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Introduction

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1.1 Introduction

The past decades have witnessed significantly irreversible resource and environmental degradation issues, especially for exhaustion and pollution due to the increasing global industrialization and indiscriminate consumption of fossil fuels, which critically restrict the sustainable development of society [1–3]. Proverbially, the sun, wind, hydro, and tides as renewable energy sources provide huge prospects to supplement electricity in an environment-friendly way to satisfy our social needs [4–6]. However, these renewable energies are restrained by the geographical conditions (longitude, latitude, altitude, geomorphology, etc.) and natural environments (alternation of sunrise and sunset, seasonal and weather variations, etc.) [7, 8]. To store these random, uncontrolled, and intermittent energies, secondary batteries can be utilized to accommodate these energies in the form of chemical energy and transform chemical energy into the electric energy required, which have presented dominant application and popularization in consumer electronics, electric vehicles (EVs), and intelligent grids for electrochemical energy storage (EES), which are attributed to their rechargeability and considerable electrochemical performance [9, 10]. However, safety, cost, and service life are plaguing their applications [11, 12]. Accordingly, it is particularly urgent to develop novel EES devices and corresponding materials with low cost, high device performance, and environmental friendliness [13].

As displayed in Figure 1.1, the number of literature published per year on batteries gradually increased from 2000 to 2022, and more than 31,000 articles were published in just 2022, demonstrating the persistent investigation enthusiasm on batteries among researchers, scientists, and experts from all over the world. Noticeably, the lithium-ion batteries (LIBs) account for a larger and larger share of these publications from 2000 to 2010. As known, LIBs have become the research hotspot for portable devices since they were commercialized by Sony in 1991 [14].

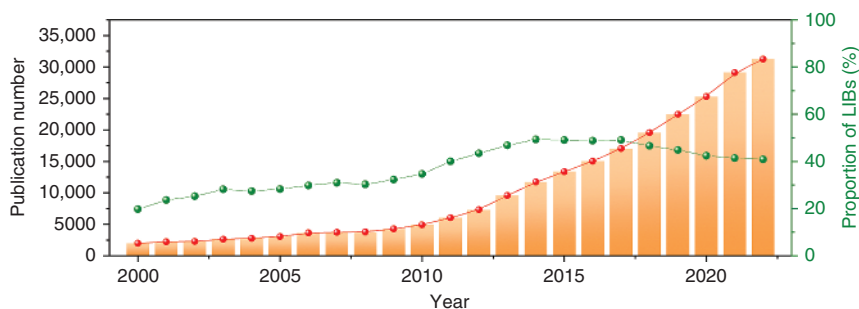


Figure 1.1 Left y-axis: the publication number of papers per year relevant to batteries from 2000 to 2022. Right y-axis: the percentage of papers published for LIBs. (Here, the results were refined from the Web of Science with “topic = Batteries” and “document types = article” for the left data and “topic = lithium-ion batteries or Li-ion batteries” and “document types = article” for the right data until 8 October 2023.)

Soon afterward, they outperformed their competition, i.e. nickel metal hydride and nickel cadmium batteries as the leading battery technology in digital cameras, laptop computers, and cell phones with an overall market share of more than 60% worldwide [15]. With the popularization of LIBs in portable consumer electronics, the next trial focuses on the application of LIBs for EVs based on the updated battery engineering in 2010s; therefore, the research attention on LIBs is a new level among batteries [16]. Although LIBs are still research hotspots in EES fields relevant to high energy density and their mature technologies on pivotal electrode materials and devices [17]. The reserves of resources such as lithium, cobalt, and nickel from cathode are cumulatively scarce and their costs are increasingly expensive, which restrict their sustainable development [18]. Therefore, novel EES devices have been investigated to complement the traditional LIBs and satisfy growing demand [19]. For example, some cost-effective cathodes can be considered, such as air, sulfur, and CO_2 [20–23]. Moreover, the construction of these batteries can be evolved into multivalent-ion or dual-ion types [24]. Perhaps, they can also be reconstituted into aqueous type, flow type, hybrid type, or flexible type [25–29]. Furthermore, fuels such as hydrogen or methanol can also be utilized as active materials to generate electric energy from chemical energy [30]. Under the circumstances, various new alternative EES devices such as metal-air batteries, metal-S batteries, metal- CO_2 batteries, multivalent-ion batteries, dual-ion batteries, fuel cells, aqueous batteries, flow batteries, hybrid capacitors, and flexible energy storage devices have been developed to achieve the aims of high energy/power density, long cycling lifetime, inherently safe, cost-effective, or environmentally benign [31]. Meanwhile, numerous scientific challenges in critical electrode materials, electrolytes, and construction of these devices and research attention concerned with the system integration for energy storage and utilization are extremely vibrant [32]. First, the reaction mechanisms of these novel EES devices should be elaborated through advanced characterization techniques and theoretical simulations [33, 34]. Second, structural and electrochemical stability of device components should be enhanced to improve the device lifetime by rational electrode design [35, 36]. Third, the

electrode activity and ion transport property in electrolytes should be optimized to promote their energy storage efficiency [37, 38]. The specific issues faced by different devices are briefly introduced and feasible solutions could be summarized and proposed as follows.

1.2 New Energy Storage Devices

1.2.1 Metal-Air Batteries

Metal-air batteries with an invincible theoretical specific energy and rich feedstock abundance of air have attracted much attention, especially, Li-air batteries with a theoretical specific energy of 3500 Wh kg^{-1} , whose anodes undergo the stripping/plating of metals and cathodes underpin the formation/decomposition of metal peroxides (M_2O_2) [39]. In 1996, Abraham et al. reported a rechargeable Li- O_2 battery constructed by a Li metal foil anode and a carbon composite cathode for oxygen reduction reaction with an organic polymer electrolyte membrane, which obtained a discharge capacity of 630 mAh g^{-1} [40]. This investigation is usually considered the pioneer of the aprotic Li-air batteries and inspires researchers to engage in metal-air batteries in the following years [41]. However, metal-air batteries are still in their infancy stage, and formidable challenges involving air cathodes, metal anodes, and electrolytes need to be addressed for practical applications [42].

As for the air cathode, especially for O_2 cathode, the solid and insulative discharge product M_2O_2 passivates the cathode surface and prohibits the diffusion of metal ions and O_2 , which results in a high overpotential, low Coulombic efficiency, and inferior discharge capacity, even the oxidative degradation of cathode materials and electrolytes [43]. Moreover, O_2 could exacerbate the side reactions. For example, M_2CO_3 , a common disreputable side reaction product, can ever-growingly deposit on the cathode/electrolyte interface because it is hard to eliminate even at a high voltage, which also degenerates the device performance and induces decomposition of the device components [44]. Noticeably, the formation of M_2O_2 might experience two different pathways of the surface-mediated and solution-mediated routes, which could influence the physical properties of M_2O_2 [45]. Usually, the solution route could alleviate the electrode passivation and contribute to a high discharge capacity by utilizing organic solvents with high solvating ability, which enhances the solvation of MO_2 intermediate [46]. However, superoxide species might lead to the solvent decomposition in the solvents with high solvating ability. In this case, designing stable electrolyte solvents without sacrificing the solvating ability can maintain the robust M_2O_2 formation property in electrolyte solution [47]. Moreover, the reactivity occupancy of superoxide intermediates is an effective strategy by incorporating the reduction mediator (RM_{disch}), which delivers electrons between the electrode and M_2O_2 , and forms a soluble intermediate, i.e. $\text{MO}_2\text{-RM}_{\text{disch}}$ complex, where the side reactions from electrode and electrolyte disintegration are restrained, increasing the M_2O_2 product upon discharge [48]. To decrease the side reactions and reduce the charging overpotential to enhance the kinetics of O_2 reduction, stable noncarbon materials such as metal oxides and nanostructured gold are

employed to replace the unstable carbon-based cathodes. Meanwhile, noncarbonate electrolytes, e.g. ionic liquid and ether, are combined with carbonate electrolytes to construct a stable metal- O_2 battery [49]. In terms of metal anodes, dendritic growth of the metallic anode is a vexing puzzle, which has been extensively investigated in metal-ion batteries, causing low Coulombic efficiency, poor cycle life, and safety issues [50]. Herein, the high-quality solid electrolyte interphase (SEI) can enhance the anode stability and improve the Coulombic efficiency via the electrolyte and additive design [51]. Simultaneously, the quasi-solid-state electrolyte and solid-state electrolyte could be applied as modified separators to protect metal anodes from ambient air, i.e. CO_2 and H_2O [52]. In addition, metal anodes could be replaced by several alternatives including silicon, aluminum, tin, and their derivatives to guarantee safety [53].

1.2.2 Li-S Batteries

Li-S batteries are considered one of the promising EES devices for applications in both EVs and stationary energy storages due to the merits of high energy density based on the conversion reaction between Li and S and abundant S with low cost [54]. Notably, the Li-S batteries could achieve a theoretical energy density of $\sim 2600 \text{ Wh kg}^{-1}$, which is four times higher than that of LIBs. In 1989, a rechargeable Li-S battery was established with an average discharge voltage of 2.1 V and an energy density of $\sim 80 \text{ Wh kg}^{-1}$ by the introduction of ether electrolyte [55]. Subsequently, the sulfur/porous carbon composition and the lithium nitrate additive were verified to be feasible in inhibiting the shuttle effect of polysulfides and enhancing the cycling robustness during the early 2000s, which ignites the researchers' enthusiasm to explore Li-S batteries in the following decades [56]. Nevertheless, formidable challenges still slow down the evolution of Li-S batteries for the practical application.

First, the S electrode faces the structural collapse after the lithiation of S to Li_2S due to the tremendous volume expansion, which is adverse to the application of high-density S electrodes [57]. Meanwhile, the electric/ionic insulating discharge products (Li_2S) and S electrode usually cause the reaction passivation [58]. Another notorious plight is the shuttle effect, where the intermediate discharge products (Li_2S_n , $4 \leq n \leq 8$) react with Li by the chemical reaction rather than by the electrochemical reaction ascribed to their dissolution, as a result of the remediless dissipation of both S and Li with the electrolyte decomposition due to the enhanced polarization [59]. Accordingly, functionalizing separators with catalytic materials are effective approaches to cope with polysulfides shuttle effect, where catalytic materials can boost sulfur redox reaction kinetics to achieve Li-S batteries with long cycle life and high energy density [60]. Besides, the multitudinous conductive matrix such as porous carbonaceous materials, conductive frameworks or polymers, and other novel materials could be introduced to alleviate the volume variation and suppress the shuttle effect by the dispersion or confinement of the active materials and to enhance the electric/ionic conductivity [61]. However, the carbonaceous hosts are inadequate to absorb Li_2S_n by physical approach. To anchor and convert

Li_2S_n to Li or S for the restraint of the shuttle effect by the catalytic methods, it is feasible to conduct the heteroatom doping or construct transition metal compounds in the carbonaceous hosts [62]. Simultaneously, the microstructure optimization of the carbonaceous hosts with aligned pores or hierarchical microstructures can decrease the porosity of S electrodes to improve the sulfur utilization and reaction dynamics [63]. Second, highly active Li anodes not only bring the safety troubles together with volatile and flammable ether-based electrolytes but also cause the depletion of the active materials derived from the shuttle effect [64]. Accordingly, it is significant to protect Li anode by introducing the protective coating or constructing effective SEI films [65]. Meanwhile, highly efficient electrolytes and solid-state electrolytes are also necessary to promote the battery performance [66].

1.2.3 Metal- CO_2 Batteries

Metal- CO_2 batteries are intriguing research hotspots in the background of global carbon neutrality due to the advantages of the real-time power input that drives CO_2 conversion and high battery energy density [67]. Compared with LIBs, metal- CO_2 batteries possess safer and higher energy density, providing cost-efficient techniques for renewable energy storage [68]. In contrast to normal CO_2 conversion and storage methods, which need energy input after fixing carbon, metal- CO_2 batteries supply electrical energy as required once recharging the battery to release the concentrated CO_2 [69]. In all, they demonstrate significant potential involving the capture of the greenhouse gas CO_2 to alleviate climate warming, the conversion of CO_2 into valuable chemicals, the storage of surplus electricity from other sustainable power systems, fossil fuels, and nuclear and carbon cycle balance [70]. Metal- CO_2 batteries are explored by supplementing CO_2 in Li/Na- O_2 batteries, and the battery capacity and energy density were greatly improved in 2011 [71, 72]. The pioneer metal- CO_2 batteries with pure CO_2 gas cathode and Li, Mg, and Al anodes in the nonaqueous electrolyte were presented in 2013 [73]. Soon afterward, the first rechargeable Li- CO_2 battery was demonstrated in organic electrolytes, which could be cycled reversibly at room temperature for several cycles [74]. Shortly after, various metal- CO_2 batteries with Li, Na, K, Mg, Zn, and Al metal anodes were dramatically developed [75]. Herein, metal- CO_2 batteries encounter several bottlenecks for commercial applications such as obscure electrochemical mechanisms, catalyst with low catalytic activity, high charge potential, poor rate capability, weak reversibility, short cycle life, and various parasitic side reactions [76].

The metal anodes in metal- CO_2 batteries contain monovalent metals (Li, Na, K, etc.), divalent metals (Mg, Zn, etc.), and trivalent metals (Al, etc.). Generally, the monovalent metal anode-based metal- CO_2 batteries select organic electrolytes due to the high metal activity [70]. In comparison, divalent and trivalent metals demonstrate weaker reaction activity; therefore, they usually utilize aqueous or ionic liquid electrolytes along with multifarious by-products [77]. Notably, various by-products involving carbonates or methanoic acid and disproportionation products such as CO or oxalate make the electrochemical mechanisms complicated; [78] therefore, it is significant to elaborate the electrochemical mechanisms via

advanced characterization techniques such as *operando/in situ* gas chromatography, Fourier transform infrared spectroscopy, X-ray synchrotron radiation spectra, and neutron techniques [79]. In the aspect of catalysts, carbonaceous materials demonstrate multiple superiorities of abundant reserves, low cost, great electrical conductivity, large surface area, and glorious porosity, whose catalytic activity could be enhanced by defect engineering, nanostructure design, or surface modifications [80]. Besides, noble metal-based catalysts generally deliver high catalytic activity and great stability, which could be promoted by alloying, composition with other metals or carbonaceous materials, etc. [81]. Transition metal-based catalysts with low cost including Cu, Fe, Co, Ni, Mo, and Mn are also research focuses, especially for the single atom catalysts and coupling with carbonaceous materials, which can enhance rate capacity, cycle life, and stability by exposing abundant active sites and promoting the utilization efficiency of active sites [82]. Besides, porous framework-based catalysts such as metal-organic frameworks and covalent organic frameworks also attract extensive consideration by virtue of the large surface area, structural adjustability, and porosity, which deliver significant potential as highly stable catalysts [83]. Moreover, the exploration of novel liquid electrolytes with low volatility and great electrochemical stability is also significant for metal-CO₂ batteries along with suitable additives to promote battery reaction kinetics and rate performance [84].

1.2.4 Multivalent-Ion Batteries

Multivalent-ion batteries involving Zn²⁺, Mg²⁺, Ca²⁺, and Al³⁺ have been regarded as potential candidates for grid energy storage due to low cost, good safety, and large volumetric energy density, which have the feasibility to fundamentally break through the energy density of current LIBs because they can achieve doubled or tripled capacities with the same number of reactive ions as Li⁺ [85]. Besides, the reserves of multivalent-ion metal elements such as Zn, Mg, Ca, and Al are distinctly more than that of Li on the Earth's crust, displaying cost-effectiveness [86]. However, commercial or mature cathode materials in LIBs cannot be suitable for multivalent-ion batteries because the guest ions suffer from serious Coulombic interaction with the surrounding lattice due to their high charge density, which causes large energy barrier. It would block the ion diffusion process and induce the irreversible phase transition, structural collapse, and electrode element dissolution, leading to inferior rate capability and cycling robustness [87]. Therefore, the development of multivalent-ion batteries still suffers from several challenges such as sluggish ion mobility kinetics, poor ion storage reversibility, dendrite growth of metal anodes, and lack of appropriate electrode materials [88].

In the case of Zn-ion batteries, they suffer from inferior reversible capacity, low Coulombic efficiency, and poor cycle life ascribed to dendrite growth and electrochemical corrosion of Zn anode, high polarization of divalent Zn²⁺, and unsatisfactory side reactions [89]. As for Al-ion batteries, although the graphite materials demonstrate a robust AlCl₄⁻ intercalation stability, the low specific capacities restrict their practical application [90]. Moreover, the electrochemical properties

are blocked by low discharge voltages and capacities, various by-products, unstable charge voltage platforms, the passivation of Al anode, inferior charge/discharge reversibility, severe corrosivity of chloroaluminate species for current collectors and battery assembling cans, and structural instability of cathode materials [91]. Mg-ion batteries and Ca-ion batteries are usually hindered by the incompatibility between electrolytes with metal anodes, and the generation of an impermeable passivation layer on surface of metal anodes, causing the poor reversibility of metal plating/stripping [92, 93]. Besides, Ca-ion batteries are usually operated at high temperature, which incurs high-security risks [94]. To overcome the above bottlenecks, nanostructure construction of domain size and hierarchical structure configuration are employed to boost the reaction dynamics by shortening the ion diffusion length, and the defect engineering can also promote the ion diffusion kinetics as well as enhance the electronic conductivity intrinsically [95]. Besides, the surface modification and metal alloying or compounds can effectively alleviate the metal anode surface passivation and corrosion, contributing to enhanced comprehensive performance [96]. The electrolytes also suffer from several crucial barriers, where the desolvation energy of cations increases with the rising of the charge density of the multivalent ions, causing sluggish mobility kinetics of multivalent ions across the electrode interface, especially for Mg-ion batteries and Al-ion batteries [31]. Simultaneously, the side reactions consume the electrolyte continuously, especially for aqueous system [97]. To mitigate these challenges, electrolyte design through altering electrolyte and additive composition is a feasible strategy along with the anode electrode optimization, which can improve the reversibility of ion deposition/stripping [98].

1.2.5 Dual-Ion Batteries

Dual-ion batteries exhibit a different working mechanism from the traditional LIBs with a “rocking-chair” mechanism, where anions (such as PF_6^- , FSI^- , and ClO_4^-) and cations (such as Li^+ , Na^+ , and K^+) are simultaneously intercalated/deintercalated into cathode and anode materials, respectively, during the charge/discharge process. This endows dual-ion battery with a competitive cutoff voltage of more than 4.5 V, contributing to a relatively high energy density [99, 100]. Besides, the energy density of dual-ion battery can be improved by regulating the concentration of electrolytes as they also serve as the active substances in this system [101]. Noteworthy, the low specific capacity, inferior rate performance, and insufficient cycling life are significant barriers for dual-ion battery to practical popularize because of the tardy ion diffusion kinetics and dissolution of electrolyte at high voltage [102].

Electrolytes as the crucial part of dual-ion battery mainly include organic liquid electrolytes, aqueous electrolytes, ionic liquid electrolytes, and solid-state electrolytes, whose energy gap depends on their highest occupied molecular orbital (HOMO) and lowest unoccupied molecular orbital (LUMO), which determines the electrochemical stability window [103]. To circumvent the electrolyte decomposition, electrolytes need to exhibit excellent compatibility with electrodes and

robust electrochemical stability at high voltage [104]. Carbonates (e.g. ethylene carbonate, diethyl carbonate, ethyl methyl carbonate, and dimethyl carbonate) are one of the promising options, which can readily dissolve cations and anions due to their low viscosity and high ion conductivity, and exhibit a wide potential window [105]. Meanwhile, the compatibility between electrodes and electrolytes could be regulated by the categories and concentrations of salts, additives, and solvents [106]. Especially, fluorinated electrolytes can enhance the oxidative robustness due to their high oxidation potentials [107]. Meanwhile, highly concentrated electrolytes are promising selection for dual-ion battery to achieve the long cycle stability [108]. As for graphite electrode materials, the design strategies are significant to promote the battery performance [109]. First, an elevated graphitization degree for electrodes could contribute to the enhanced voltage efficiency and Coulombic efficiency as well as the decreased polarization and improved ion storage capability, because the graphitization degree of electrode materials could influence ion diffusion kinetics, electronic conductivity, and structural stability [110]. Besides, the nanostructure construction of electrode materials can sustain the volume variation during the charge/discharge process, thus enhancing the battery cycling stability and rate capability, because the nanostructures can improve ion transport via shortened diffusion channels and increase ion storage sites with enlarged specific surface areas [111]. Further, adequate defects or dopants in electrode materials might introduce fresh ion storage sites, boost ion diffusion kinetics, and promote the electronic conductivity, contributing high capacity and rate capability [112]. In addition to graphite, other high conductive carbon materials, organic materials, layered transitional metal disulfides, and even metals show the potential to host the active anions [113]. Amazingly, organic materials have been investigated hotspots because of their flexible designability, rich ion storage sites, and redox reversibility, which endow them with the high theoretical capacity and great cycling stability [114]. Noteworthily, the nanostructural construction and interfacial modification are extensively conducted to promote the ion reaction kinetics and strengthen the battery robustness [115]. While for the anode materials, the metallic materials, intercalation materials, alloying materials, and conversion materials have been developed, which are similar to those in rocking-chair batteries.

1.2.6 Fuel Cells

Fuel cells have reached the early stage of commercial deployment due to their outstanding energy efficiency, high energy/power densities, potential for zero/low-emission lifecycles, quick refill ability, and versatile applications involving portable devices, automotive vehicles, and power grid stations. In the fuel cells, the chemical energy stored in fuels (hydrogen, methanol, or ethanol) and oxygen is directly converted to electrical energy through electrochemical reactions with fuel oxidized reaction at the anode and oxygen reduced reaction (ORR) at the cathode, respectively [116]. However, the widespread applications of fuel cells still suffer from several technical barriers on their electrochemical performance, durability, and cost [117]. To accelerate electrochemical processes, electrocatalysts

should demonstrate high activity and stability, especially for the cathodic ORR, which could be multielectron and multistep reactions with sluggish kinetics [118]. Usually, electrocatalysts are composed of metal nanoparticles as the active species and substrate materials as the support to facilitate the electron/mass transfer and enhance the stability [119]. Carbon-based materials as the common support deliver glorious conductivity, high specific surface area, and abundant porosity, which can disperse metal nanoparticles, contributing to high electrochemically active surface area [120]. Currently, carbon-supported Pt-based electrocatalysts have been regarded as the feasible choice to achieve the high catalytic activity and durability for the ORR [121]. Unfortunately, the scarce reserves and high cost of Pt-based electrocatalysts severely block the practical applications of fuel cells [122]. Therefore, more intensive efforts are still needed to promote the activity and durability of electrocatalysts under the condition of lowering the utilization of noble metal electrocatalysts.

To decrease the usage of Pt and reduce the cost, it is imperative to explore nonprecious metal electrocatalysts with great durability and high active site density. Recently, several types of nonprecious metal electrocatalysts have been reported, such as metal-free carbon materials, transition metal oxides, and M/N/C catalysts [123]. Notably, M/N/C represents metal atoms coordinated with nitrogen functionalities and encapsulated into carbonaceous matrix, where M denotes transition metals such as Fe, Co, and Ni or main group metals such as Mg, Al, and Ca have been regarded as the most promising catalysts [124]. However, the activity and stability of M/N/C catalysts need to be improved comparable to those of Pt-based electrocatalysts in practical operating environments of fuel cells by engineering the active sites. Generally, the metal-N₄ species were proved to be the active sites for the ORR; therefore, the design and increase of active sites are the research focus for M/N/C catalysts [125]. Besides, the aggregation of metal atoms should be considered. Noticeably, the metal-organic frameworks or metal macrocyclic compounds with metal-nitrogen coordination structures are usually utilized as precursors to evenly disperse metal ions [126]. Besides, metal single atom catalysts of the M/N/C are also research hotspots due to their maximized metal atom utilization efficiency and distinct electronic structure, where their catalytic properties can be regulated by adjusting the microenvironment of active sites involving the coordination number of M–N, heteroatom doping, and vacancy creation for the improved catalytic performance [127]. However, the scalable preparation of M/N/C catalysts for the practical application in fuel cells is also crucial adjective. Accordingly, the synthesis methods still face great challenges.

1.2.7 Aqueous Batteries

Aqueous batteries without utilization of flammable organic electrolytes demonstrate their superiority of high safety, low cost, and easy operation [128]. The early research on aqueous LIBs can be traced back to 1994, when the battery configuration involving LiMn₂O₄ as a cathode and VO₂ as an anode with 5 M LiNO₃ aqueous electrolyte presented a voltage of ~1.5 V and an energy density of 55 Wh kg⁻¹, outperforming than those of aqueous lead-acid batteries (~30 Wh kg⁻¹) and Ni-Cd

batteries ($\sim 50 \text{ Wh kg}^{-1}$) [129]. Subsequently, metal oxides, polyanionic compounds, and Prussian blue analogs (PBAs) were utilized as cathodes for aqueous LIBs, where spinel LiMn_2O_4 is one of the research focuses owing to its availability in aqueous solutions [130]. Unfortunately, most anode materials for nonaqueous LIBs are not applicable for aqueous batteries due to their low redox potentials beyond the cathodic limit of aqueous electrolytes, where hydrogen evolution reaction (HER) would occur on the anode as a competing reaction, causing the decomposition of the aqueous electrolytes [131]. Further, various metal-based aqueous batteries ($\text{M} = \text{Li, Na, K, Zn, Mg, Ca, Al}$) were investigated to enhance energy density and cycle life [132]. Noticeably, the rechargeable aqueous Zn-based battery is one of the prospective battery devices due to the rich reserve of Zn, suitable redox potential (-0.763 V vs. standard hydrogen electrode [SHE]), and high theoretical specific capacity (820 mAh g^{-1}) [133]. Although Al anode possesses larger volumetric capacities of $\sim 8046 \text{ mAh cm}^{-3}$, its instability in aqueous solutions results in the poor cycle performance [134]. Zn-based batteries demonstrate glorious reversibility in diversified aqueous electrolytes, which include alkaline Zn metal batteries and neutral or acid Zn-ion batteries [135]. However, the reactivity of water splitting influences the promotion of aqueous batteries, where the low potentials on the anode lead to water reduction, i.e. HER and the high potentials on the cathode cause water oxidation, i.e. oxygen evolution reaction (OER) [136]. Therefore, aqueous batteries also suffer from several obstacles such as self-discharge resulting in inferior battery storage and narrow electrochemical stability window leading to unavoidable side reactions in aqueous media, which cause the limited energy density and poor long-term stability [137].

Considering that the theoretical capacity of cathode materials depends on the electron transfer number and material mass, reducing the inactive mass of the electrodes is a theoretically feasible strategy to improve the battery capacity, but is quite challenging in the case of maintaining electrode structural stability [138]. Alternatively, it is a pivotal measure to increase the electron transfer numbers by introducing multiple redox reaction centers. Taking low-valence vanadium-based compounds as an example, their *in situ* electrochemical oxidations contribute to high capacities due to the rich active sites of high-valence compounds [139]. Meanwhile, the surface carbon coating is also necessary for these high-capacity oxides to keep their structural and chemical stability [140]. Besides, the poor intrinsic conductivity and weak selectivity to metal ions for electrode materials could lead to the deterioration of battery performance. Herein, it is crucial to perform optimizing strategies such as regulating the crystal structures and introducing conductive materials [141]. Noticeably, as for the layered electrode materials, preintercalating molecules or ions can weaken electrostatic interaction of the host lattice oxygen and metal ions and achieve the expansion of interlayer distance to promote the ion storage capacity [142]. Another focus is to achieve high-voltage aqueous batteries, which mainly depends on the electrochemical stability window of aqueous electrolytes, restricted by OER on the cathode and HER on the anode [143]. The working voltages of actual battery systems are usually less than 2.0 V due to the low theoretical water splitting potential (1.23 V vs. SHE). It is urgent

and effective to broaden the electrochemical stability window by weakening the correlation between electrode materials and water molecules, e.g. water-in-salt electrolytes by adopting salt-concentrated electrolyte design can provide a wide electrochemical stability window by decreasing the electrochemical activity of water molecules [144].

1.2.8 Flow Batteries

Flow batteries are promising candidates for large-scale energy storage applications due to their high safety with aqueous recyclable electrolytes, glorious environmental friendliness, and excellent cycling stability. In the flow battery, the electrolyte in the tank circularly flows across the matched electrode with the aid of the pumps, allowing redox reactions of active components from the electrolyte to occur on the electrode surface to convert the electric energy from the chemical energy by the valence state variation of ions pairs [145]. Accordingly, the power and energy densities can be adjusted respectively through individual electrode engineering. Recently, traditional vanadium flow batteries and burgeoning zinc-based flow batteries have been popularized as the megawatt energy storage devices, demonstrating the potential commercial and industrial applications [146, 147]. Meanwhile, several novel aqueous flow battery systems can enlarge the power and energy density, lower electrolyte resistance, and enhance safety and environmental benignity, such as aqueous flow batteries including quinone/bromide system, quinone/iron system, etc. and nonaqueous flow batteries containing Li/ferrocene system, Li/cobaltocene system, TEMPO system, viologen system, etc. [148, 149]. However, the HER and OER in aqueous solutions are the focus of attention, which causes the narrow work voltage window, restricting its energy density [150]. Simultaneously, the poor power density needs to be improved for the practical industrialization. As for nonaqueous solutions, the active species are generally unstable and the solvents are usually flammable, leading to poor work stability [151].

It is effective to design and optimize pivotal materials involving the electrodes, electrolytes, membranes, and bipolar plates. To decrease the battery polarization and enhance the power density, electrode materials should possess high electron conductivity, great reaction activity to redox couples, excellent chemical and electrochemical stability, and large surface area for the sufficient active sites [152]. Herein, electrode modification, especially for the carbonaceous materials, is the common strategy, involving electrochemical oxidation, acid treatment, thermal activation, defect incorporation, functional group introduction, surface coating, or depositing electrocatalysts on the surface. Usually, the energy density of flow batteries depends on the concentration and volume of the electrolyte [153]. Accordingly, to develop high-performance and highly stable electrolytes, ideal active materials in electrolytes should have great solubility and stability. Generally, the membrane can separate the positive and negative electrolytes and transport charge carriers, whose properties also quite influence the battery performance [154]. Consequently, perfect membranes should deliver excellent ion conductivity, ion selectivity, chemical and mechanical stability, and low cost, which can effectively mitigate the self-discharge

and capacity attenuation and efficiently improve the battery energy density and power density. Moreover, bipolar plates are the indispensable components to connect each cell electrically, separate neighbor cells, and guide the electrolyte flow, which should demonstrate great conductivity, stable mechanical strength, high corrosion resistance, and excellent compactness to prevent electrolytes leakage [155]. Simultaneously, the stack and assembly of flow battery are also significant to promote the industrialization and commercialization [156]. As for the non-aqueous flow battery systems, stable active materials and electrolyte optimizations are critical fields of exploration [157]. It is noteworthy that other novel systems or conceptions such as semisolid flow batteries and redox-targeting-based flow batteries also deliver the possibility to boost the battery performance.

1.2.9 Hybrid Capacitors

Hybrid capacitors deliver elevated power densities with a moderate energy density, short charge/discharge time interval, wide operating temperature range, and elongated cycle life, which are alternative technologies to decrease consumption of fossil fuels and greenhouse gas pollution [158]. Capacitors are divided into three categories involving pseudocapacitor with fast surface redox reactions, electric double layer capacitor with the ion adsorption, and hybrid capacitors [159], which can store energy via electrode materials' surface/near-surface faradaic redox reactions as well as the formation of an electrical double layer at the electrode/electrolyte interface from the purely electrostatic interaction [160]. Although their fast charge storage mechanism does not suffer from poor solid-state ion diffusion in battery electrodes ascribed to the electrode surface or near-surface reactions, their energy density and charge storage are quite inferior [161]. As for the electric double layer capacitors, they need high conductivity and large surface area to increase reaction rate at the electrode/electrolyte interface and achieve a fast charging/discharging rate [162]. However, the limited electric double layer usually brings about low energy density. In contrast, materials involving faradaic charge transfer processes such as metal oxides, sulfides, nitrides, phosphates, and oxynitrides can provide higher capacity [163]. Unfortunately, these materials often suffer from the phase/structure variation during the charge/discharge process or deliver poor electrical conductivity, causing the sluggish energy storage kinetics [164]. Therefore, there is still vast room to promote their performance.

As for the electrode materials, the reduced domain sizes of nanostructured electrodes can offer large specific surface area, increase the access of the electrode surface by electrolyte ions, shorten ion diffusion paths, and block the phase/structure variations, which tend to boost the capacitance, enhance the rate capability, and weaken cyclic degradation [165]. Simultaneously, nanomaterials provide the convenient reaction exchange on the atomic level or molecular level for the hybrid materials, which could synergistically promote electrode properties. Furthermore, it is effective in increasing the electronic conductivity, shortening the charging time, and maintaining the phase/structure stability by incorporating highly conducting carbonaceous-based materials [166]. Besides, to obtain high

energy devices, selection of materials with high capacity is necessary, where hybrid materials with the combination of surface and intercalation charge storage such as battery-type oxide/hydroxide materials can contribute to higher energy density and better cyclic stability compared to carbonaceous materials and conducting polymers [167]. Moreover, hybrid configuration adhering disparate energy storage mechanisms with faradaic and non-faradaic material components (asymmetric electrode configuration) in a single device is a sensible approach to reach the synergistic performance of each component with high energy (faradaic process) and high power (non-faradaic process) storage, such as electric double layer capacitive electrode/pseudocapacitive electrode, or electric double layer capacitive electrode/battery-type electrode [168]. Besides the electrodes, the electrolytes are also significant components, where they are expected to have robust electrochemical stability, low volatility, low solvated ionic radius, low resistivity, low viscosity, low toxicity, high ionic concentration, wide voltage window, etc. [169].

1.2.10 Flexible Energy Storage Devices

Flexible energy storage devices play a significant role in portable, ultrathin, and flexible electronic and device markets due to their good mechanical stability, miniaturization, and multifunctionalities [170]. Multitudinous flexible devices were integrated into wearable devices or human body without being perceived, such as sensors, energy harvesters, antennas, and radio-frequency identification tags, which can maintain mechanical robustness under various mechanical deformation from human movements [171]. Currently, the traditional rechargeable energy storage devices mainly including batteries and supercapacitors incline to structure failure, safety hazard power, and supply interruption once they suffer from bending, folding, stretching, twisting, etc. because of the rigid structures lacking mechanical flexibility [172]. Accordingly, it is significant to comprehensively design and fabricate flexible energy conversion and storage devices. Recently, various novel materials and structures with mechanical pliability including electrodes, current collectors, and solid-state or quasi-solid-state electrolytes have been utilized for flexible energy devices, which will unveil substantial electrochemical mechanisms and challenging investigations in interdisciplinary fields such as chemistry, physics, material science, electronics, engineering, biology, and medicine [173]. Noteworthily, several fundamental and technological bottlenecks still need to be addressed for the practical applications of flexible energy conversion and storage devices. On the one hand, the initial interfaces including active materials/conductor additives, electrodes/current collectors, and electrodes/electrolytes are gradually deteriorated, which restrict the electronic transfer and/or ionic transport channels [174]. On the other hand, the weakened internal electronic conductivity from mechanical deformation results in local short circuits and serious side reactions [175].

The flexible design strategies for materials, electrodes, and architectures need to endow energy storage devices with bendable, implantable, and wearable properties. Usually, introducing redundancy in volume such as pores can achieve high flexibility by lowering the bending stiffness and releasing strain [176]. As for the electrolytes,

fluidic properties of liquid electrolyte restrain the size and shape of batteries; therefore, plastic or rubbery ion-conductive medium is regarded as a feasible alternative to construct flexible batteries, where gel polymer electrolytes can be utilized as both the electrolyte and the separator [171]. Meanwhile, plastic crystal electrolytes combined with a polymer matrix as a flexible framework demonstrate great thermal and structure stability, good ionic conductivity, and mechanical strength [177]. Besides, the solid polymer electrolytes involving polyacrylonitrile, polyvinylidene fluoride, polyethylene oxide, and polymethyl methacrylate are utilized to eliminate the outflow of liquid electrolyte, where ceramic nanoparticles can be employed as fillers to avoid crystallization of polymers or ion conductors to enhance ionic conductivity, which are relatively ideal ion transport medium with variable shapes and controllable integration [178]. However, they still suffer from severe interfacial passivation and poor contact with active materials [179]. Furthermore, the current collector as another significant component is critical to construct flexibility batteries, where traditional Cu and Al foils have small tensile fracture strains and yield strains, which make it hard to maintain integrity in the condition of repetitive deformations [180]. Herein, carbon materials and conductive polymers as alternative current collectors can be employed due to highly conductive flexibility. Noticeably, the battery components and architecture innovation are feasible to promote flexibility such as wire, cable, or wave shapes [181].

1.3 Conclusion

This book provides a comprehensive review on promising energy storage devices including metal-air batteries, Li-S batteries, metal-CO₂ batteries, multivalent-ion batteries, dual-ion batteries, fuel cells, aqueous batteries, flow batteries, hybrid capacitors, and flexible energy storage devices. It also covers the fundamentals of energy storage devices and key materials (cathode, anode, and electrolyte). In addition, some advanced characterization techniques are introduced to achieve an in-depth understanding of the fundamentals inside the devices for the further improvement of their electrochemical performance. Moreover, the current challenges and effective strategies of new energy storage devices with high performance are also proposed. This book can attract more readers from various research fields on new energy storage and pivotal electrode/electrolyte materials, provide guidelines for those who are entering the related research fields, and enlighten readers to grasp the evolvement orientations in the future.

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