Bismuth-Based Composites for Energy Storage Systems

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Bismuth (Bi) has been prompted many investigations into the development of next-generation energy storage systems on account of its unique physicochemical properties. Although there are still some challenges, the application of metallic Bi-based materials in the field of energy storage still has good prospects.

Keywords: bismuth ; electrochemical energy storage ; alkali ion batteries ; alkali metal anode ; sulfur cathode

1. Introduction

In recent years, public opinion worldwide has been drawn toward curbing global warming and developing renewable energy. Currently, the most promising clean and renewable energy sources from solar, wind, tidal, etc., are remittent in nature and must rely on energy storage equipment to achieve full-time availability. Electrochemical energy storage devices have the advantages of short response time, high energy density, low maintenance cost and high flexibility, so they are considered an important development direction for large-capacity energy storage technology [1][2]. On the other hand, the rapidly expanding market size of various portable electronic products and electric vehicles has led to rapid growth in the demand for energy storage devices with high energy density, long lifespans, and excellent safety [3][4][5].

Among the electrochemical energy storage devices, lithium ion batteries (LIBs) have gained popularity among numerous energy storage systems owing to their high energy density, high operation potential, stable cyclability and eco-friendly nature [6][7][8]. After decades of research, LIBs have been successfully commercialized and widely penetrated into our daily lives, changing people's lifestyles. During this development process, mobile consumer electronics products promoted the rise of the LIB industry, and the promotion of electric vehicles in recent years has directly driven the rapid growth of the LIB industry. In turn, industrialization has also increased the intensity of research, leading to the rapid development of theory and application technology for secondary batteries. Traditional LIBs with graphite as anode and layered transition metal oxides as cathode are gradually approaching their theoretical energy density, and it is difficult to further improve it [9][10]. Meanwhile, with the development of the electric vehicle industry, low global reserves of lithium resources and uneven global distribution have caused lithium salt prices to soar [11][12]. Therefore, intensive investigation has been devoted to the study of next-generation energy storage systems, including sodium ion batteries (SIBs), potassium ion batteries (PIBs), magnesium ion batteries (MIBs), lithium-sulfur (Li-S) batteries, etc. Basically, these beyond-lithium ion batteries and next-generation LIBs all operate based on the "rocking-chair" principle with alkali ions shuttling between anodes and cathodes [13][14]. Thus, what is crucial for these advanced energy storage systems is to develop appropriate electrode materials with high electrochemical performance.

Bismuth (Bi) is a group VA element with a unique semi-metallic nature that has been found to have good application in the photocatalysis and electrocatalysis fields [15][16][17]. Bismuth has unique electrical properties and a layered crystal structure and has been found to be promising for use in the anodes of alkali ion batteries, mainly due to its high theoretical volumetric specific capacity and appropriate operating potential [18][19][20]. Compared with other alloying anodes, such as Si, Sn, Sb and Ge, Bi has higher theoretical volumetric capacity of 3800 mAh cm^{-3}. Zhao et al. demonstrated the application prospects for bismuth-based materials in LIBs and revealed the existence of binary Li-Bi alloys via ex situ XRD analyses in 2001 [21]. Later, owing to its high specific capacity, bismuth was explored as an anode material for MIBs, SIBs and PIBs in 2012, 2014 and 2018, respectively [22][23][24].

However, the large volume variation during the alloying/dealloying process is the main reason hindering the application of Bi-based anodes for alkali ion batteries [25][26]. Severe volume expansion directly destroys the structure of the electrode material, causing the electrode to be pulverized and fall off, which in turn causes the capacity of batteries to decay rapidly. To solve these problems, massive efforts have been devoted to Bi-based anodes by downsizing the particle size, designing innovative architectures and optimizing electrolyte configurations. For example, Dai and coworkers constructed

Bi@C core-shell nanowires in 2021, with the carbon shell accommodating large volume expansion and efficiently confining the nanowires [27]. Wang et al. proposed ether-based electrolytes for bulk Bi anode, which has interaction effects completely different from traditional ester-based electrolytes, and greatly promoted the advance of Bi-based anode materials [28].

For the purpose of breaking through the upper limit of traditional lithium-ion batteries, lithium metal batteries (LMBs) were re-proposed [29][30]. Based on similar considerations, sodium metal batteries (SMBs) and potassium metal batteries (PMBs) stand out among beyond-lithium ion batteries [31][32]. However, these battery systems with alkali metal anodes suffer from poor cycling stability because of the high reactivity of alkali metal, unlimited volume variation and dendrite growth [33][34][35]. For these issues, several strategies have been proposed to stabilize the alkali metal anodes, including constructing 3D hosts [36][37], metal–electrolyte interfacial engineering [38][39] and electrolyte optimization [40][41]. Inspired by the research progress on Bi-based anodes in alkali ion batteries, Bi-base materials were introduced to solve these key scientific issues for alkali metal anodes. For example, Cui et al. used K_4BiI_7 as an electrolyte additive for LMBs, which generated a robust and Li-ion-conductive solid electrolyte interphase (SEI) [42]. The Bi-containing electrolyte significantly improved the average coulombic efficiency and relieved Li dendrite growth.

Li-S batteries with sulfur as cathodes have the characteristics of high specific capacity, abundance and nontoxic active material, which is considered as the best choice for next-generation LIBs [43][44]. Unfortunately, the shuttle effect of the polysulfides and slow reaction kinetics restrict its practical performance. According to the reported researches, an ideal sulfur host must provide strong surface polarity, prominent electrocatalytic activity and large specific surface area [45][46]. Noteworthily, the semi-metallic nature and the typical 2D layered crystal structure of Bi make it viable as an electrocatalyst for the conversion reaction of polysulfides.

2. Fundamentals of Bismuth

2.1. Crystal Structure of Bi

Bi is the last element in the pnictogens and is considered as a nontoxic and environment-friendly metal. The crystal structure of Bi is rhombohedral symmetry, which is typical for the group V semimetals (phosphorus, arsenic and antimony) [47]. The crystal structure of Bi exhibits typical layer stacking with *c*-axis layer spacing of 3.95 Å. The interlayer spacing of Bi is much larger than the radius of many alkali elements, including Li, Na, K and Mg, demonstrating its application potential for rechargeable alkali ion batteries.

2.2. Physicochemical Properties of Bi

Bi has been classified as a semimetal that has an electronic structure very different from that of other metal elements. Thus, Bi is endowed with many unique properties owing to its special electronic structure. Bi has a highly anisotropic Fermi surface and ultrasmall band overlap energy [48]. Bi has a theoretical conductivity value of 7.75×10^5 S m^{-1} [49]. Thermodynamically, Bi can alloy with many alkali metals, including Li, Na, K and Mg, and obtain various bimetal alloying phases [50][51][52][53]. As shown in the Li-Bi alloy phase diagram, Bi can alloy with Li, forming different alloy phases, including LiBi, Li_2Bi and Li_3Bi. Similarly, Bi can alloy with Na to obtain NaBi and Na_3Bi, alloy with K to obtain KBi_2, K_3Bi_2 and K_3Bi, and alloy with Mg to obtain Mg_3Bi_2, respectively. As reported by Weppner and Huggins, the thermodynamic enhancement factor of Li-Bi alloy increases smoothly on the stoichiometry with a maximum of 360. The diffusion coefficient of Li ions in Li-Bi alloy is above 10^{-4} cm^2 s^{-1} and also increases with the content of Li. Therefore, Bi has the potential as an anode material for LIBs, SIBs, PIBs and MIBs. Moreover, Bi has been used as a photocatalyst owing to its semimetal and plasmonic characteristics. Therefore, it is worth looking forward to its application as an electrocatalyst in the catalytic conversion reaction of sulfur cathodes.

3. Application of Bi-Based Composites for Energy Storage Systems

3.1. Anode for Alkali Ion Batteries

Commercial LIBs with graphite as anodes suffer from low energy density and are gradually approaching their fundamental limits, which cannot meet the demands of the energy storage market. Commercial graphite anodes for LIBs can only provide relatively low theoretical specific capacity, which further restrains the energy densities of commercial LIBs. Nowadays, the energy storage market's requirements for energy storage systems are high specific energy density, high safety and stable cyclability. To meet the growing demands, some next-generation rechargeable LIBs and beyond-lithium ion batteries have been proposed. There are several criteria for the development of these novel energy storage devices, but the construction of high-performance electrode materials is a top priority of this technology. To achieve these targets, various anode materials based on insertion, conversion and alloying reaction mechanism are being explored and developed to meet high criteria for next-generation energy storage systems. In this context, bismuth is a typical alloy-type anode that can alloy with various alkali metal elements. As **Table 1** illustrates, Bi-based anodes deliver high specific capacity and low operation voltage in LIBs, SIBs, PIBs and MIBs. However, the higher specific capacity is also accompanied by huge volume expansion due to alloying with multi alkali atoms. The volume expansion ratio was calculated by dividing the volume of the alloyed phase by the original volume of Bi.

Table 1. The characteristics of Bi-based anodes for alkali ion batteries.

Application	Products	Mass Specific Capacity (mAh g^{-1})	Volumetric Specific Capacity (mAh cm^{-3})	Operation Voltage (V)	Volume Expansion Ratio (%)
LIBs	Li_3Bi	385	3800	0.8/0.7	208
SIBs	Na_3Bi	385	3800	0.77/0.67	352
PIBs	K_3Bi	385	3800	1.0/0.4/0.3	509
MIBs	Mg_3Bi_2	385	3800	0.25	196

In order to alleviate the damage to the electrode material structure caused by volume expansion, various strategies have been explored and applied, including the hydrothermal/solvothermal method, electrospinning, dealloying, annealing, etc. Of those, the hydrothermal/solvothermal method is an effective method to synthesize nanomaterials with unique structures. Electrospinning techniques are usually applied to synthesize Bi-based precursors embedded in 1D nanofibers, followed by an appropriate pyrolysis process to obtain final Bi-based anode materials. Dealloying is an effective method for Bi-based nanomaterials with different dimensional structures [54]. The annealing process is a common synthesis method for Bi/C composites. Owing to the unique layered crystal structure of Bi, ultrasonic techniques are often used for the synthesis of 2D Bi nanosheets.

3.2. Modification for Alkali Metal Anodes

Alkali metal anodes have gained increasing attention in recent years owing to their high specific capacities (3861, 1165 and 678 mAh g^{-1} for Li, Na and K metals, respectively) and potential for high-energy-density secondary batteries [55]. However, metal dendrites can cause short circuits between the positive and negative electrodes, leading to thermal runaway and serious safety issues. Dendritic growth is a critical issue that must be eliminated to attain stable cyclability and dendrite-free metal anodes in a wide range of operating conditions [56]. Since the discovery of lithium dendrites, much effort has been invested in studying the growth mechanism of lithium dendrites and inhibiting dendrite growth. Many well-known reaction models have been established, greatly motivating the advancement of alkali metal batteries [57][58][59]. Meanwhile, many strategies have been proposed to inhibit dendrite growth and can be subdivided into electrode structure engineering, metal−electrolyte interfacial engineering and electrolyte optimization [60][61]. Cohn et al. employed anodes of SIBs, including carbon and alloy-type anodes, as the nucleation layers for anode-free batteries, demonstrating the significant effect of Bi on sodium metal anodes [62].

3.3. Host for Sulfur Cathodes

Li-S batteries with merits of high specific capacity, low cost, and environmental friendliness have attracted considerable attention from researchers. Nonetheless, the shuttling effect of polysulfides and sluggish reaction kinetics seriously impact the electrochemical performance and commercialization of Li-S batteries. One of the effective solutions is to construct an appropriate structure by introducing an ideal sulfur host [63][64]. An ideal host for sulfur cathodes should have the following properties: strong polarity to anchor polysulfides, excellent electrocatalytic activity to accelerate the kinetics of polysulfide conversion reactions, large specific surface area to accommodate high sulfur loading and good conductivity to ensure the rapid progress of electrochemical reactions. In recent years, various catalysts have been developed for high-performance sulfur cathodes, such as metal oxides, sulfides, single-atom composites, etc. [65][66][67][68].

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