] These graphene-based coatings provide an interconnected conductive network to link the active anode particles and result in an outstanding cycling stability and superior rate capability.

2D MATERIALS AS BATTERY ELECTRODES

Poor electrochemical properties of the conventional electrode materials have led to the design and implementation of 2D materials as battery electrodes.[37,221,222] Figure 4 shows some of the typical 2D materials that have been used as electrode materials in LIBs. Different atomic architectures of these materials, *e.g.*, planar, buckling, and puckered structures, play an important role in introducing facile ion and electron transport channels.[4] Graphene is among the most used 2D material anode materials with a low lithiation voltage of 0.29 V vs Li/Li$^+$ and noticeable specific capacity of >600 mA h/g.[186−188] Other elemental graphene analogue materials such as phosphorene, silicene, and germanene have been developed for high-capacity anode materials.[223] Although there are still some major challenges to practical applications of these 2D materials, some progress has been made to diminish their capacity decay. We have summarized some of these mitigation approaches in the following section. Other types of 2D materials such as 2D transition metal oxides (TMOs), transition metal hydroxides (TMHs), transition metal dichalcogenides (TMDs), and MXenes have been widely studied for their potential applications as LIB electrodes.[224] Reports have shown that the few layers of TMO/TMH materials such as 2D V_2O_5 and $Ni(OH)_2$ demonstrate a fast charging–discharging rate, high specific capacity, and small volume expansion during Li$^+$ intercalation due to the short diffusion paths and abundant intercalation sites.[225−227] TMD nanosheets with chemical composition of MX_2 (*e.g.*, M = Mo, W, Nb, Ta; X = S, Se) are among other intriguing 2D electrode materials and have shown great potential as the anode electrode in LIBs.[228,229] On the other hand, MXene materials or 2D transition metal carbides/nitrides have been demonstrated to introduce a good electrochemical performance in Li-based battery applications. Compounds such as M_2C (M= *e.g.*, Sc, Ti, V, and Cr) are among some of this large family and have shown a high discharge capacity of >400 mA h/g.[37,230] This class of 2D materials can provide a range of working potentials, which make them applicable for the choice of anodes or cathodes.[231] However, there is still some variation in reaching the maximum capacity of these compositions depending on the synthesis and processing approaches.[230]

2D MATERIALS TO PREVENT ELECTRODE PULVERIZATION

The pulverization of electrode materials due to volume expansion and stress accumulation during cycling is a major

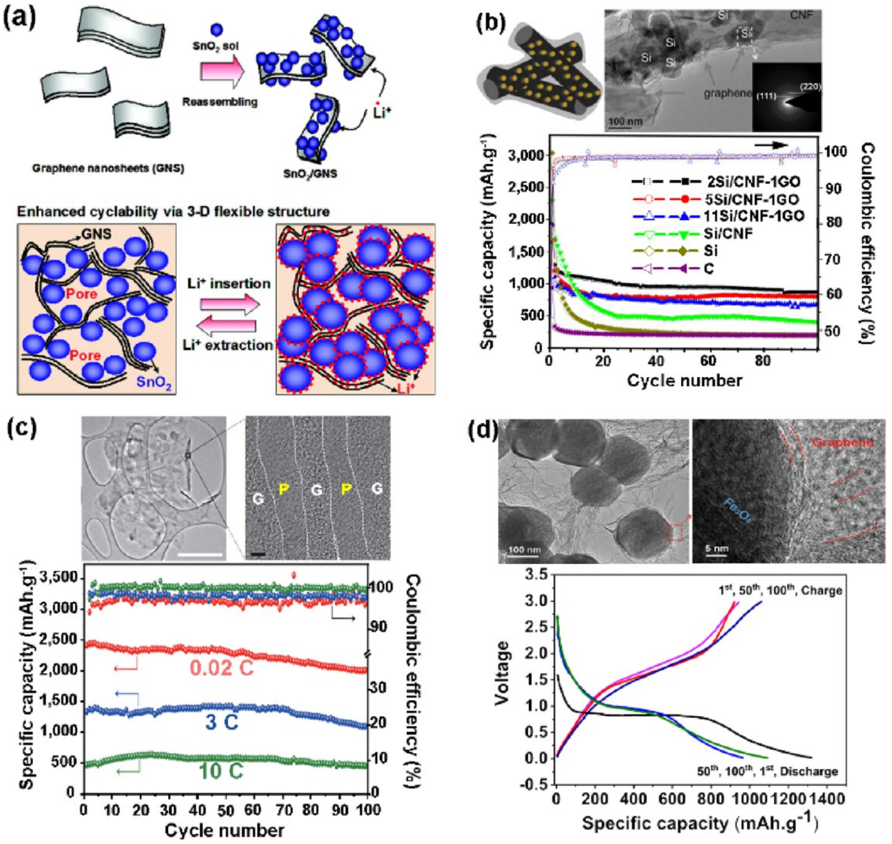

Figure 5. 2D materials to prevent electrode pulverization. (a) Schematic illustration for the synthesis and the structure of SnO_2/GNS. Reprinted in part with permission from ref 244. Copyright 2009 American Chemical Society. (b) Schematic illustration of the Si/CNF-G nanostructure and its corresponding TEM image (top). Cycling performance of the Si/CNFs, 2Si/CNF-1GO, 5Si/CNF-1GO, and 11Si/CNF-1GO electrodes at a current density of 100 mA/g (bottom). Reprinted in part with permission from ref 245. Copyright 2015 John Wiley and Sons. (c) TEM image of the phosphorene−graphene hybrid material and its corresponding cross-section (top). Reversible capacity and high Coulombic efficiency of the phosphorene−graphene anode at different current densities (bottom). Reprinted in part with permission from ref 246. Copyright 2015 Springer Nature. (d) TEM images of Fe_3O_4@GS/GF showing encapsulated particles in graphene shells and interconnected by graphene networks (top). Voltage profiles of Fe_3O_4@GS/GF at a current density of 93 mA/g (bottom). Reprinted in part with permission from ref 254. Copyright 2013 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim.

challenge for high-capacity electrodes, especially anode materials, and limits the cycle life and performance of LIBs.[232] Although the intercalation electrodes such as graphite and lithium-titanate spinel (LTO) introduce very stable electrochemical properties with very low-volume expansion upon de/lithiation (<7%), they have far lower theoretical capacity compared with other alloying or conversion-type active materials.[233,234] The high-specific-capacity group IV elements such as Si-, Ge-, and Sn-based electrodes are among the alloying-type anodes that have led to significant attention for their application in LIBs.[235,236] Si, Ge, and Sn can accommodate more than four Li^+ ions in their structure and have a theoretical capacity of 4200, 1625, and 994 mA h/g respectively.[237] However, these anode materials usually suffer from large volumetric variations (>300%) upon de/lithiation which leads to electrode pulverization, short cycle life, and considerably low capacity retention.[238]

Due to outstanding mechanical properties, a network of 2D materials encapsulating the electrode particles can prevent cracking and improve the electrochemical stability. Often 10−60 wt % of the weight of active materials has been used to boost mechanical integrity and electrochemical performance of the electrodes.[239−243] To this end, Paek *et al.*[244] prepared a composite of SnO_2 nanoparticles confined with graphene nanosheets (GNSs) as shown in Figure 5a. They have shown that the GNS confinement could improve the cyclability of the anodic half-cell of SnO_2/GNS to 570 mA h/g after 30 cycles which is significantly higher than the bare SnO_2 nanoparticle with 60 mA h/g after 15 cycles.[244] Although this method could mitigate the mechanical failure of these anodes, the performance could not reach to practical applications. Wang *et al.*[245] addressed the pulverization of Si nanoparticles during the cycling process by developing graphene-wrapped silicon/carbon nanofiber hybrids (Si/CNF-G nanostructure). Figure 5b shows the schematic illustration, TEM image of the Si/CNF-G nanosheet composite, and the corresponding cycling behavior at different ratios of silicon. This developed binder-free electrode showed a reversible capacity of 878 mA h/g over 100 cycles for the optimized sample (2Si/CNF-1GO), while the Si electrode can only retain a reversible discharge capacity of 200 mA h/g after 100 cycles. Black phosphorus and its 2D form nanosheets (known as phosphorene) are another alloying anode material that has attracted a great deal of attention due to its ultrahigh Li^+ ion mobility and high theoretical capacity of 2596 mA h/g for Li^+ ion storage.[90,246,247] However, it suffers from rapid capacity loss during cycling due to the large volume change similar to other high-capacity anode materials.[246,248]

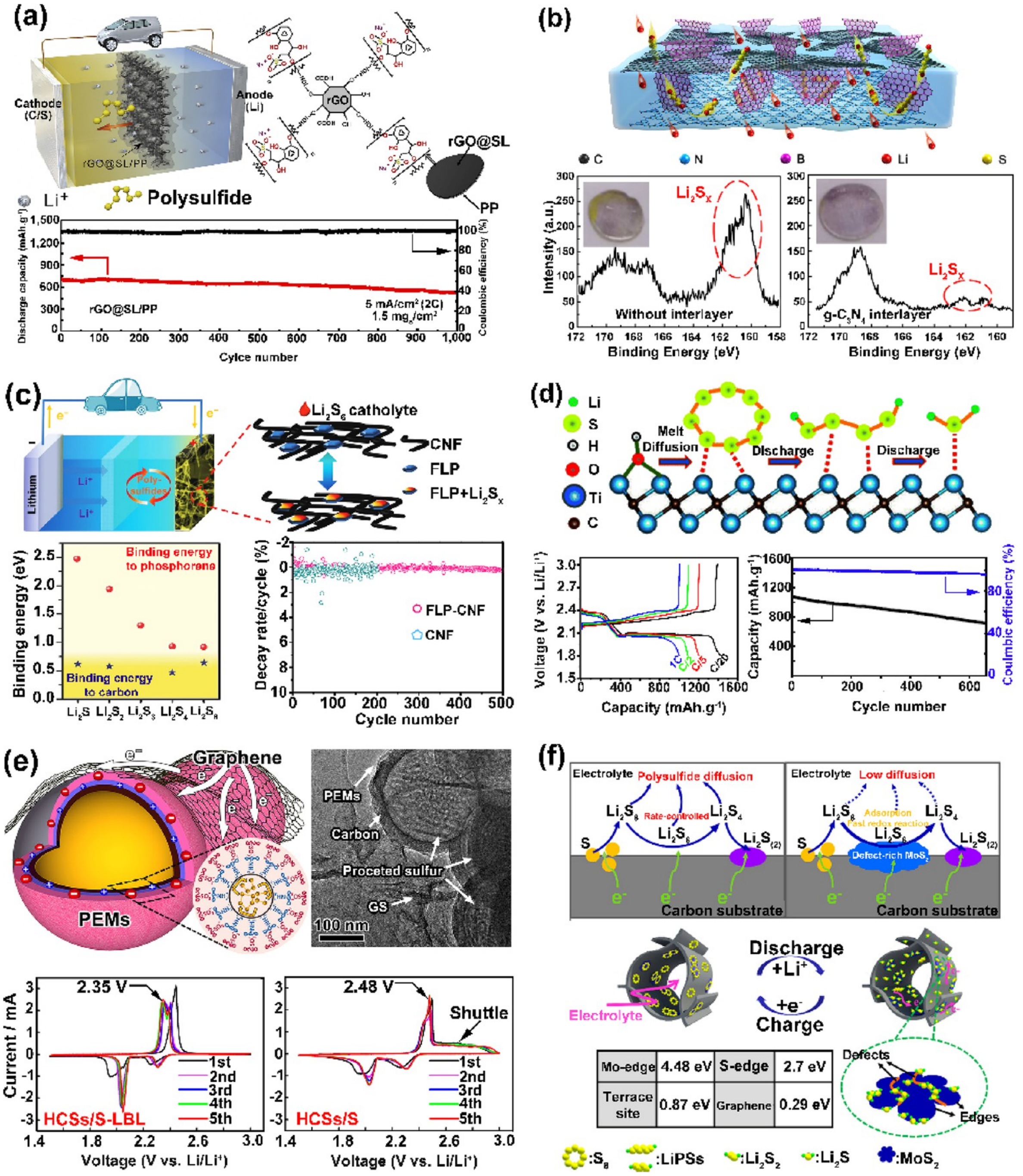

Figure 6. 2D materials to prevent shuttling of polysulfides. (a) Schematic illustrations of the rGO@SL/PP separators for inhibiting the polysulfide shuttling effect in Li−S batteries and its corresponding long-term cycling stability. Reprinted in part with permission from ref 260. Copyright 2018 Elsevier Inc. (b) Schematic illustration of the multifunctional ion-sieve constructed by three 2D materials of g-C_3N_4, BN, and graphene (top), XPS of S 2p of the Li metal anode after 100 cycles with and without the g-C_3N_4 interlayer coating on the separator, demonstrating the ability of this coating to block the shuttle of polysulfides (bottom). Reprinted in part with permission from ref 262. Copyright 2019 American Chemical Society. (c) Schematic illustration of the FLP-CNF composite cathode to accommodate lithium polysulfides (top). Binding energy between lithium polysulfides and phosphorene, and carbon hexatomic ring network (bottom left). Decay rate per cycle for Li−S batteries with the FLP-CNF electrode and pure CNF electrode (bottom right). Reprinted in part with permission from ref 91. Copyright 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (d) Schematic illustration of the replacement of the Ti−OH bond on the MXene surface with a S−Ti−C upon contact with polysulfides (top). Voltage profiles of 70 wt % S_d-Ti_2C at various rates ranging from 0.05 to 1 C (bottom-left). Long-term cycling of 70 wt % S_d-Ti_2C at 0.5 C (bottom-right). Reprinted in part with permission from ref 266. Copyright 2015 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (e) Schematic illustration (top-left) and TEM image (top-right) of the structure and functions of the HCSs/S-LBL composite including functionalized GS coating. Corresponding CV curves of the HCSs/S-LBL (bottom-left) and HCSs/S (bottom-right) composites. Reprinted in part with permission from ref 272. Copyright 2016 American Chemical Society. (f) Schematic illustration of the sulfur conversion reaction process on the carbon/S cathode and graphene surface with defect-rich MoS_2 nanosheets. The inset table represents the interactions between Li_2S molecule and different MoS_2 atomic site interactions. Reprinted in part with permission from ref 280. Copyright 2019 American Chemical Society.

In order to alleviate the poor electrochemical performance of phosphorene, Chowdhury *et al.*[249] proposed the use of hexagonal boron nitride nanosheets (BNNs) as a capping agent to improve the performance of phosphorene without perturbing its intrinsic high specific capacity. In this modeling study, it has been shown that BNN capping is able to improve the structural stability of phosphorene at ambient conditions and, also, reduce the volume change during de/lithiation to <2%.[249] Although the idea is very intriguing, adding an electrically insulating BNN layer may increase the concern of poor electrical conductivity of the developed electrode. Therefore, phosphorene−graphene hybrid material was used by Sun *et al.*[246] to address this challenge. They showed that this hybrid material with a few phosphorene layers sandwiched between graphene layers could reach a specific capacity of 2440 mA h/g at a current density of 50 mA/g (0.02 C) and an 83% capacity retention after 100 cycles for sodium ion batteries.[246] This developed anode also has shown 84% and 77% capacity retentions at relatively fast rates of 3 and 10 C after 100 cycles (Figure 5c). Although the main focus of this study was to develop phosphorene-based anode materials for Na^+ ion batteries, this methodology is expected to be applicable for LIBs as well.

Among the high-capacity electrode materials, conversion electrodes such as transition metal compounds (e.g., Fe_3O_4, MoS_2) undergo reversible redox reactions between Li^+ ions, resulting in high specific capacity and fast reaction kinetics.[250] Fe_3O_4 is a low-cost and ecofriendly material with high theoretical capacity of 924 mA h/g and has gained significant attention as Li-based battery anodes.[251] However, this anode material cannot endure large strains due to large volume expansion and leading to electrode pulverization and capacity loss similar to alloying materials.[250] Several strategies have been proposed to accommodate this volume change and improve the electrochemical properties of such materials. 2D graphitic carbon nanosheets have widely been used as a buffer layer to allow the Fe_3O_4 nanoparticles to expand upon lithiation without breaking the carbon shell and improve the electric conductivity of such electrodes.[252,253] Wei *et al.*[254] developed graphene encapsulated Fe_3O_4 nanosphere sheets and further confined within continuous graphene networks (Fe_3O_4 @GS/GF) as shown in Figure 5d. This homogeneously dispersed architecture provides a dual protection against the aggregation and volume changes and leads to a low Li^+ ion-transfer resistance and superior cycling performance of 1059 mA h/g over 150 cycles.[254] A similar study was performed by Luo *et al.*[255] where they used grafted mesoporous Fe_3O_4 nanoparticles onto three-dimensional graphene foams by atomic layer deposition. Using this approach, they synthesized a hybrid electrode that could provide a specific capacity of *ca.* 780 mA h/g at 1 C and *ca.* 350 mA h/g at 10 C up to 500 cycles.[255] In a recent work by Wang *et al.*,[256] $Fe_3O_4-Ti_3C_2$ MXene hybrid materials with different weight ratios were studied. They showed that the 2D layered structure of Ti_3C_2 could accommodate the volume change and provide fast transmission of Li^+ ion channels during cycling. In addition, their observation revealed that pulverization of the Fe_3O_4 nanoparticles can work in favor of the charge transfer kinetics in these composite electrodes. Basically, pulverization of the Fe_3O_4 nanoparticles improved the contact of these nanoparticles and the Ti_3C_2 MXene layers over cycles and led to higher Li^+ ion storage sites.[256]

2D MATERIALS TO PREVENT SHUTTLING OF POLYSULFIDES

Among the various studies that have been performed to address the Li−S issues, 2D materials were explored for their potential application as host electrodes, separator, and electrolytes.[257−259] Lei *et al.*[260] developed a negatively charged reduced graphene oxide (rGO)-lignosulfonate (SL) coating on a commercial polypropylene (PP) separator as shown in Figure 6a. This layer introduce a repulsive force to suppress the transport of the negatively charged polysulfide ions and therefore results in outstanding cycling stability, with a capacity retention of 74% over 1000 cycles at relatively high current densities of 5 mA/cm^2 (2 C).[260] A similar concept was followed by Kim *et al.*[261] to design the interphases composed of positively charged conductive nanoparticles of polyethylenimine attached rGO on the cathode facing side of the separator. Negatively charged nanoparticles of lithium terminated sulfonated TiO_2 were also applied on Li metal. These ionically shielding nanomaterial composites have been shown to stabilize Li surfaces and intercept the polysulfide shuttling.[261] To address poor electrochemical properties of Li−S batteries, Deng *et al.*[262] developed a multifunctional interlayer coating consisting of graphitic 2D materials ($g\text{-}C_3N_4$, boron nitride (BN), and graphene nanosheets) on the separator (Figure 6b). They have shown that the crystalline structure of $g\text{-}C_3N_4$ nanosheets with *ca.* 3 Å channel size blocks the polysulfides and only allows the Li^+ ions to travel freely. The XPS spectra of the separators with and without the $g\text{-}C_3N_4$ nanosheet interlayer coating after 100 cycles (Figure 6b) show the effectiveness of this interlayer in impeding the polysulfide shuttling. BN nanosheets provide excellent catalytic behavior for sulfur redox kinetics. Finally, the graphene additive promotes the electrical and ionic conductivity of the electrodes. As a result, the synergistic effect leads to a high specific capacity of *ca.* 600 mA h/g at 1 C after 500 cycles and capacity fading of <0.01% per cycle at high loading of 6 mg/cm^2 of sulfur.[262]

Li *et al.*[91] proposed the use of few layer phosphorene (FLP)-carbon nanofiber (CNF) composite material as a cathode host to address the poor performance of Li−S batteries as shown in Figure 6c. The idea of using phosphorene in the electrode design originated from the strong affinity of lithium polysulfides to these nanosheets. Figure 6c shows the binding energy of various lithium polysulfides to phosphorene (ranging from 1 to 2.5 eV), which is considerably greater than a carbon network (*ca.* 0.5 eV). As a result, phosphorene improves utilization of sulfur and lowers the polarization as an indication of enhanced electrochemical reaction dynamics in such batteries. In addition, phosphorene is demonstrated to have a catalysis effect on redox reactions of S/Li_2S and facilitates the conversion reactions. The cycling of the Li−S batteries with developed host materials has shown that the specific capacity of the battery with phosphorene is retained >660 mA h/g with a very low capacity attenuation of ≈0.053% over 500 cycles, which shows a much better performance than the battery without phosphorene (≈0.25% capacity decay in 200 cycles).[91]

Similarly, 2D transition metal dichalcogenides such as MoS_2 and WS_2 have been used to enhance the electrocatalytic activities for fast sulfur conversion reactions. Due to abundant edges, a specific chemical and electronic structure, and plentiful defects, accumulation of soluble polysulfides decreases

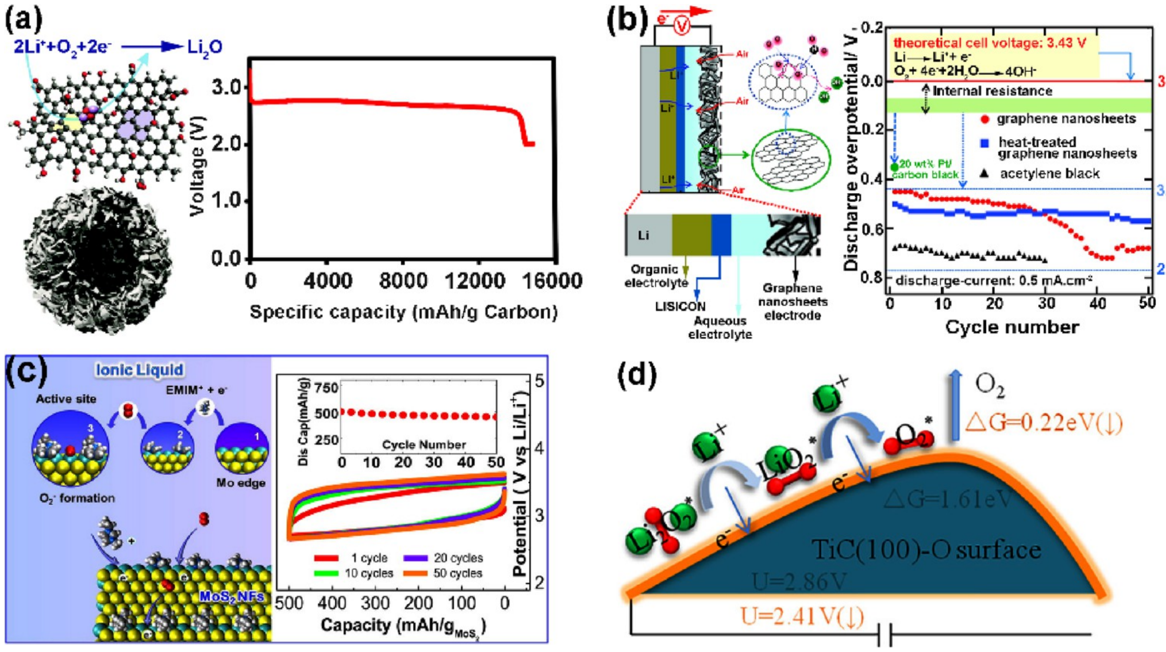

Figure 7. 2D materials in Li−O_2 batteries. (a) Schematic structure of an FGS (top-left) with a 3D network of FGS porous structure (bottom-left) and a typical discharge curve of a Li−O_2 cell using FGS as cathode (right). Reprinted in part with permission from ref 283. Copyright 2011 American Chemical Society. (b) Structure of the rechargeable Li−air battery with the porous GNS cathode (left), comparison of discharge overpotential for GNS, heat-treated GNS, and acetylene black as cathodes (right). Reprinted in part with permission from ref 286. Copyright 2011 American Chemical Society. (c) Schematic illustration of the oxygen reduction reaction on MoS_2 NFs in ionic liquid electrolyte (EMIM-BF4) (left) and voltage profiles of a battery using carbon-free MoS_2 NFs and an ionic liquid electrolyte (right). Reprinted in part with permission from ref 295. Copyright 2016 American Chemical Society. (d) Schematic of TiC(100)-O surface reactions for Li_2O_2 decomposition. Reprinted in part with permission from ref 307. Copyright 2018 American Chemical Society.

on the sulfur cathode upon adding these 2D materials.[257,263,264]

Another strategy to mitigate the irreversible dissolution and shuttle of polysulfides is to anchor the polysulfides onto the surface of 2D materials by creating functional groups and defect sites.[257,265] MXene 2D materials with functional groups on the surface are among the most studied 2D materials, which have been shown to be able to efficiently trap the polysulfides by chemical and physical adsorption.[257,266] Liang *et al.*[266] showed that the hydroxylated surface of Ti_2C 2D nanosheets reacts with sulfur and provides strong S−Ti−C bonding. As shown in Figure 6d, up to 70 wt % of sulfur was incorporated into the Ti_2C matrix by melt-diffusion and demonstrated an outstanding initial specific capacity of 1440 mA h/g at 0.05 C (83 mA/g) to *ca.* 1000 mA h/g at 1 C (1675 mA/g). In addition, this method led to a stable capacity retention of 723 mA h/g after 650 cycles at a high current density of 0.5 C (837 mA/g) with considerably low decay rate of 0.05% per cycle and Coulombic efficiency *ca.* 94%.[266]

Similarly, the C—H, —OH, >C=O, —C(=O)OH, and >C=C< functional groups that can usually appear on carbonaceous materials surfaces have been shown to provide a high affinity to bind with sulfur. Therefore, the surface modification of these materials increases the chemical adsorption of long —S_n— chains and form insoluble short-chain polysulfides. Although the formation of both short-chain and long-chain polysulfides is detrimental to electrochemical properties of Li−S batteries, it has been shown that fragmentation of polysulfide to shorter chains could mitigate the shuttling effect and enhance electrochemical properties.[267−271] To this end, Wu *et al.*[272] developed a layer-by-layer (LBL) self-assembly coating composed of polyelectrolyte multilayers (polyethylene imine (PEI)), polystyrene sulfonate (PSS), and functionalized graphene sheets (GSs) on the surface of hollow carbon spheres/sulfur (HCSs/S) composite particles as shown in Figure 6e. This study showed that functionalizing graphene sheets by hydroxyl groups improves the formation of LBL film on the HCSs/S composite, as well as acting as a blocking shield to confine the S_n^{2-} anions within the hollow carbon through strong electrostatic interactions. This method minimized the parasitic reactions at the sulfur cathode with outstanding reversibility. Figure 6e shows the CV plots of the HCSs/S and HCSs/S-LBL composites at a scan rate of 0.1 mV/s. Clearly, only the typical redox reaction peaks of the sulfur cathode were detected for HCSs/S-LBL composite with the first anodic peak at 2.35 V vs Li/Li^+. In the HCSs/S composite, these redox peaks showed much broader behavior and shifted to a higher voltage of 2.48 V vs Li/Li^+, indicating a higher polarization and increased internal resistance. In addition, the CV plot of the HCSs/S composite showed some activities with high anodic current between 2.5 and 3 V vs Li/Li^+, implying the polysulfide shuttle reactions in this unprotected C/S structure.[272]

Incorporating defective 2D materials into sulfur cathodes can prevent the polysulfides from disintegrating and have been widely investigated.[273−276] Experimental studies and density functional theory (DFT) calculations showed that the defect sites on 2D materials such as nitrogen-doping could prevent Li_2S_n diffusion and dissolution by introducing a strong synergistic effect on their adsorption.[277−279] Following the effect of structural defects in Li−S batteries, Liu *et al.*[280] performed a study on a composite of sulfur with three-dimensional porous graphene aerogel (GA) decorated with defect-rich MoS_2 nanosheets. As shown in Figure 6f, the abundant defects and edge sites of MoS_2 demonstrate higher binding energy with polysulfides, weaken their diffusion into the electrolyte, and act as an electrocatalyst to promote the redox reaction kinetics. Moreover, the polar surfaces of these MoS_2 dispersed GA provide a chemical anchoring effect to immobilize the lithium polysulfides.

2D MATERIALS IN LI–O_2 BATTERIES

Recent advances of using 2D materials as porous cathodes or catalysts provide a perspective on improving the cycling performance and voltage polarization in Li–O_2 batteries. Since the oxygen reduction reaction (ORR) and oxygen evolution reaction (OER) take place on the oxygen-induced cathode surface, the structure and morphology of these materials significantly affect the electrochemical properties of Li–O_2 batteries.[281,282] Graphene-based composites are among the most studied active materials to improve the rechargeability of Li–O_2 cells. Xiao *et al.*[283] developed a 3D network of functionalized graphene sheets (FGSs) with interconnected pore channels as the Li–O_2 cathode which is demonstrated in Figure 7a. The aggregation of the Li_2O_2 can be prevented owing to the presence of structural defects on FGSs. These defects, however, only allow the formation of isolated nanosized Li_2O_2 particles. Therefore, the hierarchical and contiguous pore structure of FGS led to a high discharge capacity of 15 000 mA h/g.[283] This structure can facilitate rapid O_2 diffusion and prevent the air blocking which mainly happens due to coagulation of byproducts on the porous cathode surface.[283−285]

Graphene nanosheets (GNSs) have also been reported to have electrocatalytic activity in the ORR and OER. Yoo *et al.*[286] reported an aqueous Li–air battery with stable discharge voltage of 3.00 V vs Li/Li^+ for heat-treated GNS and low overpotential of *ca.* 0.7 V vs Li/Li^+ which are nearly similar values to that of the 20 wt % Pt/carbon black. This confirms the exhibition of a high catalytic activity of heat-treated GNS which dissociates O_2 into atomic oxygen and forms hydroxide ions (OH^-) in the aqueous electrolyte.[286] Figure 7b summarizes this study. GNS has also been investigated as a catalyst or catalyst support for nonaqueous electrolytes.[287−290] Electrocatalytic activity of GNS was observed especially while carbon vacancies and defects are present on the surface as active sites for the ORR and OER.[291,292] Moreover, using the graphene-based host as a support to catalyst particles has shown promising electrochemical activities in Li–O_2 batteries. One of the recent works in this field is reported by Lu *et al.*,[293] whose reduced graphene oxide (rGO) decorated with iridium (Ir) nanoparticles was used as the cathode in Li–O_2 batteries. Owing to the catalytic activity of Ir particles on the rGO backbone, stable formation of lithium superoxide (LiO_2) and reversible cycling behavior with a very low charge overpotential were observed, although LiO_2 is considered to be an unstable intermediate of the ORR in Li–O_2 batteries.[293,294]

The potential of other 2D materials is also explored as cathode materials for rechargeable Li–O_2 batteries. To this end, Asadi *et al.*[295] investigated the ability of MoS_2 nanoflakes (MoS_2 NFs) for their high catalytic activity for both the ORR and OER in an ionic liquid (EMIM-BF_4). As shown in Figure 7c, O_2 strongly binds to negatively charged Mo edges which enables oxygen reduction. This eventually led to 85% round-trip efficiency and reversibility up to 50 cycles when the capacity was limited to 500 mA h/g.[295] This promising electrochemical performance of Li–O_2 batteries was also observed in other studies by benefiting the superior catalytic property of MoS_2 nanosheets.[296−298] Moreover, 2D nanosheets of metal–organic frameworks (MOFs) and covalent organic frameworks (COFs) have shown great potentials as Li–O_2 electrocatalysts. These materials are able to accelerate the OER and ORR through intrinsic open active sites and offer long lifespan batteries.[222,299,300] In a recent study, Yuan *et al.*[301] developed ultrathin 2D Mn–O frameworks as the cathode electrocatalyst in aprotic Li–O_2 batteries. This 2D MOF showed a specific discharge capacity of 9464 mA h/g and stable cyclability of >200 cycles at 100 mA/g at a limited discharge capacity of 1000 mA h/g. The ability of 2D MOFs for adsorbability to oxygen molecules, fast mass transport, and efficient decomposition of lithium oxide/hydroxide products is considered as responsible for such electrochemical performances.[301,302]

In pursuit of catalytic cathodes, the potential application of 2D materials beyond graphene for Li–O_2 batteries was investigated by computer simulation. Wang *et al.*[303] showed that the MXene-based 2D material $Ti_nC_{n-1}O_2$ ($n = 2, 3, 4$) can potentially improve the cyclability and lower the voltage polarization due to the excellent catalytic activity of this material. The theoretical studies show that the Li_2O_2 clusters have strong affinity to adsorb on Ti–O and Ti–C terminations of $Ti_nC_{n-1}O_2$, leading to uniform deposition of Li_2O_2 coating and facilitating reversible deposition and decomposition of the surface.[304−307] Figure 7d shows a schematic of TiC(100)-O surface reactions for Li_2O_2 decomposition. Meanwhile, some other 2D materials such as phosphorene-based nanosheets have been explored in Li–O_2 batteries for their high catalytic properties. The activation barrier of phosphorene for the catalytic decomposition of Li_2O_2 is 1.01 eV for phosphorene which is almost half of that of the graphene with the value of 2.06 eV.[308] Li *et al.*[309] evaluated phosphorene oxides for possible application as Li–O_2 battery cathodes with molecular dynamics (MD) simulations. MD simulation showed high diffusivity of Li^+ ions along the zigzag direction (diffusion barrier = 0.62 eV) compared to the armchair direction with the diffusion barrier = 1.22 eV. This offers a great potential for phosphorene-based cathodes for Li–O_2 batteries.

2D MATERIALS TO CONTROL LI^+ ION ELECTRODEPOSITION

Nonuniform lithium deposition/stripping is among the main issues which hinder the development of LIBs for practical high-power and high-energy applications.[310] Poor Li^+ ion transport properties at the electrode/electrolyte interface lead to an uneven charge distribution and high overpotential values during galvanostatic electrodeposition.[311] Consequently, lithium dendrites nucleate and grow in the battery and consume the electrolyte over parasitic reactions. Eventually, this may result in short circuit and catastrophic cell failure.[312] So far, the electrode/electrolyte interface remains a huge challenge due to high resistance, mechanical failure of interfacial contact, and electrolyte depletion, leading to low electrochemical performance.[313] Over the past decade, research studies introduced different physicochemical methods in designing electrolyte compositions, modifying separators, and electrode coatings to address these issues.[314] In particular, using 2D materials as electrolyte separators, surface protective films, and electrolyte additives has led battery designs to overcome the poor mechanical, thermal, and electrochemical properties of LIBs.

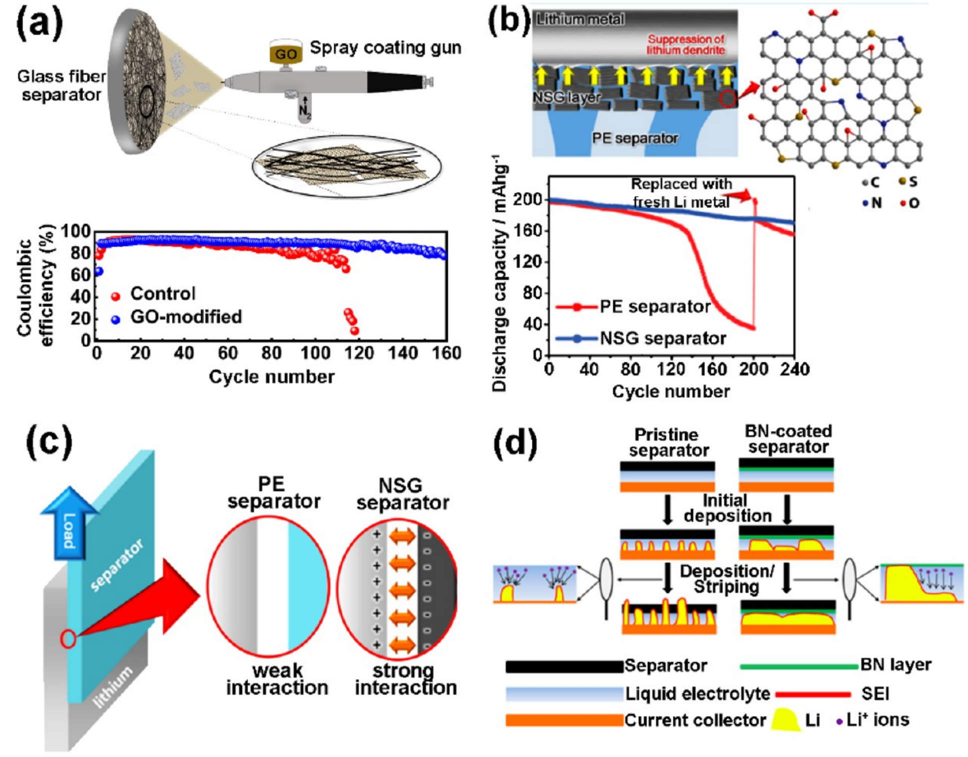

Figure 8. 2D material coating on separators to control the Li electrodeposition and dendrite behavior. (a) Schematic illustration of the GO spray coating on the glass fiber separator. The corresponding cycle life is shown compared to the bare separator as a control. Reprinted in part with permission from ref 48. Copyright 2018 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) Suppression of dendrite growth on the lithium metal electrode with the NSG-coated PE separator and its corresponding behavior on discharge capacity retention in comparison with the bare PE separator. Reprinted in part with permission from ref 317. Copyright 2015 American Chemical Society. (c) Interfacial interaction between the lithium electrode and PE separator with and without the NSG coating. Reprinted in part with permission from ref 317. Copyright 2015 American Chemical Society. (d) Li deposition/striping process with the pristine separator and BN-coated separator. Reprinted in part with permission from ref 319. Copyright 2015 American Chemical Society.

This development can substantially limit the side reactions and extend the performance and life of such batteries.

2D MATERIAL COATING ON SEPARATORS

Utilizing 2D materials as a protective coating on the electrolyte separators is among the techniques that is used for improving the electrochemical properties. 2D materials exhibit superior mechanical properties which pose a huge potential to suppress the dendrite formation and growth.[257,315] They can also act as Li^+ ion carriers which decrease the energy for Li^+ ion migration and, therefore, act as low-energy pathways for uniform Li^+ ion stripping/plating.[48,316] Depending on the type and synthesis of 2D material coatings, they can show different electrochemical behavior.

Graphene oxide (GO) is one of the most common 2D materials that has been used so far as a separator coating layer. Foroozan *et al.*[48] showed that, upon coating the glass fiber separator with GO, the voltage hysteresis of Li||Cu decreases in charge−discharge cycles and demonstrates an *ca.* 2-fold improvement in the cyclability of Li metal compared to the similar battery with a bare separator (Figure 8a). They realized that the diffusion of Li^+ ions through the GO coating mainly occurs as a result of existing discontinuities and defects on the GO structure. On the other hand, Shin *et al.*[317] deposited a nitrogen and sulfur codoped graphene (NSG) nanosheet coating on a polyethylene (PE) separator by vacuum infiltration as shown in Figure 8b,c. They have shown that the NSG nanosheets could effectively suppress the dendrite growth and provide a uniform ionic flux on the Li metal surface. These coatings were also able to decrease the Li^+ ion charge transfer resistance by 6-fold after 200 cycles compared to the similar condition with the PE separator. The reason lies with the line and point defects and structural deformations in the graphene framework which facilitate the ionic migration.[317,318] As a result of heteroatom doping, NSG nanosheets exhibit a negative charge throughout their surfaces due to lone-pair electrons which attract the Li^+ ions and decrease the interfacial resistance. Since the formation and growth of Li dendrites consume a large amount of electrolytes and can raise crucial safety concerns, one should find effective ways to mitigate this issue. Similar to GO and defective G coating on the electrolyte separators, Luo *et al.*[319] showed that the 2D h-BN nanostructure nanosheet (BNN) coating provides a uniform surface area at the electrode/electrolyte interface and minimizes the growth rate of Li dendrites (Figure 8d). Therefore, the electrolyte chemistry remains more stable, resulting in high Coulombic efficiency and cycle stability. Li dendrites are grown during initial Li deposition, gradually penetrate the typical commercial separator, and consume electrolyte due to their high active surface which leads to low Coulombic efficiency and, eventually, failure. However, BNN coating results in a uniform stripping/plating of Li^+ ions and, therefore, results in less dendritic nuclei and improves cycle performance especially at high current densities.[319]

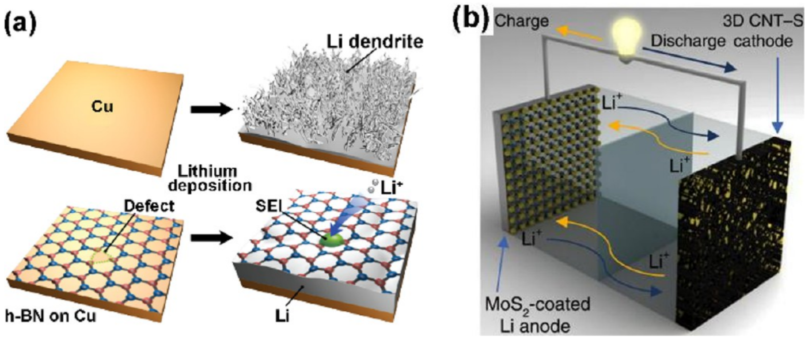

Figure 9. 2D material coating on metal anodes to control Li$^+$ ion electrodeposition and dendrite behavior. (a) Schematic illustration of Li metal deposition on a bare copper substrate with and without a defected 2D BNNS film on the surface. Reprinted in part with permission from ref 323. Copyright 2014 American Chemical Society. (b) Schematic of the Li–MoS$_2$/CNT–S cell configuration. MoS$_2$ provides efficient electron and ion channels as well as prevention of Li dendrite growth on the Li metal anode. Reprinted in part with permission from ref 326. Copyright 2018 Springer Nature.

2D MATERIAL COATING ON METAL ANODES

2D materials have been used on Li metal anodes to create artificially designed SEI (ASEI) coatings. Due to the highly reactive nature of Li metal, a layer consisting of the components of organic electrolyte evolves on the Li anode surface by electrochemical activities. This layer is also known as a solid electrolyte interphase (SEI) and can play an important role in preventing the growth of Li dendrites.[320,321] However, the thickness of the SEI film increases during cycling and is likely to break down and delaminate from the Li surface. The uneven SEI film increases the risk of electrolyte consumption and parasitic reactions.[322] One possible method to prevent such circumstances is to create ASEI coatings on the Li metal anodes. The goal of this layer is to create a stable ionically conductive coating on the Li metal anode to endure electrochemical reactions for hundreds of cycles. Therefore, the ASEI should form a protective layer with great flexible elastomeric properties that can block the Li dendrites during the deposition and dissolution of Li$^+$ ions.[312] Owing to superior mechanical properties and facile manufacturing of reduced graphene oxide (rGO), Lin *et al.*[316] developed a Li–rGO electrode *via* molten Li infusion into layered rGO. They reported a low overpotential of *ca.* 80 mV for 300 h at 3 mA h/cm^2. Benefiting from a high aspect ratio, 2D materials have been used to decrease the interfacial resistance of the battery and improve cation transport through the electrode/electrolyte interface. High mechanical properties and the insulative nature of BNNSs triggered Yan *et al.*[323] to study the cycling of the lithium metal anode by depositing 6–10 layers of this 2D material on the copper counter electrode *via* the chemical vapor deposition (CVD) technique (Figure 9a). It is known that BNNSs have a Young's modulus of *ca.* 1.0 TPa and the large band gap of 5.5–6.1 eV. Hence, this coating not only speeds up the uniform Li$^+$ ion flux but also avoids the dendritic and mossy Li formation during plating and stripping. The higher chemical stability and mechanical strength of BNN deposition on copper in Li‖Cu led to >95% of Coulombic efficiency at high areal capacities of up to 5 mA/cm^2.[323,324]

2D materials were also used in Li–S batteries to address the polysulfide dissolution at the cathode and formation of Li dendrites at the anode which accelerates the capacity fading.[325] Cha *et al.*[326] developed a thick 2D MoS$_2$ coating (~10 nm) on Li metal anodes to diminish these side reactions and improve the performance of Li–S batteries as shown in Figure 9b. The sputter coated MoS$_2$ provides a stable Li$^+$ ion electrodeposition and mitigates dendrite nucleation sites at high current densities of 10 mA/cm^2. These phenomena are attributed to the layered structure and phase transition of the MoS$_2$ layer in contact with Li$^+$ ions.[326] There are some scarce reports that indicate that these ASEI layers might compromise the electrochemical performance by increasing the electrode/electrolyte interfacial resistance of such batteries especially for the first few cycles.[326,327] However, due to the intrinsic defected structure and mechanical robustness, 2D materials can provide synergistic effects for deposition and stripping Li$^+$ ions uniformly.

2D MATERIALS FOR THERMAL STABILITY

One of the main concerns in LIBs is the vigorous parasitic electrochemical reactions during charge/discharge processes.[319,328] These side reactions may result in thermal runaway phenomena and explosions.[329] Therefore, the temperature durability of the battery components is vulnerable upon cycling.[330,331] Meanwhile, some types of 2D materials such as graphene, boron nitride, and transition metal dichalcogenides have shown specific thermal behaviors. These materials are generally used in matrices with very low thermal conductivity. High-quality graphene has the highest thermal conductivity of *ca.* 3000–5000 W/mK among all the 2D materials which makes it a great candidate for heat dissipation in batteries.[332,333] Hexagonal boron nitride (h-BN) as one of the most thermally conductive materials (*ca.* 250–400 W/mK) with its electrically insulative nature can be a great choice as an electrolyte membrane in batteries.[334,335] Experimental data and theoretical calculations have shown that there is a layer-dependent thermal conductivity in such materials.[336]

2D materials have also been used to suppress the oxygen release of cathode materials and diminish the risk of thermal runaway. Sharifi-Asl *et al.*[337] (Figure 10a) showed that a few layers of the rGO coating on LCO particles can mitigate the oxygen release under thermal abusive conditions (>300 °C). This rGO coating provides an impermeable membrane against gas evolution by forming a chemical bond between carbon and oxygen.[337] The rGO coating can also improve the structural stability of the cathode particles at high voltages (4.6 V vs Li/Li$^+$).[337]

The h-BN incorporation in batteries such as a composite electrolyte additive or coating the separator is shown to improve the thermal safety of batteries. More interestingly, the

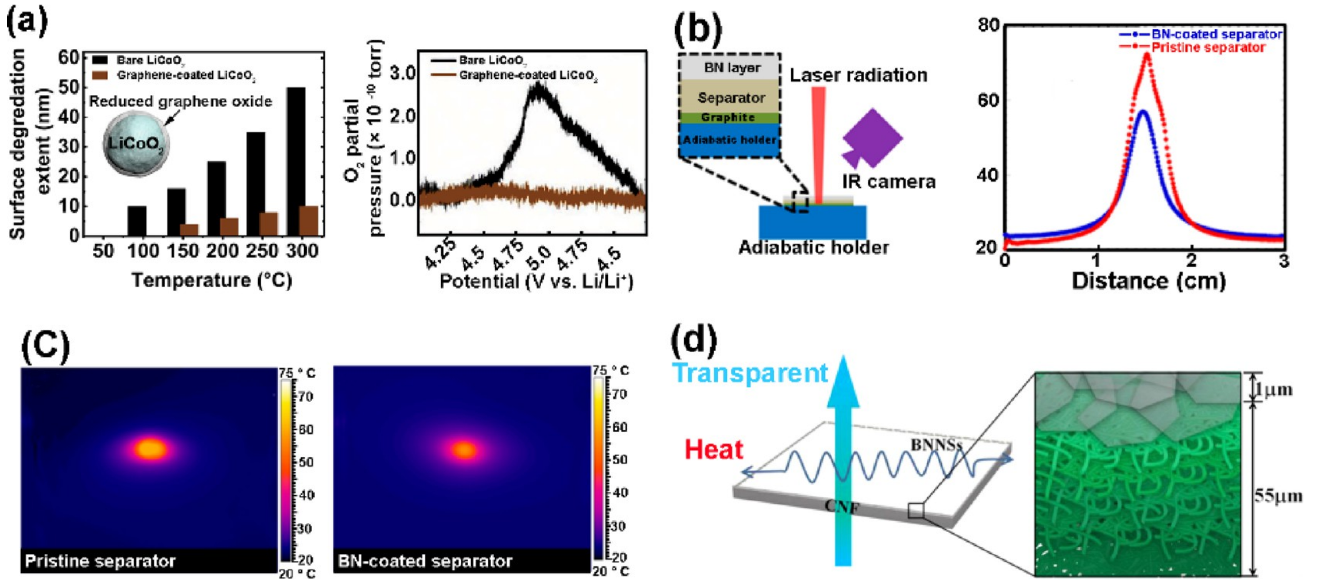

Figure 10. Controlling thermal stability of the battery with 2D materials. (a) Effect of graphene coating on the surface of $LiCoO_2$ particles at high-temperature (left) and high-voltage (right) abusive environments. The schematic illustration of the graphene coating is shown in the inset of the left figure. Reprinted in part with permission from ref 337. Copyright 2019 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) Schematic illustration of creating a hotspot on the separators by a focused laser beam (left) and corresponding temperature distribution on the PVDF-HFP separator with and without the BNN coating. Reprinted in part with permission from ref 319. Copyright 2015 American Chemical Society. (c) Mapping the temperature distribution of irradiated heat on the PVDF-HFP separator without (left) and with (right) the BNN coating. Reprinted in part with permission from ref 319. Copyright 2015 American Chemical Society. (d) Schematic illustration of the bilayer BNNS-CNF nanopaper that conducts heat along the in-plane direction. Reprinted in part with permission from ref 338. Copyright 2016 American Chemical Society.

BNNs induce a monotonic thermal profile and hinder exothermal electrochemical reactions more effectively.[319,335] Luo *et al.*[319] developed BNNs on the polypropylene (PP)/polyethylene (PE)-based commercial separator and studied the heat distribution upon irradiating a laser beam through the samples. Based on their observation, the PP/PE separator is only able to exhibit a low thermal conductivity, <1.0 W/mK, while the BNN-coated separator has a high thermal conductivity of 82 W/mK. The thermal distribution profile in Figure 10b,c shows that the BN coating improves the temperature uniformity during laser radiation from the centroid heat of about 50 to 34 °C at the PP/PE separator to its BNN-coating counterpart due to the enhanced heat spreading properties of such nanosheets.[319] The great thermal conductivity properties of the BNNs have been also used in other research studies. Zhou *et al.*[338] designed a bilayer BNN coating on the cellulose nanofiber (CNF) separators as shown in Figure 10d. They reported a significant increase of in-plane thermal conductivity of the BNN-coated CNF up to 30 W/mK which is 750 times higher than the pure CNF-nanopaper with poor thermal conductivity of 0.04 W/mK. Following the thermal management of high-performance Li metal anodes, Liu *et al.*[339] developed a 3D printed poly(vinylidene fluoride)-hexafluoropropene (PVDF-HFP) separator including BNNs. Besides the effective ability to improve the overall electrochemical performance of the Li metal battery by enhancing electrolyte wettability, once the local heat source is emitted on the separator, the localized heat can be diffused effectively along the BNN separator and yield a lower central temperature by 14 °C due to its specific thermal transport properties.[339]

2D MATERIALS AS CURRENT COLLECTORS

As key component of batteries, current collectors are responsible for allowing the electrons to flow though the electrical circuit during charge/discharge. Low electrical conductivity often leads to intensified voltage polarization and poor electrochemical performance.[340,341] The electrical conductivity is dependent on a variety of factors including synthesis methods, purification degree, and the thickness of a material.[342,343] Therefore, these materials should be tuned to have a low electrical resistance of usually < 0.001 Ω cm, *i.e.*, high electrical conductivity of >1000 S/cm.[341] In addition, current collectors should endure aggravated electrochemical environments. One of the significant issues that limits the choice of materials as current collectors is their susceptibility to corrosion and poor compatibility to different electrolyte chemistries.[344] Therefore, working potential and environmental conditions of the materials should be considered in cell designs precisely. Moreover, mechanical property requirements such as high Young's modulus and flexibility under repeated deformation stresses are among the other important factors to select materials as current collectors.[342,345]

Freestanding current collectors with the ability to be used simultaneously as electrodes have the potential to increase the volumetric and gravimetric energy densities.[346,347] This strategy leads to producing binder-free electrodes and eliminates the use of current collector. Therefore, it can reduce the mass production expenses and total weight of the cell, while benefiting from good electrochemical performance for long cycles.[348,349] Due to exceptional mechanical and electrical properties, low density, and good electrochemical stability, graphene is widely considered for this application.[350] To this end, Shi *et al.*[351] studied the graphene-based integrated electrodes for both cathodes and anodes for a typical Li

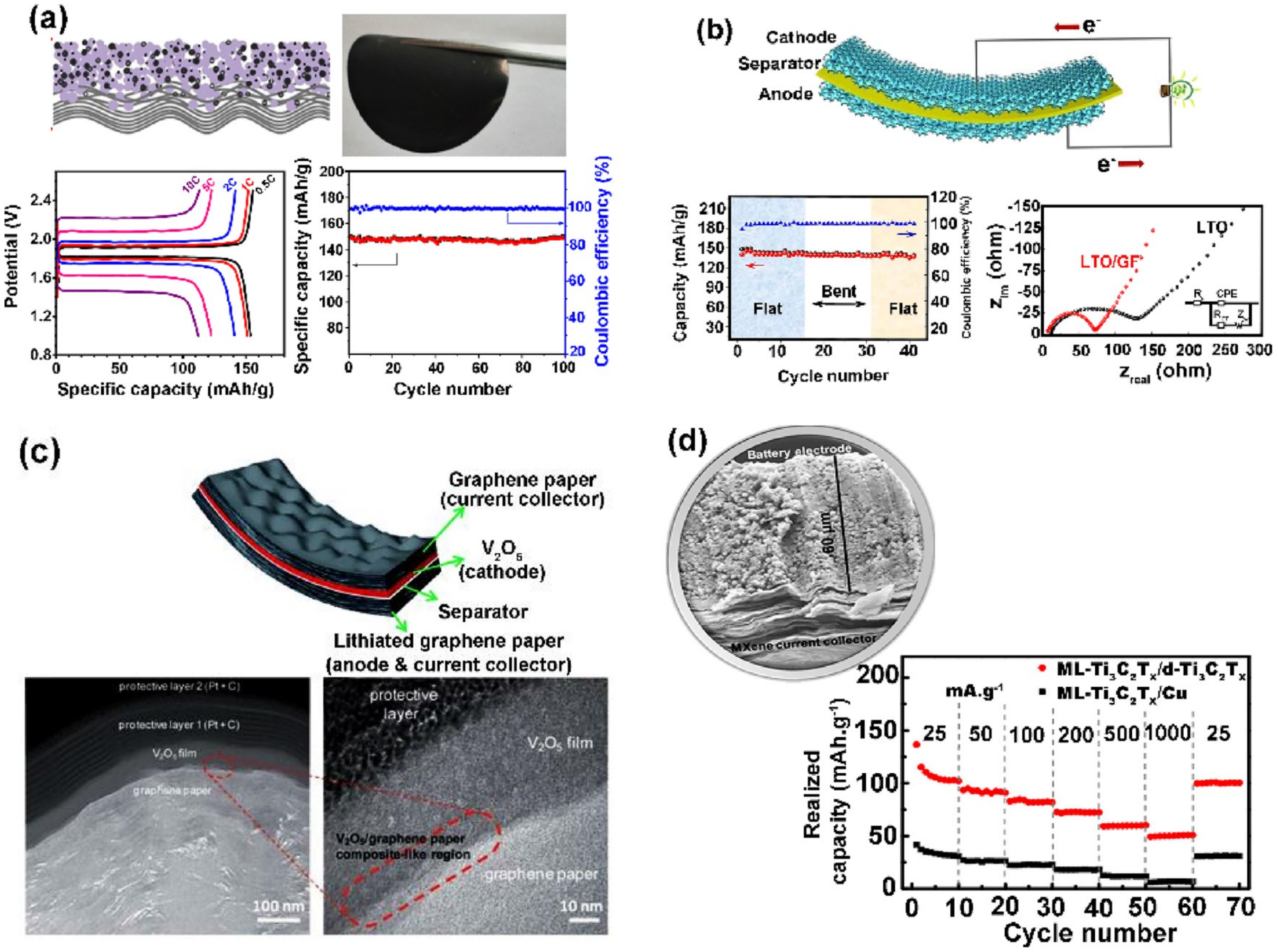

Figure 11. Application of 2D materials as the current collector. (a) Schematic illustration (top-left) and optical image of the flexible graphene (G) paper (top-right); electrochemical behavior of the G-based integrated electrodes. Charge and discharge curves of a full G-LTO/G-LFP battery at different current rates (bottom-left) and cycling performance and Coulombic efficiency of the same battery over 100 cycles at 1 C (145 mA/g) (bottom-right). Reprinted in part with permission from ref 351. Copyright 2015 IOP Publishing Ltd. (b) Schematic illustration of a flexible battery including a cathode and an anode made from interconnected graphene foam (GF) (top), cyclic performance and Coulombic efficiency of the battery under consecutive flat and bent states (bottom-left), and comparing the EIS of the LTO/GF electrode and reference LTO coated on an Al current collector (bottom-right). Reprinted in part with permission from ref 352. Copyright 2012 PNAS. (c) Schematic illustration of (PLD) grown V_2O_5 and the lithiated graphene paper as the cathode and anode, respectively (top), and a cross-sectional TEM image of V_2O_5 film on graphene paper (bottom). Reprinted in part with permission from ref 353. Copyright 2011 RSC Publishing. (d) SEM cross-sectional image of using d-$Ti_3C_2T_x$ as a current collector (top-left) and realized capacity (considering the total weight of active materials and the current collector) of ML-$Ti_3C_2T_x$ on a Cu foil and ML-$Ti_3C_2T_x$ on d-$Ti_3C_2T_x$ (bottom-right). Reprinted in part with permission from ref 18. Copyright 2018 American Chemical Society.

battery. A full Li battery cell with flexible G-LTO and G-LFP self-standing films as anode and cathode materials resulted in a great cycling behavior even at an ultrahigh rate of 10 C (1450 mA/g) of 112.1 mA h/g and negligible capacity loss over 100 cycles. Interestingly, this innovation led to lower polarization values than the conventional electrodes with Al and Cu current collectors for the cathode and anode, respectively. This is due to poor contact and adhesion of these substrates with active materials which shows more drastic changes at high current density cycling.[351] Figure 11a summarizes this study. A similar study has been done by Li *et al.*[352] who have shown that the graphene foam (GF) network current collectors can provide outstanding cyclability while bending (Figure 11b). These highly GF networks were used both for a full cell battery of LFP/GF and LTO/GF electrodes and demonstrated *ca.* 50% reduction in charge-transfer resistance. As a result, this battery led to 98% capacity retention over 500 cycles at a 10 C charge/discharge rate. Gwon *et al.*[353] developed a pulsed laser deposition (PLD) grown V_2O_5 on graphene paper as the cathode and the lithiated graphene paper anode as shown in Figure 11c. PLD helps to achieve uniform coverage of the V_2O_5 on graphene paper, while prelithiation eliminates the undesirable lithium uptake from electrolyte. Hu *et al.*[354] also prepared hybrid graphene nanosheet (GNS) and carbon nanotube (CNT) papers *via* vacuum-assisted filtration and further annealing at 800 °C. They showed that the resulting product can be used as a Li battery anode with no binder and current collector involved and exhibited a reversible capacity of >330 mA h/g over 100 cycles at a current rate of 100 mA/g.[354]

Metallically conductive freestanding films of MXenes were also explored as integrated anode-current collector films. The highest electrical conductivity of 2D titanium carbide among other MXene materials (~8000 S/cm) inspired Wang *et al.*[18] to use $Ti_3C_2T_x$ (T_x: surface functional groups) as current collectors for both anode and cathodes. This material not only has 3 times lower packing density compared to that of Cu foil, but it also has a good electrochemical stability at a wide potential range. As shown in Figure 11d, the realized gravimetric capacity of delaminated $Ti_3C_2T_x$ (d-$Ti_3C_2T_x$) is considerably higher than using a Cu foil as a current collector for the same active material. This offers a great potential application of such materials to improve the energy density of LIBs.

2D MATERIALS TO BOOST THE IONIC CONDUCTIVITY IN ELECTROLYTES

In the search for more reliable materials to tackle low ionic conductivity and limited Li^+ ion flux, using 2D materials as the electrolyte additive has shown growing interests in LIBs. 2D materials have a great potential to improve ionic conductivity by lowering the dissociation energy of Li^+ ions and their counterpart anions.[355] These electrolyte additives can decrease the cationic−anionic intermolecular interactions in electrolyte media, resulting in an improved ionic conductivity.[355−357]

Furthermore, boosting cationic mobility through 2D materials can improve the ionic conduction in the electrolyte. By definition, Li^+ ion mobility is the migration of Li^+ ions by a redox process between the electrodes and through the electrolyte and is directly related to the diffusion coefficient.[358] Limited diffusion of Li^+ ions through solid electrolytes restricts effective ion motion and avoids fast ion conduction in these electrolytes. Due to high specific surface area and atomic structure, using 2D materials can reduce the ion diffusion distance and promote fast surface redox reactions.[2,3] Moreover, 2D materials advocate directed diffusion properties and lower the ion mobility barriers for ultrafast ion transport due to their superionic conduction properties.[5] The presence of structural defects and surface charges in 2D materials can also lead to higher ionic conductivity of the electrolytes. In this hypothesis, the anions anchor to the surface of these additives by Lewis acid−base interactions and decrease the Li^+-anion dissociation energy.[359] These electrolyte additives are also reported to affect the crystalline structure of the polymer host, enhance its segmental motion, and thus facilitate the ion transport.[360]

Recent efforts show that 2D materials have great potential to be used in battery electrolytes. Graphene-analogues boron nitride nanosheets (g-BNNs) are one of the most used 2D materials in the electrolyte due to their wide energy band gap, high thermal conductivity, and great mechanical properties.[155] These outstanding properties along with the high surface area and ability to adsorb high amounts of ionic liquid have led to introducing quasiliquid solid electrolytes by confining ionic liquids by Li *et al.*[155] (Figure 12a). They have demonstrated that the ionic conductivity is as high as 3.85×10^{-3} S/cm at 25 °C, and 2.32×10^{-4} S/cm at −20 °C. The outstanding electrochemical properties of such an electrolyte were assigned to continuous and fast Li^+ ion transport channels in interlayers and out-of-layers of g-BNNs.[155]

Recently, exceptional properties of graphene-based materials have emerged as alternative electrolyte additives in the polymer backbone to acquire high ionic conductivity, thermal and electrochemical stability, and dendrite suppression robustness. 2D nanosheets of GO in this family pose electrical insulating properties, and owing to the specific geometry, it has high specific surface area. Using GO nanosheets as an electrolyte additive promotes charge carrier concentration by increasing the dissociation degree of lithium salt ion pairs based on Lewis acid−base interactions.[365] Moreover, these materials disrupt PEO crystallization by increasing the glass transition temperature (T_g) of the polymer composite. Hence, the grain boundaries are diminished after mixing the polymer electrolyte with this additive and suggest an up to 2 orders of magnitude increase in ionic conductivity (Figure 12b).[361] GO nanosheets were also embedded in solid poly(ethylene oxide) (PEO) matrix as a flexible electrolyte by Kammoun *et al.*[366] and showed high capacity retention during deformation. GO nanosheets reduce the internal and interfacial resistance between electrodes and electrolyte by affecting the host polymer crystallinity and facilitating ion motion. Reportedly, not always is the addition of GO fillers beneficial for stimulating the charge carrier ability. A high loading of GO nanosheets may increase the aggregation and reduce the open cation transport channels.[367] The strong interaction between GO and the polymer backbone by highly oxygenated functional groups of GO sheets led to present poly(ethylene glycol)-grafted GO (PGO) as an electrolyte in a study by Shim *et al.*[362] (Figure 12c). They revealed that a good dispersion of

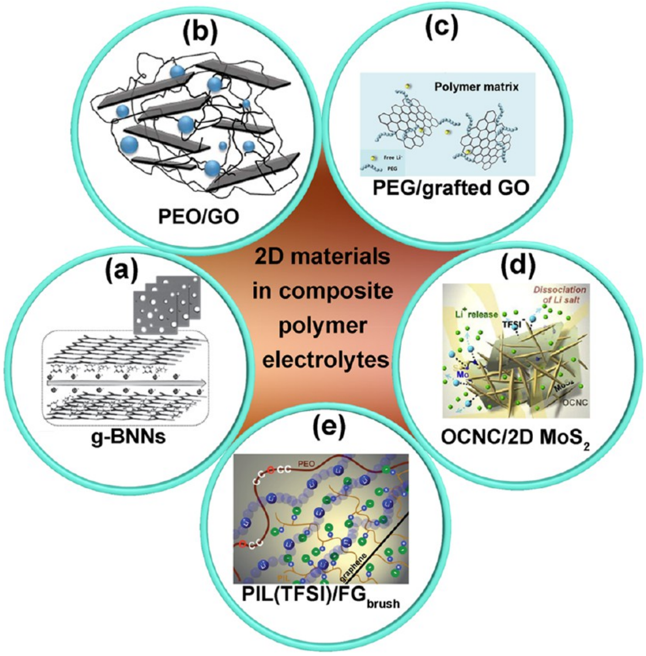

Figure 12. Schematic showing the application of some typical 2D materials in composite polymer electrolytes for Li batteries. (a) Graphene-analogues boron nitride nanosheets (g-BNNs) confined ionic liquid. Reprinted in part with permission from ref 155. Copyright 2016 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) Solid composite electrolyte consisting of poly(ethylene oxide) (PEO-GO). Reprinted in part with permission from ref 361. Copyright 2014 RSC Publishing. (c) Poly(ethylene glycol)-grafted graphene oxide (PEG-grafted GO). Reprinted in part with permission from ref 362. Copyright 2014 Royal Society of Chemistry. (d) 3D ion conducting network with oxidized cellulose nanocrystals and MoS_2 nanosheets (OCNC/2D MoS_2). Reprinted in part with permission from ref 363. Copyright 2018 Elsevier B.V. (e) Composite polymer electrolyte consisting of functionalized reduced GO-based 2D molecular brushes (FG_{brush}) with polymeric ionic liquid PIL arms (PIL(TFSI)/FG_{brush}). Reprinted in part with permission from ref 364. Copyright 2015 Royal Society of Chemistry.

Table 2. Ionic Conductivity and Transference Number Values of Typical 2D Material Additives in Polymer Electrolytes

	host material	2D material(s) additive	electrolyte type	temperature (°C)	maximum ionic conductivity (S/cm)	transference number (t_{Li^+})	ref.
1	PEO+LiTFSI	$Ti_3C_2T_x$	solid	28	2.2×10^{-5}	0.18	360
2	EMI-TFSI+LiTFSI	g-BN	quasisolid	25	3.85×10^{-3}		155
3	poly(ethylene glycol)+BCP[a]+$LiClO_4$	GO	solid	30	2.1×10^{-4}	0.99	362
4	PIL[b]+OCNC+LiTFSI	MoS_2	solid	30	0.8×10^{-3}	0.65	363
5	PIL(Br)+LiTFSI	FG_{brush}	solid	30	7.9×10^{-5}	0.68	364
6	PAN[c]+LiTFSI	GO	solid	30	1.1×10^{-4}	0.4	367

[a] Branched-graft copolymer. [b] Polymeric ionic liquid. [c] Polyacrylonitrile.

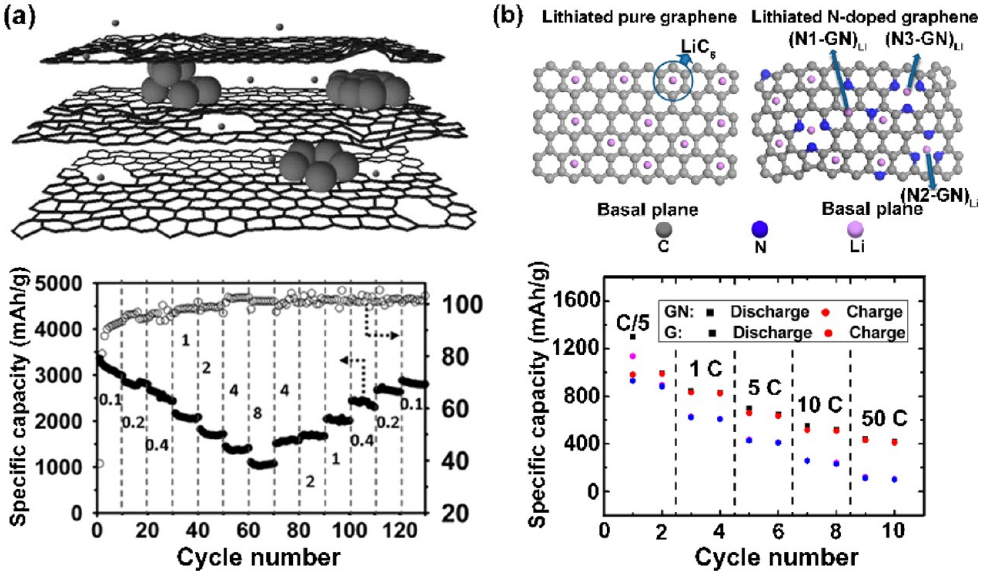

Figure 13. Role of 2D materials on improving rate performance. (a) Schematic illustration of the graphenic scaffold with aligned graphene sheets and sandwiched Si nanoparticles (top). Rate capability and Coulombic efficiency of the Si-G scaffold composite from 0.1 to 8 A/g (bottom). Reprinted in part with permission from ref 379. Copyright 2011 WILEY-VCH Verlag GmbH & Co. KGaA, Weinheim. (b) Schematic illustrations of G and GN nanosheets from the basal plane direction (top). Comparative rate capability of GN and G (bottom). Reprinted in part with permission from ref 385. Copyright 2014 American Chemical Society.

PGO facilitates the amount of dissociated free Li^+ ions resulting in an enhanced cycle performance.[362]

Following the enhancing of the electrochemical properties of solid electrolytes with 2D materials, Wu *et al.*[363] developed a composite polymer electrolyte using multifunctional MoS_2 nanosheets and oxidized cellulose nanocrystals (OCNCs) distributed in poly(diallyldimethylammonium chloride) (PDDA) as shown in Figure 12d. They reported that the hybrid network of MoS_2/OCNC acts as a physical barrier against Li dendrite growth and improves Young's modulus by 3-fold to 7.56 MPa and tensile strength by 20%. Moreover, the addition of MoS_2 nanosheets provides an ion-conducting network and increases the Li^+ ion transference number from 0.37 to 0.65 and good ionic conductivity of 0.8 mS/cm. Table 2 summarizes the ionic conductivity and transference number values of different 2D material additives in recently developed polymer electrolytes.

MXenes are another emerging family of layered 2D materials that have demonstrated promising performance in energy storage applications.[368,369] In a recent study by Pan *et al.*,[360] few-layered $Ti_3C_2T_x$ flakes were dispersed in the poly(ethylene oxide) matrix to improve its ionic conductivity. The hydrophilicity of these 2D materials leads to the interaction between MXene flakes and the ethylene oxide chains of the polymer backbone and reducing its crystallinity.[360]

In search for improving compatibility and dispensability of 2D material additives within polymeric chains, Banerjee *et al.*[370] functionalized GO with the Aza-crown functional group. They suggested that Li^+ ions can attach to the functionalized cages through the electrostatic interaction between Li^+ and oxygen atoms and intercalate in the graphene oxide structure.[370] This intercalation decreases the activation energy of Li^+ migration and improves the ionic conductivity for potential application as the solid electrolyte in Li batteries.[364,370,371] In another study, Ye *et al.*[364,371] developed a functionalized rGO composite polymer electrolyte as shown in Figure 12e. They demonstrated that functionalization of rGO by polymeric ionic liquid groups diminishes electron conduction and basically eliminates the risk of electrical shorts. Moreover, they showed that the direction of these nanosheets can potentially lead to interconnected ion conducting channels and facilitate ion mobility in resistant polymer chains through their low-energy pathways.[371]

2D MATERIALS TO IMPROVE RATE PERFORMANCE

One of the key parameters to improve electrochemical properties of Li batteries is to improve the rate performance of electrodes. Rate performance determines the charging speed and power delivery of secondary batteries.[372,373] As for electrodes, rate performance is a function of reaction kinetics of ionic motion and diffusion of Li^+ ions through the interlayers of active materials.[374] Research studies have shown that 2D materials can provide short diffusion paths and high surface area.[375] These properties decrease the required time scale for Li^+ ion diffusion and, thus, enable a high-rate capability.[376] Besides, the enlarged interlayer distance in 2D materials benefits faster reaction kinetics and higher rate performance.[377,378] To this end, Zhao *et al.*[379] developed and studied a three-dimensional scaffold of aligned graphene (G) nanosheets with sandwiched Si nanoparticles as shown in Figure 13a. The Li^+ ions can diffuse easily through a structurally integrated network of graphene nanosheets and their corresponding in-plane vacancy defects. The electrochemical studies of this composite material showed a reversible specific capacity approximately initiating from 1100 mA h/g and degraded at *ca.* 0.34% per cycle for 150 cycles at 8 A/g (approximately 8 C based on the accessible storage capacity). The rate performance of the Si-G scaffold composite from 0.1 to 8 A/g is shown in Figure 13a which confirms the high capacity retention of *ca.* 1000 mA h/g at peak current density and recovery of the *ca.* 85% of the original capacity when switched back to 0.1 A/g after 130 cycles.[379] A similar study was performed by Zhang *et al.*[7] where they developed a thick composite electrode material of Si/MXene of up to 450 μm with two-dimensional $Ti_3C_2T_x$ (MX-C) nanosheets as conductive binders. MX-C benefits from high metallic conductivity (up to 10^3 S/cm), which boosts electron transport through the electrode.[18,380] High electrical conductivity improves long-term reversible electrochemical reactions and leads to a high-rate cycling performance.[7,381] By studying S-/MX-C, this electrode could deliver an initial capacity of >2000 mA h/g combined with 98.4−99.8% of Coulombic efficiency over 280 cycles at high current densities of 1.5 A/g.[7] The ultrafast solid-state diffusion of Li^+ ions was also reported for cathode materials. Yang *et al.*[382] developed a cathode of vanadium dioxide (VO_2) nanoribbons included with graphene layers which could exhibit an ultrafast charging and discharging capability. This developed composite architecture showed a high reversible capacity of 204 mA h/g at the extremely high rates of 190 C (37.2 A/g) for 1000 cycles.

In addition, the disorder of the atomic arrangement of an electrode microstructure such as the introduction of defects can play a significant role on Li^+ ion transportation kinetics.[107,383] Defects can promote the rate performance of an electrode by providing alternative diffusion channels with low migration energies for Li^+ ion transport.[384] Wang *et al.*[385] studied the origin of the high-rate capability and capacity for the lithium storage of N-doped graphene (GN) compared to pure graphene. By N doping, the electrical conductivity of GN paper increases by 3-fold as compared to that of the pure graphene paper and reaches to 10^3 S/cm at room temperature. Interestingly, N3-GN and N2-GN point site defects enlarge edge {0002} spacings and lead to improved surface capacitive effects.[385,386] Figure 13b shows the schematic illustration of these defects and a comparison of NG and pure graphene (G) nanosheet rate capabilities. The GN electrodes demonstrate an outstanding discharge capacity of 1284 mA h/g in the first cycle at 0.2 C followed by 432 mA h/g at 50 C (*ca.* 72 s). This is 10−30% higher than that of the pure G and confirms the faster diffusivity of Li^+ ions in the presence of structural defects.[16,385]

It is worth noting that there are several research studies that reported significant improvement in rate performance of Li^+ ion batteries in the presence of 2D materials. However, the majority of these papers use coin-cells and pouch cells with low mass loadings of electrodes.[387,388] The commercially viable electrodes should be functional with high mass loading active materials and thick electrodes. Therefore, further studies of the rate performance with high mass loading of active electrode materials are inevitable.

In summary, exploring 2D materials as a main component of LIBs can pose cutting-edge advances. From this point of view, the right choice of 2D materials potentially increases the specific capacity, reduces the interfacial resistance, and introduces dominant pathways for transporting ions. Accordingly, these high-surface-area materials can alleviate the high polarization voltage, a problem that most solid-state batteries suffer,[389,390] hence ensuring high Coulombic efficiency and long-term stability.

SUMMARY

In this review, we discussed the material challenges of LIBs that can be addressed by the utilization of emerging 2D materials. These materials can be utilized as electrodes, separators, catalysts, current collectors, and electrolytes. To overcome the poor electrical properties of semiconductive electrode materials, 2D materials with outstanding tunable electrical conductivity are a promising choice since their added inactive mass to the batteries will not be significant. Compositing semiconductive active materials with high-electrical-conductivity 2D materials has been shown to significantly enhance the electron conductivity through the electrode, resulting in a better electrochemical performance.[3,7,182,183] The elemental and layered structure 2D materials have shown promising results for their application as cathodes or anodes with high specific and volumetric capacities.[64,391−393] 2D-structured architectures are proven to show high electrochemical activity and can demonstrate high electrochemical stability during cycling.

Due to enhanced mechanical properties of 2D materials, this class of materials can protect electrode materials from disintegration during cycling. The outstanding mechanical properties of 2D nanosheets can accommodate the mechanical strains (can reach 200−500%) that occur in the high-capacity electrode particles during de/lithiation. By controlling the expansion/shrinkage of high-capacity electrode materials during cycling with 2D protective layers, other electrode materials can be introduced. Moreover, due to large surface area and the ability to tune the surface of 2D materials by functionalization, these nanosheets can act as a supportive matrix for the active material particles and prevent pulverization effectively.[238,394−396]

The ability of 2D materials in blocking the lithium polysulfide species as a major bottleneck in Li−S batteries is quite intriguing. 2D materials such as MXenes, graphene, graphene-analogous, and other elemental nanosheets have an anchoring effect and can trap the polysulfides on their surface. In addition, the abundant edges, defects, and functional groups on these nanosheets act as a catalyst for Li−S redox reactions and improve the electrochemical performance of such batteries.[266,397−400]

We studied the significance of 2D materials in electrocatalytic activities in $Li-O_2$ batteries and their potential application as porous cathodes and/or catalysts for oxygen-related reactions. 2D materials have been shown to enable controlled growth and facile decomposition of reaction products such as Li_2O_2 to increase reversibility and improve the electrochemical performance of Li–O_2 batteries.[401–403]

The role of 2D materials as the electrode–electrolyte interface, separator coating, and electrolyte additives in uniform Li^+ ion electrodeposition was also covered in this Review. These materials improve the synergy of Li^+ ion deposition and diminish charge accumulation on the anode. They are also able to facilitate cationic transfer and diminish the sluggish reactions by assisting the uniform distribution of Li^+ flux through their structure. In addition, high mechanical properties of 2D materials have led their application in impeding dendrite nucleation, growth, and penetration which make the use of Li metal as an anode with an ultrahigh specific capacity of 3860 mA h/g more viable.[48,182,317]

Heat dissipation and conformal heat distribution properties of 2D materials such as boron nitride nanosheets as a coating layer on the separator and/or their use as electrolyte additives showed an improved thermal safety property.[31,319] Applying these nanosheets onto electrode materials also showed an improved thermal stability by impeding their high-temperature degradation.[404–406]

Boosting the ionic conductivity by introducing a high surface area and changing the crystal structure of the host electrolyte could be one of the many benefits of 2D material additives. These materials can increase the dissociation degree of Li^+ ions from their anion counterparts and, therefore, enhance Li^+ ion transference number and ionic conductivity.[361,407]

In addition to the above-mentioned properties, 2D materials still suffer from several challenges such as poor electrochemical properties[408] and poor chemical and structural stability at ambient conditions,[409] including expensive and often complicated processability of high-quality materials.[410,411] The high surface area of 2D materials may also participate in increasing parasitic reactions at the electrode/electrolyte interface.[3] Therefore, one should consider developing techniques to tackle these challenges such as developing composites and 2D heterostructured materials to improve electrochemical properties and mitigate undesired side reactions.[412–414] Moreover, commercial availability and cost of 2D materials are very critical factors for battery manufacturers. Some 2D materials such as graphene and boron nitride nanosheets are commercially available at relatively low cost. However, a majority of the 2D materials such as phosphorene and MXene materials require complicated synthesis processes and, yet, are only available in the small-scale experiments. Commercial electrodes often require >100 μm thickness to maximize areal capacity and energy density. However, the ability to fabricate thick electrodes from 2D materials is not yet guaranteed. Therefore, it is critical to rationalize cost-effective methods in synthesis, purification, and industrial-scale production stages to take a practical step for commercializing these materials in batteries.

FUTURE PROSPECTS

Owing to the outstanding properties of 2D materials which are discussed in this Review, the authors believe that this class of materials can lead to the fabrication of safe, long-lasting batteries for high power and energy applications. Here, we outline some of the most prominent fields in which 2D materials may lead to breakthrough applications of LIBs in the future as shown in Figure 14.

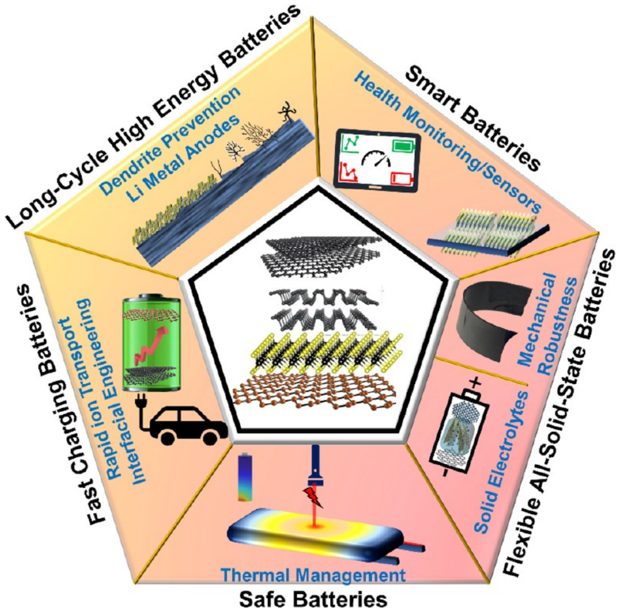

Figure 14. Future prospects of 2D materials in LIBs. 2D materials can lead to outstanding safety, electrochemical, and sensing properties.

LONG-CYCLE HIGH-ENERGY BATTERIES

The current progress in Li batteries is limited due to proliferation of metallic Li dendrites which can lead to dramatic safety concerns.[415,416] The diffusion-limited properties of Li^+ ions within the electrode/electrolyte interface[311] and mechanical stress during plating in Li metal[417] are known to be some of the major reasons for Li metal dendrites. High current densities often tend to accelerate the formation and growth of Li dendrites more significantly.[418] This is one of the key challenges that has impeded wide commercial development of fast-charging batteries to date. However, application of 2D materials can offer uniform current distribution through the electrode/electrolyte interface, lower ion transfer resistance, and increase mechanical robustness against penetration of Li dendrites. The synergistic effects of 2D materials are capable of delocalizing Li^+ ion accumulation and lead to a uniform electrodeposition and dendrite-free environment. Further investigation on material selection, structural modification, and designing engineering methods to utilize 2D materials in Li batteries are still required.

SMART BATTERIES

The importance of accurately measuring and estimating the *battery* charge, voltage, current, and temperature has led to the development of versatile methods to monitor battery health.[419,420] Due to unique physicochemical and structural properties, 2D materials have emerged for sensing applications in energy storage systems.[238] The large surface area of 2D materials combined with their tunable electrical properties provides a sensitive platform to detect safety concerns, inform the consumers and manufacturing about the on-going degradations, and adapt to the environmental changes. These are relatively less explored in LIBs.

FLEXIBLE ALL-SOLID-STATE BATTERIES

2D materials can be impactful on solid-state batteries and conformal battery designs by improving mechanical flexibility and introducing better ionic conduction at the electrode/electrolyte interface and within the electrolyte, which have been a major barrier for developing such energy storage devices. Further studies on the alignment of 2D materials in the electrode−electrolyte structures, engineering the band gap with respect to mechanical straining, and developing 2D materials with scalable manufacturing processes could be great contributions to this field. More precise atomistic and molecular models as well as *in situ* experimental validation facilitate the identification of 2D materials with proper architectures for designing solid-state batteries.

Developing heterostructure 2D materials is another promising strategy that can introduce more robust solid-state LIBs in the near future. Moreover, it is vital to make a balance with the rate of ion shuttling through the electrolyte and the diffusion rate in the corresponding electrodes. Failure to harmonize these two major phenomena will lead to fast degradation of the battery due to accumulation of charges with parasitic reactions. We believe that introducing an "all 2D materials battery", where all electrodes and electrolyte are interconnected with 2D-structured materials, may be able to resolve this issue in energy storage systems.

SAFE BATTERIES

Thermal runaway is the most catastrophic failure mode in LIBs, which can be caused by material decomposition, high uncontrolled voltages, and internal short-circuit.[421] 2D materials with superior thermal properties can prevent decomposition and increase the safety by impeding the release of oxygen[337] and offering efficient heat dissipation.[30] Therefore, developing ideas to utilize these advantages of 2D materials can push the safety of LIBs to the frontline.

FAST CHARGING BATTERIES

High-rate charging capability is one of the key factors pursued by battery manufactures. Although there have been numerous investigations to introduce rapid charging batteries, it often sacrifices specific capacity, power density, and life of such batteries. Low activation energy for diffusion of Li^+ ions in 2D materials offers a promising approach to increase the charging rates. However, this research is still in its infancy, and more investigation is required in this realm.

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Notes

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VOCABULARY

battery capacity, the maximum amount of energy that can be derived from a battery at specific environmental and electrochemical conditions; Coulombic efficiency, the fraction of available discharge capacity to actual charge capacity over a full cycle; Li^+ ion transference number, the ratio of the electrical current carried by Li^+ ions to the total electrical current; thermal runaway, a phenomenon that results in excessive heat generation due to decomposition of materials in batteries under abusive conditions; ionic conductivity, a measure that shows the ability to flow ions through a material; ionic conductive materials do not necessarily possess good electrical conductivity

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