Preparation and Assembly of Battery Materials of High Performance Lithium Metal

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Abstract: lithium metal battery plays an important role in the field of battery. The preparation and assembly of lithium metal battery materials also play an important role in lithium metal batteries. Through the introduction of the working principle of lithium-ion battery, the positive material, negative material and electrolyte in the structure of lithium-ion battery are analyzed. After describing the types, advantages and disadvantages of battery materials, the preparation method of lithium metal composite electrode, the assembly of button battery and the electrochemical test are discussed. Under the premise of doing a good job at present, it is essential to go on better and faster development and innovation.

Keywords: Lithium metal; Battery material; Preparation and assembly

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1 Introduction

With the continuous reduction of fossil energy and the control of exhaust emissions in order to protect the environment, the demand for electric vehicles and hybrid vehicles continues to increase. Although lithium-ion batteries have achieved great success, for the urgent need of clean energy from the society, lithium-ion batteries are needed to solve their own shortcomings and improve energy density. So it is necessary to study the lithium air battery and lithium sulfur battery. It is essential to solve the shortcomings of traditional lithium battery replacement scheme that cannot be widely used. Therefore, there is still a long way to go in the research of lithium-ion battery, and it needs continuous innovation and improvement to meet the new needs of the society.

2 Structure and principle of Li-ion battery

The electrolyte containing lithium salt, positive electrode and negative electrode constitute a typical structure of lithium-ion battery. The electrodes are isolated from each other through a membrane, commonly known as a micro-porous polymer film [1]. The film allows lithium ions to be exchanged between the two electrodes. Common electrolytes include polymer dielectric, liquid electrolyte, ceramic electrolyte and gel electrolyte. Take the lithium-ion battery with lithium cobaltite as the positive electrode and graphite as the negative electrode as an example: when it is charged under the action of an external electric field, the positive lithium ion is separated from the lithium element in the positive material LiCoO₂, the carbon atom of the negative electrode reacts with the lithium ion moving from the positive electrode to the negative electrode to generate LiC₆, which is stably embedded in the negative electrode of the layered graphite medium. In contrast to the way of discharge, lithium ions turn from the negative electrode through the internal electric field, return to the positive electrode along the direction of the electric field, and become lithium cobaltite again. The electrons in the external circuit move from the negative to the positive to form a discharge. That is to say, when charging, the positive reaction is: LiCoO₂→Li₁₋ₓCoO₂+xLi⁺+xe⁻; the negative reaction is: C+xLi⁺+xe⁻→CLiₓ; the battery reaction is: LiCoO₂+C→Li₁₋ₓCoO₂+CLiₓ; when discharging, it is the reverse reaction of the above reactions.
2.1 Positive material

According to the voltage and lithiation, the positive materials can be divided into: two-dimensional layered TiS2 and MoS2 are 2V positive materials; MnO2 and V2O3 are 3V positive materials; LiCoO2 is 4V positive materials, LiNiO2 is two-dimensional layered structure, olivine LiFePO4 and three-dimensional spinel LiMn2O4 are positive materials; olivine LiCoPO4, LiMnPO4 and Li2Mn2-xMoxO8 (M=Fe, Co) ridge three-dimensional structure are 5V positive materials.

2.2 Negative material

The chemical properties of lithium-ion batteries, such as charging speed, cycle performance and energy density, are significantly affected by the negative electrode materials. Since the first commercial use of carbon negative electrode, carbon still dominates in today’s commercial lithium-ion batteries. In recent years, the research focus of negative materials has shifted to the silicon-based negative electrode. Silicon has a very high theoretical capacity of 3572mAh/g (based on Li3Si) or 4200mAh/g (based on Li4,3Si), which is about four times of many metal oxides, even ten times of graphite negative electrode [3]. However, silicon also has the main disadvantages of large cyclic volume change (400%) and poor conductivity. This makes it difficult to use the silicon negative electrode. In order to overcome the above shortcomings, silicon nanostructures have been extensively studied. However, as a semiconductor material, Si is different from the metal negative electrode; that is to say, it is difficult to prepare Si particles on a large scale at the nanometer scale by a simple way. Cui and his colleagues proposed and proved that silicon nanowires have more advantages than silicon thin films and particles in lithium ion storage. The strain relaxation properties of Si nanowires are moderate. Nanowires can effectively transport electrons, reduce problems such as pulverization and contact loss. Bogart et al. also show that carbon coated silicon nanowires can improve the cycle performance and multiplying power performance of silicon nanowires in lithium storage. However, Dahn and his colleagues believe that although Si has the highest theoretical specific capacity (mAh/g), other materials have similar energy density (Wh/m3) with Si based on unit volume.

2.3 Electrolyte

The electrolyte shall be characterized by its ability to withstand the redox environment on the cathode and anode sides without decomposition or degradation within the applied voltage range. In addition, the electrolyte should be inert and stable within an acceptable temperature range. On the other hand, a solvent with low viscosity and melting point is required for high ion mobility in the operating temperature range. When the situation is ideal enough, the electrolyte liquid should also be environmentally friendly and can be produced at a low cost in the future. The organic liquid electrolyte with lithium salt is the most typical electrolyte in commercial lithium-ion batteries, but this kind of electrolyte has potential fire hazard.

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3.1 Preparation of lithium metal composite electrode tab

Carbon powder such as redox graphene (rGO) needs to be dried in vacuum for 12 hours at 110 °C, and then put into glove box for subsequent operation in argon glove box, in which the content of water and oxygen is less than 0.5ppm. The diameter of lithium metal is 16mm and the thickness of lithium sheet is 460 μm. First, a single Li sheet is put into the rGO powder and press it simply until the original silver white on the surface of the lithium metal cannot be seen, only a layer of rGO powder on the upper and lower sides of the lithium sheet can be seen. Then, the Li sheet covered with rGO is simply rolled until there is no rGO powder on the sheet surface. According to the final required thickness and number of layers, the rGO/Li/rGO sheet is rolled again. Here, the final material thickness is 460 μm with 10 layers of lithium sheet. The rGO / Li / rGO sheet is rolled to 250 μm. The above operations are repeated to prepare a number of 250 μm rGO / Li / rGO sheets. At this time, 10 pieces of rGO / Li / rGO are superposed and then rolled. First, the rolling thickness is set at 2cm (2×10^-4 μm), and then slowly reduced. After rolling, the composite lithium metal sheet with 10 layers of Li and 11 layers of rGO alternately superimposed is obtained, which is marked as 10Li / rGO here. In order to obtain the electrode sheets for button batteries, a circular stainless steel punch is used to punch 10Li / rGO sheet, and multiple circular electrode sheets can be obtained. Here, 10Li / rGO electrode pieces are all 1cm in diameter for assembly of button half battery. If
the final electrode sheet is made of 5 layers, the initial 10Li / rGO / rGO sheet is pressed to 300 μm thick. If the thickness is more than 10 layers, such as 20 layers, it is necessary to roll 5 pieces of 10Li / rGO / rGO sheet to 500 μm thick first, then stack and roll 4 pieces of 5Li / rGO sheet, and gradually decrease the thickness to 460 μm. This preparation process is mainly limited by the roller press.

3.2 Assembly of button battery

The purpose of using half-cell and symmetrical cell to test is to explore the electrochemical performance of composite lithium metal electrode. The half-cell consists of negative electrode shell, pure lithium sheet, diaphragm, composite electrode sheet, steel sheet, gasket and positive electrode shell. The symmetrical cell consists of negative electrode shell, compound electrode sheet, diaphragm, compound electrode sheet, steel sheet, gasket and positive electrode sheet. When it is assembled into a button battery, it is placed in the above order of composition. The next assembly can be carried out after adding 45 μL electrolyte liquid before and after the diaphragm is put in. The commonly used lithium sulfur electrolyte liquid is also the electrolyte liquid used here. Lithium sulfur electrolyte liquid: 1MliTFSI (DOL: DME=1:1vol%) +2vol%LiNO₃. Here the composition of the diaphragm is polypropylene (pp). Finally, after the positive shell is placed, the cell is sealed with a press and labeled well. The assembled cells are first placed at room temperature for 12 hours and then tested to make the electrolyte fully infiltrate the diaphragm and electrode tab.

4 Electrochemical testing of high-performance lithium-metal battery materials

In order to test the stability of Li/C composite electrode during the electrochemical cycle, it is necessary to use a constant current density for button battery during the charge-discharge test. The current density is 10 mAcm⁻², and the charging and discharging time are 1h, i.e. 2h is a cycle. The charge and discharge process is repeated until the over potential of the battery decreases or increases rapidly, that is, when the battery is short circuited, the charge and discharge test of the battery will be stopped. During the test, the lithium metal of the negative electrode is peeled off from the negative electrode first and deposited on the positive side. During the charging process, this process is carried out in the opposite direction. If the dendrites grow on one side of the electrode and pierce the diaphragm during the cycle, it means that the battery has a short circuit, and the cycle over potential of the battery decreases sharply. If the structure of composite electrode or lithium-metal negative electrode collapses during the test, a large number of dead lithium will appear. When the internal resistance of the battery increases, the circulating over potential will increase accordingly. In conclusion, constant current charge and discharge can be used to test the long-term stability and dendrite formation of electrode materials, especially Li/C composite materials, under the process of Li stripping and deposition.

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