Multiscale nanomaterials for electrochemical energy storage and conversion

ARTICLE INFO

ABSTRACT

High-performance electrochemical energy storage and conversion devices (EESCDs) are highly desirable for meeting sustainable development of human society. It is the assiduous goal to develop advanced EESCDs with large power/energy density and high conversion efficiency. This special issue presents a collection of the most recent advances in EESCDs including lithium ion batteries, supercapacitors, alkali ion batteries, alkaline batteries, and photo-electrochemical devices. The recent development on controllable synthesis of multiscale nanomaterials and nanostructures of EESCDs is also proposed. The electrochemical performances of different types of multiscale nanomaterials and composites are presented. Moreover, design rules and corresponding merits/demerits are discussed and future research trend is analyzed.

1. Introduction

The energy & environmental crisis is threatening the sustainable development of human society. To turn the table, green energy technology has become a primary focus of the major world powers and scientific community. There has been great interest in developing and refining more efficient electrochemical energy storage and conversion devices (EESCDs) due to their promising and potential applications in current and future consumer products, ranging from modern electronics to electric grids, and electric vehicles with arbitrarily shaped surfaces [1,2]. It is the assiduous goal to develop EESCDs with large power & energy density, and high energy conversion efficiency. Currently, different forms of EESDs including supercapacitors, lithium ion batteries, sodium ion batteries, lithium-sulfur batteries, electrochemical solar cells, alkaline batteries, chloride ion batteries and magnesium ion batteries have achieved great progress, meanwhile, they are encountered with new challenges. It is well accepted that the performance of EESCDs is mainly determined by their active electrode materials and components, which are controlled by the electrochemical activity and kinetic feature of the electrodes. Therefore, electrode materials and components must be appropriately designed and synthesized to enhance the kinetics of ion and electron transport to achieve high-performance EESCDs.

Nanoscale design of active materials has been demonstrated to be an effective strategy to achieve high performance due to decreased size with shortened diffusion path of ions and larger surface area. To date, emerging multiscale nanomaterials (from 0D to 3D) have been widely explored over the last decades and have demonstrated promise for applications in EESCDs [3–5]. For future advanced energy storage and conversion devices, it is still essential to controllably design and fabricate novel multiscale nanomaterials with optimized properties to realize/construct high-performance devices.

This special issue presents a collection of the most recent advances in EESCDs and shows the recent development on controllable synthesis of multiscale new classes of nanomaterials and nanostructures and their applications in EESCDs. It is also emphasized on developing new synthetic approaches and understanding the fundamental issues of structural assembly and growth that will enable high performance of EESCDs. In addition, the special issue gives a large overview on emerging strategy to develop advanced EESCDs with the help of novel multiscale materials.

2. Results and discussion

2.1. High-performance lithium ion batteries (LIBs)

The fast development of LIBs has greatly changed the landscape of electrochemical energy storage devices since 1990s. Despite great progress, the LIBs are facing severe development bottleneck due to the limited capacity of electrodes (anode and cathode). Therefore, the innovation or modification on the electrode materials is still on-going.

In regard to the anode research, the commercial graphite anode has no potential to be tapped and suffers from compromised high-rate performance and relatively low capacity. Hence, great efforts are dedicated to exploiting new anode materials with larger capacity and higher power characteristics. One typical alternative high-rate anode is lithium titanate (LTO). As a zero-strain insertion material, LTO can maintain initial structure well and have a fast process of phase transformation at high rates. Despite of these advantages, its electronic conductivity and ionic

conductivity are not satisfactory. Nanostructuring porous design and conductive composite design are dispensable to achieve high performance. In this special issue, for example, Saxena et al. [6] reported vanadium doped $Li_4Ti_5O_{12}$ (LTO) with enhanced high-rate performance and found that V doping could enhance the ion transfer at high working current. In addition, Pan et al. [7] designed integrated graphene foam integrated LTO anode and the discharge capacity and power density is greatly reinforced. This light-weight electrode design protocol can further improve the energy & power densities. The LTO based anode show excellent high-rate capability, about 10 times larger than the commercial graphite, up to 50-200C. But the working voltage of LTO is in the voltage range of 1-2 V (*vs.* Li^+/Li), higher than the commercial graphite. Additionally, the discharge capacity of LTO is still lower than 200 mAh g^{-1}. This would cause energy and power loss when coupled with the commercial cathode. Even so, the LTO is still considered as one of the most promising alternative candidate. Meanwhile, similar lithium ion storage is also noticed in the TiO_2 anode. Huang et al. [8] and Chen et al. [9] reported TiO_2 nanotube and nanobelt as the anodes, respectively, which exhibited higher capacity than LTO but with worse high-rate properties due to the volume change accompanied by the strain out of control leading to fading of structure.

Apart from the LTO, other high-capacity anodes spring up over the past decades. Especially, silicon based materials are considered as the most promising successor to the commercial graphite anode and demonstrated with an ultrahigh theoretical specific capacity (~ 4200 mAh g^{-1}) and low working potential (< 0.5 V vs. Li/Li^+). Nevertheless, its first coulombic efficiency and cycling life is poor because of huge volume expansion (> 300 %) during alloying/dealloying processes and poor electrical conductivity. To overcome these stubborn problems, it is essential to reduce the Si size and integrate with conductive matrix (e.g., carbon). In this issue, Shen et al. [10] designed biomass-derived carbon/silicon three-dimensional hierarchical composites as enhanced anode. Impressively, the cycling life and coulombic efficiency were greatly improved due to the uniform carbon coating on the nanoscale Si. Moreover, their work demonstrated that the Si could be organically combined with bio-mass derived carbon to obtain high performance. Without modification, the Si can not meet the practical application. Given the cost the coulombic efficiency, novel carbon/Si composite anode is the most popular choice. In recent years, another research route is to use Si as additive to modify the carbon or graphite anode. The Si usually accounts for about 5-10 % in the composite. It is proven that the Si additive can enhance the capacity of the composite anode with good cycles. As a cousin to Si, cost-effective SiO_2 is also verified with good lithium ion storage capacity. Liang et al. [11] adopted sand precursor to prepare high-quality SiO_2 anode with high capacity > 500 mAh g^{-1}. But the impurity in the sand will affect the performance leading to compromised rate capability and cycles. As compared to the Si based materials, SiO_2 anode does not have much more advantages.

In parallel with the above anode, metal oxides such as NiO, MoO_2 and MoO_3 have been widely studied since the pioneering work of Tarascon group [12]. Nevertheless, these metal oxides suffer from some common issues. Their practical application is blocked by the poor cycling stability arising from the large volume change causing pulverization and deterioration of active materials during cycling. Furthermore, their working voltage is embarrassing with a high voltage above 1 V. It is implied that the discharge working voltage would be not high as assembling full cells with commercial cathode. Even so, lots of modification strategies are still made on them to expect to obtain high performance. In this issue, Jiang et al. [13] prepared partially sulfurized MoO_2 Film for durable lithium storage. It is found that the S doping can enhance the structural stability and electrical conductivity of MoO_2 film. Liu et al. [14] reported hollow SnO_2 spheres with excellent electrochemical performance via hydrothermal synthesis. The hollow structure can alleviate the volume change and increase the contact between active material and electrolyte. Zhu et al. [15] fabricated hierarchically branched $TiO_2@SnO_2$ nanofibers as high-performance anodes for LIBs. It is reported that the core/shell structure can improve the capacity due to the positive synergistic effects such as optimized space utilization, enhanced conductivity, prolonged discharge plateau, and reinforced morphological stability. Wang et al. [16] and Ma et al. [17] constructed porous NiO films and NiO/polyaniline composite arrays, respectively. Their research demonstrates the porous design of NiO and introduction of conducting polymers can effectively provide larger active sites, better conductive network, and buffer the volume expansion with the help of polyaniline coating, leading to improved cycling performance. In a nutshell, porous conductive design plus binder-free design of arrays are the current mainstream route for metal oxides anodes.

Exploration on cathode materials never stops. It is well known that the energy density of LIBs is mainly governed by the capacity of the positive electrode. Tarascon' group did a simple calculation a nd revealed that the energy density of a cell could increase up to 57 % by doubling the capacity of the positive electrode, while only an increase of 47 % could be achieved as the capacity of the negative electrode increased by a factor of 10 [18]. In this issue, Chen et al. [19] summarized the recent progress in surface coating of layered $LiNi_xCo_yMn_zO_2$ for LIBs. The high-capacity $LiNi_xCo_yMn_zO_2$ has matured significantly and been widely used in commercial LIBs. But the cycling life should be further enhanced by introduction of different surface coating with the aid of multiscale design. On the other hand, Cao et al. [20] and Liu et al. [21] adopted porous design strategy to construct N-doped carbon/$LiFePO_4$ microspheres and $LiMn_2O_4$ hollow microspheres as high-performance cathode, respectively. Though the porous design will decrease the tap density of active materials, the transfer kinetics of ions/electrons would be greatly accelerated leading to enhanced high-rate performance. In addition, the carbon coating or hetero-atom doped carbon is necessary for construction of advanced $LiFePO_4$ cathode. Of course, the thickness of the carbon layer should be controlled lower than 5 nm. If too thick, the diffusion of Li^+ will be blocked. Meanwhile, a niche cathode such as MoO_3 is studied by Chu et al. [22], who prepared MoO_3 nanowire bundles from a self-assembled organic-inorganic precursor. The lithium ion storage of the MoO_3 nanowire is high, but the working voltage can not compete with the commercial $LiFePO_4$ and $LiNi_xCo_yMn_zO_2$. The exploration on the cathode is tough and it is highly desirable to search for high-capacity cathode with multi-electron reactions as well as high voltage. In such a context, Li-S batteries emerge and have attracted great attentions in recent decades on account of high theoretical energy density (2600 Wh kg^{-1}) and large specific capacity (1675 mAh g^{-1}), accompanied by advantages of low cost and abundant storage. However, the commercialization of S cathode is still impeded by several challenges. 1) Low conductivity. On the sulfur cathode side, both sulfur and discharge product (lithium sulfide) are insulating in nature, resulting in poor high-rate capability and low material utilization.5 2) Poor cycling life associated with the "shuttle effect". During cycles, the active sulfur will form a series of dissolved long-chain polysulfides, which can transfer freely between cathode and anode, and cause side reactions on the Li anode accompanied by continual loss of active material and rapid capacity decay. Hence, tailored strategies must be taken to tackle these headwinds to advance the commercialization of Li-S batteries. In this special issue, Li et al. [23] and Chen et al. [24] constructed ultrathin SnS_2 nanosheets decorated S cathode and N-doped carbon nanosphere/S composites as enhanced cathodes for high-performance lithium-sulfur batteries, respectively. The SnS_2 nanosheets can not only act as physical dams to suppress the diffusion of polysulfides, but also form chemical bonds with polysulfides to anchor them leading to enhanced performance. The N-doped carbon nanospheres are excellent conductive matrix for the loading of S and can provide better fast transfer path for electrons. Now, single modification on S is still not enough. The physical and chemical modification on sulfur cathode must be done simultaneously to achieve good cycles and high capacity.

2.2. High-performance post lithium ion batteries

Post LIBs ear is on the way and the research on alternatives to LIBs is in full swing due to the high cost and scarcity of lithium. Typical, alkali ion

batteries are entering into people's vision such as sodium ion batteries (SIBs), potassium ion batteries (PIBs), and magnesium ion batteries (MIBs) due to their cost-effective, non-toxic, and larger reserves on earth (about one thousand times more abundant than lithium). Their performance can not match LIBs, but has improved greatly and is close to the early products of LIBs. In this issue, several metal oxides and sulfides are constructed as anodes for applications in SIBs. For example, Yu et al. [25] reported flower-like tin Sulfide @ Reduced Graphene Oxide composites as stale sodium ion storage anode. Wang et al. [26] presented 3D foam-like $CoNiO_2$ anode of SIBs. Similar to metal oxides anodes of LIBs, their voltage is still high leading to lower discharge plateau when coupled with the cathode of SIBs. Therefore, design/fabrication of anode with low working voltage is still highly desirable for SIBs. Simultaneously, the cathode of MIBs is also a research hotspot. Cho et al. [27] reported layered $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ as a host for Mg intercalation in rechargeable Mg batteries. The capacity is not high, but the operation cycles are good. The composition of the $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ still needs to be optimized with tunable layered space for Mg intercalation. In addition, interestingly, Yu et al. [28] reported novel chloride ion battery by using layered FeOCl cathode material. The chloride ion is the working ion and can be reversibly insertion/desertion. However, the cycling stability is still not satisfactory and should be improved in the future.

2.3. High-performance supercapacitors

Supercapacitor has emerged with the potential to enable major advances in energy storage. Supercapacitors are governed by the same fundamental equations as conventional capacitors, but utilize higher surface area electrodes to achieve greater capacitances [29]. This allows for energy densities greater than those of conventional capacitors and power densities greater than those of batteries. As a result, supercapacitors may become an attractive power solution for an increasing number of applications. The advancement of supercapacitors largely relies on the innovation of active materials. In this special issue, several pseudocapacitive materials including MnO_2, Bi_2S_3 Fe_2O_3, are reported. Guo et al. [30] presented hierarchical Co_3O_4@PPy core-shell composite nanowires for supercapacitors with enhanced electrochemical performance. Lu et al. [31] reported Bi_2S_3 nanoparticles anchored on graphene nanosheets with superior capacitive performance. The PPY shell and graphene matrix can enhance the electrical conductivity and structural stability, resulting in enhanced capacitance. Xie et al. [32] designed negative electrode of Fe_2O_3/graphene oxide paper for high energy supercapacitors. This paper electrode is very light and flexible, and can be compatible with wearable equipment. Moreover, MnO_2 based cathode such as nanotubes and nanowires are discussed by Xiao et al. [33] and Kim group [34]. It is proven that nano-structuring on MnO_2 is favorable for the fast transportation of ions/electrons, leading to increased utilization of active MnO_2. To date, the working mechanism on the pseudocapacitive materials is still not clear and controversial. Especially, who dominates the capacitance contribution in the pseudocapacitive materials? Battery behavior or surface capacitive behavior? These questions are still on debate.

2.4. High-performance alkaline batteries

Hybrid alkaline batteries are one of promising energy storage devices because they use battery-type cathode and capacitor-type anode, implying that the can combine the advantages of batteries (high energy density) and capacitor (superior power density). Extensive theoretical and experimental studies have demonstrated that the performance of alkaline batteries is mainly determined by the electrochemical activity and kinetic feature of electrodes. Direct growth of nanostructured metal oxides/hydroxides on conductive substrates is an effective way to improve the reaction activity due to good electrical connection with the substrate, porous structure and high specific surface area. In this special issue, Wu et al. [35] reported Ni nanoparticles embedded into cross-linked NiO nanoflakes as enhanced cathode for alkaline batteries. In addition, Ma et al. [36] designed cobalt oxide nanoflakes grown on cobalt nanowires as composite cathode for high-performance alkaline batteries. Chen et al. [37] fabricated nickel hydroxide shell on TiO_2 nanorod core forming enhanced arrays for energy storage. These arrays architecture offers shortened ion diffusion paths and facilitates migration of the electrolyte ions, leading to improve high-rate performance. It is noteworthy that these metal oxides/hydroxides cathode must be coupled with capacitive carbon anode to achieve high performance.

2.5. High-performance optical-electrochemical energy storage & conversion

Solar energy offers a clean, abundant and unlimited energy resource to mankind and provides a green way to fulfil the global demand for carbon-free energy [38]. The solar provides us with a wide spectrum of applications in solar heating, photovoltaics, photoelectrochemical water splitting, photosynthesis, and photocatalysis. In this special issue, some of the above themes are discussed. Shi et al. [39] reported ZrO_2 quantum dots/graphene phototransistors. The introduction of graphene can improve the sensitivity. Zhang et al. [40] designed oxygen-deficient bismuth oxychloride nanosheets with superior photocatalytic performance. Yadian et al. [41] presented metal-sulfide-decorated ZnO/Si nano-heterostructure arrays with enhanced photo-electrochemical performance. In addition, Railey et al. [42] proposed metal organic frameworks with immobilized nanoparticles and applications in photocatalytic hydrogen generation. These results show that the interface structure is very important to the high absorption/utilization of light energy. The nanoscale interface heterostructure is required be tunable with different thickness and size. Yang et al. [43] used the first-principles method to investigate the metal-doped cubic $BaTiO_3$ when they are used as the photo-electrochemical catalysis. All the results indicate that the design/fabrication of interface structure or active materials should be combined with the theoretical calculation results, which can guide the synthesis of efficient photo-electrochemical catalysts and photoanode/photocathode.

3. Conclusions

The advancement of social economy is highly related to the development of electrochemical energy storage and conversion devices (EESCDs), which are largely dependent on the design/fabrication of advanced multiscale nanomaterials. In this special issue, different active materials of EESCDs including LIBs, SIBs, MIBs, Post-LIBs, supercapacitors are proposed and their merits and demerits are discussed. The deign rule is to construct structures with fast transfer paths for ions/electrons. Nanoscale design and conductive composite design strategies have been demonstrated to be the most powerful strategies to achieve high performance due to enhanced conductivity, reinforced structure and larger surface area. Meanwhile, the photo-electrochemical performance is highly associated with the interface layer between the active composites, which should be accurately controlled and tuned. In a nutshell, the current LIBs and supercapacitors are still flourishing and their potential alternatives are emerging. It is a burgeoning era for the EESCDs and their multiscale nanomaterials.

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