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# Study on sodium-ion anode material - Graphene battery

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1 School of Materials Science and Engineering, Lanzhou University of Technology, Gansu Lanzhou 730050, China Abstract:

Batteries are the most common form of energy storage. Sodium-ion batteries are gradually replacing traditional lithium-ion batteries, becoming a key factor restricting their safety, service life, energy density and capacity. With the continuous consumption of energy by human society, the increasing scarcity of mineral resources, and the increasingly severe environmental problems. It is an urgent task to seek new, renewable and clean energy. Sodium-ion battery is expected to be a new type of charge/discharge device because of its high energy conversion efficiency, high energy density, long cycle life, environmental friendliness and good economy. The development of new green and high-efficiency large-capacity energy storage technology is great significance for promoting China's green and low-carbon energy development. It is great significance for the implementation of China's "double carbon" strategy. Sodium-ion battery is the most important class of secondary batteries. Sodium-ion battery has high rate, good low temperature resistance, suitable for low salt concentration electrolyte, high safety and other characteristics. The sodium is abundant in the earth, low price, low REDOX potential. That is a class of secondary batteries with great application prospects. With the progress of society and the sustainable development of economy, people's demand for sodium-ion batteries is also increasing. The "14th Five-Year Plan" New Energy Storage Development Implementation Plan clearly points out that is necessary to carry out the test and demonstration of a new generation of high energy density storage technology represented by sodium-ion batteries.

**Keywords:** Sodium-ion battery; Graphene; Secondary battery; Sustainable development

#### 1. Overview of sodium-ion batteries

Sodium-ion batteries (SIBs) have attracted much attention because of their low price, abundant reserves and working mechanism similar to lithium-ion batteries. Due to the large radius of sodium ion, the wide application of sodium ion is limited. So the exploration of anode materials suitable for sodium ion batteries have became a research hotspot. It is expected to have excellent electrochemical properties when it is used as a negative electrode material. Graphene is an ideal nagative material for sodium-ion batteries due to its abundant source, low price and stable performance. Graphene and graphene composite materials have been widely used in lithium-ion batteries because of their high specific capacity, and have shown excellent comprehensive properties, which is a promising new negative electrode material for lithium-ion batteries. Graphene is regarded as a new nagative material for sodium ion batteries. And its preparation method not only has an important impact on its electrochemical properties, but also is closely related to the full utilization of its functions. Sodium-ion battery is composed of positive electrode, negative electrode, electrolyte, diaphragm, collector and so on. A new anode material for sodium-ion battery with low cost, simple operation and excellent electrochemical performance was developed.

#### **1.1 The composition of sodium-ion batteries**

With the large-scale use of lithium-ion batteries, the demand for lithium has exceeded the existing supply level, resulting in the rising production cost of lithium[1]. Lithium is an important metallic element and its spatial distribution in nature is extremely uneven[2]. Although there is a large amount of lithium salt in the ocean. Its preparation cost is difficult to control due to its extremely low content[3]. Sodium is a metal with a higher storage capacity than lithium on Earth, more than 400 times higher than lithium, with a reserve of 2.74%[4]. So sodium is widely distributed on the earth. In life and industrial production have a great use, such as edible salt sodium chloride, industrial salt sodium nitrate and so on. Compared with lithium, sodium has a relatively concentrated distribution and low production cost, which makes it more cost-effective than lithium in the battery field[5]. Therefore, finding a new type of energy storage device that is not constrained by resources. Clean and green development is a problem to be solved at present[6]. In order to solve this problem, sodium-ion batteries, which were developed at the same time as lithium-ion batteries but were neglected. Sodium-ion batteries have become the focus of research again. The reserves of sodium metal in the earth's crust are much larger than lithium metal. So the development of sodium ion batteries has become a new trend.

	Na	Li
Standard electrode potential(V)vsSHE	-2.7	-3.04
Crustal content(%)	2.74	0.006
Ionic radius(mm)	1.02	0.776
Specific capacity (mAh g <sup>-1</sup> )	1166	3862
Price(U.S. dollar/t)	150	15000

Table 1.1 Comparison of some properties of sodium and lithium	Table 1.	1 Com	parison	of	some	pro	perties	of	sodium	and	lithium
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In recent years, Contemporary Amperex Technology Co., Limited has launched the first generation of sodium-ion batteries for the first time, but compared with lithium-ion batteries, its commercialization progress is relatively slow. Sodium-ion battery is a new energy storage material with great potential[7]. Compared with lithium-ion batteries, sodium-ion batteries have the following advantages: 1 Rich in resources. Sodium resources are abundant, mainly distributed in land and sea, easy to develop, low cost, and stable price[8]; ②Strong adaptability. Since there is no alloying reaction between sodium and aluminum, aluminum foil can be used as the positive and negative electrode materials. Compared with the negative electrode materials of lithium-ion batteries, more expensive copper foil is required. So the cost of sodium-ion batteries is lower in terms of fluid collection[9]; ③ High security. Sodium ion has a small charge density. It can be rapidly diffused in aqueous and non-aqueous solutions. It can adapt to the high temperature environment of 30~80°C, and will not lead to energy loss of the battery, with good stability and high safety.

Nowadays, according to its different packaging methods. It is divided into square batteries, cylindrical batteries and button batteries[7]. Usually, the research batteries made in the laboratory are made of button batteries. Its structure is: positive shell, shrapnel, gasket, positive material, electrolyte, diaphragm, negative material, gasket, negative shell and so on[10]. In recent years, more and more researches have been made on anode and cathode materials, electrolyte materials. A diaphragm is a dielectric layer with tiny pores that allows sodium ions to spread rapidly between the positive and negative materials.

Electrolyte is an important part of sodium ion battery, and its electrolyte has the characteristics of ion transport, insulation, safety and stability. At present, the liquid electrolyte based on carbonate ester as electrolyte is more studied[9][11]. Such as sulfide, Na<sub>3</sub>Zr<sub>2</sub>Si<sub>2</sub>PO<sub>12</sub>[12] and

other solid electrolytes. And sodium hexafluorophosphate (NaPF<sub>6</sub>), sodium perchlorate (NaClO<sub>4</sub>) and other salt electrolytes. This kind of material is one of the most widely studied and applied materials in electrolyte field because of its good conductivity, good stability and good cycle stability.

The positive electrode material mainly bears the supply of sodium ions and the role of high potential REDOX pairs, so it must ensure a high working voltage. The positive electrode material occupies a larger mass fraction in SIBs, providing a larger space for the full battery operating voltage[9]. At present, the known positive electrode materials are: layered structure of NaxCoO<sub>2</sub>, NaFePO<sub>4</sub>[13], NaVPO<sub>4</sub>F[13][14] and similar to Prussian Blue compounds, such as Na<sub>1.81</sub>Fe[Fe (CN)<sub>6</sub>]<sub>0.83</sub>•2.04H<sub>2</sub>O[15] and so on.

The negative electrode material provides a low potential electrode for sodium-ion batteries, which needs to meet the requirements of high voltage operation. At the same time, it is not easy to react with the electrolyte. So it has a high energy density to a certain extent[16]. The negative electrode material is the carrier of sodium ions, which plays a crucial role in the loading and release of sodium ions, thus affecting the dynamic performance of the battery[10]. Due to the large atomic radius of sodium ion, its release rate in graphite-like anode materials is very low. So it must simultaneously take into account the characteristics of high specific capacity, low REDOX potential, good cycle stability, and high first loop coulomb efficiency[9].

# **1.2** The working principle of sodium-ion batteries

The structure and working mechanism of sodium-ion batteries are similar to that of lithium-ion batteries, both of which follow the principle of "rocking chair movement". In essence, sodium-ion batteries are concentrated batteries composed of sodium-removing/sodium-embedding materials with different potential. The energy storage reaction of batteries is the process of sodium-ion embedding in electrode materials[12]. The working principle of sodium-ion batteries is shown in Figure 1.2. During the charging process, sodium ions will escape from the positive electrode material, enter the electrolyte and migrate, and be stored in the electrode material through the diaphragm. At the same time, the external electrons migrate to the negative electrode, the negative electrode potential decreases, and the positive electrode potential increases, thereby increasing the potential difference between the positive and negative electrodes and maintaining the system charge conservation. In the discharge process, sodium ions will be removed from the negative electrode. They through the electrolyte, diaphragm, etc. They finally embed in the positive electrode material. And they transmitted by the external circuit to the positive electrode, providing electrical energy for the external device to complete the discharge process. The reciprocating motion of sodium ions is used to achieve energy conversion and storage[17]. Therefore, sodium-ion batteries are figuratively compared to a "rocking chair", in which electrical energy can be easily stored and released[16]. The core of this battery technology is its ability to absorb and release large amounts of electrical energy. As for anode materials, giving their performance requirements. Carbon-based materials stand out for their excellent electrical conductivity and high energy density[7][14]; Alloyed materials provide additional strength and stability; Metal oxide materials and metal sulfide materials are favored because of their good chemical stability and high theoretical capacity. The theoretical adaptability and potential of these materials in practical applications make them the preferred electrode materials for the development of new sodium-ion batteries. By optimizing these materials, the energy efficiency and cycle life of sodium-ion batteries are improved. Bringing them closer to the performance level of traditional lithium-ion batteries[5].

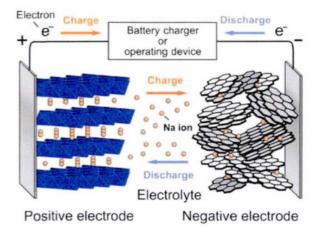


Figure 1.2 Schematic diagram of sodium-ion battery

#### 2. Sodium-ion battery anode material—Preparation of graphene oxide

Graphene oxide was prepared according to the improved Hummer's process[19]. The basic principle is as follows: Firstly, the graphite is treated with strong proton acid. And the proton acid is introduced in the interlayer to form intercalation compound between it and the graphite layer; Then, on this basis, through the introduction of strong oxidants, oxygen-containing functional groups (such as hydroxyl, carboxyl, epoxide, carbonyl, etc.) are introduced into the graphite surface layer. Due to the existence of oxygen-containing groups, the graphene oxide sheets no longer have stable  $\pi$ - $\pi$  van der Waals attraction[20], so that they are separated from each other and relatively stable. The specific method is: (1) Weigh the appropriate amount of flake graphite and sodium nitrate, mix and stir well. That need to completely mixed into the round-bottom three-neck reaction bottle. Inserting a thermometer on the left side of the round-bottom three-neck bottle, and add a mechanical mixing paddle in the middle. (2) Add an appropriate amount of concentrated sulfuric acid and stir for 20 minutes until the solid and liquid are completely dispersed; (3) Slowly add potassium permanganate, control the reaction temperature at about 65°C, about 20 minutes to complete; (4) After heating to 60°C, start temperature measurement and maintain a constant temperature of 60~70°C for one hour; (5) Reaction until the mechanical stirring cannot be stirred, stop, and pour out the reaction system; (6) Remove the slurry after the reaction and prepare twice its volume of water; (7) The drug spoon will slowly add the slurry to the water, keep stirring, and do not exceed 70°C for one hour stirring; (8) After cooling to room temperature, slowly add 30% hydrogen peroxide until there are no bubbles. (9) Before loading into the barrel, it is continuously stirred for 1~2 hours. Then centrifuged at high speed to remove the water, thereby obtaining a water graphene oxide solution, calibration. Finally ready for use.

# **3.** Application of graphene battery in sodium-ion battery and current situation at domestic and abroad

# **3.1 Application of graphene in sodium-ion batteries**

In previous graphene lithium-ion batteries, the lithium storage process is to embed  $Na^+$  into the graphene to form NaC, whose voltage platform is close to the deposition potential of lithium metal. However, the NaC negative electrode of the charged state of the graphene battery will form a solid electrolyte interface passivation film that consumes  $Na^+$  in the positive electrode material, which will negatively affect the performance of the battery[21]. Therefore, the study of graphene batteries as anode materials for sodium ion batteries is great significance to solve these problems and improve the performance of sodium ion batteries.

Specifically, studying the application of graphene batteries in sodium-ion batteries can bring the following benefits:

(1) Improving the energy density of sodium-ion batteries: Graphene has high electrical conductivity and excellent electron transport performance, which can effectively improve the energy density of sodium-ion batteries[14].

(2) Improving the charge and discharge performance of sodium-ion batteries: The high conductivity and excellent electron transport performance of graphene can improve the charge and discharge performance of sodium-ion batteries. That can increase their charging speed and discharge capacity [22].

(3) Enhance the cycle life of sodium-ion batteries[5]: The stability of graphene can enhance the cycle life of sodium-ion batteries and reduce their capacity attenuation during cyclic charge and discharge[23].

(4) Reduce the cost of sodium-ion batteries: The abundant resources and low cost of sodium ions can reduce the cost of sodium-ion graphene batteries, sodium ions have more energy reserves and lower production costs, making them more competitive in the market[24].

In summary, it is of great significance and value to study graphene batteries as anode materials for sodium-ion batteries. It can not only improve the performance of sodium-ion batteries, but also reduce their cost and promote their application and development in the field of renewable energy.

#### 3.2 Research status of graphene in sodium-ion

#### batteries at domestic and abroad

Overview of research status at domestic and abroad: At domestic and abroad, the research of graphene batteries in the field of sodium ion batteries is gradually deepening. Researchers have carried out a lot of work in the preparation, modification and matching of graphene with sodium-ion battery cathode materials.

#### 3.2.2 Domestic research status

In China, the research of graphene battery as the negative electrode material of sodium ion battery is gradually receiving attention. With the continuous development of graphene preparation technology, domestic research teams have made some important progress in the performance optimization of graphene anode materials.

(1)Preparation and modification of graphene

Domestic research teams have conducted a lot of exploration in the preparation of graphene, including chemical vapor deposition, liquid phase stripping, reduced graphene oxide and other methods. In order to improve the sodium storage performance of graphene, the researchers also try to improve the electrochemical performance of graphene by modifying methods. For example, the specific surface area of graphene can be increased, its conductivity improved, and its interaction with sodium ions enhanced by surface treatment, chemical doping, and loading with metal or nonmetallic elements[25].

(2)Composite anode materials of graphene and other materials

In order to further optimize the performance of graphene anode materials, domestic research teams are also trying to compound graphene with other materials. For example, graphene is combined with transition metal oxides, carbon nanotubes[26], polymers[27] and other materials to form a composite negative electrode material with excellent electrochemical properties. These composites can give full play to the synergistic effect of each component and improve the sodium storage capacity, cycle stability and rate performance.

(3)Research on the application of graphene battery

In addition to the research on graphene anode materials, domestic research teams are also actively carrying out application research on graphene batteries. They match the graphene anode material with the anode material of the sodium-ion battery to build a high-performance sodium-ion battery[4]. At the same time, they also studied the charge-discharge mechanism, cycle life, safety performance and other characteristics of graphene batteries to provide a basis for their promotion in practical applications[5].

#### 3.2.3 Foreign research status

In foreign countries, the study of graphene batteries as negative electrode materials for sodium ion batteries has also attracted much attention. Some well-known universities and laboratories have conducted in-depth research in the preparation, modification and performance optimization of graphene anode materials.

(1)Preparation and characterization of graphene

Foreign research teams have carried out a lot of exploration and innovation in the preparation technology of graphene, such as chemical vapor deposition, arc discharge method[28] and so on. They pay attention to the influence of structural parameters such as crystallinity, layer number and defects on the electrochemical properties of graphene[25]. At the same time, they also used various characterization methods to analyze. They study the morphology and structure of graphene in detail to understand the mechanism and law of its sodium storage properties.

(2)Composite anode materials of graphene and other materials

Foreign research on graphene and composite materials is similar to domestic research, and foreign research teams also try to compound graphene with other materials to further optimize its electrochemical properties. They combine graphene with transition metal oxides, sulfides [12][21], nitrides and other materials to form a composite negative electrode material with excellent sodium storage properties. These composites can not only increase the sodium storage capacity, but also maintain good cycle stability and rate performance.

(3)Study on performance optimization and mechanism of graphene batteries

Foreign research teams are also concerned about the performance optimization and mechanism research of graphene batteries by optimizing electrode preparation process and adjusting electrolyte composition and other methods. They change the internal intercalation mode and arrange structure of graphene batteries. Thereby, these ways can improve the electrochemical performance of graphene batteries[29]. At the same time, they also use the situ X-ray diffraction, transmission electron microscopy and other means to conduct in-depth research on the reaction mechanism and electrode reaction process of graphene batteries to provide theoretical support for further optimization of their performance[30].

In general, some progresses have been made in the research of graphene batteries as anode materials for sodium-ion batteries at domestic and abroad. However, research in this field still faces some challenges and problems, such as the preparation cost of graphene, stability, and matching with other materials. Therefore, future research workes need to further explore and optimize the properties of graphene anode materials, reduce costs, and improve stability to promote their practical application and development in the field of sodium-ion batteries[32].

## 4. Discussion and analysis

#### 4.1 Technical feasibility

Graphene has excellent conductivity, high specific surface area and good mechanical properties. That has great potential as a negative electrode material for sodium-ion batteries. At present, the synthetic technology of graphene has been relatively matured and can meet the needs of large-scale preparation. At the same time, the technology of components such as cathode materials, electrolyte and diaphragm of sodium-ion batteries also become more and more mature, which provide a good technical basis for the research and development of sodium-ion graphene batteries[31]. Therefore, the development of sodium-ion graphene batteries is feasible by a technical point of view. (1)Economic feasibility

The manufacturing cost of sodium-ion graphene batteries mainly comes from raw materials, preparation process and equipment. With the continuous progress of graphene synthetic technology and large-scale production. Expected cost will be further reduced. At the same time, sodium salt as a raw material for sodium ion batteries, rich resources, the price is relatively low. In addition, the preparation process of sodium ion batteries is relatively mature, which can reduce the production cost. Therefore, from an economic point of view, sodium-ion graphene batteries have certain competitive advantages[33].

(2)Environmental implication

The environmental impact of waste disposal and production of sodium-ion graphene batteries is an important factor to consider. Compared with lithium-ion batteries, the sodium salt in sodium-ion batteries is less toxic and has less impact in the environment. In addition, graphene can be synthesized in environmentally friendly ways, such as chemical vapor deposition. Therefore, from an environmental point of view, sodium-ion graphene batteries have better environmental performance[33].

#### (3)Resource sustainability

The abundance of sodium resources in the Earth's crust is higher than that of lithium resources. At the same time, the raw material of graphene. Graphite is a sustainable resource. It is mining and processing processes that are relatively environmentally friendly. Therefore, from the perspective of resource sustainability, sodium-ion graphene batteries have good development prospects.

#### (4)Safety performance

The safety performance of sodium ion graphene battery is an important part. Compared with lithium-ion batteries, sodium-ion batteries have higher safety performance. In addition, graphene has high thermal and chemical stability, which can further improve the safety performance of batteries. Therefore, from the perspective of safety performance, sodium-ion graphene batteries have good safety.

# **5.** Conclusion

This study summarizes the advantages and principles of graphene as a negative electrode in sodium ion batteries. It takes graphene oxide as a specific object to describe its preparation methods and principles. The application of graphene in sodium-ion batteries. As the specific anode material of graphene sodium-ion batteries. I analyze the current situation of the research and development of graphene sodium-ion batteries at domestic and abroad. By the way, I fully explained that sodium-ion batteries for the positive battery will gradually replace lithium-ion batteries one day. The development of today's battery industry, as early as the previous lead-acid battery has gradually withdrawn from the historical stage. A new generation of graphene batteries will gradually enter people's vision. Sodium ions in the earth on which people rely for survival has a higher storage capacity. It will appear in a more environmentally friendly and economical way that be applied in human's daily life. In the meantime, but also in response to China's call to achieve the goal of "double carbon". Our strive to create an economic system of energy conservation and emission reduction to achieve a community of human destiny.

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