



# BATTERY SYSTEM MODELING

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# Lithium-ion battery characteristics and applications

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## 1.1 Introduction to lithium-ion battery technology

The lithium-ion battery industry is a great direction of global high-tech development. The lithium-ion battery has advantages of high specific energy, high specific power, high conversion rate, long cycle life, and no pollution. It is widely used in electric vehicles and various energy storage devices due to its good electrochemical stability, high energy density, long battery life, and no need for maintenance. At present, the application range of lithium-ion batteries is more extensive. Its application mainly includes five fields: transportation, electric energy storage, mobile communication, new energy storage, and aerospace/military. Its application in electric vehicles could not only replace oil with electricity and reduce greenhouse gas emissions, but also store excess electricity from the grid. As lithium-ion batteries gradually enter the market, the use and consumption of lithium resources in the world are increasing substantially, so the derived industrial chain has great development potential and broad prospects.

### 1.1.1 Development history

Along with the rapid development of new energy vehicles, the power battery industry chain has gradually shifted from policy-driven to market-driven conditions. As can be known from the perspective of market demand for power batteries, the installed capacity might no longer explode but stabilize. Under such circumstances, the power battery industry has overall excess capacity and insufficient structural performance. However, the rising industry-leading enterprises are accelerating the expansion of power battery capacity, which makes the market competition intense for industry differentiation. Due to lithium energy density and other requirements, new material and technological breakthroughs are imminent. From the perspective of power battery preparation, there is a huge development space of the current power battery equipment, which is expected to improve the localization



rate of equipment. The current power battery equipment development space is huge, and the equipment localization rate is expected to be improved.

When the lithium-ion battery was invented, it was found that many compound atoms in laminates interacted with each other in strong covalent bonds. The layered or columnar chemical substance can be provided with intermolecular forces, such as clay, silicate, phosphate, and so on. Lithium-ion batteries can be formed by the reversible reaction against lithium metal. In the early 1970s, the layered structure was proposed as the most representative cathode, and lithium metal was used as the anode of the battery system. Its reliability was also confirmed when conducting the system level application. And then, Exxon took a closer look at battery systems with hopes of commercialization. The system soon revealed many fatal flaws. First of all, the active metal lithium can easily lead to the decomposition of organic electrolytes, leading to internal pressure on the battery.

Due to the uneven potential distribution of the lithium electrode surface, lithium metal is deposited in the cathode, resulting in lithium dendrites. When the crystal deviates from equilibrium conditions, it is easy to grow like a branch and form dendritic crystals. It causes a reversible capacity loss of the embedded lithium. The dendrites can penetrate the diaphragm and connect the negative electrode. This can cause a short circuit of the battery, instantly absorbing a large amount of heat and causing an explosion, which may lead to serious safety hazards. These factors can lead to the deterioration of the recycling and safety performance of lithium metal batteries, so the system was not commercialized.

In 1980, Armand first put forward the idea of a rocking chair battery [1]. In the charge-discharge process, the lithium-ion is in the motion of a pole-negative and pole-positive state, in which both ends of the rocking chair are battery poles. Lithium compounds with low embedded lithium potential are used instead of lithium metal as the anode, and lithium compounds with high embedded lithium potential are used as the cathode. In the same year, Professor Goodenough of the University of Texas proposed a series of lithium transition metal oxides as battery anode materials such as Co, Ni, or Mn. In 1987, the researcher Auburn successfully assembled a concentration difference and proved the feasibility of the rocking chair battery idea.

Due to the high embedding potential for the negative electrode material of 0.7–2.0 V vs the Li/lithium-ion with low lithium capacity, it did not have high specific capacity advantages for the high-voltage lithium-ion secondary battery. In 1987, Sony used lithium embedded coke  $\text{Li}_x\text{C}_6$  instead of lithium metal as the anode through a battery system, using reversible embedded carbon materials for the lithium negative. It was used to prolong the service lifespan and at the same time maintain high voltage stability. The lithium-ion secondary battery cycle life is low with a poor safety performance.

Pure lithium-ion batteries started in 1989 from the invention by Nagura in Japan with petroleum coke as the positive electrode and lithium-ion cobalt as the negative electrode. In the same year, the company officially launched the first-generation market structure of the commercial batteries as coke, which adopted the battery concept for the first time. Since then, this graphite positive battery has been commercialized with the continuous deepening of the material and systematic research. Due to the advent of the rapid battery development era, it has taken the largest share in the small secondary battery market of cameras, mobile phones, notebook computers, power tools, and so on. In recent years, the lithium-ion battery has also achieved rapid development in electric vehicles.

In lithium-ion battery development history, three development characteristics are found in the world industry. First, the green environmental protection battery grows rapidly. Second, battery transition is a sustainable development strategy. Third, photovoltaic cells are becoming ever smaller and thinner. In the commercialization process of batteries, the proportion of lithium-ion batteries is the highest, especially polymer batteries, which can realize thin rechargeable batteries. It grows rapidly because of its small size, high energy, light weight, rechargeability, and pollution-free advantages. In recent years, the development of electronic information has brought many market opportunities. Because of its unique safety advantage, the lithium-ion battery has replaced the traditional battery gradually into the mainstream. The polymer type of this is called the 21st-century battery. Its development prospects are very promising.

The energy structures with fossil fuels, oil, and natural gas as the main energy sources have caused increasingly serious environmental pollution, so the resulting global warming and ecological environment deterioration have attracted more attention. Therefore, the development of renewable energy has become one of the most decisive influences in the future technological field and the future economic world. The lithium-ion battery is used as a new secondary clean energy and renewable energy. It has the advantages of high working voltage, light mass, and high energy density. It has been used widely, which shows a strong trend of development.

### 1.1.2 Energy storage technologies

Lithium-ion batteries are mainly used in a variety of portable electronic products, the application range of which continues to expand along with its application progress, material performance, and design technology. According to the cathode material, it can be classified into different types such as lithium-iron-phosphate, lithium-cobalt acid, lithium-manganese acid, lithium-nickel acid, and ternary materials. All have advantages and disadvantages, and they also have suitable application scenarios correspondingly. This chapter introduces different types of lithium-ion batteries and summarizes their advantages and disadvantages. Their working characteristics are then analyzed.

#### (1) Applications in electronic products

Due to high volume-specific energy, lithium-ion batteries can be made smaller and lighter, and they have been widely used in portable electronics. With the popularity of mobile phones, digital cameras, cameras, laptops, and handheld game consoles, the battery market has maintained rapid growth. With the high current charge-discharge performance improvement, lithium-ion batteries have also expanded to the field of wireless phones and power tools.

#### (2) Applications in electric bicycles

Public transportation has been recognized as a main method of urban transportation development in the future. However, transportation can form a broad network, and it is hard to meet different points of service. Besides, electric bicycles (e-bike) have more advantages and practicability by analyzing the objective factors of national conditions. Electric bicycles no longer produce pollution in the application process. The industrial development of electric vehicles is in line with national conditions. Especially in recent years, the global development boom has been rising again to solve energy and pollution

problems. Therefore, the lithium-ion battery plays an active role in solving social problems with a shortage of oil resources and the aggravation of environmental pollution.

(3) Applications in electric vehicles

Promoting the development of electric vehicles can reduce greenhouse gas emissions, in line with the scientific outlook on development, which is a strategic opportunity for the automobile industry. Therefore, the key to electric vehicles should be the research focus. Promoting the industrialization of electric vehicles is a strategic choice of national conditions, and an important way to ensure energy security.

At the Beijing Olympic Games, 50 electric buses claimed a record of zero breakdowns and zero failures, showing the charming style of the green Olympics. At the World Expo in Shanghai, more than 1000 new energy vehicles were used for the first time, including fuel cell vehicles, hybrid electric vehicles, ultracapacitance vehicles, and pure electric vehicles. It is estimated that this expo can save 10,000 tons of traditional fuels, eliminating 118 tons of harmful gas emissions and 28,400 tons of greenhouse gas emissions. Also, electric vehicle charging stations and other related facilities have been built with practical applications, according to which the development of the electric vehicle industry is increasingly mature.

(4) Applications in aerospace

Lithium-ion batteries are used on rovers and are planned for future missions. NASA considers lithium-ion batteries for its space missions, in addition to other space agencies. Currently, lithium-ion batteries in aviation are primarily used for launch, flight correction support, and ground operations. Battery efficiency and night operation support have also improved.

(5) Application in energy storage devices

A storage plant stores the electricity generated by a power plant from a low peak period when the electricity price is low. When selling the stored electricity during peak hours, the electricity price of the storage electric field has certain advantages over the gradient electricity price peak electricity of the power plant. Peak-valley regulation is a difficult problem. More power plants are usually needed to ensure peak demand, but it increases the cost of investment energy by keeping power plants running when demand is low. When selling electricity during peak hours, the electricity price of the energy storage field has certain advantages over the peak electricity price of the power plant.

Therefore, some companies put forward the idea of investing in the construction of energy storage power stations, reducing the purchase of large and medium-sized energy storage equipment. The charged electricity in the peak period of low power consumption and in time-sharing forms a win-win situation. As a kind of green power supply object, the lithium-ion battery is recognized as an ideal choice for high-power batteries because of its good cycling performance, high energy density, and high charge retention.

## 1.2 Battery working mechanism

The lithium-ion battery system is complex, integrating chemical, electrical, and mechanical characteristics. Consequently, the requirements of various characteristics must be considered for the battery system design. The safety and life attenuation characteristics cannot be

measured directly, as they are contained in the battery chemical characteristics, which are also not easy to predict in a short time. Therefore, when designing a battery system, it is necessary to adopt battery technology, group technology, and management system technology. This also takes into account battery safety, reliability, and durability. Lithium-ion batteries usually come in cylindrical and rectangular shapes.

### 1.2.1 Characteristic analysis

The internal cylindrical battery uses a spiral-wound structure, which is made of a very fine and highly permeable thin septum isolation material. It is spaced between the positive and negative electrodes. The main materials are polyethylene, polypropylene, and a composite material [2–4]. The rectangular lithium-ion battery is formed by laminate sheets, placing a separator on the positive electrode, then placing the negative electrode, and successively stacking. The positive pole consists of a lithium-ion collector consisting of lithium-containing materials such as lithium-cobalt, lithium-manganate, Ni-cobalt-manganese oxide, and current collector materials consisting of the aluminum film.

The negative electrode is composed of a lithium-ion collector composed of layered carbon material and a current collector composed of a thin copper septum. The battery is filled with organic electrolyte solution and equipped with a safety valve together with positive temperature coefficient components. This has the advantages of small thermal resistance, high heat-to-transfer efficiency, noncombustion, safety, and reliability. It can effectively prevent the battery from being hurt when it is in an abnormal state or when the output is a short circuit.

The lithium-ion battery is affected by abnormal factors such as short circuits, overheating, and overcharging. High-pressure gas is likely to be generated inside the battery, which causes a deformation of the battery shell and even the risk of an explosion. As for safety application, the battery must be equipped with safety valves, which is used to avoid its abnormal discharge as well as the explosion [5]. When the pressure in the battery container rises to an abnormal state, the safety valve can quickly open and expel the gas, providing protection in case of an abnormal situation [6]. Because the positive temperature coefficient element is in a low resistance state at a normal temperature, the current can be adjusted to prevent the battery from overheating when it is abnormal.

When the battery overheats due to an unusually large current caused by a short circuit or overcharging, the positive temperature coefficient element is converted into an extremely high resistance state to reduce the current in the loop [7]. Therefore, it is usually used to prevent the overcurrent of the battery and overheat caused by it to protect the battery. A single lithium-ion battery generally has a voltage of 2.8–4.20 V with a capacity of 1.5 Ah while a large-capacity one generally has a single capacity of 2–200 Ah. For new energy vehicles, several hundred volts of voltage should be introduced to meet the current range requirements [8]. However, a single battery cannot provide high voltage and energy. Therefore, the single battery cells are often processed in series or parallel to form battery packs to meet the voltage and power requirement from the power supply system.

### 1.2.2 Components and working principle

The lithium-ion battery is mainly composed of four parts: the positive electrode material, the negative electrode material, the diaphragm, and the electrolyte. Anode materials provide

lithium ions for batteries. Common materials include lithium manganate, lithium cobalt oxide, and lithium nickel cobalt manganate [9]. The cathode material is mainly graphite. The main function of lithium-ion batteries can be introduced to store lithium ions, which realizes embedding and disembedding in the charge-discharge process [10]. The diaphragm is a special composite mode that prevents electrons from moving freely between positive and negative electrodes in a battery, but lithium ions can move freely in the electrolyte.

The electrolyte is generally composed of lithium salts together with organic solvents, which conduct lots of lithium ions. Electrons cannot exist independently on a carrier, and the diaphragm is essentially an insulator that cannot contain free electrons. Consequently, it cannot conduct electricity. In a battery, elements are ions that can easily pass through the membrane while electrons escape from elements to a new carrier no matter whether its positive or negative [11]. When in contact with the membrane, it cannot absorb free electrons from the electrode, thus preventing electrons from passing through.

The common materials of the diaphragm are single-layer polypropylene, polyethylene, and composite three-layer polypropylene-polyethylene-polypropylene septum. The electrolyte realizes the conduction of the lithium ions between the positive and negative electrodes of the battery. Currently,  $\text{LiPF}_6$  is the most widely used electrolyte. It is an objective representation of its internal structure. There are lithium ions, metal ions, oxygen ions, and carbon layers inside the battery [12]. The lithium-ion batteries are mainly composed of compounds. Internal reactions take place through the movement of ions in the battery. And then, the diaphragm of the battery acts as a barrier to keep the two poles of the battery apart as shown in Fig. 1.1.

The lithium-ion battery is an indispensable portable energy storage element. Its performance is characterized by many external parameters such as voltage, current, and internal

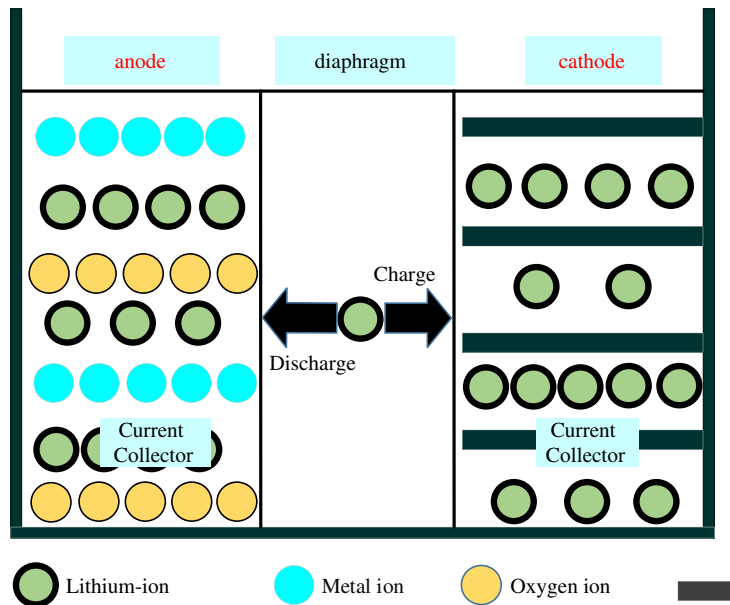


FIG. 1.1 Schematic diagram of the lithium-ion battery structure.

resistance. The reason it is used more than other batteries is that it has many advantages compared with other batteries. First, it has a high heat of combustion, which is the amount of heat given off per unit. Second, it is more environmentally friendly and can meet the requirements of green social development [13]. Third, it has a long cycle life. Under normal conditions, it can be charged and discharged hundreds of times. Consequently, it can be used for a long time [14]. Finally, it has no memory effect. The battery working process causes the battery capacity to be lost, resulting in less and less capacity; this is the memory effect that does not exist on lithium-ion batteries [15]. It has many other advantages for its widely used conditions, such as good safety performance, low self-discharge, fast-charging, and wide operating temperature range.

The internal chemical reaction of the lithium-ion battery is a redox reaction, which is also the working principle of the battery in the application process. It converts electrical energy into heat energy through a chemical reaction. According to the chemical reaction equation, the battery charge-discharge process is the embedding and disembedding process of the lithium ions. When a battery is charged, the positively charged lithium atom undergoes an oxidation reaction, losing electrons and becoming lithium ions. Numerous lithium ions are produced by the oxidation reaction of the positive electrode. These lithium ions start from the positive electrode and pass through the electrolyte solution to the carbon layer of the negative electrode.

The battery capacity is related to the number of lithium ions that are produced in the positive electrode reaction. It is related to the number of lithium ions that are exchanged with a negative electrode through the electrolyte [16]. In the discharge process, an oxidation reaction occurs in the negative electrode. In this process, lithium ions embedded in the negative carbon layer come out and move back to the positive electrode. The more lithium ions returning to the positive electrode, the higher the discharge capacity [17]. Similarly, when charging, the lithium ions are generated in the positive electrode of the battery, which moves to the negative electrode through the electrolyte. The lithium ions in the negative electrode can be embedded in the pores of the carbon layer [18]. When more lithium ions are embedded, the charging capacity becomes higher. The internal chemical reaction process of the lithium-ion battery is described as shown in Fig. 1.2.

Electrolytes are dissolved organic solutions to lithium salts [19]. In general, the electrochemical reaction process of the lithium-ion battery is the exchange of the lithium ions by the back and forth transformation from positive to negative poles. The positive and negative electrode reaction, according to which the total reaction equations are described, is as follows. The positive electrode reaction, negative electrode reaction, and the total battery response can be described as shown in Eq. (1.1):

$$\begin{cases} \text{P: } \text{LiM}_x\text{O}_y = \text{Li}_{(1-x)}\text{M}_x\text{O}_y + x\text{Li}^+ + xe^- \\ \text{N: } n\text{C} + x\text{Li}^+ + xe^- = \text{Li}_x\text{C}_n \\ \text{T: } \text{LiM}_x\text{O}_y + n\text{C} = \text{Li}_{(1-x)}\text{M}_x\text{O}_y + \text{Li}_x\text{C}_n \end{cases} \quad (1.1)$$

In the above three equations, M can be Co, Mn, Fe, and Ni, respectively, representing lithium-cobalt-oxide, lithium-manganese oxide, lithium-iron-phosphate, and lithium-nickel-oxide batteries. Its operating principle is different from the oxidation-reduction process of ordinary batteries, but the embedding stripping process of the lithium ions can be reversibly

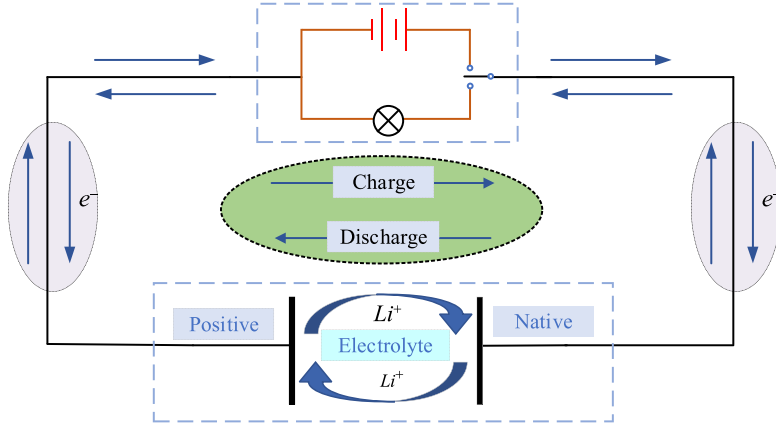
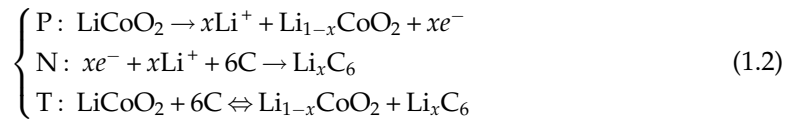


FIG. 1.2 Lithium-ion battery working principle.

embedded or extricated from the main material. In two stages of charge-discharge, they are embedded and deembedded from the positive to negative electrodes. In the charging process, it is first deembedded from the positive pole through the electrolyte to the negative pole, which is then embedded into the negative pole. Its negative electrode realizes a lithium-rich state at this time point.

Rich lithium is a positive electrode material made by doping a small amount of lithium with positive electrode active substances such as  $\text{LiMn}_2\text{O}_4$ . It can make the volume changeless in the charge-discharge process of cell contraction, which improves the structural stability and cycling performance of the material. The charge-discharge processes are opposite to each other. The cathode material of the lithium-ion battery is composed of a lithium embedded compound. If there is an external electric field, ions in the cathode material can be released and embedded from the lattice under the action of the electric field. The battery reaction takes place against its positive pole and negative electrode at the same time, according to which the general equation can be described as shown in Eq. (1.2).



It has three important parts called the diaphragm, the positive electrode, and the negative electrode. Its work mainly relies on the movement back and forth between negative ions, which is caused by the lithium-ion concentration difference between ions on both ends. During the charging process, the lithium ions are disembedded from the positive electrode and embedded into the negative electrode through the corresponding electrolyte. After a series of chemical reactions, the positive electrode is in a state of less lithium and the negative electrode is in a state of more lithium [20]. Meanwhile, it compensates for the charge from the external circuit of the negative electrode. In the discharge process, the lithium ion is dislodged from the negative electrode and inserted into the positive electrode again by the action of the electrolyte.



### 1.2.3 Lithium-ion battery construction

The composition of lithium-ion batteries is some compounds. The reaction is completed by the movement of ions by using the diaphragm as a barrier. The diaphragm separates the two electrodes of the battery effectively as shown in Fig. 1.3.

The segmented charging of the lithium-ion battery can ensure that it can be filled quickly but not charged and play a certain repair role of the battery that cannot be discharged over a long period. At present, there are two main charging modes for it: constant-current and constant-voltage charging strategies [21]. Whether it is a constant-current or constant-voltage condition, the charging mode can be mainly divided into five stages: trickle charging, low-voltage precharging, constant-current charging, constant-voltage charging, and termination of charging.

### 1.2.4 Charge-discharge strategies

The charging method is a constant-current and voltage limit, most of which are controlled by an IC chip. Typical charging methods are introduced. First, the voltage to be recharged is tested. If the voltage falls below 3.00 V, it needs to be recharged. At this time, the charging current is generally one-tenth of the set current. Until the voltage rises steadily to the terminal voltage, it enters the standard charging process, which is described as follows. First, constant-current charging is conducted at the set current. When the battery voltage rises to 4.20 V, it is changed by using the constant-voltage charging treatment. The charging voltage continues to be 4.20 V for charging. After charging for a period, the charging current gradually drops. When it drops to one-tenth of the set current, the charging process is finished.

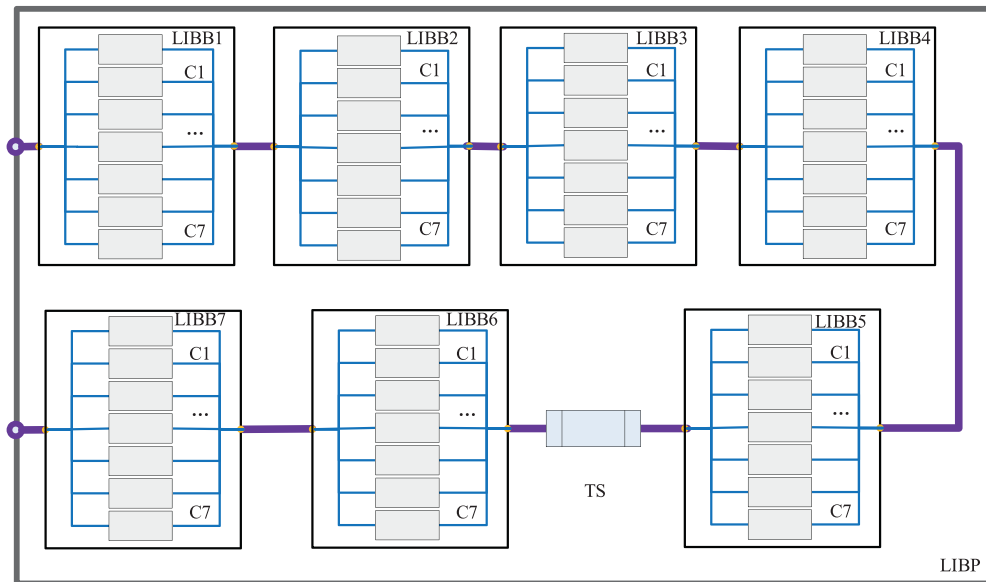


FIG. 1.3 Lithium-ion battery packing structure.



The first stage is the trickling charge, which is mainly used for precharging recovery charging or fully discharged battery cells.  $C$  is a representation of the nominal capacity of the battery of current. For example, if the battery currently has a capacity of 1000 mAh, then  $1C$  is the charging current of 1000 mA. Trickle charging is used when the battery voltage is lower than 3.00 V that is set as the limited value [22]. The trickle charging current is one-tenth of the current in constant-current charging mode, namely  $0.1C$ . If the constant charging current value is 1.00 A, the trickle charging current currently is set as 100 mA for the terminal value.

The second stage is the constant-current charging. When the voltage value of the battery rises above the trickle charging threshold, the charging current at this time is increased to the constant-current charging. In general, the current value of constant-current charging should be varying from  $0.2C$  to  $1.0C$ . The voltage of lithium-ion batteries gradually rises with the constant-current charging process. Generally, the voltage value set by a single battery is 3.00 V to 4.20 V. The whole structure of the lithium-ion battery packing system is described in Fig. 1.4.

The third stage is the constant-voltage charging. When the voltage value of the lithium-ion battery rises to 4.20 V, the constant-current charging stage ends and the constant-voltage charging stage begins. At this time, the change of current value is determined according to the saturation degree of the cell. With the charging process, the charging current gradually decreases from the maximum value. When it decreases to  $0.05C$ , the charging is considered to terminate [23].

The fourth stage is the charging termination, in which there are two typical methods of charging termination. The minimum charging current is used to distinguish, or the timer is used, or both are combined. The minimum current method is used to monitor the current value in the constant-voltage charging stage and the charging current value is terminated when it decreases to  $0.05C$  or ranges from  $0.02C$  to  $0.07C$ . The second method can be adopted as the timing treatment approach. The time of the constant-voltage charging stage is the initial time point and the charging process is terminated after 2 h of continuous charging.

The above four-stage charging method takes approximately 2.5–3 h to complete the charging of a fully discharged battery. After charging, if it is detected that the battery voltage is lower than 3.89 V, it is recharged. In the charge-discharge process, there is a certain pressure in the battery of empirical data onto 0.30–0.60 mPa. Under the same pressure, the larger the stressed area, the more serious the deformation of the battery wall of the shells. In the first charge-discharge process of the liquid lithium-ion battery, the electrode material and electrolyte react to the solid-liquid interface, forming a passivation layer covering the surface of the

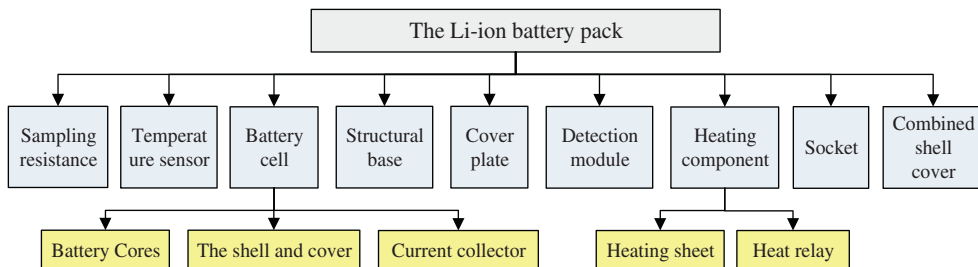


FIG. 1.4 Lithium-ion battery packing system.

electrode material [24]. The formed passivation film can effectively prevent the passage of solvent molecules. Meanwhile, the lithium ions can be freely inserted and extracted through the passivation layer. This passivation film has the characteristics of a solid electrolyte, so it is named as Solid Electrolyte Interface (SEI) film.

## **1.3 Lithium-ion battery chemistries**

According to the classification of positive electrode materials, lithium-ion batteries can be classified into lithium-iron-phosphate, lithium-cobalt, lithium-manganate, lithium-nickelic, and ternary materials.

### **1.3.1 Lithium-ion battery family**

The phase-changing material could change its physical state in a certain temperature range [25]. It is used as the thermal conductivity medium, which is attached to the surface of the single cell; the heat dissipation effect has been greatly improved. Besides, there are also plans to combine the heat conduction materials with water cooling, so that the water cooling system heats the conduction materials absorbed by the heat transfer of the outside of the system. As for the lithium-ion battery system to prevent the thermal runaway problem, the ideal situation is to be able to directly detect the temperature, voltage, and current parameters of each cell. Therefore, even if there is no new type of sensor with high quality, low price, and good performance, the early warning and prevention of thermal runaway conditions may be successful. The number of battery cells in the system is small, and it is an important competitiveness of square batteries.

Compared with soft-pack and square lithium-ion batteries, cylindrical 18,650 batteries were the earliest available commercially. They are the most automated and cheapest power battery type. With years of support from Tesla, it is a three-way race against soft packs and square batteries. The cylindrical battery family also has one more star after Tesla announced that Model 3 uses the 21,700 cylindrical lithium-ion battery. The following is a brief description of several technical points related to the cylindrical battery of those not specifically stated in the process, and the cylindrical batteries are specifically the 18,650 battery type.

The cylindrical battery is the most studied and discussed type. It is mainly composed of the positive electrode, negative electrode, diaphragm, safety valve, overcurrent protection device, insulating parts, and shell [26]. There is more steel shell in the early stage and the aluminum shell is the main one at present. Each manufacturer cell overcurrent protection device design is not the same. The price is completely different according to different security requirements. General safety devices mainly have positive temperature coefficient resistance and fuse two categories. The excessive current, resistance heating, and temperature accumulation promote the rise of the positive temperature coefficient resistance value.

When the temperature exceeds a threshold, the positive coefficient resistance value suddenly increases, which can separate the fault cell from the overall circuit effectively to avoid its further thermal runaway conditions. In principle, the fusing device is a fuse. In the case of excessive current, the fuse fuses, and the circuit is disconnected [27]. The difference between the two types of protection is that the former can be restored. The latter protection can only be