First-principles Calculation and Mesoscopic Simulation of New Energy Battery Materials

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Abstract: With the growth of global energy demand and the pursuit of sustainable energy, new energy batteries, as important technologies in energy storage and electric vehicles, have received widespread attention. In order to improve the energy density, cycle life, and safety of batteries, it is necessary to have a deep understanding of the properties and working mechanisms of battery materials, which requires the use of first-principles calculations and mesoscopic simulations for research. First-principles calculations and mesoscopic simulations can provide theoretical simulations and predictions of the structure and chemical reaction experiments of these battery materials, providing guidance for the optimization and improvement of battery performance. Through relevant research on the types of new energy battery materials and their performance effects, and by analyzing the adsorption of lithium atoms on the single-layer surface of the battery, combined with first-principles calculations and mesoscopic simulations, this article discussed the following conclusions based on the data results: When additives A and B were used, the energy density of new energy batteries increased compared to the situation without additives at the same temperature. At the same time, when the temperature increased from 25°C to 40°C, the energy density of new energy batteries decreased regardless of whether additives were used, and the cycle stability showed a decreasing trend. The experimental results indicated that in order to optimize battery performance, appropriate additives and operating temperatures needed to be selected based on specific applications.

Keywords: New Energy, Battery Materials, First-principles, Mesoscopic Simulation

1. Introduction

There are many research theories on new energy battery materials and their first-principles calculations and mesoscopic simulations. Based on the rapidly growing electronics industry and the design of rechargeable batteries with higher storage capacity and lifespan, Makaremi M used first-principles electronic structure calculations to study the feasibility of using this nanosheet as an anode material for Li/Na/Ca/Mg/Al ion batteries [1]. Yang H conducted ab initio molecular dynamics simulations and density function theory calculations on three designed electrolytes to

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improve battery stability through the formation of anionic derived solid electrolyte interfaces in salt concentrated non-aqueous electrolytes. The simulation results showed that the concentrations of each electrolyte were high [2]. For rechargeable magnesium ion batteries as a promising alternative to commercial lithium-ion batteries, Fiesinger F studied the atomic properties of metallic magnesium using Density Functional Theory (DFT) [3]. Zhang X Y studied the modification of Fe/Co based diatomic catalysts on nitrogen doped graphene substrates through first-principles calculations to improve the electrochemical performance of lithium sulfur batteries [4]. Ertural C proposed the first first-principles calculation study based on plane wave derivation, and also explored the stability of dicyandiamide salts in the removal of Li and Na [5]. With the in-depth study of first-principles calculations and mesoscopic simulations, the complementary combination of the two can obtain comprehensive information on battery materials from atomic to macroscopic scales, thereby better understanding and optimizing the performance of batteries [6-7]. The above studies have explored the micro performance of new energy batteries using various theoretical methods, but there is a lack of experimental analysis of specific materials.

The application analysis of first-principles calculation in lithium atomic materials was a major focus of this paper. In this article, the key factors affecting the performance of new energy batteries were analyzed using the type description of new energy battery materials. Combined with the adsorption of lithium atoms on the single-layer surface of the battery, design research was conducted with the goal of improving the performance of new energy batteries. Finally, first-principles calculation methods and parameter optimization, as well as mesoscopic simulation models and performance indicator results data, were obtained.

2. Exploration of the Types and Performance of New Energy Battery Materials

Different battery materials have different chemical composition and structural characteristics, which directly affect the energy density, cycle life, safety and other performance indicators of the battery [8-9]. By exploring the types and properties of battery materials, important guidance can be provided for battery design and optimization, while laying the foundation for subsequent first-principles calculations and mesoscopic simulations [10-11]. There are various types of new energy battery materials, and the following would analyze several common new energy battery materials in detail, as shown in Figure 1:

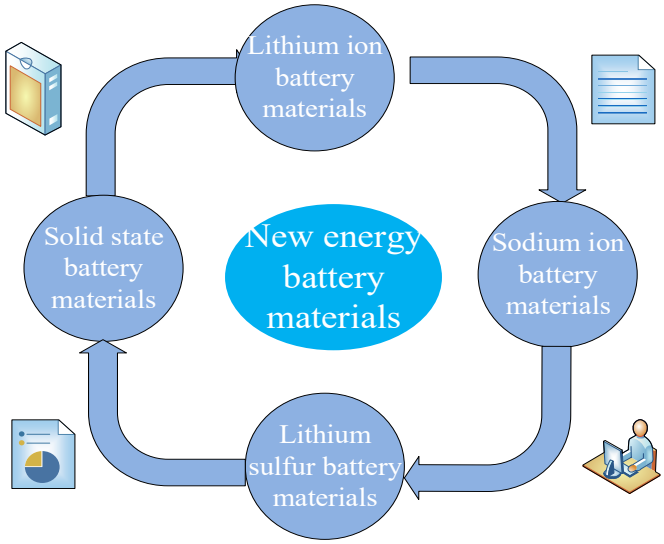

Fig 1. Type description of new energy battery materials

As shown in Figure 1, new energy battery materials mainly include four types: lithium-ion battery materials, sodium ion battery materials, lithium-sulfur battery materials, and solid-state battery materials. Among them, lithium-ion batteries are currently one of the most widely used battery technologies, with the main positive electrode material being lithium cobalt oxide (LiCoO2), and the negative electrode material usually using graphite or graphene. Sodium ion batteries and solid-state batteries are emerging battery technologies, while lithium sulfur batteries have high energy density potential. In addition to the common new energy battery materials mentioned above, there are also other emerging battery materials, such as magnesium ion battery materials and aluminum ion battery materials [12-13]. The research and development of these new energy battery materials aims to improve the energy density, cycle life, safety, and sustainability of batteries, in order to meet the performance requirements of different application fields for batteries. Materials are one of the key factors affecting the performance of new energy batteries. Based on relevant knowledge and theoretical achievements around the world, the specific research content on the factors affecting the performance of new energy batteries is shown in Table 1:

Table 1. Key Factors Affecting the Performance of New Energy Batteries

Major factor	Specific instructions
Electrode material	Having different electrochemical properties - voltage, capacity, and cycling stability
Electrolyte	Ionic conductivity directly affects the internal resistance and discharge rate of batteries
Diaphragm	The chemical stability of the diaphragm determines its reliability under high voltage

Battery design and manufacturing process	Battery assembly affects the energy density and power density of the battery
Battery management system	Monitor and manage the charging and discharging, temperature, and health status of batteries

The interaction of electrode materials, electrolytes, separators, battery design and manufacturing processes, and battery management systems collectively determines the performance, lifespan, and safety of new energy batteries [14-15]. Therefore, conducting battery performance analysis before conducting first-principles calculations and mesoscopic simulations of new energy battery materials can study and predict the impact of these key factors on battery performance. The development and optimization of batteries need to comprehensively consider these factors to achieve optimal performance and lifespan.

3. Application of First-principles Calculation in Lithium Atomic Materials

3.1 Adsorption of Lithium Atoms on the Monolayer Surface of Batteries

From the previous section, it can be seen that lithium-ion batteries are the most widely used type of new energy batteries. The adsorption and migration process of lithium atoms on the electrode surface has a significant impact on battery performance. First-principles calculations provide a way to understand this process from a quantum mechanical perspective [16-17]. The process of concentrating molecules or atoms at the upper and lower electrodes of a battery onto the surface of another adsorbent material is called adsorption. Different electrode materials have different surface properties, electronic structures, and chemical activities, as well as the structure of the electrode surface, including surface morphology, defects, and surface modification, which can affect the adsorption of lithium atoms. The adsorption of lithium atoms and their impact on battery performance are shown in Figure 2:

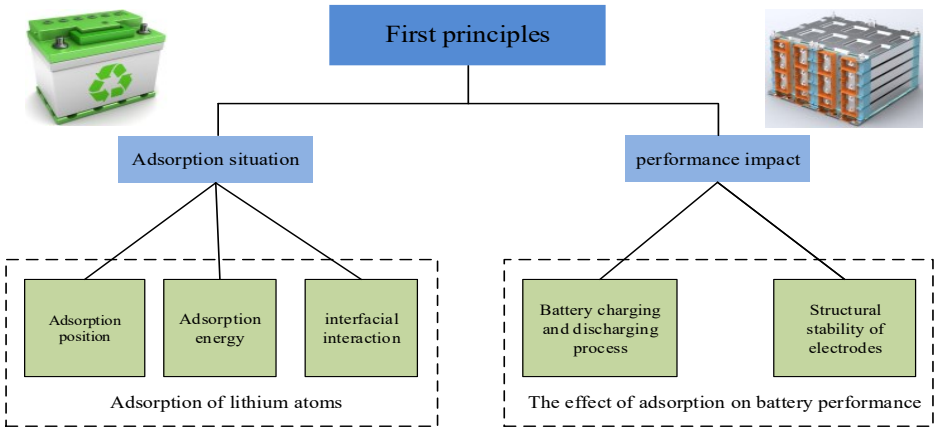

Fig 2. Structure diagram of lithium atom adsorption and its performance impact

In summary, lithium atoms usually adsorb on surface vacancies, defects, or specific active sites of electrode materials. The adsorption energy is the energy change when lithium atoms adsorb from the gas phase to the solid surface. Generally, the lower the adsorption energy, the more stable the adsorption of lithium atoms at that surface position. The first-principles calculation of new energy battery materials is based on quantum mechanics, especially density functional theory, used to calculate electronic structure and energy. This calculation does not rely on any empirical parameters, so it has high predictive accuracy. By calculating the energy changes during the adsorption of lithium atoms through first-principles, it is possible to predict the electronic structure changes when lithium atoms adsorb onto the electrode surface and evaluate the stability of the adsorption process. Therefore, it is necessary to study the adsorption of lithium atoms on the single-layer surface of batteries [18].

3.2 Calculation Process and Parameter Optimization

The adsorption and desorption of lithium atoms are important steps in the battery charging and discharging process. The adsorption of lithium atoms on the electrode surface can affect the charging and discharging rate, energy density, and cycle stability of the battery. The affinity of lithium atoms on the electrode surface determines the stability of lithium ions on the electrode surface. If the adsorption energy is too high, the lithium atoms may tightly adsorb on the electrode surface, making it difficult to desorb, thereby reducing the charging and discharging rate of the battery. On the contrary, if the adsorption energy is too low, lithium atoms may easily desorb from the electrode surface, which may lead to a decrease in the energy density of the battery. The basic steps of first-principles calculation are shown in Figure 3:

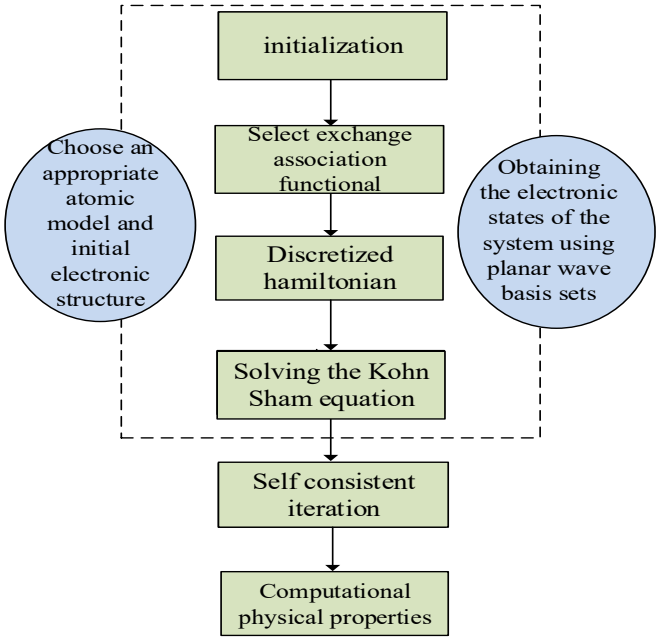

Fig 3. Flow chart of first-principles calculation for new energy batteries

From the basic step structure of the first-principles calculation established above, it can be seen that for new energy battery materials, the first step is to select a suitable atomic model and initial electronic structure, and then select the exchange correlation functional to add Hubbard U correction. Subsequently, the Kohn-Sham formula is solved using a plane wave basis system or a local orbital basis system until the energy and charge density converge to a self-consistent iteration, and finally the physical properties of the material and its performance effects are calculated. When it comes to scheduling optimization of virtual power plant systems, it is often necessary to solve simultaneously. Therefore, when constructing a scheduling model, it is necessary to consider its scheduling flexibility.

In order to effectively avoid the interaction between lithium atoms, a single-layer MoS2 supercell substrate with 4×4×1 was used to obtain two highly symmetric and stable localization points. By calculating the binding energy, the stability of adsorption sites can be easily distinguished. The specific calculation formula is as follows:

$$E = (E_0 - NE_1 - E_2) \div N \tag{1}$$

Among them, E_0 is the total energy of lithium atoms adsorbed on the surface of MS2 layered structure; E_1 is the energy of a single lithium atom in a vacuum crystal cell; E_2 is the total energy corresponding to the original 4×4×1 MS2 supercell; N is the number of lithium atoms adsorbed on the surface. Based on the determined optimal lithiation adsorption sites, the binding energy changes of lithium atoms on different sizes of lithiation substrate surfaces were further studied. The lithiated lithium atomic layer does not need to be used for lithium ion migration, so the formula for calculating the storage rate of lithium atoms on the two-dimensional material surface is as follows:

$$C = \frac{1000nF}{3600M}(mAh/g) \tag{2}$$

Among them, C is the theoretical specific capacity, n is the maximum number of moles embedded in lithium, F is the Faraday constant, and M is the molecular weight of the substance. After calculating the combined energy and storage rate, the first-principles of new energy lithium batteries were further optimized for parameters. According to the characteristics and performance of new energy batteries, there are four key parameters, namely cutoff energy, k-point sampling, exchange correlation functional, ion position and lattice parameters. For battery materials containing transition metals, high cutoff energy may be required, while the Brillouin region of the battery material may be complex and require dense k-point sampling. For new energy battery materials, it may be necessary to try various functional methods to obtain the best results, while ion position and lattice parameters are optimized through energy minimization.

4. Mesoscopic Simulation of New Energy Battery Materials

4.1. Explanation of the Steps for Mesoscopic Simulation

The mesoscopic simulation and first-principles calculation of new energy battery materials are two different calculation methods, which differ in scale, accuracy, and application fields. However, they are closely related and often combined to obtain more comprehensive information on battery material properties. The first-principles calculation is based on quantum mechanics, while mesoscopic simulation is between the micro and macro scales, mainly focusing on the collective behavior of materials, such as electrode cracking, electrolyte ion conductivity, and electrode-electrolyte interface reactions. This simulation typically uses some empirical parameters and can handle larger systems and longer time scales. First-principles calculations can provide the parameters required for mesoscopic simulations. Conversely, the results of mesoscopic simulations can also provide macroscopic environmental information for first-principles calculations. By combining their advantages, the performance of batteries can be better understood and optimized [19-20].

The mesoscopic simulation of new energy battery materials is mainly divided into four steps. The first step is to choose a suitable model and choose a suitable mesoscopic model based on the specific problem being studied; The second is parameterization. Micro simulation or experimental data are used to parameterize mesoscopic models; The third is simulation. Appropriate calculation tools are used for simulation; The fourth is verification and calibration. Experimental data is used to validate and calibrate the model. Mesoscopic simulation provides a powerful tool for the research of new energy battery materials. It bridges micro and macro scales, enabling experimenters to better understand and predict the behavior of materials in practical applications.

4.2. Effects of Additives and Temperature on Performance Indicators

After analyzing the mesoscopic simulation steps of new energy battery materials, the effects of additives and temperature on the performance indicators of battery materials were further explored through experiments. The experimental purpose is to study the changes in performance indicators of new energy batteries under different additives and temperature conditions. The experimental equipment mainly consists of a new energy battery testing system, a temperature control box, and an electrochemical workstation. The experimental materials are basic electrolytes, different types of additives A and B, and new energy battery samples. Additive solutions of different concentrations are prepared, and battery samples are placed in a temperature control box and set to the specified temperature. The battery is tested for charge and discharge using an electrochemical workstation, and performance data is recorded. The above steps were repeated, and the types of additives and temperature were changed. The final experimental data results are shown in Table 2:

Table 2. Experimental results of additives and temperature on battery performance

Serial Number	Additive	Temperature(°C)	Energy Density(Wh/kg)	Cycling Stability(%)
1	Use A	25	140.8	95.3
2	Use A	40	130.1	90.1
3	Use B	25	143.4	95.4
4	Use B	40	133.5.	90.3

5	Not used	25	125.3	95.0
6	Not used	40	114.7	89.1

From the Table 2, it can be seen that when additives A and B are used, the energy density of new energy batteries increases at the same temperature compared to the situation without additives, and the increase amplitude exceeds 15Wh/kg. This may be because the additives alter the properties of the electrolyte, thereby improving the performance of the battery. At the same time, when the temperature increases from 25°C to 40°C, the energy density of new energy batteries decreases regardless of whether additives are used, and the cycle stability decreases from about 95% to about 90%. This may be because temperature changes affect the ion conductivity of the electrolyte and the reaction kinetics of electrode materials. Therefore, in order to optimize battery performance, it is necessary to select appropriate additives and operating temperatures based on specific applications.

5. Conclusions

New energy batteries face challenges such as insufficient energy density, limited cycle life, and safety. To address these issues, it is necessary to conduct in-depth research on the structure and chemical reactions of battery materials, as well as key processes such as interface reactions and charge transfer between electrodes and electrolytes. First-principles calculations and mesoscopic simulations can provide theoretical simulations and predictions of these key processes, providing guidance for optimizing and improving battery performance. This article was based on the description of the types of new energy battery materials and the key factors that affected the performance of new energy batteries. By analyzing the adsorption of lithium atoms on the single-layer surface of the battery, and using the first-principles calculation process and mesoscopic simulation steps to explain and conduct performance testing experiments, it was concluded that additives changed the properties of the electrolyte to improve the performance of the battery, as well as the extent to which temperature affects the performance of the battery. This article aimed to provide first-principles calculations and mesoscopic simulation experiments for new energy battery materials in China through theoretical and empirical research. Due to the higher computational complexity of complex battery material systems and interface reactions, as well as the greater demand for computing resources, large-scale calculations and simulations may not be possible due to the limitations of computing resources, thereby limiting the comprehensive understanding and optimization of battery material performance. Further improvements and improvements would be made in future research.

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