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Technology and Development of Cathode Materials for Secondary Cell Batteries Review

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ABSTRACT

Lithium cell batteries have experienced explosive market growth recently, and the rate of expansion is expected to accelerate in the future. Lithium cell batteries were first commercialized in 1990, and have experienced steady development and market growth since then. As the secondary cell battery market increases, many companies and research institutes are researching various anode materials and materials to secure technological competitiveness. The demand for highly efficient lithium secondary cell batteries is expected to increase rapidly due to the rapid increase in demand for energy storage systems. The anode material is one of the core materials of lithium secondary cell batteries, accounts for the largest proportion of material costs, and is a key factor that determines the performance of the battery. In this review paper, we will describe the background of various major anode materials from the perspective of secondary cell batteries and describe the advantages and disadvantages of each material.

Keywords: Secondary cell battery, Rechargeable battery, Battery, Storage battery, Cathode

1. Introduction

With the development of human civilization, the use of fossil fuels as an energy source has steadily increased. These fossil fuels emit a lot of greenhouse gases, including carbon dioxide, through combustion reactions, accelerating global warming. The various climate change phenomena observed recently and the problem of global warming is becoming a reality. A significant amount of carbon dioxide generated on Earth is emitted from the combustion of gasoline and diesel in automobiles. Therefore, a transition from existing fuel-burning vehicles to new types of vehicles is needed, and therefore electric vehicles are receiving a lot of attention as next-generation vehicles. In electric vehicles, lithium cell batteries are mainly used as electric energy storage devices. Lithium cell batteries have experienced explosive market growth recently, and the rate is expected to accelerate.

The anode material of cell batteries has been using the traditional LiCoO material, which has not changed much from the early lithium cell batteries. However, the cell battery market, which is rapidly increasing recently, is experiencing new problems, including raw material supply. (Luo et al., 2016; Nzereogu, Omah, Ezema, Iwuoha, & Nwanya, 2022; Yoshio et al., 2002) As the secondary cell battery market increases, many companies and research institutes are studying various anode materials and materials to secure technological competitiveness. In this review paper, we will describe the background of various major anode materials from the perspective of secondary cell batteries and describe the advantages and disadvantages of each material.

2. Cathode material technology and development

Because lithium is highly reactive, it exists in the form with an oxide in the positive electrode of a battery. The materials which participate in the battery electrode reaction at the positive electrode, such as lithium oxide, are called active materials. The number of electrons that can be stored varies depending on which cathode active material is used, which determines the capacity and voltage of the battery. Many types of lithium oxide can be created by adding other materials to lithium and oxygen, and there are broadly known five types of active materials that provide performance suitable for batteries. (Luo et al., 2016; Nzereogu et al., 2022; Yoshio et al., 2002) The positive electrode active material is composed of a combination of lithium and metal components, and has different characteristics depending on the type and ratio of the metal. Nickel, Ni, improves high capacity characteristics, manganese, Mn, and cobalt, Co, improve safety, and aluminum, Al, improves output characteristics. The appropriate combination of these metal materials is important to satisfy battery performance requirements in various applications. Since the ultimate goal of an electrode is to achieve maximum capacity within a given space, it is important to increase electrode density by maximizing the amount of anode material loaded within a range that does not impair performance. The electrode density is also greatly influenced by the characteristics of the cathode material. In particular, particle size, size distribution, and particle density are important factors, so attention should be paid to these factors from the research stage. The positive electrode active material must meet the following characteristics.

1. Since irreversible phase transition in the crystal structure shortens the lifespan of the battery, insertion/desorption of lithium ions during charging and discharging must be reversible and not cause phase transition.

2. Side reactions unrelated to the insertion/desorption reaction of lithium ions should be eliminated as much as possible. If a side reaction unrelated to the circulation of lithium ions occurs in the anode or cathode, the efficiency of the battery decreases.

3. In order not to shorten the life of the battery, it must not cause a chemical reaction with other elements of the battery such as electrolyte.

4. The particle size must be relatively constant. This is because it improves electrical conductivity as well as the contact efficiency between particles, and does not damage the aluminum thin film when making the electrode plate.

LCO batteries are lithium cobalt oxide-based batteries, a material used in lithium secondary cell batteries, and currently occupy the largest portion of the battery market. The LCO battery has a layered structure, a nominal voltage of $3.7V$, and a rated capacity of $150mA/g$, making it a high energy density. NCM batteries are ternary batteries using Ni, Co, and Mn. They have a higher energy density than other active materials, making them suitable for large-capacity batteries, and can be charged and discharged thousands of times, making them widely used in electric vehicle batteries. Recently, research is being actively conducted to develop NCM batteries with high energy density by increasing the proportion of Ni to 80%. NCM battery types include NCM111, NCM523, NCM622, and NCM811, and each number represents the mixing ratio in order of Ni, Co, and Mn. (de Biasi et al., 2019; Fan et al., 2020; Kasnatscheew et al., 2016)

The NCA battery is a ternary battery using nickel (Ni), cobalt (Co), and aluminum (Al) and is suitable for manufacturing large capacity batteries, but it can be charged and discharged only a few hundred times, so it is used for power tools in the small battery market. It is mainly installed in cylindrical batteries and is the second most widely used after NCM batteries. (Li, Lee, & Manthiram, 2020; Purwanto et al., 2018)

LMO batteries do not use cobalt (Co), so their raw material costs are low and they have high-efficiency charge/discharge characteristics. (Tran, DaCosta, Mevawalla, Panchal, & Fowler, 2021; Wu & Lee, 2017) However, a rapid decrease in capacity occurs at the beginning of the charge/discharge cycle, and a decrease in capacity also occurs at high temperatures. It is reported that this is due to structural instability that causes loss of oxygen or elution of manganese during charging.

The LFP battery is a battery using $FePO_4$ and has an olivine structure rather than a layered structure, so it is very stable. It is characterized by a very long lifespan as the crystal structure does not soften even when lithium ions escape. (Tran et al., 2021) Because it uses iron instead of cobalt, it is very inexpensive and eco-friendly, so research is being actively conducted to apply it to electric vehicles. However, due to its structural characteristics, lithium ions do not move actively and have low electrical conductivity, so research and development on the diffusion path of lithium ions is necessary.

LiCoO cathode material - LiCoO, which has a rhombohedral structure, is easy to manufacture, mass-produced, and has high reliability. Although it is an expensive raw material, it is currently used as the main cathode material for commercialized lithium secondary cell batteries. (Y. Jiang, Qin, Yan, & Sui, 2019; Wang et al., 2022) When synthesized above 800°C, a layered structure of LiCoO is formed, and for commercial applications, the particle size is grown to 10um or more by sintering at a high temperature above 1,000°C for a long time. When synthesized at about 400°C, a spinel structure is formed, and electrochemical properties are poor due to defects inside the crystal and low crystallinity. Therefore, LiCoO, which has a layered structure synthesized at high temperatures, is mainly used as an anode material. LiCoO cathode material has a density of $5.1g/cm$, which is the highest among lithium secondary cell battery cathode materials. In addition, it is the most widely used cathode material because it has excellent lifespan characteristics and high temperature performance. However, due to the high density, the strength is high, and there is a high possibility of foreign substances being mixed in due to wear of the equipment during the process of pulverizing the LiCoO lump obtained during the synthesis process, so careful attention must be paid to managing impurities. In addition, due to the recent rapid rise in raw material prices, it is losing some of its appeal. LiCoO's charge and discharge characteristics improve when surface modification is performed. Surface modification appears to be not a simple coating effect because the coating material does not completely cover the entire surface of LiCoO, but only a certain portion, but rather the coating material prevents the elution of Co by changing the stability of the surrounding crystal structure. It is expected that surface modification technology to increase the capacity of LiCoO will continue to develop in the future.

LiNiO cathode material - LiNiO is structurally very similar to LiCoO, but Ni tends to prefer divalent atoms rather than trivalent atoms, making it difficult to implement an accurate phase during the synthesis process. (Bianchini, Fauth, Hartmann, Brezesinski, & Janek, 2020; Kalyani & Kalaiselvi, 2005; Xu et al., 2016) In the case of LiCoO, Co is stable, so LiCoO is formed relatively stably depending on the Li/Co ratio during the synthesis process. However, in the case of LiNiO, LiNiO is formed, and some Ni changes to divalent, deteriorating electrochemical properties. In particular, Ni present in the lithium layer not only hinders the movement of lithium, but also greatly increases irreversibility, thereby reducing reversible capacity. Additionally, because LiNiO undergoes various types of phase changes during the charging process, its structural stability is greatly reduced and its lifespan characteristics are poor. This problem can be partially solved by replacing part of Ni with Co. Even a small amount of Co doping can form a significant amount of Ni. However, because Ni is still formed in the lithium layer, large-scale synthesis is not easy and excessive lithium must be added or heat treated in an oxygen atmosphere for a long time. In general, as the Ni content increases, the formation of Ni increases, so the stability of the layered structure depending on temperature decreases. Therefore, the sintering temperature is lowered and the diffusion rate of ions is reduced, so not only does it take a long time to form a layered structure, but there is a high possibility that unreacted substances will remain even after synthesis. However, despite these shortcomings, Ni-based materials have a high energy density, which is advantageous for high capacity, and are being continuously studied. Recently, with the development of surface modification technology, Li[NiM]O(M=Co) with high capacity is being developed to overcome these shortcomings. Similar to what was applied to LiCoO, surface modification with oxides, etc. can improve electrochemical properties. Because surface modification stabilizes the surface structure, thermal structural stability is also expected.

Li[NiMnCo]O - A three-component cathode material using Ni, Mn, and Co, showing the individual advantages of LiNiO, LiMnO, and LiCoO. It is a material that is being actively researched because it has many advantages in terms of safety, lifespan, and price. (Ecker et al., 2014; Gao et al., 2019; J. Jiang, Gao, Zhang, Zhang, & Jiang, 2019) Three-component anode materials have different properties and structures depending on the content of each component. In particular, Li[NiMnCo]O, in which each component is added equally, is a material that has recently been successfully commercialized and used. Structurally, this material has the same layered structure as LiCoO, but it is composed of a large structure called superlattice, so it is structurally stable and can be driven at high voltage. Although this material does not have a very high electrode density, it has been actively studied recently as applications requiring high power have increased. Even when driven at a high voltage of 4.5*V*, it shows stable lifespan characteristics. At 4.6*V*, there is a decrease in lifespan, which is probably due to the decomposition of the electrolyte. It is likely that various compositions will be developed in the future, and various materials will likely be developed. Another material with potential for development is Li[NiMn]O, and like Li[NiCoMn]O, the ratio of nickel to manganese is 1:1, so it shows similar characteristics. If electrolytes that can withstand high voltage are developed in the future, their utilization is expected to increase.

LiMnO spinel compound anode material – Mn is an abundant resource, so it is inexpensive and environmentally friendly, and its structural stability provides excellent battery safety, making it a suitable material for high-safety, high-capacity batteries such as electric vehicles. (Bruce, áRobert Armstrong, & Gitzendanner, 1999; Kong et al., 2015) There are several problems in using spinel-structured LiMnO as an anode material. The biggest problem is Mn elution. This problem can be solved by doping with a monovalent or divalent element to increase the average valence of Mn. However, in this case, the capacity decreases because the amount of Mn participating in the oxidation/reduction reaction decreases. The reason for the dissolution of Mn is that Mn is generated due to an unbalanced reaction on the electrode surface during discharge. As the eluted Mn dissolves in the acidic electrolyte solution, the amount of LiMnO active material decreases. In addition, the eluted Mn is deposited as a metal on the cathode, preventing the movement of lithium ions or causing reduction decomposition of the electrolyte solution, drastically reducing performance. In particular, the reason why capacity decreases significantly when overcharging at high temperatures is because this catalytic reaction is promoted. Recently, like LiMnAlO, some of the Mn is replaced with Al, etc. to further increase the oxidation number of Mn and improve high-temperature performance, so it is being applied to HEV batteries that require high output characteristics. In addition, methods of coating heterogeneous elements on the surface to reduce capacity reduction have been developed and applied, but are not commercially utilized. An important feature of the LiMnO spinel compound cathode material is that its structure is relatively stable and the battery has good safety even when overcharged. These advantages are the reason why this material is expected to be one of the most suitable materials as a cathode active material for industrial batteries in the future.

Fe-based olivine anode material - The representative Fe-based olivine anode material is LiFePO, and iron is one of the most abundant metals and is much cheaper than Co and is environmentally friendly. (Tan et al., 2014; Xiang, Yin, Zhang, Hou, & Xu, 2021) The theoretical density of LiFePO is 3.6*g/cm*, which is relatively low compared to other cathode materials, and the theoretical capacity is 170m*Ah/g* (2.0~4.2*V*). If a defect exists in the structure, the lithium movement channel is blocked, causing extremely poor charging and discharging. Therefore, when synthesizing LiFePO, it is important to create a defect-free crystal structure. One of the biggest problems with LiFePO is that its electrical conductivity is quite low. Additionally, if the uniformity of the conductive material within the electrode is not secured, the capacity rapidly deteriorates. To increase the conductivity of LiFePO, methods such as particle size control, surface coating of carbon film, and doping with elements such as Nb are being applied. The solid-state reaction method is most commonly used as a synthesis method, and since it is synthesized at high temperature, the particle size of the sample increases rapidly, reducing the surface area, and the diffusion of lithium ions is reduced, leading to a decrease in battery performance. Therefore, methods for synthesizing at low temperatures need to be developed to minimize particle growth.

Solid solution compound anode material - A solid solution compound is a case where compounds with two or more structures form a solid solution. A representative example is Li[NiLiMn]O, which is a solid solution of LiMnO and Li[NiMn]O. (Atlung, West, & Jacobsen, 1979; Jin et al., 2020; Park, Park, Kim, & Kang, 2022) In addition, there are (1−x)LiMnOLiMO(M=Ni, Co, Cr), LiMO-LiMnO, and LiMO-LiMnO-LiMnO. In particular, oxidation/reduction reactions of Mn occur in existing manganese oxides, but since the oxidation state of Mn4+ is maintained in these solid solution materials, the JahnTeller effect due to Mn does not appear. This composition is known to be a solid solution, but depending on the composition change, it shows the characteristics of a solid solution and a composite. It is a solid solution up to a certain composition, but beyond that it exhibits the characteristics of a composite. When 0.3LiMnO·0.7LiMnNiO, a representative solid solution, is charged to a charging voltage of 4.8*V*, the charging and discharging capacities are 352 and 287*mAh/g*, respectively, and a flat voltage appears in the voltage region around 4.5*V*. This plateau voltage is related to the initial irreversible capacity and is caused by the reaction in which LiMnO forms LiO during charging. For this material, it is important to select an appropriate cut-off voltage because the cut-off voltage of charging determines the amount of residual LiMnO, which determines the stability of the structure. However, solid solutions have the disadvantage of low electronic conductivity, and various studies have recently been conducted to overcome this.

LiFeSO anode material - LiFeSOF has very low activation energy for lithium diffusion, resulting in a fast lithium diffusion rate. (Luo et al., 2016; Nzereogu et al., 2022; Yoshio et al., 2002) Due to these characteristics, LiFeSOF has higher discharge voltage and ionic conductivity compared to LiFePO. However, LiMSOF is unstable at high temperatures (600-700°C) and cannot be synthesized through general solid-state reactions. Therefore, synthetic processes using ionic and solvothermal methods that can be synthesized at low temperatures are being studied. LiFeSOF anode material manufactured through ionic and solvothermal methods has a reversible capacity of ~130*mAh/g* (2.5-4.2*V*, @0.1*C*), and the discharge voltage is 3.6*V*, which is higher than that of LiFePO (3.45V). LiFeSOF has a higher voltage and excellent conductivity compared to LiFePO, so there is no need for particle nanoization or coating processes.

LiFeBO anode material - LiFeBO has a lower molecular weight than PO, a theoretical capacity of 220mAh/g, which is higher than LiFePO, and a density of 3.46g/cm, showing a large energy density per volume similar to LiFePO (density:3.50g/cm3). (Bo et al., 2012; Tao et al., 2014) However, the low discharge voltage (~2.9V) is a disadvantage. To date, LiFeBO has been mainly produced using solid-state methods. The mechanochemical method has the advantage of reducing particle size by grinding during the mixing process, and can be coated using a carbon precursor. LiFeBO manufactured by mechanochemical methods has a usable capacity of ~120mAh/g (1.0-4.8V, 5mA/g), and has recently been manufactured by adding KB, ketchen black, and VGCF, vapor grown carbon fiber. A usable capacity of ~200mAh/g (1.5-4.5V, @0.1C) was achieved, which is higher than that of LiFePO. However, there is a disadvantage that electrochemical properties deteriorate when left in the air due to an increase in Fe_3^+ due to moisture absorption.

LiFeSiO anode material - Compared to LiFePO, LiFeSiO has a high theoretical capacity (~330mAh/g) and energy density, and also has good chemical stability, so it has the advantage of excellent thermal safety and low price. However, there are problems with low rate capability, electrical conductivity, and shortened lifespan due to crystalline degradation during the charging process. (Larsson, Ahuja, Nytén, & Thomas, 2006; Togashi, Honma, & Komatsu, 2014) To overcome output characteristics due to low electrical conductivity, there are methods of reducing particle size or coating conductive carbon.

3. Conclusion

In this paper, we reviewed the types of positive electrode active materials and their characteristics. In the future, the demand for highly efficient lithium secondary cell batteries is expected to increase rapidly due to the rapid increase in demand for mid- to large-sized energy storage systems, including small-sized energy storage systems, and economical manufacturing technology and advancement of manufacturing technology are urgently needed. In particular, the anode material is one of the core materials of lithium secondary cell batteries, accounts for the largest proportion of material costs, and is a key factor that determines the performance of the battery. In particular, various types of batteries are being developed as their application in new fields such as electric vehicles and power storage expands in addition to existing batteries for mobile phones and laptops. Because these new application fields require characteristics different from those required for existing small information and electronic devices, the materials applied to batteries must also be differentiated. Therefore, a three-component cathode material that shows better performance at high power and high temperature has been developed from the LiCoO2 cathode material used for small batteries, and in particular, Fe-based cathode materials with excellent safety are expected to be used in new application fields in the future. In conclusion, high capacity, safety, and high output in lithium secondary cell batteries are characteristics that must be continuously developed in the future, and technology development in new fields will also be determined depending on the speed of technology development.

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