

Judicious Design of Electrode Materials for Highly Efficient Lithium-Ion Batteries

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Abstract. The global climate change, which is intensifying yearly, makes more countries realize that mankind needs to take effective actions as soon as possible. Eliminating carbon emissions from diesel vehicles has become the research target under the spotlight in recent decades; that is to say, electric vehicles are expected to be accessible and popularized by more people shortly. Batteries are the key component of electric vehicles. However, they are also the major impediment to the development of electric vehicles for their high production costs, safety issues, durability, long charging time and low working efficiency, making them less attractive to consumers than traditional fossil fuel-powered cars. Enhancing the performance of electrodes is the most direct and effective way toward highly efficient lithium-ion batteries. This paper will introduce the mainstream electrode materials commonly used in the lithium-ion battery industry and the possible methods of improving the properties of electrode materials through compositing and nanotechnology.

Keywords: Lithium-Ion Batteries, positive electrode materials, anode materials, graphene, silicon nanowire.

1. Introduction

With the deteriorating global climate change, in which the annual temperature rises steadily by 3 degrees Celsius on average, the temperature increase causes dramatic damage to the natural environment and leads numerous species to extinction. The world is urgent to shift into a carbon-neutral world. Many countries have agreed on a carbon neutral plan, that is, achieving the balance between the number of carbon pollutants emitted and treated by the year 2050 and transforming into a carbon-clean world. Reducing carbon emissions from vehicles undoubtedly becomes one of the key segments of reaching the goal. Thus, electric vehicles (EVs) gained more attention for sustainability than ever.

Batteries, an essential component of electric cars, have become the current research highlight. In order to promote sales of EVs and encourage more consumers to choose this type of car, cutting down prices is the most effective way. However, the main obstacle to production cost reduction is batteries, which are extraordinarily pricy to manufacture due to the highly demanding materials and production methods. Lithium-ion batteries successfully beat other types, considering the range of energy storage devices developed, such as alkaline batteries, nickel metal-hybrid cells, metal-air batteries, atomic batteries, etc. It currently occupies the major niche market of EV batteries, owing to lithium ions' high energy density and abundance (low cost). In LIBs, lithium metal oxide composites are used at the positive electrode—there are already many cathode materials have already been discovered and used at a large scale during the past decades--mainstream cathode materials mainly incorporate LiFePO_4 , LiCoO_2 and LiMn_2O_4 . Graphene, as the traditional anode material, loses its competitiveness after the discovery of using silicon for the anode. In 2014, a research group from Stanford University applied nanotechnology to the production of batteries. They invented the silicon nanowire, which ideally coped with the pulverization and short life span problem when using pure silicon to LIBs anode.

Despite this, materials for electrodes seem to be the bottleneck of the progress of LIB. Several main concerns for a well-performing LIB in electric vehicles, including its safety performance, high charge capacity and long life span, that is, a great number of cycles, are found hard to achieve an ideal balance between them all. Enhancing these properties by researching and redesigning electrode materials brings accountable promising outcomes worth further exploration and investment to

promote EV battery functioning. Composite materials are regarded as the biggest hope of improving cathode performance by LIB researchers recently, and as for anode material, though silicon nanowire gained wide recognition, it still has a long way to go—reducing the production cost.

In this paper, the first section will introduce the mainstream positive electrode materials in terms of their energy density, the number of cycles, safety and costs, and also analyze how these properties are affected based on the materials' crystal structure. The second section illustrates the history, structure, properties and current development of three popular anode materials—graphene, silicon nanowire and alloys anode. There is also a further exploration of the problems encountered and the future development of silicon nanowires.

2. Cathode materials

2.1. LiFePO₄ (LFP)

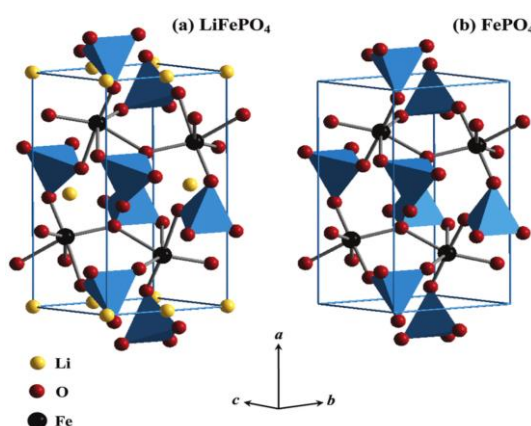
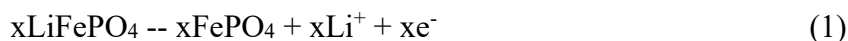


Figure 1. Crystal structures of LiFePO₄ and FePO₄ [1]

Lithium iron phosphate crystal, a composite of FeO₆, PO₄ and LiO₆, has an olivine structure belonging to the orthorhombic crystal group (Figure 1). Common vertices connect FeO₆ and its surrounding four FeO₆ crystals to form a zigzag plane layer. (This transition metal layer is sufficient to transmit electricity). PO₄ tetrahedrons connect the parallel plane layers formed by FeO₆ (each PO₄ has a common point with one FeO₆ layer, and the other FeO₆ layer has a common edge and a common point with each PO₄). There is no connection between PO₄. The structure is stable because LiFePO₄ has strong three-dimensional P-O-Fe bonds, and it is not easy to decompose bonds formed by oxygen [2]. In a crooked hexagonal closed pack lattice, the O atoms serve as the fundamental structural framework of this unit cell. At the vertex, oxygen atoms directly bond with the FeO₆ octahedron, and the LiO₆ octahedron is linked by the common side to build up a chain. Covalent bonds exist between O²⁻s and P⁵⁺s. P contributes to the stabilization of the entire skeleton because of the high bond energy possessed by P-O. The material possesses significant overcharge resistance and excellent thermal stability in this approach. The electrode reaction is as follows [3]:



In practical application, however, the actual conductivity of LFP turned to be far below the experimental estimation, mainly due to the crystal structure. In the olivine type LFP structure, the diffusion barrier of lithium ions in some directions is too high, so it can only diffuse along a specific direction where the diffusion barrier exists in less amount. Thereby, in the LFP electrode, the diffusion channel of lithium ions is restricted to one-dimensional only, and lithium ions can only diffuse along the c-axis direction (corresponding to the [010] direction of the crystal). Additionally, because the FeO₆ octahedron has no common edges and is solely connected by its common vertices, no continuous network structure is created, contributing to the material's low conductivity of electrons [3]. Considering all the chemical and physical properties above, the performance of LiFePO₄ in LIB can be concluded: high energy density, good safety performance, long life span but low conductivity.

2.2. LiMn₂O₄ (LM)

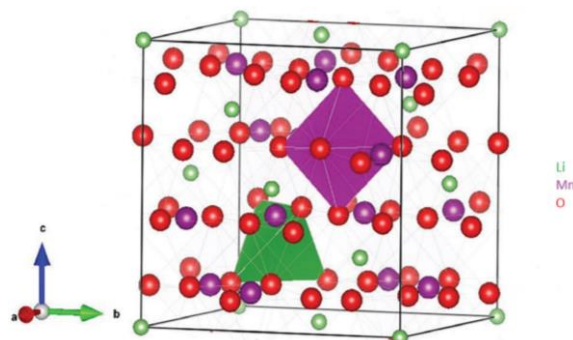


Figure 2. The crystalline structure of the spinel LiMn₂O₄ (Li, Mn and O atoms) [4]

M Thackeray, J Goodenough et al. found that LM may be applied in cathode for LIB in 1983 [5]. As part of the cell skeleton of the LM lattice, O is stacked in a cubical pattern, with Li⁺ dominating 1/8 of the oxygen tetrahedron and Mn ions dominating 1/2 of the oxygen octahedron, as shown in Figure 2. Mn³⁺ and Mn⁴⁺ each make up 50% of the structure's manganese oxidation states. In the LM lattice, the empty tetrahedrons of O are bonded in a coplanar manner. In three dimensions, these empty spaces form lithium-ion diffusion channels. By this way, Li⁺ is moved through it easily with a diffusion coefficient of 8–10 cm²/s. As seen below, the electrode reaction is [6]:



When Li⁺ intercalation and deintercalation occur, the Mn atoms in the structure can stabilize the O in the cubic close pack and support the whole skeleton. Therefore, the structure of LM is relatively firmer. The major issue related to LM electrodes is that the charge capacity decays rapidly. The main causes for such decay are: (1) LM will be transformed to tetragonal Li₂Mn₂O₄ during deep or high-rate of charging and discharging, and at the same time, Mn⁴⁺ is reduced to Mn³⁺. This valence state variation will cause the deformation of the material caused by the Jahn teller effect, causing the cell volume to increase by 6.5% [7], distorting its original lattice shape and causing capacity attenuation. (2) During the reaction, Mn³⁺ is disproportioned to form Mn⁴⁺ and Mn²⁺ and then dissolve in the electrolyte, leading to the missing of useful substances and the number of lattices that can store lithium ions [8]. Considering all the chemical and physical properties above, the performance of LM in LIB can be concluded: as safe, low cost, and stable, but its energy density is low.

2.3. LiMO₂ (M=Co, Ni, Mn)

The layered LiMO₂ cathode material is designed concerning layered LiCoO₂. It is constructed by substituting certain proportions of cobalt with nickel and manganese metals. The layered crystal structure is remained unchanged, where Li⁺ is situated between the uniform octahedral planes. Hence, Li⁺ can diffuse two-dimensionally between planes during the charging and discharging process, and its intercalation and deintercalation speeds are faster. Here is how electrochemistry works [9]:



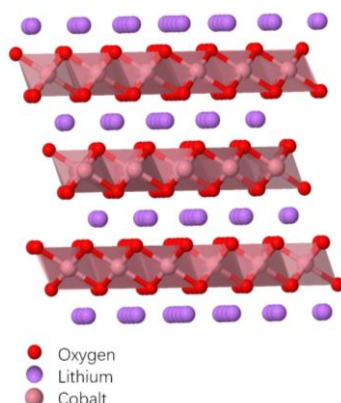


Figure 3. Crystal structure of LiMO_2 [10]

As for the layered structure of LiMO_2 (Figure 3), the following list summarizes the electrochemical and synthesis characteristics of various transition metal materials: In LiCoO_2 , the quantity of Li ions that may be intercalated and deintercalated reversibly is only 0.5 units. If this amount is exceeded, the material would undergo an irreversible phase shift and lose capacity. LiCoO_2 has poor overcharge resistance as a result. The real capacity of LiNiO_2 is between 190 to 200mAh/g as opposed to the theoretical 275mAh/g [11]. However, since Ni^{2+} have a smaller radius than Li^{+} , they frequently move into lithium ions' place during charge and discharge. This causes the dislocation of cations, leading to the crystal lattice's collapse and lowering the LIB's capacity. LiNiO_2 material also has many defects, including weak thermal and overcharge resistance. LiMnO_2 has a distinct crystal shape from LiCoO_2 . Although it can reach high experimental capacity, its life span is short [12]. The removal of Li^{+} destabilizes the structure, shifting it to a spinel LiMn_2O_4 lattice, causing capacity fade [13].

Considering all the chemical and physical properties above, the performance of LiMO_2 in LIB can be concluded: high energy density and good conductivity, but it is not durable and relatively unsafe.

3. Composite Materials

3.1. LiMO_2 /Graphene

The capacity of LiMO_2 cathode material is seriously attenuated during the long-term cycle, and the capacity is low at high current density, which seriously limits the further application of LiMO_2 . For this reason, Luo and others synthesized graphene/ $\text{LiNi}_{0.8}\text{Co}_{0.15}\text{Al}_{0.05}\text{O}_2$ with three-dimensional nanostructure by template self-assembly method [14]. Among them, the three-dimensional network formed by graphene stabilizes the skeleton of the composite cathode material, greatly improves the surface area and conductivity, and improves the composite material's rate capacity and cycle stability [15]. It can be seen from the ratio performance comparison diagram of LiMO_2 , and composite positive electrode material given in Figures 4 and 5 that the reversible capacity of composite positive electrode material is 153.6mAh/g, and the capacity retention rate is 82.1%. By contrast, the reversible capacity of NCA is only 125.3mAh/g, and the rate is 77% at 5-coulomb current density [15].

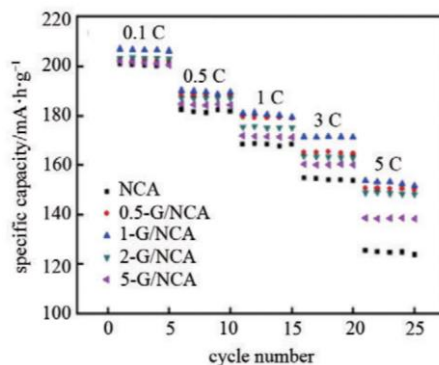


Figure 4. Specific capability at the different current rates, NCA, represents LiMO_2 [14]

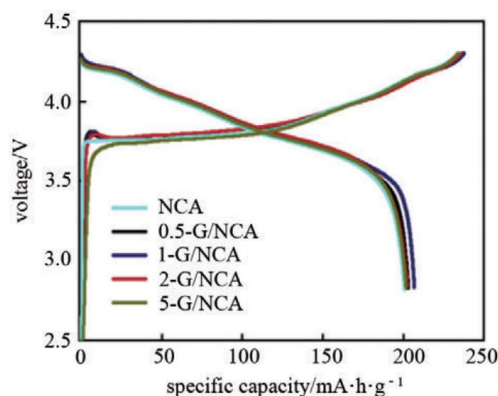


Figure 5. Initial charge-discharge curves at 0.1 C rate [14]

3.2. LFP/Carbon Nanofiber

Based on the excellent conductivity and high dimension ratio of carbon nanofiber (CNF), the electronic conductivity of LFP can be greatly enhanced by combining CNF with LFP. Adepoju et al. incorporated CNF into LFP to obtain LFP/CNF composite cathode material [16]. The composite cathode material has a higher specific capacity (150mAh/g at 0.1 C current density) and good cycle stability (98.4% capacity remained after 200 cycles at 5-coulomb current density). The long conductive paths created by the bridge built by CNF are responsible for the high-rated performance of LFP/CNF, which points out the direction for the development of LIB in further applications and commercialization.

3.3. LiMn₂O₄/Oxidized Multi-Wall Carbon Nanotube (ox MWCNT)

Liu et al. formed LiMn₂O₄/ox MWCNT composite cathode material with a three-dimensional wiring network structure by connecting spinel LiMn₂O₄ cathode material with ox MWCNT and prepared LiMn₂O₄/acetylene black (AB) for experimental comparison [17]. The charge-discharge test showed that the initial discharge capacity of LiMn₂O₄/ox MWCNT composite cathode material was 119.4, 110.6, 105.5 and 91.4mAhg⁻¹ at 0.1, 0.5, 1 and 2 C rate, respectively, which was higher than that of LM/AB composite cathode material with identical composition. This proves that ox MWCNT can significantly improve the material's rated capacity and cycle efficiency. The fitting results of electrochemical impedance show that the charge transfer resistances of LM/ox MWCNT and LM/AB are 34.3 Ω and 53.2 Ω . This is attributed to the 3-dimensional network structure formed by ox MWCNT, which increases the surface area of the cathode material, reduces the charge transfer resistance, and effectively improves the electron transfer rate and electrochemical activity.

3.4. LiMO₂/Polymers

Polypyrrole (PPY), polytriphenylamine (PYPAN), polyaniline (PANI) and other polymers have excellent electrical conductivity and unique electrochemical stability. They can greatly enhance the conductivity and contribute to stabilizing the structure of positive electrode materials by monolayer composite with layered/olivine/spinel positive electrode materials [18-20]. The results show that the composite of polymer and LiMO₂ cathode material can increase the cathode material's capacity and cycle life by effectively inhibiting the reaction of electrode and electrolyte. By ultrasonic dispersion, Yang et al. synthesized a series of LiNi_{1/3}Co_{1/3}Mn_{1/3}O₂/PTPAN cathode materials [21]. Among them, the composite cathode materials containing 5% PTPAN have better electrochemical performance than other composite cathode materials. After 100 cycles of the current density of 0.2 C, the initial discharge capacity can reach 223.7mAh/g, and the capacity retention rate is 84.39%.

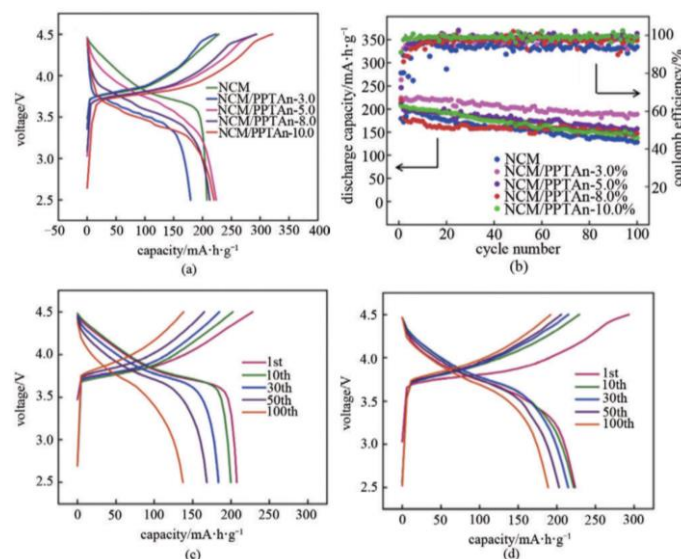


Figure 6. Initial charge-discharge curves (a) and cycling performances (b), and charge-discharge curves of different cycles for pristine NCM (c) and NCM/PTPAN-5.0% composite cathode material (d) [21]

Compared with the original $\text{LiNi}_{1/3}\text{Co}_{1/3}\text{Mn}_{1/3}\text{O}_2$, NCM/PTPAN and NCM/PPY composite positive electrode materials have a better rate and cycle performance. The performance comparison between NCM positive electrode materials and NCM/PTPAN composite positive electrode materials is shown in Figure 6. In order to improve the charge storage capacity, Puthirath et al. mixed 15% lithium substituted polyaniline (LiPo) and LFP/LM in the ratio of 9:1 to synthesize the composite cathode material of LiPo-LFP/LM and compared its performance through the cyclic voltammetry curve and charge-discharge test of assembled batteries [22]. It was found that the theoretical capacity of polyaniline was 142mAh/g after 100% lithium substitution [23]. The maximum discharge capacity of the 15% lithium substituted polyaniline LiPo-LFP/ LiMn_2O_4 composite cathode material is 37mAh/g, 12mAh/g greater than the expected experimental one--25mAh/g, and the coulomb efficiency is 99.99%. After 50 cycles, it still maintains excellent cycle stability. This shows that the composite cathode material of LiPo-LFP/ LiMn_2O_4 can be prepared by treating polyaniline with n-butyl lithium in n-hexane to form LiPo and then compositing LiPo with LFP and LM. The composite cathode material prepared has a low cost and is eco-friendly. Also, the inherent Jahn teller effect of LM is reduced to the maximum extent so that a LIB with high specific density, large capacity and eco-friendly can be obtained.

4. Anode Materials

4.1. Alloys

Alloy anodes can effectively cope with the problem of low capacity and instability possessed by electrodes made of graphene. Alloy anodes have a specific capacity of 2-10 times greater than carbon anode. Additionally, alloy anodes with greater onset voltages than Li/Li^+ , such as Tin/Sn alloys, can aid in preventing lithium deposition [24]. These characteristics make alloy anodes an intriguing subject with potential for LIB development. A concept known as alloy negative materials is also used in alloy anodes as an addition. They are merely very pure metals or alloys made of many components that have a substantial ability to store lithium ions. The insertion procedure is at the heart of the fundamental ideas behind metal anode conduction. The chemical reaction is another important idea. The active metal and lithium ions combine to form the complex Li_nM . (M represents metal). The value of n is more than 1. This is the main justification for why alloy anodes have a higher potential lithium capacity than carbon anodes, which are much higher.

Nevertheless, alloy anodes have a few associated disadvantages. Reports state that the LIB properties are substantially compromised by the enormous volumetric expansions that form a new phase [24]. Again, issues like significant capacity attenuation are caused by drawbacks like low conductivity combined with significant fluctuations in the volume of lattice while inserting/extracting lithium ions. Thereby, to heighten the performance of LIB, there are need to improve the number of cycles and capacity during loads of charging and discharging.

The two problems above are supposed to be handled by modifying and reconstructing the surface structure of alloy anodes. For instance, a layer of nanoparticles can be coated on the surface of the anode to prevent oxidation. Also, researchers are also making the effort of using carbon materials in anode--Zhang et al. created TNHCs--Tin nanoparticles with a diameter under 100nm enclosed in a hollow carbon spherical shell with a diameter close to 20nm [25]. Tin nanoparticles can expand in an ideal way according to the morphology and form of the hollow carbon spherical shell (Figure 7).

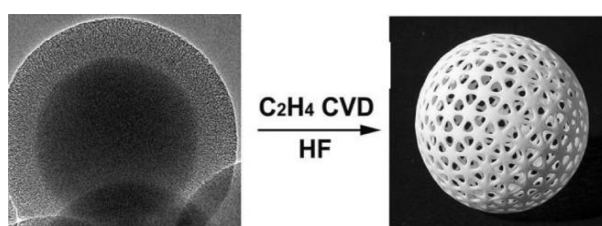


Figure 7. A hollow carbon sphere shell [26]

This end product's anode has cycling retention of over 800mAhg⁻¹, which was maintained after 100 cycles. Additionally, it demonstrated high working potential. The results of numerous studies conclusively demonstrate that the lifespan of the secondary battery is closely correlated with the amount of graphite present. The specific capacity did, however, significantly diminish, and the irreversible capacity loss increased.

4.2. Graphene

Graphene is a single layer of graphite, and carbon atoms are systematically arranged in hexagonal rings. This material has a large surface area and high conductivity (as each C is bonded to 3 other Cs, leaving one extra atom in its outer shell unbonded; this electron is free to move to cross the layers). Compared to graphite, graphene has a higher charge capacity—372mAhg⁻¹ and 744mAhg⁻¹ [27], respectively—because the two sides of the surfaces of graphene can be exposed to ions. Despite these benefits mentioned above, graphene has some drawbacks, including low density due to its low stacking efficiency. Also, the lithium ions' transportation length increases, owing to the graphene's high aspect ratio. These demerits undermine graphene's intrinsic merits: large surface area and high conductivity. The previous concerns can be addressed via functionalized graphene, also called heteroatom-doped graphene [28]. Making different kinds of three-dimensional porous C layers and two-dimensional (2D) holey C layers is another technique to overcome the aforementioned difficulties. In-plane carbon vacancy defects can be deposited onto graphene nanosheets to produce 2D holey graphene materials [29].

4.3. Silicon Nanowire

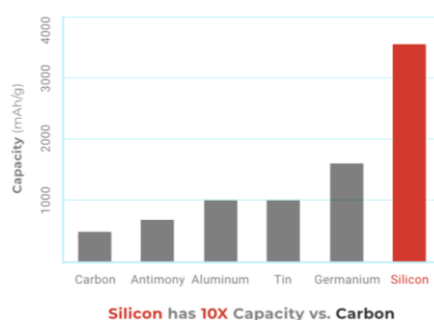


Figure 8. Silicon Anode Capacity VS. Carbon [30]

Considering the high capacity (Figure 8), abundance and similar chemical properties to carbon electrodes, silicon has become the new research target this decade. The bottleneck of using pure silicon as the anode is that the lattice will pulverize within a few cycles because of the deformation led by intercalation and deintercalation of Li^+ .

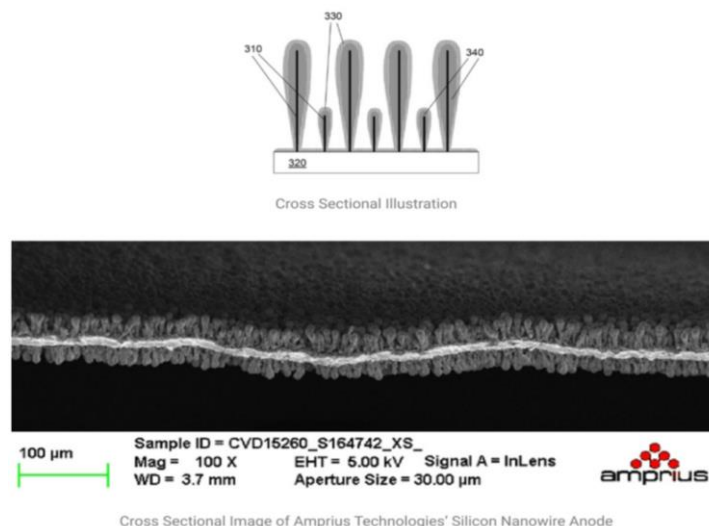


Figure 9. Cross-Sectional Illustration and Cross-Sectional Image of Amprius Technologies' Silicon Nanowire Anode [30]

The swelling issue with silicon anodes was resolved in 2007 by researchers at Stanford University. Silicon nanowires have been demonstrated to tolerate swelling and resist cracking. Amprius has mastered this process and created the first LIB anode made entirely of silicon nanowire [30]. The structure of a silicon nanowire under electron microscope can be seen in Figure 9. The established worldwide supply chain for LIB is used to manufacture the remaining battery parts and processes. Building the structure of silicon wire on the nanoscale helps to shape the inter structure better, leaving more spaces for the wire to expand when silicon is stored and enabling silicon to withstand greater stress and strain, reducing pulverization takes place. By controlling molten silicon's flow rate and deposition rate of molten silicon, the structure in the nanoscale is formed just about the target—uniformly and with enough spacing. However, the production of silicon nanowires needs to be controlled strictly and precisely, which makes the process extremely expensive. There is still a long way to go to reach the mass production goal.

5. Conclusion

The lithium-ion battery is a cylindrical container that stores chemical energy when being charged by a power supply, which is then converted into electrical energy when appliances are connected to the external circuit. It is an indispensable device for humans, and the demand for portable energy storage vessels is expanding yearly. Therefore, there is an urgent need to improve lithium-ion battery performance. The current development of LIB is stagnated because of the limitation of electrode materials. Hardly can a material be found that can possess the balanced characteristics required to be a well-performing battery: high energy density, high charge capacity, safety, long life span and low cost. Concerning cathode materials, LiFePO_4 , LiMn_2O_4 , and LiMO_2 ($\text{M}=\text{Co}$, Ni , Mn) are currently widely used in the lithium-ion battery industry, while drawbacks exist amongst them all. Compositing the existing cathode materials with carbon can obtain a product with a stable structure, but the capacity and other properties might be undermined. Anode materials, such as graphene, alloys and silicon, face the same dilemma as cathode materials. Amongst them, silicon nanowire has the strongest development potential, as silicon has superior charge capacity, enabling the battery to store and release more energy, and also has a better structure under the modification of nanotechnology. In general, all electrode materials have their highlights and defects. To achieve advancement in LIB

performance, there are needs to consider the issues at atomic scale to improve the structure to accommodate the expected properties.

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