Research Progress of Lithium-Ion Battery Current Collectors

Yaopeng Li
Changsha University of Science & Technology, China

Abstract. The current collector is one of the important components of a lithium-ion battery. It can not only carry the electrode active material, but also collect the current generated by the electrode active material to form a larger current output, which improves the charge / discharge efficiency of the lithium-ion battery. This article reviews the current research progress of single or composite current collector materials such as copper, aluminum, nickel, stainless steel, carbon, and carbon-coated aluminum foil in lithium-ion batteries.

Key words: Li-ion battery; current collector; current output; energy efficiency.

1. Introduction
With the development of the times, lithium-ion batteries continue to expand in the application market. People have put forward higher requirements for the high power charging and discharging, high safety and long service life of batteries. Current collectors are foils used in positive and negative electrodes of lithium-ion batteries. Since 2010, functional coating modification of current collectors has become an effective way to improve battery performance. Some manufacturing companies, such as the German Henkel Group (referred to as "Henkel"), the Japan Showa Denko Corporation (referred to as "Japan Showa Denko"), Shanghai Zhongxing Pano Energy Technology Co., Ltd. ("Shanghai ZTE Pano"), etc. The company developed a carbon-coated aluminum foil. These functionally modified current collectors have improved the performance of the battery to a certain extent. The degree of improvement in battery performance of functional currency collectors formed by different carbon materials is also different. This article will first introduce current collectors, and then introduce the research and development of various coatings functional modified current collectors, and briefly comment.

2. Introduction to current collectors
In a lithium-ion battery, a current collector refers to a base metal to which a positive electrode or a negative electrode of a battery is included with an active material. Generally, aluminum foil serves as the positive electrode current collector, and copper foil is used as the negative electrode current collector. The current collector comes into contact with the active material, and plays a role of collecting the current generated by the active material and outputting a large current to the outside. It can be seen that the pros and cons of contact between the current collector and the active material are an important factor affecting the charge and discharge performance of the battery.

The schematic diagram of the positive and negative electrodes of a lithium-ion battery is illustrated in fig.1. Battery positive electrode composition: 1) positive electrode current collector:2) aluminum foil; positive electrode material: 3) active material, conductive agent, binder; positive electrode tab: aluminum ribbon. Battery anode composition: 1) anode current collector: copper foil; 2) anode material:
active material, binder; 3) anode pole: nickel strip. Facts show that the current collector, as an important part of the lithium-ion battery, plays a major role in the electrochemical performance of the battery.

Fig.1 Schematic diagram of positive and negative electrodes for lithium-ion batteries

In realistic applications, different current collector materials still have such problems, so they cannot fully meet the above multi-scale requirements. For example, copper is easily oxidized at higher potentials and is suitable for use as a negative electrode current collector; aluminum is more serious as a negative electrode current collector and is suitable as a definite electrode current collector [1, 2]. Materials that can be invoked as current collectors for lithium-ion batteries include metal conductor materials such as copper, aluminum, nickel, and stainless steel, semiconductor materials such as carbon, and composite materials. This article summarizes and compares several commonly used lithium-ion battery current collector materials.

3. Current collector materials

3.1. Copper current collectors
Copper is an excellent metal conductor with an electrical conductivity second only to silver, and has many advantages such as precious resources, cheap availability, and good ductility. However, considering that copper is easily oxidized at higher potentials, it is typically used as a current collector for negative electrode active materials such as graphite, silicon, tin, and cobalt-tin alloys [3-5]. Common copper current collectors are copper foil, copper foam and copper mesh, and three-dimensional nano copper array current collectors.

3.1.1. Copper foil current collector. According to the production process of copper foil, copper foil can be further divided into rolled copper foil and electrolytic copper foil. Compared with electrolytic copper foil, rolled copper foil has higher electrical conductivity and better extension effect, but its production process is difficult to control, soaring raw material costs and foreign monopoly on key technologies also limit the application of rolled copper foil. The rude materials of electrolytic copper foil can be re-refined from waste materials such as waste copper and waste cables. The cost is quite low, which helps the sustainable development strategy and reduce the environmental pressure [6]. Therefore, for lithium-ion batteries that do not require extreme bending, electrolytic copper foil can be selected as the negative electrode current collector.

Studies have shown that increasing the roughness of the copper foil surface is beneficial to increase the bonding strength between the current collector and the active material, and reducing the contact resistance between the active material and the current collector. Accordingly, the rate discharge performance and cyclical stability of the battery are also improved. Compared with the glossy copper foil, the force required to peel the active material silicon from the matte copper foil has been doubled, as showed in Figure 2a. The initial surface of the matte copper foil/silicon electrode when discharging at a rate of 0.1C. The discharge specific capacity is as high as 1951.2 mAh/g, which is 594.2 mAh/g higher than the burnished copper foil/silicon electrode. When discharging at 0.3C and 0.5C rates, the matte copper foil/silicon electrode also shows a higher specific discharge capacity. This is illustrated in figure 2b. Correspondingly, after 40 charge/discharge cycles, the discharge specific capacity of the matte copper foil electrode is still 1080 mAh/g, which is higher than the 600 mAh/g of the glossy copper foil/silicon electrode [3] (Figure 2c). On the other hand, the electrolyte is more likely to undergo a
reduction reaction on the surface of the matte copper foil and decrease the battery performance [7]. Therefore, it is necessary in order to optimize and strictly control the surface roughness of the copper foil.

Fig.2 (a) The load-displacement Curves of the electrodes with smooth Copper foil and Crude Copper foil Current Collectors; (b) initial Charge/discharge Curves of the electrodes with smooth Copper foil and Crude Copper foil Current Collectors at 0.1C, 0.3C and 0.5C; (c) Cycle performances of the electrodes with smooth Copper foil and Crude Copper foil Collectors

3.1.2. Copper foam current collector. Copper foam is a three-dimensional network material similar to sponge, which has many advantages such as lightweight, high strength and toughness, and large specific surface area [8]. Although silicon and tin anode active materials have high theoretical specific capacities and are considered to be one of the promising anode active materials for lithium-ion batteries, they also have large volume changes and powder during the cycle charge/discharge. Such shortcomings seriously affect battery performance [9]. Studies have shown that foamed copper current collectors can suppress the volume change of silicon and tin anode active materials during charge and discharge, slow down their pulverization, and improve battery performance [10]. For instance, Tang et al. [4] studied the electrochemical performance of foamed copper/silicon anodes and compared them with copper foil/silicon electrodes. The results show that after 40 charge/discharge cycles, the specific discharge capacity of copper foil/silicon electrodes from 800 mAh/g to 400 mAh/g, the discharge specific capacity of the foamed copper current collector electrode is still 780 mAh/g, and the Coulomb efficiency is 97%, which is significantly better than that of the copper foil/silicon electrode. Li et al. [10] found that when the foamed copper/silicon electrode prepared by the electrodeposition method was discharged at 0.2C rate, after 30 charge/discharge cycles, its discharge specific capacity was still as high as 2900 mAh/g.

3.1.3. Other current collectors. The three-dimensional copper narrower current collector is composed of an array of nanometres grown on the surface of a copper foil, and has the advantages of good electrical conductivity and large specific surface area. For example, Chen Xin et al. [11] prepared a three-dimensional copper narrower array/tin electrode by electro-oxidation and subsequent chemical reduction, and tested its charge/discharge performance. The results show that compared with copper foil/tin electrodes, a tin electrode using a three-dimensional copper narrower array as a current collector not only has a higher discharge specific capacity for the first time, but also has a higher discharge specific capacity after 30 charge/discharge cycles. 199.3 mAh/g.

Hao Wenjun et al. [12] used liquid phase reaction method to directly prepare nanometer zinc cobaltite anode on a copper grid current collector. The negative electrode not only has a large specific surface area, but also has a tighter combination of the current collector and the active material, which help reduce resistance. Compared with the copper mesh current collector negative electrode prepared by the traditional coating and pressing process, the in situ growth process reduces the problem of pulverization and volume expansion of the negative electrode active material during the charge/discharge process, thereby improving the lithium-ion battery’s rate discharge performance and Charge/discharge cycle performance.
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3.2. Aluminum current collectors

Although the conductivity of metallic aluminum is lower than that of copper, the mass of aluminum wire is only half that of copper wire of the same amount of electricity. Undoubtedly, the use of aluminum current collectors can help improve the energy density of lithium-ion batteries. In addition, aluminum is cheaper than copper. During the charging/discharging process of lithium-ion batteries, a dense oxide film will be established on the surface of the aluminum foil current collector, which improves the corrosion resistance of the aluminum foil. It is typically used as the current collector of the positive electrode in lithium-ion batteries. Energetic materials include LiCoO$_2$, LiCo$_{1/3}$Ni$_{1/3}$O$_{1/3}$, and LiFePO$_4$ [13, 14].

Like copper foil current collectors, surface treatment can also improve the surface characteristics of aluminum foil. After DC etching, a honeycomb structure is established on the surface of the aluminum foil, which is more tightly combined with the positive electrode active material, and improves the electrochemical performance of the foil current collector. For example, the first discharge restricted capacity of the aluminum foil/LiCoO$_2$ electrode was increased from 138.1 mAh/g before processing to 146.2 mAh/g after processing. Accordingly, cycle stability was also improved [13]. When the aluminum foil matches the positive electrode active material LiCo$_{1/3}$Ni$_{1/3}$O$_{1/3}$, under low-rate discharge conditions, the discharge of the smooth aluminum foil/LiCo$_{1/3}$Ni$_{1/3}$O$_{1/3}$ electrode and the matte aluminum foil/LiCo$_{1/3}$Ni$_{1/3}$O$_{1/3}$ electrode is discharged. The specific capacity is not much different, but as the
discharge rate is further increased, the discharge specific capacity of the matte aluminum foil/LiCo$_{1/3}$Ni$_{1/3}$O$_{1/3}$ electrode is significantly higher than that of the smooth aluminum foil/LiCo$_{1/3}$Ni$_{1/3}$O$_{1/3}$ electrode, as shown in Figure 3. [14]

![Fig. 3 Discharge Curves of LiCo$_{1/3}$Ni$_{1/3}$O$_{1/3}$ Coated on (a) smooth Al foil and (b) Crude Al foil at different discharge rates](image)

However, in fact, aluminum current collectors are often severely corroded by the destruction of the surface passivation film, and the performance of lithium-ion batteries is also reduced. Yang et al. [15] studied the effect of the particle size of the polishing agent on the corrosion of aluminum foil in LiN$_x$(SO$_2$CF$_3$)$_y$/PC solutions. It was found that the larger the particle size of the polishing agent, the more severe corrosion of the aluminum foil surface, indicating the degree of corrosion of the aluminum foil surface. It is related to its surface roughness, that is, the higher the roughness, the more severe corrosion of the aluminum surface in the electrolyte [16]. Morita et al. [17] studied the effect of mechanical polishing and electrolytic polishing on the corrosion resistance of aluminum foil, and the results showed that the surface of mechanically polished aluminum foil was more corrosive than the surface of electrolytic polishing. Therefore, in order to increase the corrosion resistance of the aluminum foil after etching, the surface thereof needs to be optimized to form a more stable passivation film.

### 3.3. Nickel current collectors

In contrast, nickel is a base metal, which is relatively inexpensive, has excellent electrical conductivity, and is relatively stable in acid and alkaline solutions. Therefore, nickel can be used as both a progressive current collector and a negative current collector. The matching is not only positive electrode active material lithium iron phosphate [18], but also negative electrode active materials such as nickel oxide. Sulfur and carbon-silicon composite materials [19-22].

Nickel current collectors are generally available in both foamed nickel and nickel foil shapes. Due to the developed pores of the nickel foam, the contact area of the active material is large, thereby reducing the contact resistance between the active material and the current collector. When using nickel foil as the electrode current collector, as the number of charge/discharge cycles increases, the functional material easily falls off, which affects the battery performance. Similarly, the surface pretreatment process is also applicable to nickel foil current collectors. For example, after the surface of the nickel foil current collector is etched, the bonding strength of the active material and the current collector is significantly enhanced, and the internal resistance of the electrode is also decreased by 1.3 Ω [23].

Nickel oxide has the benefits of stable structure and low price, and has a high theoretical specific capacity (800 mAh/g). It is a widely used anode active material for lithium-ion batteries [19]. Take into account this, Wang Chong et al. [20] prepared a layer of nickel oxide in-situ on the surface of nickel foam by solid-phase oxidation to prepare a nickel oxide negative electrode with nickel foam as the current collector. Compared with the nickel foil/nickel oxide negative electrode, the initial discharge specific capacity of the foamed nickel/nickel oxide negative electrode has been greatly increased, and the discharge is performed at a rate of 1C. The discharge exclusive capacity of the nickel foil/nickel oxide negative electrode is only 290 mAh/g, while the foamed nickel The discharge specific capacity of the nickel/nickel oxide negative electrode has doubled. The reason is perhaps that, compared with the
two-dimensional current collector, the three-dimensional structure current collector reduces the interface polarization phenomenon and improves the charge/discharge cycle stability of the battery. LiFePO$_4$ is considered as an ideal positive electrode active material for power lithium-ion batteries due to its good safety and a wide range of raw materials. Coating it on the surface of formed nickel current collector can increase the contact area between LiFePO$_4$ and nickel foam, reduce the current density of the interface reaction, and then improve the rate discharge performance of LiFePO$_4$ [18].

3.4. Stainless steel current collectors

Stainless steel refers to alloy steel containing nickel, molybdenum, titanium, niobium, copper, iron and other elements. It has good conductivity and stability, and can withstand weak corrosion media such as air, steam, water and strong corrosion such as caustic, alkali, and salt. Chemical erosion of the medium. The stainless steel surface is also easy to form a passivation film, which can protect its surface from corrosion. At the same time, stainless steel can be processed thinner than copper, which has the benefits of low cost, simple process and large-scale production. Austenitic stainless steel and ferritic stainless steel is two types of single-phase stainless steel materials divided according to different microstructures, while duplex stainless steels contain both microstructures of austenite and ferrite. Compared with the two single-phase stainless steel that makes up for it, the duplex stainless steel is harder and more tough [26]. Therefore, stainless steel can be used as the current collector of the positive or negative electrode. The positive active material supported by it is spinel-type LiMn$_2$O$_4$, and the negative active material is MnO$_2$ [25, 26]. Common types of stainless steel contemporary collectors are stainless steel mesh and porous stainless steel.

3.4.1. Stainless steel mesh current collector. The texture of the stainless steel mesh is dense. When it is used as a current collector, its surface is covered by the active electrode material, which basically does not directly contact the electrolyte, and is not prone to side reactions, which is beneficial to improve the cycling performance of the battery. For example, when a stainless steel mesh is used as the current collector of the spinel LiMn$_2$O$_4$ cathode active material, the original discharge specific capacity and the coulomb efficiency are 112.9 mAh/g and 97%, respectively. However, due to the poor conductivity of LiMn$_2$O$_4$, the electrode impedance is large, and the electrode polarization is also worrying [24].

3.4.2. Porous stainless steel current collector. In order to make full use of the active material and improve the discharge exact capacity of the electrode, a simple and effective method is to use a porous current collector. Li et al. [25] used a three-dimensional nonporous stainless steel as a current collector to carry the MnO2 negative electrode active material. After 0.2C rate discharge, after 100 charge/discharge cycles, the discharge specific capacity was as high as 1387.1 mAh/g; At 0.5C, the discharge specific capacity is 492.9 mAh/g, which indicates the potential advantages of three-dimensional nonporous stainless steel current collectors.

3.5. Carbon current collectors

When carbon material is used as the positive or negative electrode current collector, the corrosion of the metal current collector by the electrolyte can be avoided [27], and it has the advantages of rich resources, easy processing, low resistivity, no harm to the environment, and low price.

Carbon fiber cloth can be used as a current collector for flexible lithium-ion batteries due to its superior softness, electrical conductivity, and electrochemical stability [28]. Carbon nanotubes are another type of carbon current collector. Compared with metal current collectors, their obvious advantages are lightweight and large increase in battery energy density [29]. For example, Hu et al. [30] used carbon nanotube film as a current collector, coated Li$_4$Ti$_5$O$_12$ on the surface of carbon nanotube film, and assembled an ultra-thin and flexible lithium-ion battery. After 300 charge/discharge cycles, the discharge specific capacity Still up to 140 mAh/g. Although the lithium imide electrolyte has excellent safety, stability, and electrical conductivity, it is a better electrolyte system; however, some metal current collectors are easily corroded during charging/discharging, and flexible pyrolysis Graphite
Polyimide film (PGF) is stable. For example, Han et al. Respectively coated the positive electrode active material LiMn$_2$O$_4$ on aluminum foil and PGF current collector, and tested its electrochemical performance in lithium imide electrolyte. The results show that due to the severe corrosion of aluminum foil, the discharge specific capacity of LiMn$_2$O$_4$/aluminum foil electrode after 10 charge/discharge cycles is almost reduced to zero, while the capacity retention rate of LiMn$_2$O$_4$/PGF electrode after 1000 charge/discharge cycles is still as high as 89%, it shows that PGF has better corrosion resistance than aluminum foil in lithium imide electrolyte system as a current collector [31-32].

3.6. Composite current collectors
In addition to single current collectors such as copper current collector, aluminum current collector, nickel current collector, stainless steel current collector, and carbon current collector, etc., in recent years, composite current collectors have also attracted scholars' research interests, such as conductive resins, coatings Carbon aluminum foil and titanium-nickel shape memory alloy.

3.6.1. Conductive resin current collector. Polyethylene (PE) and phenolic resin (PF) current collectors are compounded with a conductive filler and a polymer resin matrix. Shi Huijuan et al. [33] used PE and PF as matrix materials and mixed them uniformly with conductive fillers (graphite, carbon black) to prepare composite current collectors and studied their physical and chemical properties. The results show that the resistivity of the PF-based current collector is approximately 0.05 Ω · cm and the flexural strength is close to 20 MPa; while the resistivity of the PE-based current collector is greater than 0.1 Ω cm and the flexural strength is less than 10 MPa. Obviously, the flexural strength and electrical conductivity of the PF-based current collector are considerably better than those of the PE-based current collector. Graphene is a unique novel two-dimensional carbon functional material formed by sp2 hybridization of carbon atoms. It has a lot of advantages such as high electrical conductivity, specific surface area, and mechanical strength. It can replace graphite as the anode of lithium-ion batteries. Operational materials can also be used as current collector materials [34, 35]. For example, Wang et al. [35] prepared a graphene-covered plastic film composite current collector, in which graphite acts as a conductive agent and the plastic film acts as a soft matrix. Self-discharge experiments show that the battery can be kept for 30 days at room temperature. The loss of discharging specific capacity is less than 1%.

3.6.2. Titanium-nickel shape memory alloy is current collector. Titanium-nickel shape memory alloy is a binary alloy composed of nickel and titanium, which can be transformed between two different crystal phases with the change of external temperature or pressure. Studies have demonstrated that the pressure range for the transition from austenite phase to martensite phase of titanium nickel shape memory alloy is 0.2 to 0.6 GPa, while the pressure induced by lithium-ion insertion is about 3.0 GPa [36]. Therefore, the titanium nickel shape memory alloy can suppress the volume change of the active material during the charge and discharge process by changing its own phase state, and improve the cycle life of the battery.

3.6.3. Carbon-coated aluminum foil current collector. The carbon-coated/aluminum foil current collector is a multifactorial current collector in which a carbon-containing composite layer is coated on the surface of an aluminum foil. Among them, the carbon-containing layer is composed of carbon fiber and conductive carbon black particles treated with a dispersant, and can be closely combined with aluminum foil to improve the conductivity and corrosion resistance of the electrode. As showed in Figure 4, the internal resistance of the carbon-coated/aluminum foil/LiNi$_{1/3}$Mn$_{1/3}$Co$_{1/3}$O$_2$ electrodes is much lower than that of the aluminum foil/LiNi$_{1/3}$Mn$_{1/3}$Co$_{1/3}$O$_2$ electrode. When discharged at different rates (2C, 5C, and 10C), the discharge capacity of the battery is much greater. Table 1 compares the electrochemical performance of carbon-coated/aluminum foil/LiNi$_{1/3}$Mn$_{1/3}$Co$_{1/3}$O$_2$ electrodes coated with carbon layers of different thicknesses (2 μm and 5 μm). According to Table 1, it can be seen that increasing the thickness of the carbon-coated layer appropriately Helps improve the explicit discharge
capacity of the electrode [37]. In addition, covering the surface of the aluminum foil current collector with an insulating graphene oxide material also greatly improved the corrosion resistance of the aluminum foil current collector [38].

Tab.1 The discharge capacities of electrodes deposited on Al foil and on Carbon/Al foil in 2C, 5C and 10C

| Current collectors                  | Discharge capacity (mAh /g) |
|-------------------------------------|-----------------------------|
|                                     | 2 C | 5 C | 10 C |
| Pure Al foil                        | 94  | 68  | 35   |
| Al foil with 2 μm carbon layer      | 103 | 80  | 51   |
| Al foil with 5 μm carbon layer      | 115 | 90  | 60   |

Fig.4 Nyquest plots of LiNi$_{1/3}$Mn$_{1/3}$Co$_{1/3}$O$_2$ electrodes coated on Al foil and Carbon/Al foil

4. Conclusion
The current collector is one of the indispensable and vital parts in the lithium-ion battery, and has multiple functions of carrying electrode active materials and collecting output current.

The performance of current collectors prepared by different materials and different production processes is altered, and the impact on lithium-ion batteries is also different. Undoubtedly, optimizing and exploring current collector materials that can meet the multi-scale requirements of lithium-ion batteries is of profound significance for improving the charge/discharge performance and cycle life of lithium-ion batteries.

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