How to Achieve Next-Generation High-Performance Lithium-Ion Batteries for Electric Vehicles

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Abstract. Due to severe environmental issues, rapidly increasing attention has been paid to electric vehicles (EV) in the past decades. However, in the choice of batteries for EVs, nearly all car makers select lithium-ion batteries (LIBs) to power their vehicles for their great properties. LIB generally consists of cathode, anode, and electrolyte; these components greatly determine the performance of LIBs together. Therefore, analyzing the property of electrodes and electrolytes is extremely critical to achieving next-generation high-performance LIBs. Based on this, this review first summarizes several popular cathode materials, with their remaining challenges, such as degradation issues and provides some viable improvement strategies. Then, two categories of typical anode materials are discussed in detail, including their bottleneck issues and some protective method such as surface modification. In the end, this paper compares the merits and drawbacks of liquid and solid-state electrolytes and offers some possible solutions, respectively. This paper aims to provide a comprehensive reference to LIBs and direct further development for next-generation LIBs.

Keywords: Lithium-ion battery, Electric vehicle, Electrode material, Electrolyte.

1. Introduction

Because of the energy crisis and increasingly serious environmental problems, people pay more attention to renewable energy. Thus, as the main source of fossil fuel consumption and environmental pollution, traditional vehicles have been gradually replaced by electric vehicles (EV). As the most important part of EV, batteries have developed rapidly in the past few decades, and there have been many different battery systems developed to satisfy different requirements of people, including lead-acid batteries, lithium-ion batteries (LIBs), alkaline batteries (disposable), nickel-cadmium batteries (NiCd), zinc-carbon batteries (disposable), nickel-metal hydride batteries (NiMH), and Li-metal batteries, etc. Among these, LIBs are advantageous in the application of EVs due to their outstanding energy density (160% greater than NiMH, 220% greater than NiCd), high operation voltage (about 3.8 V) and long cycle life(lower discharge loss than NiMH and NiCd). Although Li metal battery can provide higher energy density, its safety issues cannot be effectively tackled, for which almost all car manufacturers still take LIBs as their first choice for the power source of their EV. A complete lithium-ion battery comprises two Li^+ intercalation electrodes (anode and cathode) and an electrolyte responsible for Li^+ conduction. The energy conversion between chemical energy and electric energy in LIBs is via the reciprocating movement of Li^+ and electrons between electrodes, as is exhibited in Figure 1 [1]. However, nowadays, the most advanced LIBs cannot satisfy people's expectations for ideal EVs (long-distance per charge, short charge time, long lifespan and great stability in extreme conditions). To meet this increasing EV demand, LIBs must further improve energy density, power density, and cyclability, especially at elevated temperatures. For this purpose, these components' property needs further investigation to enhance their performances. In this article, the commonly commercialized electrodes' materials (cathode: NCA, NMC, LMO; anode: graphite and silicon) and electrolytes (liquid-state electrolyte and solid-state electrolyte) will be introduced, with their challenges and some effective solution. With all these topics covered, this review paper is expected to function as an introductory beginning point for new researchers on lithium-ion batteries and provide some useful information and function as a reference book for experts.

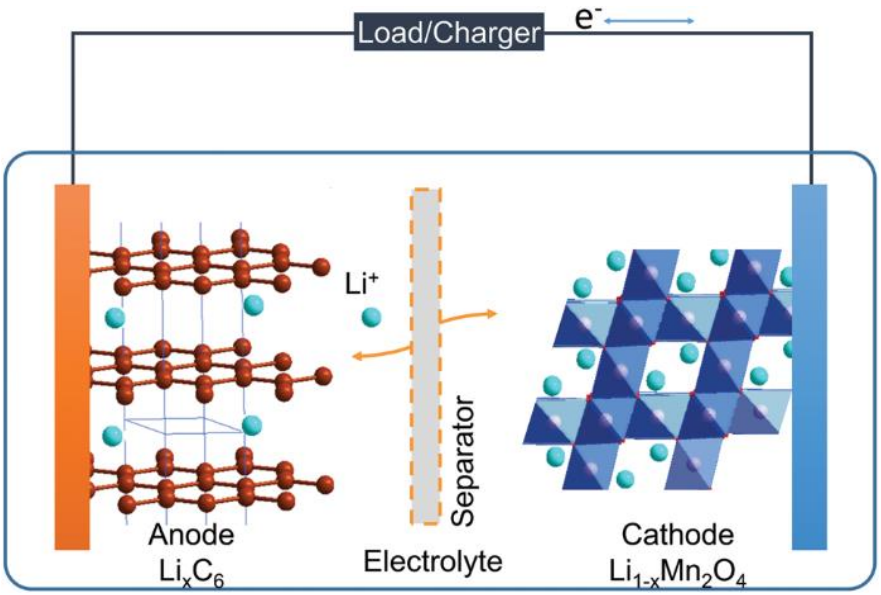

Figure 1 The basic principle of a LIB [1].

2. Cathode materials

In a LIB system, the cathode plays an important role and is responsible for providing the Li^+. Therefore, the battery's performance, to a great degree, depends on the cathode property. This section will introduce the advantages and disadvantages of three commonly used cathode materials and some solutions to tackle their issues.

2.1. NCA

Since Tesla firstly used NCA chemistry as cathode material on their EVs (model S), this particular chemistry has received increasing attention because it can offer great energy density, cycling performance, charge performance and potentially low cost. Despite such advantages, improving NCA materials' storage property, cyclability, and safety remains challenging. Further efforts are needed to overcome the above challenges of NCA materials.

Nano-coating was widely adopted as an effective strategy to improve NCA's thermal stability and electrochemical performance. In general, some stable materials were chosen to cover the surface of NCA, and this stable layer can prevent the electrode from CO_2 and H_2O, which can enhance the storage property of NCA chemistry. By reducing detrimental chemical reactions between the electrolyte and NCA, this approach can improve the cyclability of NCA chemistry [2]. According to Zhou's research, SiO_2 is a golden choice for coating for it is cheap and environmentally friendly [3]. More importantly, this material can prevent electrode material from dissolution in electrolytes by reacting with HF, lowering polarization and increasing the stability of electrode structure during charge/discharge cycles. As exhibited in Figure 2 (a-b), SiO_2-coated NCA has better electrochemical performance than pristine NCA. Especially at high rates (5 C), SiO_2-coated NCA can provide almost 40 $mAhg^{-1}$ with higher reversible capacity meaning better cycling stability. In addition, due to the protective role of the SiO_2 layer, SiO_2-coated NCA can tolerate higher temperatures than before, meaning superior thermal stability, as exhibited in Figure 2 (c) [3].

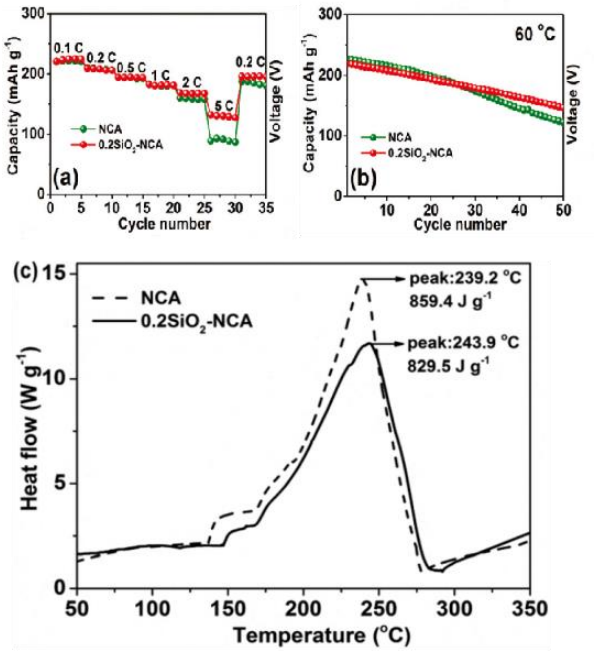

Figure 2 Comparison of (a) rate capacity, (b) cyclability and (c) thermal stability between SiO_2-coated NCA and pristine NCA [3].

2.2. LMO

In recent years, using doped lithium manganese oxide spinel as cathode material of LIBs has attracted more and more attention from many car manufacturers such as NISSAN and Chevy corporation. As a cathode, $LiMn_2O_4$ spinel can provide high energy density and great specific capacity (250 $mAhg^{-1}$, 2.5~4.5 V) [4]. In addition, to its high content of Mn, this kind of cathode material has the property of low cost and hypotonicity. However, one of the biggest challenges for LIBs with $LiMn_2O_4$ spinel is the severe fading of battery performance, including the capacity, cyclability, and power density caused by Mn^{2+} dissolution and deposition. Mn^{2+}, like transition metal ions, is supposed to maintain its original state in the cathode during the process of lithiation and delithiation. However, some Mn elements migrate to electrolyte, even solid electrolyte interphase (SEI), which changes the composition of SEI and deteriorates the protective role of SEI [5]. Nowadays, one of the most convincing dissolution and deposition models of Mn^{2+} is that Mn^{2+} dissolves in electrolytes, similar to the solvation manner of Li^+, then pierces the exterior SEI surface (ii), and deposits in the SEI interior structure, even on the surface of the graphite anode (i), as is shown in Figure 3(a) [6].

To mitigate this issue, many strategies have been studied to suppress the dissolution and deposition process of Mn^{2+}. Firstly, suppressing the dissolving process requires a stable cathode structure to defend against the acid attack from the electrolyte. Surface coating, generally, is regarded as an effective way to reduce manganese dissolution. Nano-coating with various oxides, lithiated metal oxides, fluorides, polymers, or phosphate on the cathode surface has been shown to provide a longer cycle life than pristine $LiMn_2O_4$ cathode since the coating materials can protect $LiMn_2O_4$ spinel from the attack of HF [1]. Additionally, adding some functional additives is another method used to suppress the dissolution of Mn^{2+}, which is conducive to forming a protective film on the NMC surface. LiBOB, for instance, is proposed to be an efficient addition to mitigate the Mn^{2+} dissolution due to the chemical reaction between Mn^{2+} and BOB^-, which form a chemically stable layer on the cathode [5]. As for the approaches to suppress the deposition of Mn^{2+}, According to Cho's study, vinylene carbonate (VC) additive can suppress manganese deposition. The schematic of the VC additive's protective role is exhibited in Figure 3(b) [7]. By adding VC, the components of SEI alter, with some polymer compounds forming, which can effectively defend against the attack of Mn^{2+} in the electrolyte. Moreover, choosing other functional additives, including cyclic ethers and NH^{4+}, capable

of forming Mn^{2+} complex, thus decreasing the manganese deposition potential and suppressing the deposition of Mn^{2+} [8].

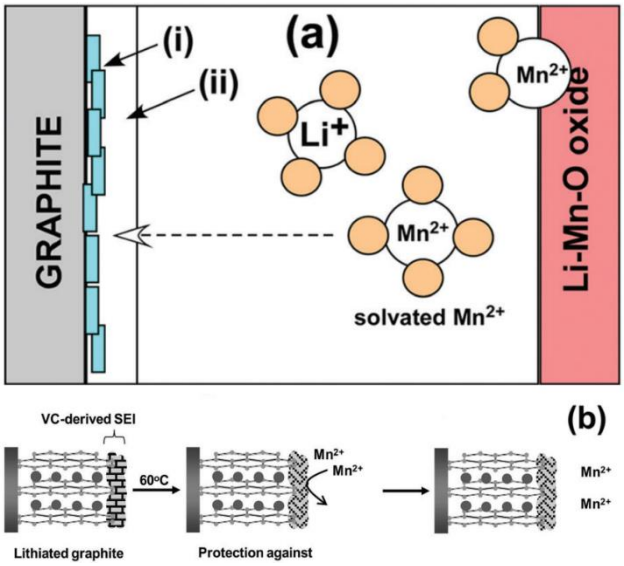

Figure 3 A schematic of (a)dissolution and deposition model of Mn^{2+} and (b) protective mechanism of VC [6].

2.3. NMC

Another class of promising cathode material is nickel-rich NMC ($LiNi_xMn_yCo_{1-x-y}O_2$, $x \geqslant 0.5$), layered oxides, which have the property of a great energy density of 220 $mAhg^{-1}$ and thermal stability. It is, therefore, advantageous to achieve commercialization in the EV industry. However, the degradation of NMC materials is the bottleneck issue, which results in the problem of severe capacity fading. The commonly proposed mechanism of NMC degradation is categorized into micrometer and atomic scales [9]. On the micrometer scale, in the Li^+ insertion/extraction process, the expansion and contraction of crystal lattices cause forceful micro-strain, which promotes the formation of micro-cracks, thus leading to secondary particles cracking. On the atomic scale, in the process of charge/discharge, there are a large number of high-valence ions generated, such as Ni^{4+}, $Mn^{3+/4+}$ and Co^{4+}, which induces the surface reconstruction and some parasitic reaction on the cathode surface, including the oxidization of non-aqueous electrolytes, O^{2-} self-redox reaction and NMC dissolution. The NMC degradation in both scales results in the loss of active materials and produces some detrimental substances such as corrosive compounds and surface impurities, thus severely aggravating the aging effect of LIBs.

Several promising strategies are applied to protect NMC from degradation, including dopants, gradient layers, coatings and carbon matrixes. Among these, dopants and coatings have efficiently mitigated NMC degradation. Cationic dopants such as Al^{3+}, Cr and Zr can enhance the cyclability of LIBs [10]. In addition, doping some anionic substances such as fluorine [11] can mitigate the side reactions and avoid the attack of HF. Coating Li_2SiO_3 onto the NMC cathode surface can also prevent electrode materials from detrimental reaction and strengthen its structural stability, thus significantly reducing capacity fading [12]. As is shown in Figure 4 [12], it is clear that compared with pristine NMC, Li_2SiO_3-coated NMC shows higher capacity retention and lower capacity loss proportion (about 12%) after 100 cycles. Meanwhile, this coating material can also facilitate the diffusion of Li^+ because Li_2SiO_3, as a great ionic conductor, can provide Li^+ with more channels to travel, which is a key point in decreasing the polarization of electrodes.

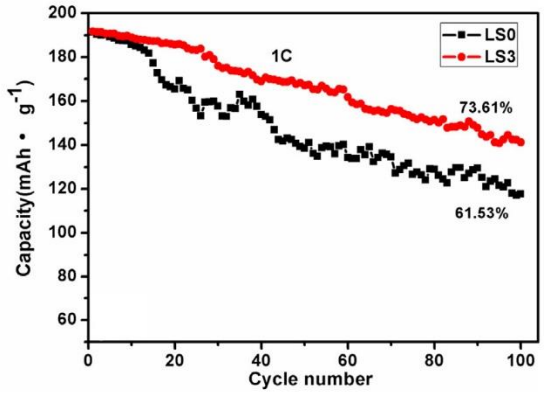

Figure 4 The comparison of cyclability between Li_2SiO_3-coated NMC and bare NMC [12].

3. Anode materials

3.1. Graphite-Based Anodes

Since 1994, almost all car makers have used graphite or its compounds as anode material for their EV's LIBs. The specific structure of graphite makes it a suitable and applicable host material. The intercalation of Li^+ into the graphite host structure abides by the "staging mechanism" [13]. Figure 5(a) exhibits the schematic illustration of the 'staging mechanism'. This model was initially created by Hofmann and Rudorff in 1938 and modified by Daumas and Herold afterward. This model shows that LiC_6 ($6C + Li^+ + xe^- \longleftrightarrow LiC_6$) is the highest possible Li capacity. In this situation, graphite's theoretical energy density can achieve a good level of 372 $mAhg^{-1}$ which can satisfy the demand of many electronic devices. In addition, the main advantage of graphite is its low de-/lithiation potential (only 0.15-25 V) which makes it superior to other alternatives.

However, there are still some issues with graphite. One major drawback of graphite is the first cycle's irreversible capacity (C_{irr}) caused by the reductive electrolyte decomposition. This harmful reaction results in the consumption of Li^+ and graphite, thus reducing LIB reversible capacity. One approach to reduce C_{irr} is surface modification. This method can be classified into three sorts, including inorganic, organic, and carbonaceous coatings. For inorganic coatings, many metals such as Ni, Al, Cu, Au, Pb, Ag, Zn, Pd and some metal fluorides and metal oxides--AlF_3 [14], Al_2O_3 [15] are used as the coating materials of the graphite particles' surface. As shown in Figure 5(b) [15], Al_2O_3-coated graphite is demonstrated to improve capacity retention effectively from 31.2% to 91.8% after 200 cycles. As for the carbonaceous coatings, numerous researchers are investigating them. They investigate that by employing spherical graphite particles, the carbonaceous coatings with optimum content and suitable graphite particle morphology can efficiently enhance electrochemical performance such as C_{irr} and energy density [16]. The polymer coating is also widely used in surface modification, which has two main advantages: high chemical diversity and mechanical flexibility.

Another problem with graphite is its low-rate capacity. Although adding some addictive into electrolytes can improve graphite's rate capacity by reducing the activation energy of the lithiation process, it still cannot completely meet people's expectations. Therefore, further research on improving graphite's rate capacity is needed to enhance LIB performance.

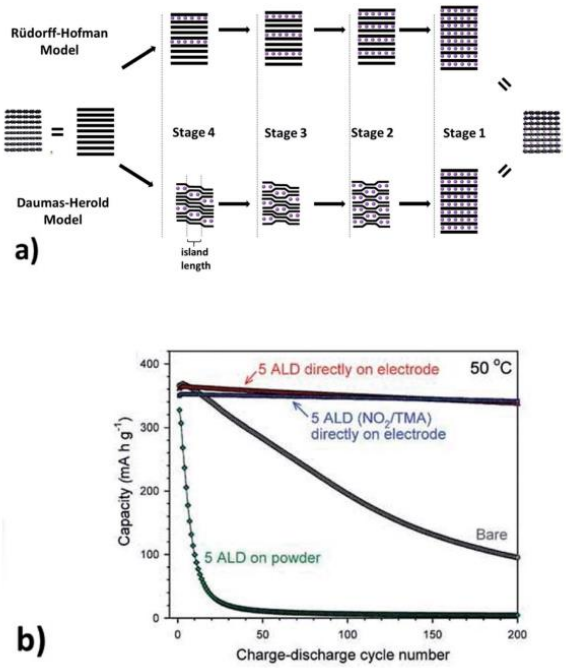

Figure 5 (a) the schematic of the 'staging mechanism' and (b) the cyclability of Al_2O_3-coated graphite and bare graphite [13, 15].

3.2. Si-Based Material

In recent years, car manufacturers and researchers have received great attention from the silicon-based anode. The theoretical capacity of common anode materials is exhibited in Figure 6 [13]. Si has an extremely high theoretical capacity of 3590 $mAhg^{-1}$ (nearly ten times more than graphite) and low operation potential (0.4~0.5 V), making silicon the most prospective material for next-generation LIB anode. Nevertheless, the huge volumetric change (about 370%) in the Li^+ insertion/extraction process is the bottleneck for commercializing LIBs with the silicon-based anode, which aggravates particle cracking and structural pulverization of silicon anode, thus resulting in severe capacity fading.

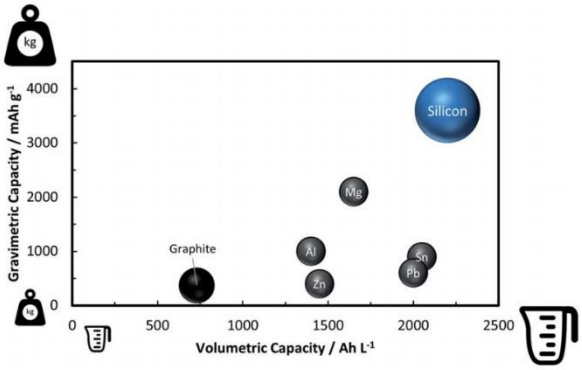

Figure 6 The comparison of theoretical capacity between several common anode materials [13].

Many strategies have been studied to mitigate this issue. Among these, some silicon nanostructure for anodes has been developed. In the one-dimension aspect, Si nanowires and nanotubes can provide a better cyclability with an excellent reversible capacity at 2000 $mAhg^{-1}$, but this nanostructure has been limited in several aspects such as poor electron conductivity and unstable SEI layer [17]. Adding chemical additives such as boron and phosphorous doping can provide better electron conductivity since these additives can drastically accelerate the electron transfer rate to the collector and anode plate by increasing surface area. Surface coating is a viable approach to protecting the SEI layer from breakdown. For example, the aluminum coating can create an artificial SEI layer with good

mechanical strength during the initial few charge/discharge cycles. In addition, the core-shell nanostructures have been reported to have a superb physical and electrochemical performance because the shell material can protect the core from the attack of other substances and limit core expansion, thus maintaining structural integrity. There are many categories of core-shell structure, such as solid core-shell, Yolk-shell, pomegranate-like structure and core-mesoporous shell. Among these, the pomegranate-like structure is one of the most promising structures (Figure 7(a)), which contributes to inhibiting the dramatic volume change and remains a great reversible capacity after long cycles, as is exhibited in Figure 7(b) [18].

4. ElectrolyteS

The electrolyte is another key component in the LIBs system. The main function of electrolyte is not only to allow Li^+ to crossover between electrodes but also to avoid electron conduction and short-circuit. This section will focus on several sorts of common electrolytes and introduce these materials' advantages, drawbacks and some possible solutions.

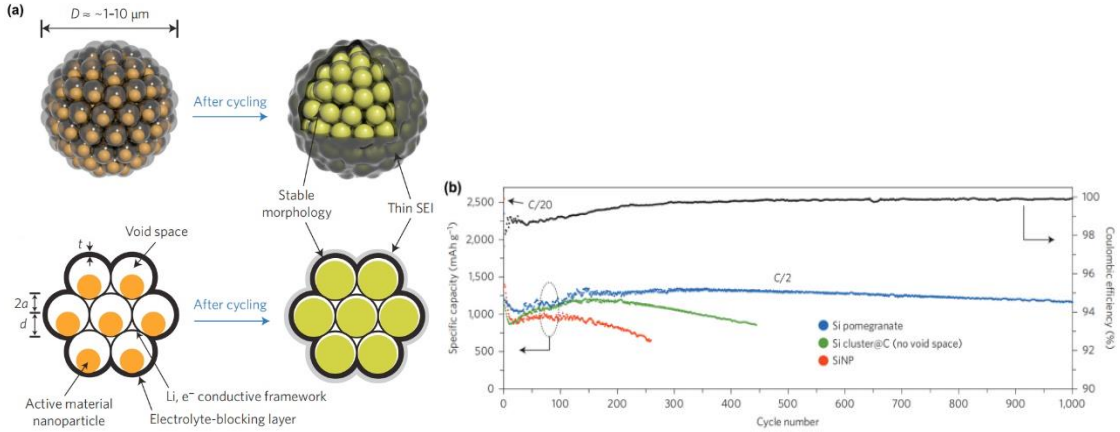

Figure 7 (a) The schematic of pomegranate-like structure and (b) reversible capacity of Si pomegranate and other structures after the first 1000 cycles [18].

4.1. Liquid Electrolytes

4.1.1 Organic Liquid Electrolytes

Generally, the most common organic liquid electrolytes are made of Lithium hexafluorophosphate ($LiPF_6$) salt dissolved in an organic carbonate solvent such as EC or PC. Traditional organic liquid electrolytes face the main issue of low voltage stability, which limits the voltage increase of the battery. Thus, scientists and researchers have been making numerous efforts to develop a new-type organic liquid electrolyte that can work in high voltage conditions. It has been reported that using organic fluoro-compounds as the solvent can effectively enhance electrochemical stability since fluorine atoms possess an extremely powerful electron-withdrawing effect, increasing fluorinated compounds' oxidation potentials [19]. In addition, SEI is crucial for the performance of LIBs with organic liquid electrolyte, which is generated on the electrode surface in several initial cycles. The principal responsibility of SEI is to protect electrodes from the chemical reaction with organic electrolytes, thus reducing the consumption of electrode and electrolyte materials. Therefore, the properties of SEI, including thickness, structure, and constitution of SEI, greatly determine the performance of LIBs, such as cyclability, irreversible charge loss, thermal stability, rate capability and safety. It has been reported that adding some additives can facilitate controlling SEI, thus improving the performance of LIBs to meet the different needs. For example, using HiFP as an additive can effectively improve the stability of organic electrolytes in slightly higher voltage conditions (about 5 V class) [20]. However, the safety issue of organic liquid electrolytes is still a challenge, which greatly limits the further development of LIBs with organic liquid electrolytes.

4.1.2 Aqueous and Ionic Liquid Electrolytes

Other promising liquid-state electrolytes are aqueous electrolytes and ionic liquid electrolytes. Generally, the aqueous rechargeable lithium-ion battery (ARLB) is divided into several categories according to different lithium salts solubilized in water, such as $LiNO_3$ and Li_2SO_4. Compared with organic liquid electrolytes, aqueous electrolytes have the excellent property of being non-flammable, allowing the application of larger-scale LIBs. But the aqueous electrolytes are facing a serious issue: the narrow electrochemical stability window (ESW) of liquid water, only 1.23 V. This narrow ESW limits the commercialization of ARLB. It has been reported recently that dissolving LiTFSI at extremely high concentrations in water can greatly expand the ESW to ~3.0 V, as shown in Figure 8 [21].

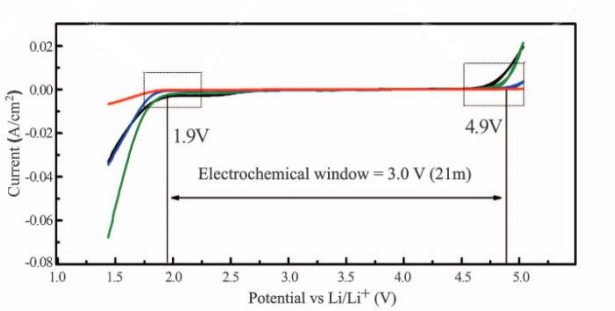

Figure 8 The ESW of LiTFSI-H_2O electrolytes [21].

As for ionic liquid electrolytes, due to their excellent non-flammable, non-volatile, large ESW, stable electrochemical stability, and remarkable ion conductivity [22], more and more attention is paid to the research on ionic liquid electrolytes. But the main drawbacks of ionic liquid electrolytes are their high viscosities and high cost. The former greatly decreases mobility and conductivity, severely limiting the application of ionic liquid electrolytes. Searching for a new combination of cations and anions is a potential approach to tackle this problem. Lastly, balancing the property and cost of ionic liquid electrolytes is key to achieving its commercialization.

4.2. Solid Electrolytes

Compared with liquid-state electrolytes, solid-state electrolytes have better thermal stability, which facilitates thoroughly tackling the safety issue of LIBs. This advantage makes solid-state electrolytes more promising in the next-generation high-performance LIBs. Compared with other solid-state electrolytes, Garnet-type, perovskite-Type and NASICON-type electrolytes are the most prospective for commercialization, especially for EV applications [23]. The following paragraphs will discuss their advantages and issues, respectively.

4.2.1 Garnet-Type

In the late 1960s, garnet-type of solid electrolytes were firstly prepared. With higher requirements for electrolyte electrochemical performance, many garnet-type solid-state electrolytes developed, including $Li_3Ln_3Te_2O_{12}$ and $Li_5La_3M_2O_{12}$ (M = Ta, Nb). Recently, an advanced garnet-type of solid-state electrolyte, $Li_7La_3Zr_2O_{12}$ (LLZO) [24], comes into people's minds, which has an extremely high ionic conductivity (about 3×10^{-4} S cm^{-1}). It has been discovered that the cubic phase shows greater ionic conductivity compared to the tetragonal phase. Therefore, maintaining the cubic phase of LLZO in a wide temperature range is critical to keeping ionic conductivity high. According to S. Ohta's study, doping some elements such as Al^{3+} and Nb^{5+} into LLZO can further enhance ionic conductivity and widen ESW because these elements can stabilize the cubic crystal structure of LLZO [25], but LLZO lacks stability in the air because of moisture and CO_2 [26], which limits the application of LLZO in the EV industry. So, improving LLZO's stability in the ambient atmosphere is the key to achieving commercialization.

4.2.2 NASICON-Type

One promising NASICON-type solid-state electrolyte is $Li_{1+x}Al_xGe_{2-x}(PO_4)_3$ (LAGP), which has great ionic conductivity. Through the process of melt-quench and recrystallization, LAGP glass-ceramics can greatly improve its ionic conductivity to an excellent level (about 4×10^{-4} S cm^{-1}) [27] because compared with the traditional manufacturing process, this technique can form denser microstructure, which provides closer contacts between grains, thus increasing the ionic conductivity.

However, the preparation of LAGP costs a lot because GeO_2 is expensive, which is one of the precursors of LAGP [28]. Therefore, using other cheaper elements to reduce the content of GeO_2 in the LAGP needs further research, but it is necessary to keep high ionic conductivity while replacing Ge with other cheaper elements. Therefore, how to balance the cost and performance is an urgent problem to be tackled.

4.2.3 Perovskite-Type

Since the first kind of perovskite-type solid-state electrolytes, LLTO [23], was developed by Inaguma, perovskite-type solid-state electrolytes have received great attention from researchers. Although LLTO has a high bulk ionic conductivity, its low grain boundary ionic conductivity limits its total ionic conductivity. Poor stability in low voltage conditions is another limitation for LLTO, meaning lithium and other low potential anode materials are incompatible with this electrolyte. With deeper research on perovskite-type solid-state electrolytes, more categories of perovskite-type solid-state electrolytes have been developed. LSTZ/LSTH [23]are two promising perovskite-type solid-state electrolytes with higher total ionic conductivity and wider ESW, meaning better electrochemical performance. But they still have many issues. The main challenge is that the preparation condition is too harsh, which requires an exceedingly high preparation temperature (about 1300 ℃); thus, optimizing its manufacturing technology is necessary to achieve the commercialization of perovskite-type solid-state electrolytes.

5. Conclusion

To popularize the EV industry, people raise further requirements for next-generation LIBs. It is expected to have a higher capacity, better cyclability and more reliable safety. To this end, as the main components of LIBs, cathode, anode and electrolyte are proposed to have better electrochemical performance and structural stability. This article lists the challenges and some solutions for these critical components. Firstly, commonly used cathode materials usually have degradation problems due to the harmful chemical reactions with electrolytes, which rapidly reduce the cycling stability of LIBs. Whether nano-coating or adding some additives are effective strategies to tackle this issue, in the future, more effective coating materials and addictive will be developed for the next-generation LIBs. Then graphite and silicon, as two sorts of potential anode materials, are discussed in detail. Focusing on their respective strengths and weaknesses, combining graphite with silicon or silicon oxide is a key point in preparing the next-generation LIBs anode to achieve higher energy density. Accordingly, making high-performance anode material requires further investigation to balance the volumetric change and energy density. Besides, as for electrolytes, several promising liquid-state electrolytes and solid-state electrolytes are introduced in this paper. Compared with conventional organic liquid electrolytes, advanced solid electrolytes are deemed to improve battery safety and volumetric capacity, which is a great choice for next-generation LIBs. But the capacity between solid-state electrolytes and electrodes should be further studied. Recycling LIBs is another critical challenge for next-generation LIBs, which needs further efforts to convert discarded batteries to environmentally friendly and reusable materials efficiently.

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