Rechargeable Battery Electrolytes

Electrochemical Energy Storage from Liquids to Solids

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Preface

Energy is an important part of our daily life, and we could not do without it. Our dream is to use electronic equipment readily at all times, even when playing sport or climbing mountains. For this we need batteries. At present, lithium-ion batteries are dominant for energy storage devices. While other novel devices have also been developed, they still cannot be used in our daily lives as the technology is not yet safe and robust.

Electrolytes are a crucial part of batteries. Although little attention has been paid to electrolytes in the past, their importance in improving the electrochemical performance of batteries has been realized. Electrolytes influence the two electrodes due to the electrochemical reactions, and also determine the interphase behavior between the electrolyte and the cathode and the electrolyte and the anode, called the solid-electrolyte interphase (SEI) and the cathode-electrolyte interphase (CEI). The SEI affects the performance of the Coulombic efficiency and the cycling life, while the CEI influences not only the Coulombic efficiency and cycling life, but also the tolerance of high voltage, which controls the energy density. Thus, electrolytes are important for battery performance, and this is one of main reasons for writing this book.

Currently, there are few books that deal specifically with electrolytes, which is another reason why this book is needed. To make the book as useful to students and researchers as possible, I have included many electrolyte topics, such as aqueous electrolytes, organic electrolytes, gel electrolytes, polymer electrolytes, solid electrolytes, and ionic liquid electrolytes. These are used for lithium-, sodium-, potassium-, zinc-, and magnesium-ion batteries, and others. The book covers a wide range of topics and I welcome readers' comments on coverage and content to improve the book in later editions. Finally, I wish to express my thanks to my students, Gaoxue Jiang, Huaping Wang, Shihan Qi, Chuan Wang, Xi Tang, Daxiong Wu, Yulu Yang, Xin Li, and Fang Li for their help with writing this book.

Jianmin Ma

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Author Biography

Dr Jianmin Ma is a professor at Tiangong University, Changsha, China. He serves as an Associate Editor of Chinese Chemical Letters and an Academic Editor for Rare Metals and is an editorial board member for the *Journal of Energy Chemistry*, *Chinese Chemical Letters*, *Nano-Micro Letters*, *Journal of Physics: Condensed Matter*, *Journal of Physics: Energy, Chemistry – An Asian Journal*, and others. His research interests focus on energy storage devices and components including metal anodes and electrolytes, and theoretical calculations from density functional theory and molecular dynamics to finite element analysis.

Dr Zhouguang Lu is currently a full professor in the Department of Materials Science and Engineering at Southern University of Science and Technology. He received his PhD degree from the City University of Hong Kong in 2009. He was elected a Fellow of the Royal Society of Chemistry (FRSC) in 2018. He has published more than 300 SCI papers with total citations of >13 000 and an H-index of 65. He joined the editorial board of *Nano Research* in 2013. His research is focused on the capture and mechanism investigation of electrochemical intermediates in advanced battery materials.

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CHAPTER 1

Introduction to Electrolytes for Rechargeable Batteries

1.1 Background

Since the appearance of lead-acid batteries, various rechargeable batteries have been studied due to their wide range of applications in various fields over the past 100 years.[1–5] Many rechargeable batteries have been developed and explored,[6] *i.e.*, nickel metal hydride batteries, nickel-cadmium cells, zinc manganese batteries, lithium batteries,[7–13] lithium-ion batteries,[14–16] lithium-sulfur batteries,[17] lithium-air batteries,[18,19] sodium-ion batteries,[20–22] zinc-ion batteries, zinc-air batteries,[23] potassium-ion batteries,[24–26] calcium-ion batteries, aluminium-ion batteries, and so on. The development of batteries has driven scientists to explore new types and structures of electrode materials, which has contributed to progress in battery technology.[27] In particular, over the past three decades, great progress has been made in materials technologies, which can be attributed to the large demand for new materials in many fields. Therefore, research and development have received significant attention from the viewpoint of materials science. In addition, the pursuit of green energy also drives the development of batteries as energy storage devices (Figure 1.1).[28]

The basic components of batteries include the cathode, anode, separator, and electrolyte. The typical structure of a lithium-ion battery is illustrated in Figure 1.2.[29] The cathode and anode are very important, and determine the voltage of the as-constructed batteries. The separator is necessary to block interaction between the cathode and the anode and thus prevent short circuits. The electrolyte is also an extremely important component, since it provides ion transport between the cathode and the anode, not only influencing the performance of the battery, but also the interface between the electrolyte

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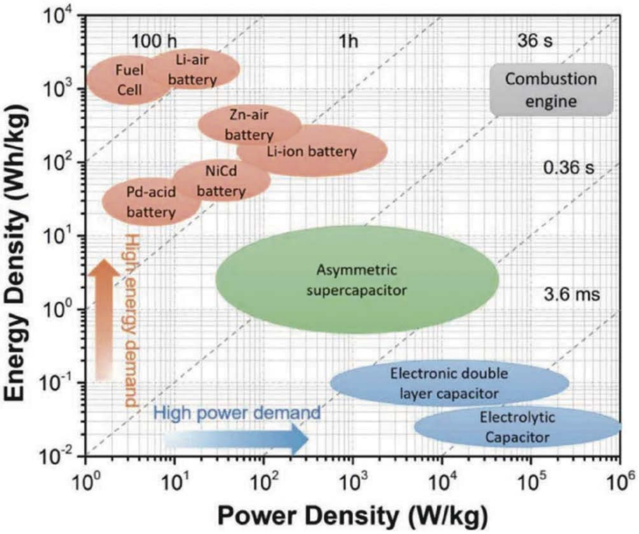

Figure 1.1 Ragone plot illustrating specific power *versus* specific energy for various types of battery. Reproduced from ref. 28 with permission from American Chemical Society, Copyright 2018.

and the cathode, the anode, and the separator. Therefore, electrolytes are considered to be multi-functional components of batteries.

1.2 Electrolytes

Traditionally, electrolytes can be divided into three types: aqueous (salts, acid, alkaline),[30–33] non-aqueous (organic solutions, ionic liquids),[34–39] and solid-state electrolytes (inorganic, gel, and polymer electrolytes).[40–46] Theoretically, any battery can employ the above-mentioned electrolytes, however, this is influenced by the limits of current technology. Thus, different batteries may have different kinds of electrolytes. For example, due to the maturity of the technology for lithium-ion batteries, various types of lithium-ion batteries have been developed using aqueous electrolytes, non-aqueous electrolytes (including ionic liquid electrolytes), inorganic electrolytes, gel electrolytes, polymer electrolytes, and their hybrids. This book covers all these electrolytes, providing a comprehensive description of aqueous and non-aqueous electrolytes for lithium-, sodium-, and zinc-ion batteries; non-aqueous electrolytes for potassium-, calcium-, magnesium-, and aluminium-ion batteries; solid electrolytes for lithium- and sodium-ion batteries; ionic liquid electrolytes for lithium-, sodium-, potassium-, zinc-, and aluminium-ion batteries, and lithium metal batteries; gel and polymer electrolytes for lithium-, sodium-, potassium-, zinc-, magnesium- and aluminium-ion batteries; and high-concentration and local high-concentration electrolytes for lithium-, sodium-, potassium-, and zinc-ion batteries.

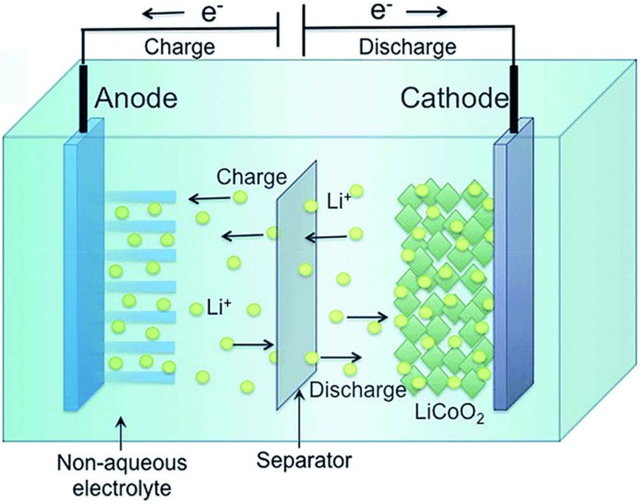

Figure 1.2 Structural illustration of a typical lithium-ion battery. Reproduced from ref. 29 with permission from the Royal Society of Chemistry, Copyright 2015.

1.3 Characteristics of Electrolytes

Electrolytes play an important role in determining battery performance, which is also influenced by the electrode materials. In this book, we discuss the battery performance only from the viewpoint of the electrolyte. In particular, we take the electrolyte chemistry of lithium metal batteries as an example.[47] The electrochemical performance of lithium metal batteries (cycling performance and rate capacity) is determined by many factors, *i.e.*, solvation energy barrier,[48,49] the components and structures of the solid electrolyte interphase/cathode electrolyte interphase,[50,51] the wetting ability of the electrolyte on the separator,[52] and ionic conductivity. To improve the overall performance of batteries, these factors need to be adjusted, *i.e.*, lowering the solvation energy barrier, improving the Li^+ conductive properties of the solid electrolyte interphase/cathode electrolyte interphase through adjusting components and structures, increasing the wetting ability toward the separator, and enhancing the ion conductivity of the electrolyte. The properties of electrolytes are influenced by the electrolyte salt and solvent, which can also be tailored with the use of additives.

The cost of electrolytes is an important index, and is determined by the cost of the salts, solvents, and additives used. Thus, aqueous electrolytes, for various rechargeable batteries, are considered to be cost-effective, while ionic liquids are considered to be high cost.[53,54] Organic compounds are also expensive, so the cost of most additives will also be considered when they are used in the electrolyte.[55,56] For some electrolytes, the balance between cost and performance should be considered when deciding on their use.

Safety is also important for rechargeable batteries.[57] Safety is influenced by the cathode, anode, and electrolyte. Aqueous electrolytes are considered to be safe due to their characteristics of non-flammability. For non-aqueous electrolytes, the exploration of non-flammable electrolytes is considered for safety reasons. Inorganic solid electrolytes and ionic liquids are also considered to be safe, but their cost needs to be reduced. As they are expensive, their performance and safety need to be balanced with cost when considering their use.

1.4 Trends in Electrolytes

Battery electrolytes can be considered to be one of the most important technologies in applied sciences. Research and development into electrolytes should receive enough attention. For future electrolytes, the native characteristics of the electrolyte should first be considered, *i.e.*, safety, cost, green characteristics. In addition, performance characteristics should also be considered, *i.e.*, high voltage electrolytes, low-temperature and high-temperature electrolytes, and wide electrochemical window. Finally, for some purposes, a crucial factor is the use of well-tailored additives for the electrolyte.

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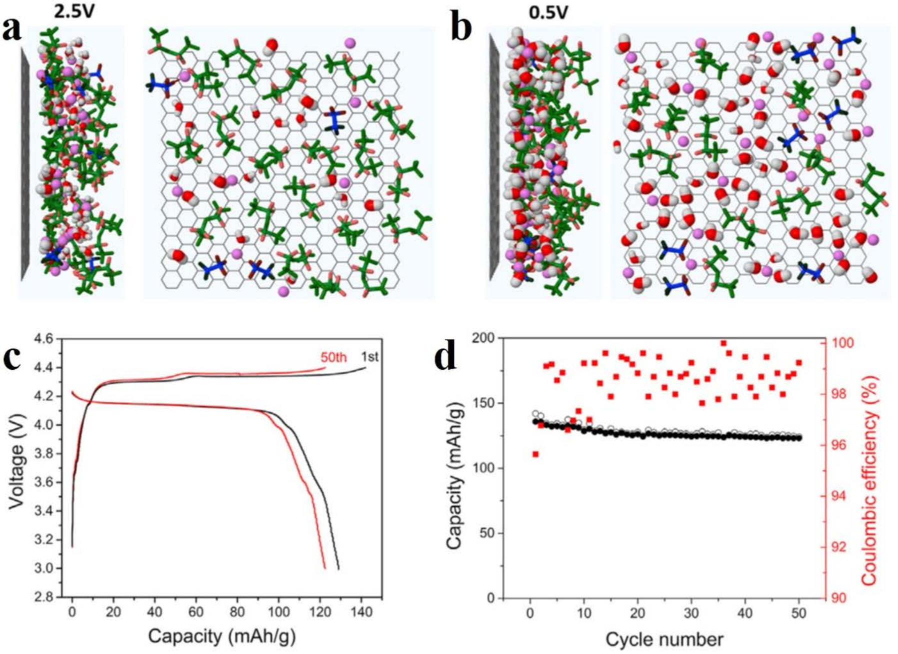

Figure 2.20 (a and b) Snapshots of inner-Helmholtz interfacial regions of the anode surface in WiBS. Reproduced from ref. 108 with permission from National Academy of Sciences. (c) The charge–discharge voltage profiles and (d) cycle performance of various 4.0 V aqueous Li-ion batteries. Reproduced from ref. 109 with permission from Elsevier, Copyright 2017.

large databases or artificial intelligence are some of the possible solutions. In addition, models with predictive capability for the interactions between electrolytes and electrodes should be developed. Finally, solid electrolytes with high safety performance are also an important direction for battery development. Although there is no satisfactory electrolyte with high energy density and high safety performance at present, we believe that these issues could be resolved eventually with the development of advanced theoretical calculation tools and novel electrolyte systems.

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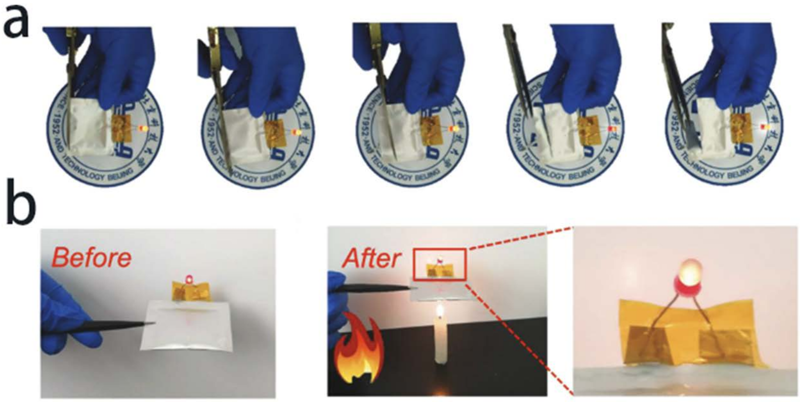

Figure 4.22 (a) Cutting off part of a solid-state aluminium-ion battery can steadily power a light-emitting diode (LED) lamp. (b) Solid-state aluminium-ion batteries for lighting LED lights when burning in a fire. Reproduced from ref. 135 with permission from John Wiley & Sons, Copyright 2019.

greatly reduced, and the energy density is comparable to that of Li-ion batteries. These ion batteries show good prospects for applications in energy storage equipment. Here, we have introduced non-aqueous electrolytes for these metal-ion batteries, first explaining the working principles of the batteries, and then comparing the advantages and disadvantages of types of non-aqueous electrolytes for different ion batteries. Although non-lithium-ion batteries have developed rapidly in recent years, there is still a long way to go before they are actually put to use. The reaction mechanisms in some ion batteries are not yet fully understood, so principles for an adjustment strategy to design electrolytes cannot be adopted. Therefore, the development of non-aqueous electrolytes for non-lithium-ion batteries requires more basic work to reach a higher level of battery development.

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In addition to the above, the performance of Zn-ion batteries can be disrupted by multiple factors. For example, variation in temperature has a great effect on Zn-ion batteries. During battery operation, a portion of the energy will be converted into heat and released, resulting in temperature rise and electrolyte volatilization. When the ratio of electrolyte to zinc salt is lower than the critical value for solubility, zinc salt precipitates, resulting in a sharp decline in battery performance. This cannot be ignored as when the temperature of the battery rises sufficiently the consequences are potential safety hazards and even the risk of explosion, which threatens life and property. On the contrary, when the ambient temperature is too low, the electrolyte viscosity increases, and solvents with low melting points will partially solidify, and so the Zn^{2+} conduction impedance (R_{ct}) increases significantly. When the ion velocity in the electrolyte cannot keep up with the electron conduction speed in the external circuit, the battery will become seriously polarized. Therefore, the key aim for research in low-temperature environments is to find solvents with low viscosity, high melting point, and high conductivity. The preparation of electrolytes with a wide temperature range, to ensure flexibility of use and mechanical stability at low temperature, is a general direction for Zn-ion battery research in the future. In addition to temperature, environmental factors, such as humidity and external force, have a great impact on Zn-ion batteries in practical application scenarios. At present, the interfacial behavior between the electrolytes and the electrodes is unclear. Research can address this through in-depth study of the internal behavior of the electrolyte, and thus provide theoretical support for the development of a new generation of Zn-ion batteries.

In conclusion, Zn-ion batteries are promising substitutes for lithium-ion batteries, with rich sources of raw materials, low price, high safety, and environmental friendliness. Compared with the current Li-ion battery (about \$300 kW^{-1} h^{-1}), the price of the Zn-ion battery (\$65 kW^{-1} h^{-1}) is much lower, even being equivalent to a lead-acid battery. Thanks to the high density and double electron reaction mechanism of zinc, the volume density of the Zn-ion battery (5855 mA h cm^{-3}) is more than twice that of the Li-ion battery (2061 mA h cm^{-3}). The high reversibility of zinc coating/stripping means that Zn-ion batteries have an ultralong cycling life. The five types of electrolytes discussed in this chapter have different characteristics and are suitable for different application environments. Exploring new electrolyte systems and combinations, such as ionic liquids with water, and gel and solid electrolytes, promotes the development of electrolytes for Zn-ion batteries. As an important part of the battery, the electrolyte needs more attention, to meet the requirements for a new generation of energy storage devices.

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Much research has been carried out on solid electrolytes, especially to enhance the ionic conductivity of the inorganic solid electrolyte and reduce its interface impedance. Researchers have studied different types of inorganic solid electrolytes. The conductivity of some solid electrolytes is about the same as that of liquid electrolytes. Common inorganic solid electrolytes mainly include oxide, sulfide, and phosphate solid electrolytes. Among these, LiPON solid electrolyte has low ionic conductivity and is generally used as an electrolyte material for film cells; perovskite solid electrolyte has higher ionic conductivity, but its stability is poor; garnet solid electrolyte has better ionic conductivity, better chemical stability, and lower electronic conductivity, but its environmental stability is poor; LISICON solid electrolyte, with a similar structure to γ-Li_3PO_4, can exhibit improved ionic conductivity through doping with various species. In addition to these common inorganic solid electrolytes, there are halide solid electrolytes and borate solid electrolytes. The characteristics of halide solid electrolytes are that high conductivity and high stability cannot be achieved at the same time. Among all of these solid electrolytes, the lithium-rich anti-perovskite halide solid electrolyte has high ionic conductivity, but it is extremely sensitive to air humidity, which results in extremely poor environmental stability. Superionic conductor-type halide solid electrolytes have excellent stability, but their Li^+ conductivity is relatively low. Borate is mainly used as an additive during sintering to lower the sintering temperature.

Through analysis of research progress on inorganic solid electrolytes, it can be seen that solid electrolytes have received a great deal of attention from researchers all over the world. The advantages of the inorganic solid electrolytes are that they can not only essentially solve the safety problems of batteries, but also further improve battery performance. Although current solid electrolytes still have some problems and challenges, with continuing research, high-performance solid electrolytes that consider ionic conductivity, mechanical properties, and stability will be obtained. The preparation of high-performance solid-phase electrolytes is the basis for the development of all-solid-state Li batteries. It is hoped that solid electrolytes will continue to break through the technical bottlenecks in the future, and realize the industrialization of all-solid-state lithium-ion batteries and so contribute to clean and safe use of energy for humans.

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can be improved from two perspectives, namely, grain conductivity and grain boundary conductivity. Using appropriate substitute anions and cations, the concentration of the transferable sodium ions and the bottleneck size of the transmission channel can be adjusted, thereby improving the conductivity of the crystal grains. The improvement in grain boundary conductivity can reduce the generation of impurity phases and grain boundary pores by improving phase purity and compactness. Improvement in conductivity and interface modification of the NASICON structure solid electrolyte are still the main directions for improving the performance of solid sodium-ion batteries in future.[53] Since the NASICON structure solid electrolyte has good overall performance, it has broad application prospects in the fields of room-temperature sodium–sulfur batteries, sodium–air batteries, and seawater batteries.[54,55]

Sulfide solid electrolytes have high ionic conductivity, and their synthesis mainly includes mechanochemical methods, solid-phase sintering, and chemical liquid phase methods, with the first two of these being the most common. The sulfide reacts with the polar solvent to form a miscellaneous phase. Among sodium-ion sulfide electrolytes, Na_3PS_4 shows good compatibility with metallic sodium. However, the material is unstable in the air, and ion doping can increase the concentration of sodium-ion vacancies and improve the ion conductivity. Research on sodium-ion sulfide electrolytes at the electrode interface mainly focuses on reducing the solid–solid contact resistance between the electrolyte and the sulfide positive electrode, active material, and improving the electrochemical stability of the metal negative electrode. Anion and cation doping optimizes sodium-ion distribution and structural symmetry. However, there are few existing sulfide electrolyte systems, research into new materials is slow, and the known material defects have yet to be resolved.[56] On the basis of further improving the ionic conductivity, improving the air stability of sulfides, and improving the electrolyte interface layer, we need to reveal the mechanism through experiment and advanced characterization techniques, thus combining theoretical calculation and experiment to develop clean and efficient new energy storage batteries. There are still many challenges in the basic science and key technologies of sodium-ion batteries that require continuous research and further exploration.

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