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Silicon: a wonderful anode material for Li-ion batteries

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Abstract— Lithium-ion battery is widely used energy storage equipment. It is well known that due to low theoretical capacity (372 $mAhg^{-1}$) commercial graphite anode cannot meet the challenges of energy storage. Among advanced materials, Silicon, because of its high specific capacity (4200mA/g) and low discharge voltage (0.4VLi/Li^+) is one of the most promising anode materials for next-generation lithium-ion batteries. However, Si anodes face pulverization issue due to large volume expansion during battery operation. In this review the core issues of the silicon anode and effective strategies to overcome from these issues are discussed.

Keywords— Li-ion battery, Silicon, Anode Material, Energy Storage, Secondary Battery

I. INTRODUCTION TO LITHIUM-ION BATTERIES

Energy demand and supply are crucial factors for the civilization. Storage devices are required to store electricity which is produced from different renewable sources like solar and wind. To store electricity, electrochemical energy storage devices like batteries play significant role. There are two types of batteries on the basis of their operation and characteristics i.e. primary and secondary batteries. In primary batteries the electrochemical reaction is irreversible, and these are designed for one time use such as Leclanché, alkaline MnO_2 and silver oxide. On the other hand, the secondary batteries are reversible and thus more attractive for long term use. Secondary batteries can be charged/ discharged for a number of cycles and known as rechargeable batteries. Among all rechargeable batteries, Li-ion batteries are found best due to highest specific energy, cell voltage, good capacity retention, negligibly small self-discharge and lower maintenance. A lithium-ion battery (LIB) has electrical cells which are capable to store energy in the chemical form and produces electricity by electrochemical reaction.

Li-ion batteries have three components i.e. positive electrode, negative electrode and an electrolyte/separator layer in between them. The commercialized battery system consists of lithium-cobalt oxide ($LiCoO_2$) as positive electrode, carbon (graphite) as negative electrode whereas the electrolyte varies. The liquid electrolyte of a battery consists of acids, soluble salts or other bases in liquid formats. In LIB electrolyte provide path to lithium ions to travel between the electrodes and through a separator anode and the cathode keep away from making direct contact [1-3]. Working of a LIB with graphite as anode is represented as follows:

Discharging process of Li-ion batteries (spontaneous)

At Anode: $LixC_6 \rightarrow Graphite + xLi^+ + xe^-$

At Cathode: $CoO_2 + LixC_6 \rightarrow Li_xCoO_2$ ($Co^{4+} \rightarrow Co^{3+}$)

Charging process of Li-ion batteries (non-spontaneous)

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At Anode: Graphite + $xLi^+ + xe^- \rightarrow Li_xC_6$

At Cathode: $Li_xCoO_2 \rightarrow CoO_2 + x\ Li^+ + x\ e^-$ ($Co^{3+} \rightarrow Co^{4+}$)

During charging process, Li^+ moves from Li_xCoO_2 to carbon through the electrolyte which resulted into the oxidation of Co^{3+} to Co^{4+} and in discharging process Li^+ moves from carbon to Li_xCoO_2. During charging and discharging, electrolyte act as a medium for the transfer of ions between the two electrodes.

However, the graphite anode is not capable to meet increasing demand due to its moderate gravimetric capacity of 372 $mAhg^{-1}$ [4, 5] and safety issue due to the lithium dendrites formation. As a consequence, new generation anode material is needed, which have large charge discharge potential, safe and low cost maintenance. Silicon (Si) is one of the best anode materials to replace graphite.

II. Si as anode material

Among all the potential lithium-ion batteries, Si is recognized as promising anode candidate by which we can replace the graphite. In comparison of graphite, silicon has high specific capacity 4200 mAh/g [6], which is much higher than the theoretical capacity of graphite (372 mAh/g) and no lithium dendrites develop during battery operation. Silicon is environment friendly, low cost and large reserves in the earth. Silicon has moderate electrochemical working potential approx. 0.4 V vs Li^+/Li [7,8]. Silicon also has high energy density and nontoxicity. However, during lithiation and delithiation Si undergo enormous volumetric changes to accommodate large amount of Li [9-11]. The volume of silicon expands 400% and conformed and recorded different Li-Si phase with increasing lithium content [12-14]. The consequence of large change in the volume of Si anodes internal resistance increases. This results to the loss of contact area between Si and conductive materials, hence rapid capacity fading occurs. In battery operation most of the material loses contact with the current collector, resulting in poor transport of electrons. Due to large volume expansion, morphology transformation occurred throughout the battery life cycle and results in cracking of Si particles, formation of an immense surface stress and as a result, adhesion problems [15,16]. It is also observed that silicon is not the only high-capacity material which suffers from large volume expansion. Other high-capacity material like tin (Sn), germanium (Ge), and antimony (Sb) also suffer from large volume expansion as silicon [17-21].

Table 1 shows comparison of Si with other similar metals.

TABLE I
THE COMPARISON OF DIFFERENT ELEMENTS IN TERMS OF THEIR GRAVIMETRIC / VOLUMETRIC CAPACITY, TOXICITY & SAFETY

Element	Gravimetric capacity (mAh/g)	Volumetric Capacity (mAh/cm^3)	Toxicity	Safety
Si	4,200	2,400	No	High
C	372	890	No	Low
Ge	1,568	2,300	High	High
Sn	990	990	No	High
P	2,600	2,250	High	Low
Sb	660	1,890	High	Low
Pb	549	1,790	High	Low

As silicon is a brittle material, it is difficult to recover to its original state. Continuous cracking of Si particles during cycling leads fresh Si surface for further reaction with electrolyte. As a result, continuous formation/repair of the passive layer known as the solid electrolyte interphase is developed [22-23]. In turn, the columbic efficiency of the silicon-based electrodes is typically lowered in comparison of graphite electrodes. Continuous formation of SEI film leads to the continuous consumption of electrolyte hence result into the increase of interface impedance, which leads increased diffusion distance for lithium-ion and hinders the transfer of lithium-ion [24]. Also, the inherent low electrical conductivity ($\sim 10^{-5}$ Scm^{-1}) and sluggish ion diffusion kinetics ($\sim 10^{-14}$ cm^2s^{-1}) exert much pressure on the realization of wide-scale usage of Si as anode in Lithium-ion Batteries.

III. Strategies to Improve Performance Silicon as Anode

A. Structure Design

Structure designing are effective way through which large volume expansion of Si anode accommodate during discharging/charging process. It includes different nanostructures of Si as nanoparticles [25], nanolayers [26], nanowires [27], nanotubes [28] and morphological structures of Si as core–shell [29], yolk–shell [30], and porous [31]. It also includes atomic-scale structures design of Si doping [32], lattice deformation [33], and heterostructure [34]. Nanomaterials having genuine potential showed influence on the electrochemical

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performance of Si anode [35] due to reduced dimensions and provide higher intercalation/deintercalation rates. The impact of nano-sized Si on performance of battery was showed by several groups. Li et al. measured that a nano-Si (78 nm) powder anode showed better capacity retention than bulk Si powder [36]. Cui et al. synthesized Si nanowires and grown directly on the metallic current collector substrate [37]. The limited nanowire diameter enables better accommodation of the large volume expansion and provides 1D electronic pathway that allows efficient charge transportation. Hollow and yolk–shell structures of Si composites which are coated with conductive materials, have shown to be an efficient way to stabilized SEI related problems [38-39]. These nanosized materials and differently designed structures accommodate large volume expansion of silicon, increase electrical conductivity, and provide pathway for ion transportation. Figure 1 shows the galavanostatic charge discharge curves of differently prepared Si nanostructures. The sample prepared by metal-assisted chemical etching (MACE) showed the best performance. This was suggested to be due to increase in pore size, those werehelful for easy lithiation.

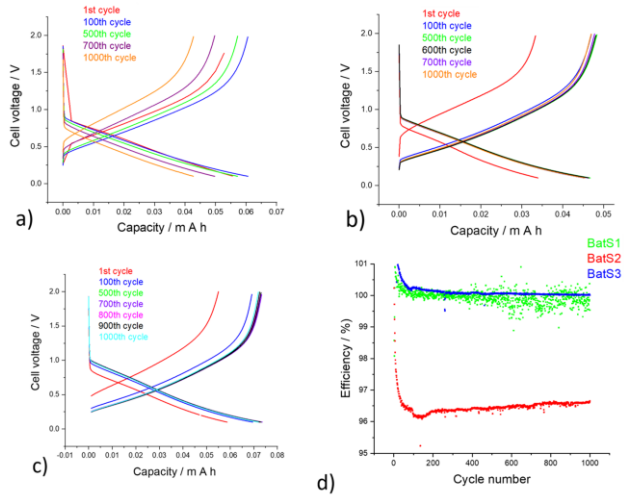

Figure 1: Galvanostatic charge-discharge curves of 3 different Cells of Si nanostructures as anode.

B. Binder's effectiveness

The active material, conductive additive and current collector are influenced by binders. Binder plays a pivotal role in the whole battery, especially in Si-based anodes [40]. A binder must have wide electrochemical window, chemically stable as well as electrochemically stability. In LIBs, the role of binder is an influencing factor [41]. In LIB, stability and irreversible capacity losses, are critically dependent on the binder's properties [42-43]. The traditional binder PVDF cannot accommodate the volume expansion of the silicon material because of its weak Vander Waals forces [44]. Various new polymer binders are reported now which includes binding actions of volume change alleviation [45], dispersing function [46], SEI contribution [47], electronic conduction [48] and ionic conduction [49]. It has been demonstrated that polymers which contain carboxy groups, such as polyacrylic acid (PAA) and carboxymethyl cellulose (CMC) are promising binders for Si-based anodes [50-51]. During the battery operation the polar hydrogen bonds between the carboxy groups of the binder and the SiO_2 on the Si surface exhibit a self-healing effect and reform if locally broken.

C. Electrolyte Additives

Electrolyte additives have been widely used for extending the cycle life of LIBs while preventing electrolyte decomposition at the electrodes. During the initial lithiation process, electrolyte additive plays a significant role in passivating the Si/electrolyte interphase. To stabilize SEI layer on the Si surface and improve the cycle life of Si, electrodes additives/cosolvents like dimethylacrylamide [52], succinic anhydride [53], citric acid [54], lithium difluorooxalatoborate (LiFOB) [55], and lithium bis(oxalate) borate (LiBOB) [56] have been investigated for Si-based materials. Carbonate electrolytes with fluoroethylene carbonate (FEC) and/or vinylene carbonate as additives shows enhanced cycle performance in Si-based anodes [57]. Recently next-generation synthetic additive approaches were investigated that allows the formation of highly stable electrode-electrolyte interface architecture from fluorinated and silylated electrolyte additives [58].

D. Prelithiation

In LIB, an irreversible electrochemical reduction of electrolytes occurs during the forming of SEI on anodes in the initial cycle. This lead to consumption of the Li-ions originally loaded in the cathode prior to cycling and coulombic efficiency (ideally 100%). The lower Li-ion concentration results in decrease of the fractional utilization of active materials. It causes net capacity losses in the following cycle's results and significantly limiting the available energy density. There are three methods to store lithium in anodes (chemical prelithiation, electrochemical prelithiation, and mechanical prelithiation). In mechanical prelithiation method microscale stabilized lithium metal powder in Si anodes is directly used through

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mechanical activation [59]. In second approach, chemical prelithiation is applied to LixSi nanoparticles from chemically synthesis as potential prelithiation reagent [60]. In third approach, electrochemical prelithiation is done by lithium foil with electrolyte-wetted anodes [61].

E. Areal capacity

For practical applications, high areal capacity is a basic requirement to achieve reduced weight percentage of inactive material that results high energy density [62]. During lithiation/delithiation, large volume changes of Si anode makes it challenging to acquire stable cycling at high areal capacity. In lithiation/delithiation, Si particles expand/contract that results in the electrode peeling off, especially in thick electrode. To achieve enhanced cyclic stability under high areal mass loading there are few methods that includes utilization of polymer binders, free-standing electrode design, fabrication of secondary microscale structure, porous materials with high tap density and microsized materials with inactive buffer [63].

F. Initial Coulombic Efficiency

In LIB, poor ICE due to increased surface area is a challenging issue [64]. To achieve enhanced ICE strategies that can be applied are coating [65], structural design [66], prelithiation [67], carbonization of binder [68]. With effective structure design, the formation of SEI in the initial lithiation reaction of Si-based anodes can be optimized to improve the ICE. In one of the surface-engineering strategy, dense silicon skin is deposited onto each mesoporous silicon microparticle to significantly improve both the ICE and Coulombic efficiency [65]. Here mesoporous Si microparticles has been coated by shell-like Si and encapsulated with graphene cage. Such design is capable to prevent the electrolyte from diffusing into the interior and thus restricts SEI formation to the outer surface by coated Si and results the improvement in ICE from 37.6% to 87.5% before and after coating Si.

IV. CONCLUSIONS

In summary Si-based anode suffer from large volume expansion that occurs during lithiation. Due to large volume expansion, the silicon nanoparticles suffer from pulverization and create poor conductivity results electrode unstable SEI films. In this Review, the effectiveness of structure, binders, electrolyte systems, additives, prelithiation, ICE and areal capacity have been discussed. After reviewing the literature, we reached a better understanding of approaches to these problems.

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