Advanced Electrode Materials for Lithium-ion Battery: Silicon-based Anodes and Co-less-Ni-rich Cathodes

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Abstract—The development of higher-performance rechargeable lithium-ion batteries (LIBs) is critical to the substantial development of electric vehicles and portable electronic devices. The cost of lithium-ion batteries needs to be decreased more and the specific energy as well as recycling degradation rate needs to be enhanced further. Silicon anodes and cobalt-free nickel-rich cathodes are widely regarded as promising materials for the next generation of lithium-ion batteries. This review discusses the current state of research on silicon anode nanomaterials and nickel-rich cathode materials without cobalt.

1. Introduction
Due to their high energy density and good storage capacity, lithium-ion batteries (LIBs) have become the most competitive battery type in the world. Nevertheless, in the current era of LIBs, when there is a growing demand for higher energy density mobile power, lithium-ion batteries still require significant improvements, notably to prolong the operating time of portable devices and the range of all-electric vehicles. Moreover, the cost and environmental impact of batteries will also determine the further development of lithium-ion batteries.

Rechargeable lithium-ion batteries consist of two electrodes, a diaphragm and the electrolyte. Conventional graphite anode and cobalt-containing cathode materials have many defects that need to be improved. Graphite is used commercially as electrode material in anodes because of its high conductivity, good reversibility, and relatively low cost. But graphite has a limited energy density because it can hold only one lithium atom for every six carbon atoms. On the other hand, silicon is usually regarded as one of the most promising anode materials since each silicon atom can bind to four lithium ions, giving it ten times the capacity of a graphite anode. However, during repeated charge/discharge cycles, the volume of silicon lattice changes dramatically, and the electrolyte interface film on the anode surface is unstable. Over the past decade, various silicon nanostructures have been successfully demonstrated to address the significant challenges of electrolyte interface crushing and cementation associated with reversible silicon anodes. In cathode, the traditional lithium-ion battery cathode material contains the
metal cobalt, which is closely related to the cost of the battery and causes great harm to the environment. Without improvements in cathode materials, cobalt consumption will increase fourfold in the next 40 years because of the demand for lithium-ion batteries, which means that the pollution involved in cobalt mining will cause even more trouble. In Congo’s cobalt mines, for example, miners have high levels of cobalt in their blood, which dramatically increases their risk of cancer. In addition, global cobalt prices are highly volatile, which can hamper the development of lithium-ion batteries and the market for electric devices and electric vehicles. Because of the above reasons, the research of low cobalt-nickel-rich batteries has become the mainstream direction.

This review will introduce the research status of silicon anode nanomaterials and nickel-rich cathode materials without cobalt. The part of nanostructured silicon-based anodes mainly focuses on two methods to solve the volume expansion of silicon materials: the construction of nanoscale structures and the preparation of silicon/carbon composites. In the nickel-rich cathode section, we will review the early LiCoO$_2$ batteries, introduce three kinds of low cobalt cathode (NMC, NFA, NCA).

2. Nanostructured silicon-based anodes

The lithium storage capacity of the silicon anode (3579 mAh g$^{-1}$ Li$_{15}$Si$_4$ at room temperature) is approximately ten times that of the carbon anode (372 mAh g$^{-1}$ LiC$_6$) used in commercial lithium-ion batteries. Nevertheless, during repeated charge/discharge cycles, the volume of silicon changes dramatically ($\geq 300\%$), and the surface electrolyte interface (SEI) film on the anode is unstable, resulting in easy crushing, low cycle efficiency and permanent productivity loss of silicon as an anode material. There are two main ways to solve these problems. The first approach is to build silicon nanoscale structures that can prevent volume expansion; the second is to mix silicon with other stress-releasing materials, such as metals, oxygen-containing compounds, and carbon materials.

2.1 Pristine silicon nanomaterials

2.1.1 Silicon nanoparticles

Generally, silicon nanoparticles larger than the critical particle diameter ($\approx 150$ nm) will produce large volume changes during lithium insertion/extraction. This large volume expansion/contraction produces large stresses, which leads to the cracking and crushing of silicon, resulting in electrical contact loss and decreased capacity (Fig.1). However, the production cost of silicon nanoparticles with particle diameters less than 150 nm is so high that it is hard to achieve industrial production. This is an important factor hindering the widespread use of silicon nanoparticles in lithium batteries. Over the past decade, various silicon nanomaterials have been successfully demonstrated to address those problems.

Fig.1 Schematic diagram of failure modes of silicon anode during repeated lithium/evaporation process: (a) pulverization of silicon particles and fracture of the electrode; (b) silicon formation and growth of solid electrolyte interface on Si surface [6]
Binders can alleviate the influence of the volume effect of silicon nanoparticles on the overall structure of the electrode. Silicon interface exposed to air will form a micron-scale oxide layer (SiO$_x$) and eventually become SiO$_2$, preventing the inside silicon from further oxidation, which results in the formation of many hydroxyl groups on the surface of silicon particles. Binders such as sodium alginate (SA) and sodium carboxymethyl cellulose (CMC) contain many carboxyl groups, which can produce esterification reaction with hydroxyl groups on the surface of silicon materials. The process uses binders to bond the material with hydrogen bonds. In repeated expansion and contraction, binders reduce particle pulverization and ensure the close connection between the silicon particles, the conductive agent, and the fluid collector. The stability of the silicon anode is enhanced, and the SEI film on the silicon surface becomes stable as well.

Inspired by SA and CMC, researchers found that amic acid, polyacrylic acid (PAA), and citric acid can also be used as anodic binders for silicon. Among them, PAA materials have attracted extensive attention because of their simple linear chemical structure. The mechanical properties of PAA are better than CMC and SA materials, and PAA is conducive to the design of cross-linking networks. He. D et al. designed in situ cross-linking PAA with D-sorbitol to generate a 3D network bonding agent (C-PAA-DS) (Fig.2), which has excellent mechanical properties and can constrain the influence of volume expansion of silicon material on electrode structure.

Besides, recently, some scientists have found some more straightforward and cheaper ways to synthesize silicon nanoparticles. Xu et al. developed a simple and industrially scalable process for fabricating silicon nanoparticle electrodes with high specific and area capacities and cyclic stability. During this process, silicon nanoparticles were deposited directly on a porous carbon collector by plasma-enhanced chemical vapor deposition (PECVD) (Fig.3). The obtained additive/binder-free electrode can be used as anode for lithium-ion batteries without further treatment. Experiments with dis-/charge for 100 cycles at 0.8 A g$^{-1}$ confirmed that the composition and structure of the electrode were retained after the first cycle and remained largely intact during the cycle.

Fig.2 Schematic representations of active materials/binders' configurations for c-PAA-DS during lithiation/delithiation of Void@SiOx@C [11]

Fig.3 Silicon nanoparticles: (a) Top view SEM of nanostructured silicon layer at 200 nm magnification; (b) TEM images of an individual particle as-synthesized from PECVD; (c) Nyquist plots of the Si NP anode in a half cell with Li metal counter electrode and liquid electrolyte: before cycling and after 1, and after 100 cycles [12]
2.1.2 Silicon nanowires (Si NWs) and porous silicon

In addition to binders, structural engineering of silicon nanomaterials to improve their cyclic performance is also a valuable direction. Silicon nanowires and porous silicon nanoparticles provide enough space for expansion, also promising candidate materials for future lithium-ion batteries. Silicon anodes with nanowire structures are more resistant to fracture than other larger silicon structures. Because the total elastic energy of nanowire structures is usually insufficient to drive crack initiation and propagation during deformation, for smaller structures, the stress release volume associated with crack growth is insufficient to overcome the surface energy penalty associated with crack growth. In addition, Si NWs have a continuous one-dimensional electron path that allows for efficient charge transfer, so the anode no longer requires carbon additives and polymer binders. This contrasts with the inefficiency with which electrons jump between particles in traditional grout-coated battery electrodes. In 2007, Yi Cui’s team first proposed the preparation of silicon anodes for silicon nanowires by gas-liquid-solid synthesis method, and then expanded supercritical fluid-liquid-solid (SFLS) method was used to prepare silicon nanowires. Schulz et al. demonstrated that silicon nanofiber anodes could also be synthesized by electrospinning. Flake et al. also reported the preparation of silicon nanowires anodes by metal-induced chemical etching of silicon wafers. In the past, most Si NWs were synthesized by a gas-liquid-solid method with an Au catalyst. Recently, some scientists have used sensing technologies to demonstrate higher characteristics of Ag-Au catalyst systems, such as an increase in the number of Si NWs per unit area, a decrease in the thickness of Si NWs, and an improved diameter distribution (Fig.4).

Moreover, porous silicon, which also effectively reduces the volume expansion and is widely used in lithium-ion batteries, could be obtained by etching, metal reduction, wet chemistry, and other methods. Li et al. synthesized large (>20 μm) mesoporous silicon sponges (MSS) by electrochemical etching method. They proved their excellent performance as lithium-ion battery anode. During the charge/discharge process, the volume change of the silicon wall is mainly adjusted by the internal pores of the MSS, resulting in the expansion of the total particle size only around 30%. Furthermore, it does not shatter even after a thousand charge and discharge cycles (Fig.5d). On the other hand, the preparation of porous silicon nanoparticles usually requires hydrogen fluoride, which is highly corrosive and may lead to a significant reduction in silicon content in the material and may cause considerable pollution to the environment. Based on magnesium thermal reduction, Du et al. of Singapore University of Technology and Design developed a green pathway to synthesize silkworm cocoon-like Si@C yolk structure (P-Si NRs@void@NC) (Fig.5e) by selecting Al2O3 as a sacrificial template, avoiding the use of HF [20].
Fig.5 Porous silicon materials: (a), (b) and (c) SEM and TEM images of the MSS at 20 μm, 100 nm, and 100 nm, respectively; (d) Typical charge-discharge profiles of the MSS anode with 46 wt.% MSS loading; (e) TEM image of the p-Si NRs@void@NC composite, showing a silkworm cocoon-like morphology (inset); (f) Nyquist plots of the p-Si NRs@void@NC composite and bare p-Si NRs electrodes after cycling at 0.5 A g⁻¹ [19,20]

2.2 Silicon-based composites

Compared to other silicon-based composites, silicon/carbon material composites have great cost and technical difficulty advantages. Carbon materials generally have decent conductivity and ductility, making them ideal for improving the conductivity of composites and mitigating the volume change of silicon. Carbon materials include two kinds of materials: graphite and pyrolytic carbon or polymer precursor. Take silicon/graphite composites as an example. The size of silicon in composites is usually smaller than that of graphite because the pores formed between the graphite particles are like the size of expanded silicon particles, which is conducive to accommodate the volume change of silicon materials. Silicon/carbon composites usually have the embedded and core/shell (cladding) structures. S.Jung et al. reported that the silicon nanoparticles could be embedded in the concave graphitic film so that the silicon nanoparticles were confined in the nano space of engineering nanocarbon film. The discharge capacity is maintained at 2500 mAh g⁻¹ under the condition of 0.5 A g⁻¹ for 200 cycles, with a coulomb efficiency of over 98% (Fig.6). X. Dong et al. prepared silicon nanoparticles wrapped in carbon matrix by using polyacrylonitrile (PAN). To wrap the silicon nanoparticles in the carbonaceous matrix thoroughly, PAN/Si films were made by uniformly dispersing the silicon nanoparticles in excess PAN solution through mechanical stirring. Then carbonization was used to created Si nanoparticles wrapped in PAN-based carbonaceous matrix (Si@C) (Fig.7). Wu et al. synthesized PDA/GO-Si composites using polydopamine as a coating and applied them to lithium-ion battery electrode materials (Fig.7d). The prepared silicon is mixed with the GO and stirred, and then PDA is added to the system and finally get the PDA coated GO/silicon composite. PDA/GO-Si has excellent capacity and stability, maintaining a capacity of 1300 mAh g⁻¹ after 450 cycles. Besides, recently, Shi et al. created a three-dimensional
hierarchical porous carbon framework prepared by a self-assembly CaCO₃ template method and finally got Si@Voids@PC composites that exhibit an improved rate capability.

Fig.6 Silicon core/shell (cladding) structure: (a) Schematics of Si/nanocarbon-based electrode; (b) Image of the silicon nanoparticles impregnated in the concave nano space; (c) Coulombic efficiency and specific capacity for 30 charge/discharge cycles for the concave graphitic electrode [21]

Fig.7 Silicon nanoparticles wrapped in carbon matrix: (a), (b) TEM image of the Si@C material the in different magnification observation; (c) Cycling performance and rate capability of Si@C; (d) Charge/discharge cycling performance of GO-Si and PDA/GO-Si composite electrode at the current density of 500 mA g⁻¹ [22,23]
3. Nickle-rich cathode

With the continuous development of battery technology, the research of low cobalt-nickel-rich batteries has become the mainstream direction.

Since John B. Goodenough first proposed applying LiCoO$_2$ as a cathode material in lithium-ion batteries in 1980, LiCoO$_2$ has been a commercial lithium battery material for a long time. The layered structure with the strongest thermal stability is shown in Fig.8.

![Fig.8 Layer LiCoO$_2$ structure [26]](image)

LiCoO$_2$ has many advantages, such as high working voltage, stable charge, discharge voltage, simple production process, etc. However, with the development of electric vehicles and other fields, the energy demand is increasingly urgent. The disadvantages of LiCoO$_2$ as a cathode material are gradually amplified:

1. Actual specific capacity is lower than ideal. At a typical charge cutoff voltage of 4.2 V, the actual capacity is only 50% of the theoretical value. In addition, because its over-charging performance is poor, the actual specific capacity is rapidly reduced at a continuously high charging voltage.

2. Its service life is short. After repeated charge and discharge, the cathode material structure will change: Its internal resistance may increase, resulting in increased voltage, causing overcharging, making the lithium branch crystals of negative poles increase rapidly. Eventually, the diaphragm is punctured, resulting in an internal short circuit, the cycle life will be significantly reduced.

3. The thermal stability is poor. As the battery reacts, a large current is generated inside the cell, causing the temperature to rise continuously. The diaphragm may be melted at a high temperature of 150 °C, when the electrolyte breaks down to produce the gas, increasing the internal pressure and eventually causing the battery to explode. Especially under high voltage, the stability of the interface of the material becomes worse, and the side reaction between the material and the electrolyte intensifies, which seriously affects the safety of the battery.

4. Metal elements are expensive. The average content of cobalt in the earth's crust is 0.001% (by mass), making it expensive compared to other metals. The cost of LiCoO$_2$ has been an important issue after its commercialization. Although the cathode's reliance on cobalt has been a great success in the portable electronics market, the high price of battery packs which it is led to, has seriously hampered the rapid growth of the electric vehicle market.

Therefore, increasing research has focused on the development of a nickel-rich cathode, cobalt materials are gradually abandoned.

3.1 Ni-rich lithium nickel cobalt aluminium oxide (NCA) cathodes

Ni-rich NCA batteries have a higher capacity and lower Co usage. Increasing the nickel content in the electrode material can increase the energy density and reduce the cost of the battery. However, the high-capacity nickel-rich LiMO$_2$ electrode's cycling performance is lacking due to the formation of
microcracks in the cathode particles in the deeply charged state. In the deeply charged state, there will be microcracks formed in the cathode particles. These microcracks grow from the inside of the particle to the surface of the particle from the inside out (Fig.9), allowing the electrolyte to penetrate the particle and limited depth of discharge (DOD), which causes a significant reduction in energy density, thus increasing the cost of EVs.

![Fig.9 Growth of microcracks](image)

Ryu et al. doped NCA with boron improved the electrochemical performance. They synthesized P-NCA and B-NCA by calcination with separate doping of B and P. The microstructure calculations and experimental comparison of their properties concluded that B-NCA has better electrochemical properties. Although the raw materials used to synthesize the two cathode materials are the same, the microstructure of the two cathode materials is completely different, as the P-NCA particles are made up of polygonal particles. In contrast, the B-NMC is made up of needle-like particles (Fig.10). This is because B doping changes its surface energy, allowing the formation of needle-like particles. The advantage of this needle-like structure is that the cathode particles are stronger. Only a few tiny cracks nucleate in the center of the cathode particles during charging and discharging. The tiny cracks are barely visible under fully charged conditions. Thus, the recyclability of the battery is enhanced (Fig.11). The B-NCA cathode increases the battery's energy density to a large extent and extends the battery's life. The cycle stability of Ni-rich NCA cathodes can be significantly improved by B-doping the NCA, which indicates that changing the surface energy is an excellent way to improve cell performance.

![Fig.10 SEM images of as-synthesized (a) P-NCA89; (b) B-NCA88 cathode particles](image)

![Fig.11 Long-term cycling performance of P-NCA89 cathodes cycled at DOD 60% and 100%, and B-NCA88 cathode cycled at DOD 100%](image)
3.2 LiNi<sub>x</sub>Co<sub>y</sub>Mn<sub>z</sub>O<sub>2</sub> (NCM) cathode

High nickel battery has become the development direction of the lithium-ion battery industry. The battery’s energy density depends to a greater extent on the cathode material. The high nickel ternary material NCM has the advantage of high energy density, which is in line with the development of the electric vehicle industry. At present, high-Ni materials have the most obvious energy density and more mature technology. Here we discuss high-Ni NCM (x = 0.8, NCM811), as well as its Zr modified materials.

As the content of nickel increases, the energy density of the cell becomes greater. However, NCM811 has the following defects, (1) the first-coulomb efficiency is less than 90%, (2) bad thermal stability, (3) Nickel-rich materials react easily with water and carbon dioxide in the air to form Li<sub>2</sub>CO<sub>3</sub> and LiOH, which largely causes severe capacity degradation and structural instability. In addition, Ni<sup>4+</sup> and Co<sup>4+</sup>, which are easily oxidized, are formed during the charging process and react with the electrolyte, deteriorating the rate performance and cycle. (4) NCM811 is a secondary spherical particle formed by the agglomeration of primary particles, which limits the energy density of NCM811 because the secondary particles are easy to break under higher pressure solids.

Shuang Gao et al. doped NCM by using zirconium to achieve higher cycle durability. They synthesize pristine and Zr-modified LiNi<sub>0.8</sub>Co<sub>0.1</sub>Mn<sub>0.1</sub>O<sub>2</sub> (ZrNCM811) by the sol-gel method. There is no difference in particle shape of ZrNCM811 compared to NCM811 (Fig.12). The 1% Zr doping performs the best among the various ratios and improves the circulation efficiency to some extent.

![Fig.12 (a) SEM images of NCM811s with 1% Zr concentrations; (b)Rate performance of NCM811s with different Zr concentrations][29]

Kang Wu et al. introducing non-electrochemical activity Zr with high valence and strong binding energy of Zr-O into the Li layers. Zr-O stabilizes the structure, and Zr<sup>4+</sup> is used to replace part of the Li<sup>+</sup>, which improves the thermal stability of the material and thus the electrochemical properties. Likewise, they performed different ratios of Zr doping. The Li<sub>1−x</sub>Zr<sub>x</sub>Ni<sub>0.8</sub>Co<sub>0.1</sub>Mn<sub>0.1</sub>O<sub>2</sub> (x = 0, 0.01, 0.03, 0.05) cathode materials were obtained by a coprecipitation and solid-state sintering method. They obtained a similar conclusion to Shuang Gao et al. that 1% Zr doping exhibited superior electrochemical performance in cycle life (Fig.13).
Doping Zr can increase the Li layer space, which widens the channel for Li$^+$ during charging and discharging, which is related to the excellent rate of performance of the modified material. The Zr doping also restrains the side reactions between the electrode and electrolyte and improves the material's thermal stability, so the ZrNCM811 electrode has excellent electrochemical performance.

3.3 lithium iron aluminium nickelate (NFA)
To achieve Co-free cathodes, the high nickel-containing LiNiO$_2$ material is also a good choice. However, this kind of material performs instable in electrochemistry. Muralidharan et al. provided a novel class of nickel-rich, cobalt-free Li-ion battery cathode material with general formula LiNi$_x$Fe$_y$Al$_z$O$_2$ ($x \geq 0.8$, $x + y + z = 1$), termed as the NFA class of cathodes. This kind of Co-free material not only remains a high capacity but also enhances structural stability and safety ability. NFA cathodes have as viable candidates in next-generation cobalt-free lithium-ion batteries. In addition to nickel, iron and aluminium are also incorporated into the crystal lattice in this battery. By a sol-gel method, three kinds of nickel-rich cathode material NFA. NFA 811, NFA F, NFA A. The difference lies in the different proportions of various elements (Table.1).

### Table.1 The determined elemental compositions of NFA 811, NFA F, NFA Am [31]

| Element   | NFA 811 At% | NFA F At% | NFA A At% |
|-----------|-------------|-----------|-----------|
| Oxygen    | 68.61       | 67.21     | 70.99     |
| Nickel    | 19.47       | 19.77     | 20.09     |
| Iron      | 3.87        | 10.78     | 2.47      |
| Aluminium | 3.68        | 2.24      | 5.85      |
| Carbon    | 4.37        | 0.00      | 0.00      |

Although cobalt-containing NCA and NCM cathode materials are already in the market, the price of cobalt has nearly tripled in the past few years, which means that the use of cobalt in batteries must be reduced if cheap lithium-ion batteries are to be produced. Cobalt plays a significant role in enhancing battery performance. To achieve the same performance of cobalt-free materials as cobalt-containing materials, the ratio of various elements must be adjusted to achieve better performance. This NFA cathode reduces the cost and increases the battery's capacity so that the replacement of NCA and NCM electrodes is achieved. Due to the high nickel content and the variation of the rest of the composition, the three materials have different shades of green and appear granular under SEM (Fig.14).
The X-value affects the electrochemical properties of the NFA cathode. We can find that the NFA A cathode with higher aluminium content performs better (Fig. 15).

NFA material is a more cost-effective cathode material, and the data also indicates that NFA A has more potential for the future development of cobalt-free lithium-ion batteries.

NFA electrodes show good electrochemical performance, yield a specific capacity of about 200 mAh g\(^{-1}\) and have high cycle efficiency. Although NFA is still at an early research stage, its excellent electrochemical performance and complete cobalt-free status give it great potential to become the next generation of cobalt-free lithium batteries.

Fig. 15 Capacity retention of the NFA variants compared with that of NCA material synthesized using the same methodology [31]

4. Conclusions
In general, different types of attempts have been made to solve the problem of significant changes in the volume of silicon caused by repeated charge-discharge cycles, such as the design of nanoscale silicon with different structures (nanowires and porous particles), the development of new binders and the combination of carbon materials to form matching materials. These processes are generally relatively simple. Those materials have good cyclic performance, but most of the anode materials development based on pure silicon remains confined to the laboratory and cannot easily be scaled up. Silicon/carbon matrix composites seem to be more promising in practical applications because they combine the advantages of silicon (high capacity) and graphite (high-capacity retention, high conductivity, and low volume variation) and have the advantages of more straightforward preparation methods, lower cost,
and ease of scaling. Cost-effective raw materials and scalable manufacturing routes are essential for producing materials with low-cost production and high commercial margins.

For the cathode, LiCoO$_2$, as the earliest commercial application of lithium-ion battery cathode material, has been in-depth studied for a long time. Its process has been relatively mature, especially because it is widely used in small electronic devices. However, with technological advances and emerging markets like electric vehicles growing, we urgently need to study better alternative materials from safety, capacity, and cost. Thus, the concept "rich nickel and cobalt free" is proposed and endorsed. At present, the main research directions in the world are the new cobalt-free synthesis method and LiCoO$_2$ modification by P, B, Ni, and so on. From a tech point of view, these ways will help extend battery life and increase energy density. From the perspective of life, this battery will be cheaper, less toxic, and more ethical than ever. In summary, although this stage is still being explored, it has great development potential. At the same time, its market and practical value cannot be ignored.

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