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Progresses of Research in Anode Materials for Lithium-Ion Batteries

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Abstract. Long term of excessive relay on fossil fuels has led to a large number of critical issues, such as energy shortage and environmental pollution. In this article, new energy storage technologies have emerged as a pivotal direction, and lithium-ion batteries (LIBS), as an ideal choice, have gained significant attention. Also, the anode materials play an important role in LIBS performance. This paper outlines the structural compositions and working principles of LIBS, reviews the research status of carbon-based and non-carbon-based anode materials, and compares their advantages and limitations. Furthermore, this paper summarizes the main modification methods and prospects of the development directions in the future. It aims to provide a reference for the advancement of high-performance anode materials for LIBS. The research on advanced anode materials is of great significance for enhancing the energy density and safety of lithium-ion batteries. This research supports the sustainable development of the energy storage system.

1 Introduction

Energy serves as a fundamental material basis for the development of human civilization. For a long time, the heavy depend on fossil fuels had a negative impact on the global ecological environment and posed a serious challenge to the long term sustainable development of society. As a substitute, new energy sources are pivotal in driving the transition towards low-carbon, clean, safe and efficient energy systems. Under the current strategic imperatives of energy transformation and sustainable development, advancing new and efficient energy storage technologies plays a crucial and central role all around the world. From this paper, new energy storage technologies are increasingly prominent, hold significant importance for optimizing the energy structure and fostering green social development [1].

As a component of new energy storage, LIBS are vital in the fields of new energy technologies. They have achieved large scale commercial success and have become the predominant choice due to new energy storage devices. LIBS offer notable advantages which includes high safety, high power density, high energy density and long cycle life. As a result, LIBS have become more and more essential in dominant market. Consequently, they are widely utilized in power batteries, consumer electronics and grid-scale energy storage [2].

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With the LIBS systems, anode materials provide the significant structure for lithium-ion intercalation and their performances are a crucial determinant of overall battery performances.

This paper focuses on the compositions and working principles of LIBS. Based on the classification of anode materials, the paper systematically reviews the current research status of both carbon-based and non-carbon-based anode materials, analyzes their performance characteristics and existing challenges. In addition, future development trends are predicted, aims to provide a reference for the research and advances in high-performance anode materials for LIBS.

2 Overview of lithium-ion batteries

As a rechargeable new energy storage device, LIBS primarily consists of cathodes, anodes, separators and electrolytes. The operational principle of LIBS is based on the reversible intercalation and deintercalation of lithium ions between the anode and cathode. During charging, lithium ions are extracted from cathode materials, migrate through electrolytes and are subsequently intercalated into anode lattices. Conversely, during discharging, lithium ions are deintercalated from anodes, migrate back to cathodes and release electrical energy. This reversible “shuttle” of lithium ions enables LIBS to undergo numerous charge and discharge cycles and establishes them as an efficient type of secondary battery.

2.1 Cathode

The cathode materials are almost the most expensive component of LIBS and account for approximately 40% of the total cost. They directly determine battery performance metrics, such as energy density, cycle life and operational voltage. Common cathode materials include layered lithium transition metal oxides ($LiMO_2$, M = Ni, Mn,Co), lithium iron phosphate ($LiFePO_4$), spinel lithium manganese oxide ($LiMn_2O_4$) and various ternary materials [3].

2.2 Anode

The anode materials are another essential component and constitutes roughly 10% of the battery cost. Anode materials for LIBS are primarily classified into carbon-based and non-carbon-based materials. Carbon-based materials encompass hard carbon, soft carbon, natural graphite and artificial graphite. Non-carbon materials mainly include silicon-based anodes, titanium-based anodes and tin-based anodes.

2.3 Separator

The separator are primary internal component, which represents about 10% to 20% of the battery mass and 10% to 14% of the cost. Between cathodes and anodes, the main functions are to prevent physical contact (thus avoiding short circuits) while allowing the free passage of lithium ions during operation. The properties of the separator significantly influence capacity, cycle performance, internal resistance and charge/discharge current density of batteries [4]. The most commonly used separators are polyolefin materials.

2.4 Electrolyte

The electrolyte constitutes approximately 15% of the battery mass and 6% to 8% of the cost. It plays a vital role by providing high ionic conductivity for efficient ion transport, which ensures smooth internal current flow. In addition, it exhibits excellent chemical and thermal stability to maintain safe and stable operation over a wide voltage range and throughout long term cycling, without undergoing detrimental side reactions [5]. Electrolytes are mainly categorized into organic and inorganic types.

3 Anode materials' classification for lithium-ion batteries

Based on chemical composition, LIBS anode materials are primarily divided into two groups: carbon-based materials and non-carbon-based materials.

3.1 Carbon-based materials

3.1.1 Hard carbon

Hard carbon refers to carbonaceous precursors that resist graphitization even after heat treatment above 2500 ℃, such as resin carbon and pyrolyzed organic polymers. It possesses three-dimensional cross-linked structure and exhibits high diffusion coefficient, wide lithium intercalation potential window, good rate capability, high specific capacity, low cost and excellent safety. These attributes make it promising for high-power LIBS. Regardless of hard carbon suffers from significant drawbacks, which includes a large irreversible capacity loss during the initial cycle, the absence of a distinct charge/discharge voltage plateau, and suboptimal overall electrochemical performance [6].

3.1.2 Soft carbon

Soft carbon refers to amorphous carbon which can be graphitized at high temperatures above 2500 ℃. Soft carbon materials include coke, mesocarbon microbeads (MCMBs) and carbon nanotubes. Coke encompasses petroleum-based and coal-based varieties, with common types like petroleum coke, needle coke and pitch coke being easily graphitizable. MCMBs feature a spherical morphology, high tap density and substantial lithium storage capacity per unit volume. Their layered structure facilitates lithium-ion intercalation/ deintercalation and their small outer surface area minimizes side reactions. Nevertheless, complex and costly production processes limit their industrial-scale application [7]. Carbon nanotubes offer advantages such as small particle size, high electrical conductivity, high mechanical strength, large specific surface area and strong interfacial effect. They exhibit higher discharge capacity and better lithium insertion stability at high current densities. Their market share in power batteries may exceed 60%.

3.1.3 Graphite Anode

Graphite anode materials include natural graphite and artificial graphite. Graphite possess layered hexagonal structure where each carbon atom is bonded to three carbon atoms, form a two-dimensional honeycomb lattice. The primary advantages of graphite include low cost, abundant resources, favorable physical properties, long cycle lives, high energy densities and excellent overall electrochemical performances as LIB anodes. However, challenges such as moderate rate capability, limited theoretical capacity and structural anisotropy still persist.

Natural graphite features a high crystalline structure, which does not require graphitization sintering, thereby reducing energy consumption. Its low cost and resource abundance grant a significant market in LIBS for consumer electronics. Although it faces issues like particle exfoliation caused by lithium-ion intercalation during cycling and low initial Coulombic efficiency, which due to severe side reactions on defective surfaces under polarization voltage [8].

Artificial graphite is produced by high-temperature graphitization of chemically treated coke precursors. It shares similar advantages with natural graphite and finds broader application in power batteries. This is because artificial graphite typically has a higher compacted density (beneficial for volumetric energy density) and smaller exhibits, more isotropic expansion, which enhances long term cycling stability. In addition, its adjustable interlayer spacing and particle structure contribute to superior rate performance [9]. But the vital drawback is lower rate capability associated with generally larger particle size, which leads to suboptimal high-current discharge performance.

3.2 Non-carbon-based materials

3.2.1 Titanium-based materials

Lithium titanate ($Li_4Ti_5O_{12}$, LTO) is the most representative titanium-based intercalation anode material. Its unique spinel structure ensures exceptional structural stability and it offers advantages such as low manufacturing cost, high safety, environmental friendliness and excellent electrochemical performance. Although its theoretical capacity (175 mAh/g) is modest, LTO demonstrates outstanding reversibility with minimal structural change during cycling and result in negligible capacity fade [10]. Its charge and discharge plateau at approximately 1.55 V avoids the formation of solid electrolyte interphase (SEI) layer, leading to high initial Coulombic efficiency, good electrolyte compatibility, prevention of lithium dendrite growth and enhanced safety. Its main limitations are low lithium-ion electronic conductivity and diffusion coefficient, which constrain its rate performance and especially under high-power charging and discharging.

3.2.2 Silica-based material

At present, there are mainly two kinds of silicon-based anodes: silicon-based system and silicon-carbon system. The silicon dioxide system is a silicon oxide and its carbon composite anode material. The silicon-carbon system is a silicon-carbon composite material prepared by co-sintering carbon precursors such as metallurgical silicon and petroleum coke pitch. The theoretical specific capacity of silicon-based anode material is as high as 4200 mAh / g, which is about 10 times that of graphite anode and has great commercial application potential. Its advantage is that silicon is rich in reserves and low in cost; silicon forms high-lithium alloy after lithium ion insertion, which provides insertion and extraction channels for lithium ions in all directions and improves the fast charging performance of the battery. The working potential of silicon is slightly higher than that of graphite, which alleviates the problem of lithium evolution [11]. However, its commercialization is facing difficulties, mainly due to the low intrinsic conductivity of silicon and poor high-power discharge performance ; in the process of charging and discharging, anisotropic expansion and contraction occur, resulting in serious pulverization of materials, spalling of surface active materials, poor contact of current collectors, which seriously affect the performance of batteries and the film of SEI formed on the silicon’ surface is unstable, which results in continuous consumption of electrolytes and low coulomb efficiency of batteries.

3.2.3 Tin-based materials

Tin, as one of the earliest studied anode materials for lithium batteries, has high specific capacity and was once considered to be a promising candidate to replace graphite. Tin-based alloy materials, which are compounds of tin with metals inactive towards lithium, can exhibit improved cycling performance and extended battery life. However, inherent shortcomings have hindered their progress. For instance, during cycling, tin atoms tend to agglomerate and cause severe volume expansion that rapidly diminishes battery capacity. Materials like tin oxide (SnO_2) offer benefits such as low-cost raw materials, eco-friendly preparation process, high theoretical capacity and good cycling stability. Nonetheless, issues of volume expansion and particle agglomeration lead to electrode pulverization, rapid capacity fade and poor cycling stability [12].

4 Performance optimization of anode materials for lithium-ion batteries

4.1 Progress in modification of carbon anode materials

The graphite material in the carbon anode material is cheap and abundant, which can make the lithium atom easily embedded and detached during the charging processes, it has the disadvantages of high initial irreversible capacity and poor cycle performance. In order to change its performance, it has a lower reversible capacity and better cycle performance. The commonly used modification methods are [13]: one is the physical grinding method, which reduces the size of graphite particles and increases the specific surface area, such as crushing and mechanical ball milling. The second is the chemical oxidation method, which forms a film of oxide on the graphite’ surface to reduce the decomposition of electrolyte, such as soaking in an oxidant for a long time. The third is the surface coating method, which reduces the natural graphite’s specific surface area, inhibits the SEI film’ formation, and makes the cycle more stable. For example, amorphous carbon and metal oxide are used for coating. Fourth, non-metallic elements are added to change the way lithium atoms are embedded in carbon materials, such as phosphorus, boron, sulfur, and silicon.

4.2 Progress in modification of non-carbon anode materials

4.2.1 Progress in modification of titanium-based anode materials

Lithium titanate has a good commercial prospect because of its unique spinel structure. However, lithium titanate has poor cycle performance and a small specific capacity at high current. In order to enhance its cycle performance and specific capacity, it is necessary to synthesize lithium titanate electrode materials at low temperature and short sintering time. Therefore, there are some modification methods for lithium titanate anode materials [14]: First, lithium titanate is synthesized by the hydrothermal method. Nano-sized lithium titanate materials are synthesized at low temperature and magnesium elements are doped to improve the cycle stability of lithium titanate batteries. The second is to increase the cut-off voltage of discharges/charges and improve discharges specific capacity of the material. Third, doping magnesium element replaces the lithium position of some crystals and releases compensation electrons to strengthen the conductivity of the material. The fourth is to improve the conductivity by doping metal oxide with good conductivity.

4.2.2 Progress in modification of tin-based anode materials

Tin alloy materials have two dominant problems: low efficiency of the first charge and poor cycle performance. To commercialize these materials, these two problems must be solved. For solving these problems, the commonly employed modification strategies include: first, the introduction of active substances to enhance lithium storage capacity and cycling stability; second, the incorporation of matrix materials to improve structural stability and cycle life; third, the utilization of nanomaterials to optimize electrochemical cycling performance.

5 Conclusion

The design of high performance for anode materials represents a crucial strategy for boosting the energy density of LIBS. Currently, graphite-based materials continue to dominate in commercial markets; nonetheless, the application of silicon-based anodes is advancing rapidly. Future researches are expected to focus on the following major directions: firstly, developing high-performance, low-cost composite anode materials; secondly, conducting in-depth investigations into the interfacial electrochemical mechanisms within all solid-state battery systems; thirdly, accelerating the research and development cycle through advanced methodologies such as materials genomics. These efforts are essential for driving the sustainable development of the energy industry.

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