

Patterns and Mechanisms of Graphene Interface Layers in Regulating Lithium-Ion Transport Pathways at the Electrode/Electrolyte Interface in Solid-State Batteries

Sipeng Zhang

School of Materials Science and Engineering, Hebei University of Technology, Tianjin, 300400, China

15512252320@163.com

ABSTRACT

The development of high-energy-density solid-state batteries is an important way to solve these problems because of the unstable solid-solid interfaces, high interfacial impedance and low density. It is used to adjust the physical and chemical effects of the boundary between electrode and electrolyte. Review systematically examines the role of graphene middle layer in anode, cathode and electrolyte. We divide its main functions into three parts. At the anode end, graphene with additional functions inhibits lithium dendrite and makes the flow of Li uniform through the guidance of surface properties and structure. At the cathode end, graphene forms a mixed conductive network as a buffer layer, which reduces the burden on the interface and improves the charge movement. In electrolyte, the nano-filler of graphene improves the ion channel and enhances the mechanical strength. This series of effects laid a theoretical foundation for multi-dimensional interface optimization, and paved the way for rational design of fast-charging, durable solid-state batteries.

Keywords: Solid-State Batteries, Graphene Interface Layer, Lithium-Ion Transport Pathways, Lithium Dendrite Suppression, High-Rate Performance.

1. INTRODUCTION

Portable electronic devices, electric vehicles and large energy storage system are developing rapidly, and society needs “more energy and safe energy storage technology”. The liquid lithium-ion battery in the past may be dangerous because of out-of-control heat. Solid-state battery (SSB) is a kind of solid-state battery that transforms flammable organic electrolyte into nonflammable one, and it is an important technology in the next era beyond the current energy storage limit. In particular, solid-state system can be used together with metallic lithium anode, which may greatly improve energy.

However, it is difficult to sell solid-state batteries as commodities, because there is a big interface problem. Poor “solid-solid” interface contact will lead to higher interface impedance. On the other hand, if the flow distribution of lithium ions is distorted, lithium dendrites will grow easily, the capacity will be reduced immediately and the cycle life will be shortened. Therefore, in the strategy of interface engineering, it is an important task that must be solved immediately to do well the ion channel and stabilize the electrode/electrolyte interface.

Among many modification strategies, graphene and its partner (GO, rGO) are very attractive because of their unique structure and physical and chemical properties. Graphene has high mechanical strength and flexibility, which can well alleviate the volume deformation in circulation. Its excellent conductivity and two-dimensional layered structure help to form a continuous and fast ion/electron transportation network. In addition, functionalized graphene derivatives and substances with defects adjusted have good affinity with lithium, which can reduce the nuclear overvoltage and promote the uniform accumulation of metal lithium. According to these characteristics, it can be said that it is an ideal material for making the interface layer of solid-state batteries.

Although there are many studies in this field, there are few studies on graphene’s planned adjustment of lithium-ion transportation behavior on the full-cell scale, and most studies only focus on one component. Therefore, the purpose of this paper is to systematically investigate the law and mechanism of graphene interface layer adjusting lithium-ion transportation path. Investigate from the following three aspects. (1) At the anode end, the resistance of the interface is reduced and the appearance of dendrite is suppressed through the action of mechanical support and conductive network. (2) At the cathode end, it is mixed with the cathode material to form an efficient mixed conductive network, which shortens

the transportation distance. (3) On the side of electrolyte matrix, the ideal artificial SEI is made by using the property of selective ion passage. The purpose of this study is to provide a widely used theoretical basis for the interface engineering when designing high-output and durable solid-state batteries.

2. REGULATION MECHANISMS OF GRAPHENE INTERFACIAL LAYERS AT ANODE, CATHODE AND SOLID ELECTROLYTE INTERFACES

2.1 Graphene-Mediated Interface Regulation on the Anode Side: From Uniform Lithium Deposition to Stable SEI Formation

There are two big problems when using solid-state batteries, especially products using lithium metal anodes. One is that lithium dendrites cannot be stretched uniformly, and the other is that the solid-solid interface contact is unstable. If lithium sticks unevenly, dendrites may break through solid electrolyte, resulting in internal short circuit. Moreover, if there are few places where anode and solid-state electrolyte physically stick together, the resistance of the interface will become greater and the action of the battery will become worse. In recent research, adding functional interface layer between lithium metal anodes and solid-state electrolyte has attracted much attention. Graphene and its companions know that due to its special physical and chemical properties, it may be very helpful to adjust these things. In this chapter, we will learn in detail how Graphene uses surface chemistry and small structure to improve the adhesion and interface stability of lithium together.

2.1.1 Surface Chemistry Modulation and Homogenization of Lithium Ion Flow

The reason of needing anode interfacial engineering is the essential limitation of bare solid-solid contact. For example, when garnet-type LLZO comes into contact with air, the lithiophobic Li_2CO_3 layer of lithium will be formed. Therefore, Li metal can only contact with the point, and the interfacial resistance can reach $1235 \Omega \text{ cm}$, which is much larger than the system using liquid [1]. If Space-charge accumulates or the stress is concentrated due to defects, it is easy to produce Li locally. As a result, even the oxide of shear moduli up to 50 GPa may penetrate dendrite [1]. In order to solve these problems, graphene interlayers is introduced [11]. Graphene interlayers is not only a conductive additive, but also plays a multifunctional role in regulating the uniform flow of Li and absorbing volume changes.

The surface properties of graphene, especially electronegativity, have great influence on the movement of lithium ion when it is first attached. The surface of pure graphene is chemically stable and elastic, so it is not suitable for uniformly producing lithium. If chemical modification such as partially reduced graphene oxide (RGO) [2] is carried out, there are oxygen-containing functional groups such as $-\text{COOH}$ or $-\text{OH}$, and the structural defects [3] show that these oxygen-containing functional groups are slightly negative. Will attract positively charged lithium ions (Li^+), so the concentration of the boundary lithium ion is more uniform. This effect is called “electrostatic shielding” and “ion flow homogenization”, which eliminates the local current hot spots caused by microdefects and the electric deviation of the solid electrolyte surface. Then, the lithium ions are guided to spread and accumulate evenly on a wide surface, which lays the foundation for forming a flat lithium accumulation layer in the future. For example, Guo et al. cultivated the sequence of nitrogen-doped vertical graphene on copper current collector by CVD. Many edge defects and nitrogen doping greatly improve the adsorption energy of lithium ions on the surface, so even if the current density is as high as 3 ma cm , it can be piled up into non-dendritic lithium.

2.1.2 Structural Design Guidance and Control of Lithium Deposition Morphology

In addition to surface chemistry, the microstructure of graphics, such as vertical alignment, polling network and component structure, are all physical structures that determine the location of lithium. The traditional horizontal graphene layer may interfere with the upward and downward movement of ions, while the vertically arranged graphene nanoplate (VGNs) provide an open ion channel [4]. The contact area between electrode and electrolyte is greatly increased. More importantly, the gap between layers of VGNs will become a nano-sized “lithium storage place”. When installing lithium, first enter the gap, not pile it on the surface, but install it in a three-dimensional structure. This “closed attachment” structure can not only inhibit the outward extension of dendrite, but also greatly alleviate the huge change of volume during repeated charging and discharging. Moreover, if graphene is mixed with soft polymer (PEO, etc.) or inorganic nanoparticle (Li_3N , etc.) to make a soft composite intermediate layer, the flexibility of the intermediate layer and the force to cooperate with the volume change of lithium will become stronger, and the interface will become stronger.

2.1.3 Inducing the Formation of a Stable Solid Electrolyte Interface (SEI)

Stable SEI, through which ions often pass, is very important for lithium dendrite and inhibition of side reactions. On the other hand, graphene with special functions (such as fluorinated graphene) participates in the interfacial reaction at the beginning of electrochemical cycle, which helps to form SEI containing a large number of inorganic components, such as LiF and Li₂O^[5]. Such inorganic SEI has high surface energy and mechanical strength, and Li can easily pass through, so it can prevent the continuous decomposition of electrolyte and inhibit the penetration of dendrite. On the other hand, graphene is well electrified and chemically stable, so it becomes the basis of SEI uniformity. The results show that the SEI formed on the graphene modified interface is thinner, more uniform and more stable than that directly formed on lithium metal or uneven copper current collector. For example, Pervez et al. commented on rGO/LiF composite interface layer^[6]. rGO network can not only establish electron channels and make ions flow evenly, but also the existing LiF seeds can cause rich and uniform SEI of LiF, thus forming a symmetrical structure.

2.2 Interface Regulation of Graphene on the Cathode Side: Constructing a Hybrid Ionic-Electronic Conductive Network

On the cathode side of solid-state batteries, the main challenge stems from the complex multiphase interfaces within composite cathodes. Traditional composite cathodes consist of a simple mixture of active material, solid-state electrolyte, and conductive agent particles; ion and electron transport pathways are tortuous and discontinuous. Particularly under high active material loading, solid-solid point contacts lead to extremely high interfacial impedance and slow reaction kinetics. The core role of graphene here is to construct a long-range, continuous, and highly efficient hybrid ion-electron conductive network, thereby fundamentally optimizing charge transport pathways.

2.2.1 As a Three-Dimensional Hybrid Conductive Scaffold

A major kinetic bottleneck at the cathode side is the space-charge layer (SCL) formed at the active-material/solid-electrolyte interface. Due to the chemical potential difference, Li⁺ migrates from the electrolyte (e.g., Li_{6.4}La₃Zr_{1.4}Ta_{0.6}O₁₂) to the layered cathode (e.g., LiCoO₂), leaving a Li-depleted layer that elevates interfacial resistance^[1]. Conventional solutions rely on ALD or sol-gel inorganic coatings (Al₂O₃, LiNbO₃, Li₃PO₄) to block side reactions and narrow the SCL effect—Hood et al. They reported that after adding ALD-Al₂O₃ to the Weibo of Li₆PS₅Cl, the ionic conductivity of pressed particles increased by two times^[1]. The network mixed with Graphene transforms this idea into a channel of 2D ions/electrons that continuously contact at scattered points, making it more progressive.

Due to the special two-dimensional layered structure and good conductivity of Graphene, effective materials can be wrapped and connected together to form a three-dimensional conductive network, which replaces the traditional carbon black conductive additive. As a supplement, more importantly, if Graphene is mixed with ion-impermeable substances (such as nanoparticles of solid-state electrolyte and coatings that often penetrate ions), its layers will be very good “nano-mixer” and “scaffold”. When solid-state electrolyte nanoparticles (LLZO, LLTO, etc.) is uniformly placed on the surface of graphene or together with polymer electrolyte, the graphene layer will have ionic conductors, so that lithium ions quickly pass through the network of “graphene- ionic conductors”, avoiding non-conductive polymer binder and useless contact points, and directly reach the surface of active material particles^[7], and electrochemical reactions occur. In Wan et al.’s research^[10], the cathode particles of LCO are put together with graphene nanosheets and sulfide electrolyte. graphene is not only a fast way for electrons to pass through, but also flat, and the electrolyte particles are closely arranged between graphene sheets. It is found that the twists and turns of the path through which ions pass become very few, and the composite cathode will show very good rate performance at room temperature.

2.2.2 Optimizing Interfacial Contact and Reducing Impedance

Graphene is soft and has a wide surface, which helps to improve the solid-solid interfacial contact. First, the soft graphene flakes closely adhere to the rough surface of active material particles, thus increasing the actual contact area. Secondly, the buffer layer of graphene is added between active material and bulk solid-state electrolyte, which can alleviate the pressure caused by lattice mismatch and volume change during charging and discharging, thereby preventing interfacial contact failure.

Electrochemical’s view is that this close contact and continuous conductive network will directly reduce the charge transfer resistance. It is found in the experiment that the interfacial impedance of composite cathodes is usually reduced by an order of magnitude after the interfacial layer of graphene is added. In addition, to some extent, graphene can improve the interfacial chemical stability by inhibiting the side reactions between unstable electrolyte such as high voltage cathode materials and sulfides.

2.3 Graphene as a Functional Additive in Solid-State Electrolytes

This troubled situation is not just polymers. This represents a common trade-off between solid electrolytes (choosing one will lead to one failure). Sulfides is as fast as liquid, and ions pass through it, but it is not resistant to humidity, and its tolerance to oxidation is very small (< 2.3 V vs. Li^+/Li). Oxides (such as LLZO) are resistant to Li metal, but they will produce ionic films that are difficult to be electrified. Halides can withstand high voltage (> 4 V), but it cannot be used with Li anodes. Although nitrides are resistant to heat and Li, their resistance to oxidation is very weak [8]. In order to solve these problems without damaging the manufacturability of polymers, it is considered a good method to mix multifunctional 2D nanofillers, especially graphene oxide (GO) [8]. GO not only dilutes the content of polymer, but also can be expected to re-create the path through which ions pass and strengthen the solid framework. This design concept is completely consistent with the research priorities of today's national solid-state electrochemistry seminar. As emphasized in the recently published authoritative review on the development of solid electrolyte, the paradigm shift is now taking place in this field. From the progress of loose materials to the change of summarizing the overall strategy. This strategy includes optimizing ion movement with the help of machine-learning, designing lattice-dynamics-guided crystals, self-repairing polymer interfaces, and mass-produced sulfide films [9]. Very importantly, the summary of this study points out that the primary bottleneck preventing the laboratory from becoming a commercially available battery with high energy density is interface compatibility and uneven ion movement. Therefore, the composite electrolyte structure using graphene-oxide introduced this time is not only an academic game, but a clear decision and answer to a recognized topic.

In addition to serving as an independent interfacial layer between the electrode and the electrolyte, graphene and its derivatives can also function as nanofillers, directly incorporated into polymer solid-state electrolytes to enhance their overall performance. Pure polymer electrolytes (such as PEO-LiTFSI) suffer from issues such as low ionic conductivity at room temperature, poor mechanical strength, and weak lithium dendrite suppression capabilities. The introduction of graphene oxide (GO) or functionalized graphene can bring improvements on multiple levels.

2.3.1 Enhancing Ionic Conductivity

In addition to electrode-facing interlayers, it is also important to change bulk electrolyte. Cationic doping (Al/Ga/Nb of LLZO; Zr/Cl doping of LiPS) stabilized the cubic phase and increased the critical current density from 0.55 to 1.7 Ma cm^{-1} . F-doping of anion will expand electrochemical window to obtain high voltage cathode. In situ polymerization of monomer or PEO precursor of DOL/DME can form a conformal interface chemically bonded with two kinds of electrode, and [1] Graphene/GO nanofiller can also be used in existing cell-assembly production lines, which can be said to be a structural evolution of these methods. Because (like plasticizer) will destroy the crystallinity of polymer, (like ceramic filler) will enhance the mechanical hardness. There is a functional group (epoxy group, hydroxyl group, carboxyl group, etc.) containing many oxygen on the surface of GO sheet. They can be closely linked with polymer chains (such as ether-oxygen bond of PEO) and anions of lithium salts (such as TFSI⁻) [6]. Thanks to this connection, the crystalline part of the polymer chain collapsed and the amorphous part increased. Therefore, there can be more free space and fast channels for lithium ions to move. At the same time, the functional groups on the GO surface also serve as the location of Lewis acid. Combined with the anion of lithium salt, it helps lithium salt to disperse and increases the number of free lithium ions. The research shows that the difficulty of ion passing through polymer electrolyte is usually increased by 1~2 orders of magnitude by adding appropriate amount of GO.

2.3.2 Enhancement of Mechanical and Electrochemical Properties

Graphene is very hard and strong, which is very suitable for mechanical reinforcing agent. Graphene flakes mixed in polymer will establish physical contact points. This can improve the mechanical strength and modulus of electrolyte and prevent the penetration of lithium dendrite. If graphene is added, the electrochemical stability window of electrolyte will be expanded, especially the oxidation strength of lithium metal will be improved.

More importantly, evenly distributed graphene flakes can establish an electrified network in electrolyte. When the front end of dendrite begins to extend and contact graphene, the electricity gathered in the narrow part will spread to the whole graphene network. In this way, the "tip charge dispersion" is realized, and the extension speed of dendrite will become slower. Due to the effect of physical strength and electrochemical stability working together, the time and safety of using polymer electrolyte's solid-state batteries can be greatly improved [12].

2.4 Comparative Discussion

The interfacial layer of graphene can produce very good results in all parts of solid-state batteries (anode end, cathode end and electrolyte matrix). However, there are obvious differences in the structure of its main function, the shape of material and the purpose of use (recorded in Table 1). In order to better understand what role graphene played in various occasions and what effect it played together, we will compare and discuss it in this part.

Table 1. Comparison of Graphene Interface Layers at Different Locations in Solid-State Batteries.

Dimension	Anode-Side Interlayer	Cathode-Side Interlayer/Additive	Electrolyte Matrix Additive
Core Function	Modulate Li plating; passivate anode surface.	Facilitate charge transport; accommodate strain.	Optimize ion transport; stabilize bulk phase.
Key Mechanism	Surface functional groups homogenize Li^+ flux; vertical arrays guide uniform deposition.	3D scaffolds provide continuous e^- pathways; hybrid phases bridge interparticle contacts.	Nano-fillers disrupt polymer crystallinity; reinforce mechanical strength against dendrites.
Representative Materials	fGO, VGNs, Graphene/LiF composites.	Graphene/active-material hybrids; interfacial buffer layers.	GO/functionalized graphene in PEO matrices.
Performance Target	High Coulombic efficiency; long-term cycling stability.	High areal capacity; enhanced rate capability.	High RT ionic conductivity; wide electrochemical window.
Interfacial Synergy	Enables stable operation under high-current conditions.	Reduces internal resistance of the composite electrode.	Provides a robust matrix for effective interfacial engineering.

Although it is used in different places, the function of graphene basically comes from its unique two-dimensional structure and adjustable physical and chemical properties. In anode, the function of graphene is to focus on the chemical aspects and the adjustment of fine structure, so it is very important to solve the inherent instability of lithium metal. In cathode, focusing on physical affairs and manufacturing large structures, it is a subject to ensure the smooth transfer of charges. In electrolyte, it has the function of manufacturing and changing the properties of nanocomposites, with the goal of improving the properties of electrolyte itself.

It should be noted that these three functions are different. To make an ideal solid-state battery system, it is necessary to coordinate the three well together. For example, only by combining the anode interface layer which can make lithium uniformly pile up and the cathode composite electrode which can quickly transport charges on the electrolyte which has changed its properties with graphene, can the battery achieve high energy density. The next interface design should have the perspective of “the whole battery” and consider how to match the functions of graphene on different interfaces well.

3. CONCLUSIONS AND OUTLOOK

3.1 Summary

This paper, graphene interface engineering solves the important interface problems of solid-state batteries, and carefully investigates the methods of lithium ion passing and the rules of adjustment. The conclusions of the center can be summarized as follows:

On the anode side, graphene can adjust the chemical properties of the surface (such as functional group containing oxygen) to make the lithium ion flow evenly and reduce the force of scattered precipitation. In addition, by considering the microstructure (such as longitudinal arrangement), a physical closed space is created for lithium metal, which can guide uniform precipitation and alleviate the volume change. At the same time, the artificial SEI film with more LiF and other components is induced, which has high ionic conductivity and mechanical strength. Working together, these mechanisms can effectively reduce the interfacial resistance between anode and electrolyte, greatly inhibit the stretching of lithium dendrites, and improve the stability and safety of lithium metal anode charging.

On the cathode side, the core value of graphene lies in establishing a mixed network through which ions and electrons can pass efficiently. Through its amazing two-dimensional conductivity, the particles of active substances are wrapped in it,

and fast electrons are formed on the whole composite cathode. At the same time, it acts like a nano-scaffold, which is combined with the particles of solid electrolyte, and the bending is not so tortuous. The structure of the “double continuous network” is continuous, which greatly shortens the passing distance of lithium ions and the charging movement of the boundaries between solids.

On the basis of electrolyte, graphene partners like graphene oxide play the role of nano-fillers. Because it is related to the part of the polymer and the anion of lithium salt, it will prevent the polymer from crystallizing and make the lithium salt easy to separate. Therefore, at the room temperature (room temperature) of the polymer electrolyte, because of its amazing mechanical properties, the electrolyte is not easy to puncture, and the structure of the conductive network they made generates current (current) at the front end of dendrites. From the physical and electrochemical aspects, the stability of the electrolyte itself and the safety of the battery have become better.

3.2 Outlook

First, regarding mechanistic studies and the expansion of material systems, exploration is needed at deeper levels and across broader dimensions. Future research should fully leverage in situ and operating-condition characterization techniques to observe, in real time under actual electrochemical conditions, the dynamic behavior of lithium ions at the interface, the evolution of deposition morphology, and the formation and evolution of interfacial phases, thereby providing direct evidence to support existing mechanisms. At the same time, research should move beyond a focus on graphene alone to systematically study and compare the performance and mechanisms of other two-dimensional materials, organic-inorganic hybrid layers, or biomimetic interface structures. By establishing a structure-property relationship map linking “material characteristics—interface behavior—electrochemical performance,” researchers can identify superior or complementary interface materials.

Secondly, many engineering and scientific problems need to be solved quickly for practical problems and full-cell integration. Cost and scalable fabrication are the main obstacles. Therefore, it is very important to develop a cheap and high-quality manufacturing method and coating technology for graphene films or composite slurries, which is suitable for the current battery manufacturing methods. Long-term stability and compatibility of interfaces are other important issues. In order to avoid new side reactions and failure modes, the chemical and electrochemical compatibility between the interface layer of graphene and various solid-state electrolyte should be improved under various conditions of voltage, temperature and cycle times. The most important thing is that most of the current research only stays at the level of symmetric cell and half-cell. In the future, the long-term cycle performance, safety and energy density must be evaluated as a whole through the work of confirming the composition of the actually used full-cell.

Finally, at the level of cross-scale design and manufacturing integration, cooperation in various fields is needed. Understanding the interaction mechanism of interface at atomic scale and predicting the precipitation of lithium and the change of interface at intermediate scale-by combining this multi-scale calculation and simulation, the “reasonable design” of interface layer is realized. At the same time, it is necessary to find specific technical ways to integrate the interface layer of graphene into the new battery manufacturing process, solve the problems of uniformity, adhesion and fit with the whole process in mass production, and finally fill the gap between new technology and industrial application in the laboratory.

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