

SiO₂ Based Composites For Lithium Ion Battery Anodes: A Review

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Abstract

The application of SiO₂ as a lithium-ion battery anode material has attracted attention due to its higher theoretical capacity compared to commercial carbon, low discharge potential, and abundance in nature. However, SiO₂ has limitations that restrict its widespread use as an anode due to its low electrical conductivity and volume expansion during cycling. Modifying the properties of SiO₂ using other materials as composites is an effective strategy to overcome these limitations. In this review, the progress and role of SiO₂-based composites in improving electrochemical performance will be discussed. First, we briefly discuss the development and sources of SiO₂ material as a lithium-ion battery anode. In addition, the mechanism of lithium storage and the challenges faced in the application of SiO₂ anodes are discussed by reviewing solutions to overcome them, including modifications to the nano and porous structure of SiO₂. Furthermore, the review focuses on the application and development of SiO₂-based composites in improving the electrochemical performance of lithium-ion batteries to provide an overview of the challenges and prospects for the development of SiO₂ anodes in lithium-ion batteries.

Keywords: Lithium-Ion battery, SiO₂ materials, SiO₂ anode, SiO₂/C composite, SiO₂/metal composite

Introduction

As time goes by, the use of lithium-ion batteries in electronic devices and electric vehicles is becoming increasingly necessary [1]. **Figure 1** illustrates the current upward trend in cumulative demand for lithium-ion batteries. Lithium-based batteries have the advantage of being able to produce large amounts of energy, high efficiency, and a relatively long life cycle. Compared to sodium and zinc ion batteries, lithium ion batteries produce more energy density (150 - 350 W/kg) which allows for longer operating times or smaller battery packs for the same capacity for applications in electric vehicles and portables although safety and cost

issues in lithium ion battery production are still being developed [2]. The performance of lithium-ion batteries is largely determined by the characteristics of their electrodes [3]. The energy capacity stored in a lithium battery system depends on the number of ions that can be stored in the electrode material structure during the charging and discharging processes. Graphite is commonly used as the anode material in commercial lithium-ion batteries because it has a long life cycle and low cost. However, graphite has a specific capacity of 372 mAh/g, which makes it unable to meet the requirements for high-energy-density battery anode materials [4].

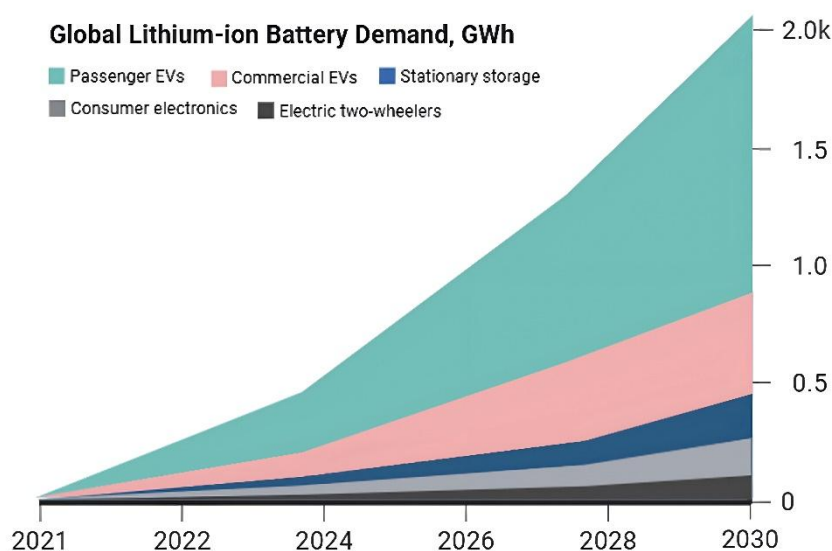


Figure 1 The growing demand for lithium-ion batteries [5].

Si material is a candidate for lithium-ion battery anode alternatives due to its low lithium potential and high theoretical specific capacity (3,587 mAh/g vs $\text{Li}_{15}\text{Si}_4$) [6]. However, large volume expansion (>300%), mechanical pulverization, the presence of an unstable solid electrolyte interface (SEI) layer during the charge-discharge process, and electrode peeling from the current collector hinder its practical application. Recently, silica (SiO_2) has been proposed as an alternative material for lithium-ion battery anodes due to its high theoretical capacity (1,965 mAh/g), lower volume expansion (100%) compared to Si, and low discharge potential (0.2 V vs. Li^+/Li) [7]. SiO_2 is also one of the most abundant materials in nature, inexpensive, and has low toxicity. In addition, this material can be synthesized more easily than Si. To date, various silica materials have also shown good electrochemical performance as anodes for lithium-ion batteries. However, poor electrical conductivity due to the strong Si-O bonds in SiO_2 and low initial coulombic efficiency (ICE ~40%) pose challenges for the application of SiO_2 as a battery anode [8]. Currently, various strategies are being employed by researchers to improve the electrochemical performance of SiO_2 anodes, focusing on structural modification, particle and pore size control, SiO_2 composition optimization, and improvements in synthesis methods to achieve higher performance and good cycling stability. SiO_2 -based composite materials are widely investigated as an

effective approach to enhancing the electrochemical performance of SiO_2 anodes. The incorporation of carbon materials to form composites not only improves the electrical conductivity of SiO_2 anodes but also serves as a buffer to accommodate volume expansion during the charge/discharge process. In addition, the incorporation of metallic materials to enhance mechanical integrity and cycle life has been applied in several studies. Efforts to improve Li-ion transport and to form a stable solid electrolyte interphase (SEI) have also been pursued through the fabrication of SiO_2 composites with 2-dimensional materials such as MXene [9].

This review focuses on analyzing the role of various SiO_2 -based composites in improving the electrochemical performance of SiO_2 anode materials. SiO_2 composites will be grouped into SiO_2/C composites and $\text{SiO}_2/\text{metal}$ composites. This review also explains the mechanism of lithium ion storage and the challenges faced in the application of SiO_2 materials as lithium-ion battery anodes.

Silica material

Silicon dioxide (SiO_2) is one source of Si, which is the second most abundant element in the Earth's crust after oxygen, as shown in **Figure 2(a)**. It is naturally found in quartz sand, various types of rock, sand, and soil [10]. SiO_2 is a quasi-metallic oxide that exists in the form of a silicate polymer with interconnected

tetrahedral SiO_4 units. In its general structure, Si atoms are bonded to 4 oxygen atoms, and each oxygen atom is bonded to 2 Si atoms, so that the silica surface consists of silanol and siloxane groups. SiO_2 exists in several

forms, each with its own structural, chemical, and physical characteristics. Silica can be found in amorphous forms, as shown in **Figure 2(b)**, gels, and crystals [11,12].

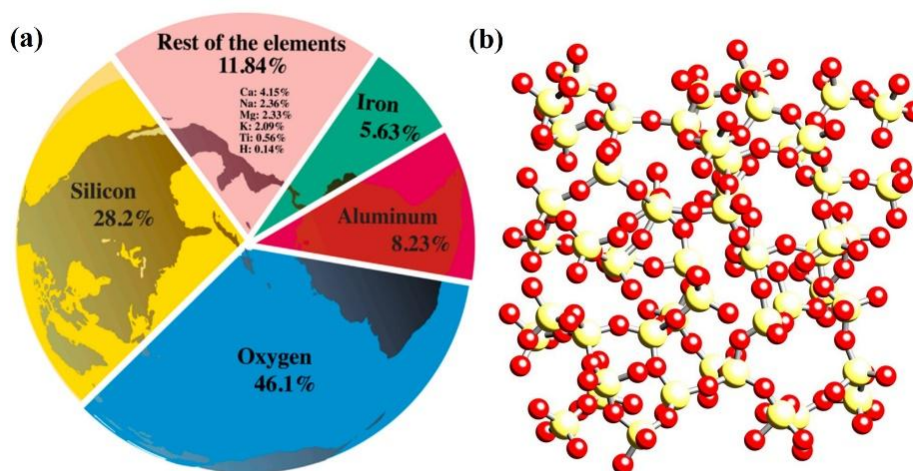


Figure 2 (a) Abundance of various materials in the Earth's crust [10] and Chemical structure of amorphous silica [13].

The chemical structure and physical properties of silica make it applicable in various industries. Some of the physical properties of silica include a high melting point of around $1,700\text{ }^{\circ}\text{C}$, non-conductivity and diamagnetism, and maximum density at high temperatures [14]. Silica is widely used in biomedicine [15], *oil water separation* [16], *water pollutants adsorption* [17], catalyst [18] and sensor [19].

Several studies have been conducted, such as modification of nano structure and porosity [20], Composition, particle size, use of amorphous phase, and synthesis methods of SiO_2 to improve the electrochemical properties of SiO_2 . Based on its superior electrochemical properties, thermal stability, and mechanical structure, silica can provide high theoretical capacity as a battery anode. Researchers have significantly used silica material in its application as a battery anode due to its high lithium storage capacity.

Silicon-based materials such as pure silicon (Si), silicon monoxide (SiO), and silicon dioxide (SiO_2) are promising candidates for lithium-ion battery anodes [21]. However, Si anodes have a high volume expansion ($>300\%$) during the alloying/dealloying process and can even peel off from the current collector [22]. In

recent years, silica has become increasingly popular as an alternative to Si anodes due to its excellent properties, such as high lithium storage capacity ($1,950\text{ mAh/g}$), lower volume expansion (100%), low release potential ($0.2\text{ V vs Li}^+/\text{Li}$), abundant availability, and low cost as an anode material for lithium-ion batteries [23]. Gao *et al.* [24] stated that commercial SiO_2 with a diameter of 7 nm can react with Li in the range of 0.0 to 1.0 V (vs. Li^+/Li) and produce a reversible capacity of 400 mAh/g . However, silica has low initial coulombic efficiency and poor electrical conductivity, which limits its application as a battery anode. Several studies have modified the properties of silica to overcome these weaknesses, such as SiO_2 nanomaterials and SiO_2 -based composites. Wang *et al.* [25] synthesized mesoporous silica nanoparticles using a sol-gel/emulsion approach and produced a stable specific capacity of $1,060\text{ mAh/g}$ at a current density of 100 mA/g after 90 cycles [25].

Sources of SiO_2 materials

SiO_2 for lithium-ion battery anodes can be obtained from biomass, quartz, chemical precursors, and industrial waste. The selection of precursors is important in determining the structure and performance of SiO_2

anodes [26]. The effects of precursors used on morphology, porosity, and the formation of active silicon species during synthesis will impact battery performance. A large surface area will provide ample storage space for lithium ions, potentially increasing anode capacity. High porosity can improve ion transport for good rate capability [27]. Therefore, optimizing the selection of precursors in material synthesis can significantly improve battery performance.

One of the most widely used methods in silica synthesis is the sol-gel method using Tetraethyl Orthosilicate (TEOS) as a precursor. TEOS is one of the chemical precursors used in the synthesis of SiO₂ nanoparticles. The use of TEOS as a silica precursor offers advantages such as the ability to produce nanoparticle structures that can contribute to increased anode storage capacity. In addition, silica produced from TEOS exhibits good thermal stability [28,29]. For example Cao *et al.* [30] synthesized amorphous SiO₂/C composites using TEOS precursors and resin, while Tu *et al.* [31] also synthesized SiO₂ nanospheres using the sol-gel method and TEOS precursors, producing good electrochemical performance.

Biomass is one source of silica for battery anodes due to its abundance and availability. For example, Askaruly *et al.* [4] synthesized SiO₂/C composites from rice husks. Rice husks are a type of agricultural waste from rice production that is not fully utilized, while annual rice husk production reaches 100 million tons [32]. Most of the rice husk content, such as cellulose,

lignin, and inorganic silicon compounds, allows this material to be used as a source of carbon and silica for anode materials [33]. Rice husks through processes like hydrothermal carbonization, chemical activation, or magnesiothermic reduction to create porous carbon fibers, activated carbon or silicon nanoparticles that have high capacity and good cycle stability. In addition to rice husks, Su *et al.* [34] used corn leaves to synthesize amorphous Si/SiO_x as a lithium-ion battery anode, and Xu *et al.* [35] synthesized SiO₂/C nanocomposites from bamboo leaves and produced good anode performance. Utilizing biomass as a source of lithium-ion battery anode material offers a sustainable and cost-effective alternative to commercial materials, making it an attractive option for large-scale applications [36].

Recently, by-products or waste from industry have been widely used as a source of SiO₂. Zhang *et al.* [37] synthesized SiO₂/graphite composites from sewage sludge, Jumari *et al.* [38] synthesized SiO₂/C composites from fly ash combustion, Prasath *et al.* [39] used laboratory glassware waste for the synthesis of nanostructured SiO₂, and Widiyandari *et al.* [40] used geothermal waste as a source of silica in the synthesis of SiO₂/Mg. The use of industrial waste as a source of silica can overcome environmental problems and has the potential to reduce the cost of lithium-ion battery production. Various sources of SiO₂ for lithium-ion battery anode applications are shown in **Table 1**.

Table 1 Source of SiO₂ for lithium-ion battery anodes.

Source	Product	Method	Initial Discharge Capacity (mAh/g)	Initial CE (%)	Reversible capacity (mAh/g)	Cycle	Ref.	
Biomass	Rice Husk	SiO ₂ /C	Carbonization	922	45	450	50	[4]
	Corn Leaves	Si/SiO _x	Aluminothermic reduction	2,100	-	2,100	300	[34]
	Bamboo leaves	SiO ₂ /C	Thermal Decomposition	586.2	-	294.7	190	[35]
	Diatom microalgae	SiO ₂ /C	Thermal Decomposition	~465 - 700	~40	661	100	[41]
Quartz		SiO ₂	Ball milling	~928	37	~800	200	[23]
Precursor	TEOS	SiO ₂ /C	Pyrolysis	3,288	38	1,024	100	[30]
Industrial Waste	Sewage Sludge	SiO ₂ /graphite	Ball milling	711	66	433	100	[37]
	Geothermal Sludge	SiO ₂ /Mg	Metallothermal	761.16	-	-	-	[40]

Source	Product	Method	Initial Discharge Capacity (mAh/g)	Initial CE (%)	Reversible capacity (mAh/g)	Cycle	Ref.
Glass Waste	SiO ₂	-	270	53	144	50	[39]
fly ash combustion	SiO ₂ /C	Sol-gel, Ball milling	586	76	586	20	[38]

SiO₂-based anodes and their importance in LIBs

Over the past few years, numerous studies have explored SiO₂-based materials as alternative anodes for lithium-ion batteries. SiO₂-based materials are considered alternative anodes due to their abundance in nature, low release potential, lower volume expansion compared to silicon anodes, and environmental friendliness. Therefore, the lithium storage mechanism, factors affecting electrochemical performance, and challenges associated with SiO₂ anodes will be discussed in this section.

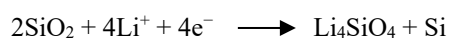
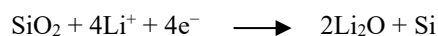
The storage mechanism of lithium

The electrochemical mechanism of silica has been widely discussed due to its potential application as a lithium-ion battery anode material. SiO₂ anodes have a high lithium storage capacity of around 1,950 mAh/g. However, SiO₂ anodes have low initial coulombic efficiency (ICE) due to volume expansion during charge-discharge cycles [23]. Therefore, it is necessary to re-examine the lithiation mechanism that occurs in the SiO₂ anode. Initially, researchers believed that SiO₂ was an inert phase during the lithiation process because the strong Si-O bond could not be broken by Li insertion, thus maintaining a stable structure [42]. However, recent research claims that SiO₂ can be reduced by Li thermodynamically [43,44].

Several studies have been conducted to determine the mechanism of lithiation on SiO₂ anodes. Zhang *et al.* [45] observed the lithiation process of SiO₂-coated SiC nanowires using Transmission Electron Microscopy (TEM) and theoretical studies. This research also systematically investigated electronic conduction, ionic transport, structural evolution, and lithiation patterns of SiO₂. SAED patterns showed that crystalline Li₂O formed during initial lithiation. Further reduction reactions will produce a mixture of Li-Si-O with Li₄SiO₄ and Li₂O crystals inside. Ostadhossein *et al.* [46] used ReaxFF Reactive Force Field Modeling to analyze the structural properties during Li⁺ discharge on SiO₂.

Structural evolution analysis shows that the lithiation reaction on the SiO₂ anode consists of 2 steps: Partial reduction of silica with Li⁺ through the conversion of SiO₂ to Si and Li₂Si₂O₅, and reduction of Si reacting with Li⁺ through a reversible reaction. Similar research using ab initio molecular dynamics shows that the mechanism and structural evolution occur, where Si-O bonds break and Li-O bonds form [47].

Wang *et al.* [8] synthesized a graphene-wrapped SiO₂ nanotube network (SiO₂-NT/G network) and obtained good cycling performance. The first cycle of the cyclic curve of SiO₂-NT/G shows a cathodic peak at 0.6 V, indicating the formation of a solid electrolyte interface (SEI), and a peak below 0.3 V associated with the formation of Li₂O and Si and the formation of Li-Si alloys. Tu *et al.* [31] analyzed the lithium storage performance of SiO₂ nanospheres supported by XPS data. The XPS pattern in **Figure 3(a)** shows that the peak at around 102.3 eV remains relatively unchanged after charging to 2.5 V, indicating that the formation of Li₄SiO₄ is irreversible. Furthermore, **Figure 3(a)** also shows that Li is present in the form of Li₂O. This indicates that both Li₂O and Li₄SiO₄ are formed simultaneously in the electrochemical reaction between SiO₂ and Li ions. The CV curve shows an anode peak around 1.2 V that appears in every cycle. The Li-Si alloy/dealloy reaction only appears at potentials below 1.0 V, which indicates that it is most likely caused by the partial reversibility of Si to SiO₂. The electrochemical reactions that may occur during the lithiation process on the silica anode are described as follows:



During the lithiation and delithiation processes, the structure of the SiO_2 anode undergoes significant changes related to the electrochemical reaction between SiO_2 and Li ions. The reaction mechanism at the SiO_2 anode during the lithiation/delithiation process is closely related to battery capacity. Jiang *et al.* [48] describe in detail the relationship between the reaction mechanism that occurs at the SiO_2/C anode and the increase in battery capacity. **Figure 3(b)** shows the CV curve of the SiO_2/C anode. During the lithiation process, Li^+ diffuses into SiO_2 , causing the formation of $\text{Li}_2\text{Si}_2\text{O}_5$, Li_4SiO_4 , Li_2O , and Li_xSi . The products resulting from the reaction between Li^+ and SiO_2 are divided into 2 categories, namely Li-poor phase and Li-rich phase. In

the CV curve, there is a reduction peak around 0.82 V after 2 scan cycles, indicating a reaction that produces Li silicate and proceeds slowly. Cycling performance shows 2 stages of the process, namely a decrease in capacity at the beginning of the cycle and a stage of capacity increase, as shown in **Figure 3(c)**. The decrease in battery capacity at the beginning occurs due to the contribution of SEI formation and the reaction between Li^+ and SiO_2 , which produces Li silicate. The conversion between the Li-poor phase and the Li-rich phase, accompanied by the formation of Si, will result in an increase in capacity because Si contributes to the reversible capacity of the battery.

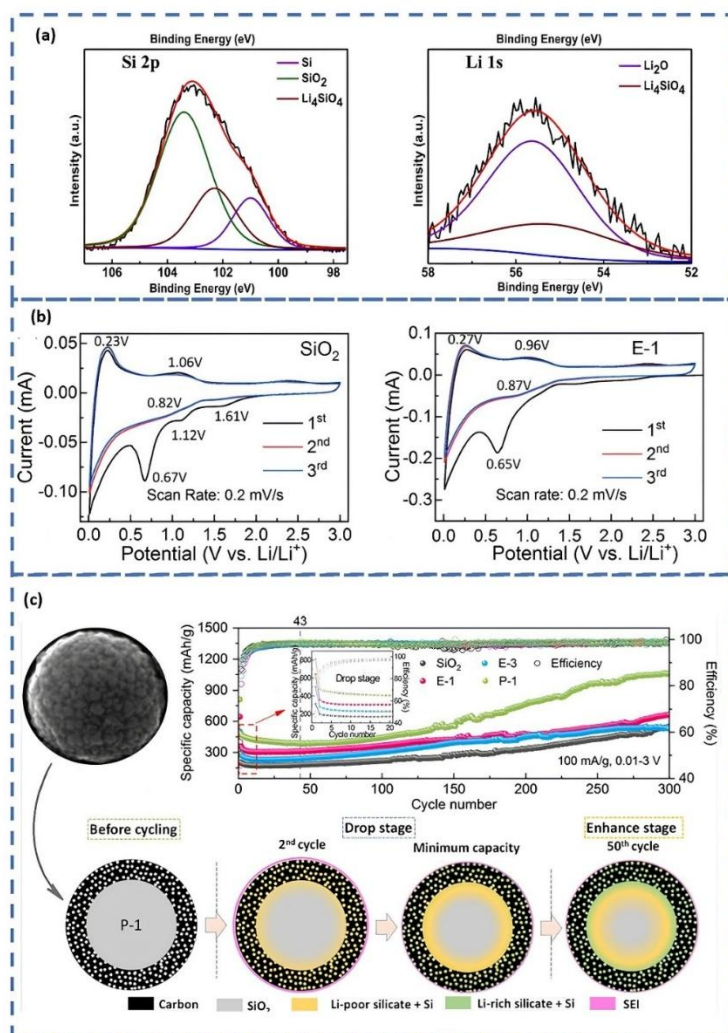


Figure 3 (a) XPS spectra of Si 2p and Li 1s SiO_2 anodes after charging to 2.5 V [31], (b) Cyclic voltammogram of SiO_2 nanospheres in the first 3 cycles, (c) Cycling performance and schematic diagram of the reaction process between Li^+ and SiO_2 in different cycling states [48].

Challenges in SiO₂-based anode

Over the past few years, silica has been extensively researched as an alternative candidate for lithium-ion battery anodes because it has a high theoretical capacity (1,965 mAh/g) and lower volume expansion compared to silicon anodes. SiO₂ also has a scale and crystallinity that affect its electrochemical activity [1]. In addition, SiO₂ is one of the most abundant materials in nature and is more cost-effective than metal-based materials [29]. Gao *et al.* [24] reported that commercial SiO₂ nanoparticles can react with Li to produce a reversible capacity of 400 mAh/g.

However, one of the drawbacks of silica that poses a challenge in its application as an anode material is its poor electrical conductivity and low initial coulombic efficiency due to volume expansion that has the potential to damage the SiO₂ structure during long cycles [49] as depicted in **Figure 4**. During the chemical reaction process, SiO₂ will be converted into Si and irreversible phases Li₄SiO₄ and Li₂O will form during the initial discharge process, causing large amounts of lithium consumption and resulting in low ICE values. ICE measures the ratio of discharge capacity to charge capacity in a battery's first cycle, and it directly ties to irreversible capacity loss that reduces the effective

battery capacity. Lower ICE means more lithium is consumed in side reactions like SEI formation on the anode, leaving less active lithium available for reversible capacity in full cells. In contrast, a high ICE value keeps more lithium available for charge-discharge cycles, thereby maximizing the battery capacity and energy density [50,51]. After a long cycling process, the silica anode undergoes damage and particle destruction between the active material and the current collector, causing slow diffusion of lithium ions into the active material [52]. The formation of the solid electrolyte interphase (SEI) and electrode destruction continuously consume electrolyte and lithium ions, causing a decrease in energy density. Xia *et al.* [53] synthesized SiO₂/C composites as battery anodes and showed a low ICE value of 59%. Research conducted by Wu *et al.* [54] in synthesizing Silica@Zn nanoparticles also produced a low ICE value of 45%. This shows that the low ICE value in SiO₂ anodes remains a challenge that has not been adequately addressed. Various strategies have been employed to overcome this problem, such as pre-lithiation, a method of introducing excess lithium ions into the active material before cycling so that it can refill the irreversible lithium vacancies during cycling [55].

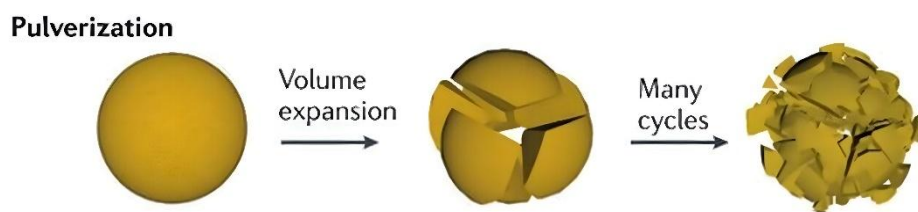


Figure 4 Schematic diagram of the degradation mechanism in Si-based materials due to volume expansion [56].

The SiO₂ anode also has poor electrical conductivity ($>10^{-1} \text{ Sm}^{-1}$), which results in low electron transport at the electrode and disrupts battery performance [57]. The low electrical conductivity of the anode material affects the battery's performance rate because charge distribution occurs slowly [58]. Strong covalent bonds in SiO₂ form a lattice with localized electrons. In addition, the absence of delocalized electrons in its structure results in a lack of mobile ions to facilitate charge transfer [59].

Structural modification to improving SiO₂ Performance

Several approaches have been taken to address volume expansion, electrochemical properties, inconsistent kinetic reactions, and low conductivity in improving the performance of SiO₂-based anode materials for lithium-ion batteries. The structure of the anode material greatly affects the electrochemical performance of the battery. Modifying the structure of the material to a nano scale and making it porous is one strategy that is still being developed today.

Porous SiO₂ anode

The pore structure of SiO₂ anode material is one of the factors that can change its electrochemical performance. The pore structure in SiO₂ facilitates electrolyte diffusion and lithium ion transport in the anode material. Porous particles can reduce the diffusion path of lithium ions to improve electrochemical performance, such as rate performance and cycleability, due to their large surface area. In addition, the porous structure also acts as a buffer to prevent volume expansion of silica during the lithiation and delithiation of Li ions and provides ample storage space for Li⁺ to increase the contact area between the electrode and electrolyte, thereby improving the reversible capacity of the battery [60,61]. For example, Li *et al.* [62] synthesized SiO₂/C nanocomposites with dual porosity and produced uniform pore distribution, large pore volume, and high surface area. Evaluation of the composite as an anode for lithium-ion batteries showed a reversible capacity of 635.7 mAh/g at 100 mA/g after 200 cycles and good rate capability. The improvement in battery performance was partly due to the dual-porosity structure, which increased the contact area and facilitated Li⁺ diffusion at the interface between the electrolyte and the active material. Yan *et al.* [29] also synthesized hollow porous SiO₂ nanocube anodes that yielded a reversible capacity of 919 mAh/g after 30 cycles. The porous structure of the anode accelerates Li⁺ transport, contributing to the formation of Li₂O and Si. In addition, the porosity of the anode material also influences SEI formation. Controlled and stable

diffusion of Li ions within the anode can improve battery performance efficiency by facilitating the formation of a stable SEI layer during cycling [63]. The SEI layer is able to block interactions between electrons and the electrolyte, thereby preventing further decomposition reactions. Anode porosity significantly affects SEI growth, often leading to the formation of more complex and dynamic layers. Higher porosity can promote initial SEI reactions with the liquid electrolyte, resulting in composite structures with lower resistance. However, excessive porosity can also hinder this process by trapping the electrolyte and promoting uneven SEI growth [64].

Suh *et al.* [65] synthesized SiO₂/C composites using SBA-15, which has high porosity, as a template and pore-forming agent. The composite structure formation process in **Figure 5(a)** shows that PTFE as a pore-forming agent will disappear during the carbonization process. The composite has a large surface area, indicating that the pore structure in the composite has been successfully formed, which will be beneficial for Li ion diffusion. **Figure 5(b)** shows many micro-sized pores formed in the composite. The pore structure in the material can shorten the Li ion diffusion path, resulting in lower resistance. In addition, the composite also shows good cycling performance of 294 mAh/g after 300 cycles. Huang *et al.* [66] also successfully prepared a porous SiO₂/C composite with a honeycomb structure that showed a reversible capacity of 1,109 mAh/g during 100 cycles and good rate capability.

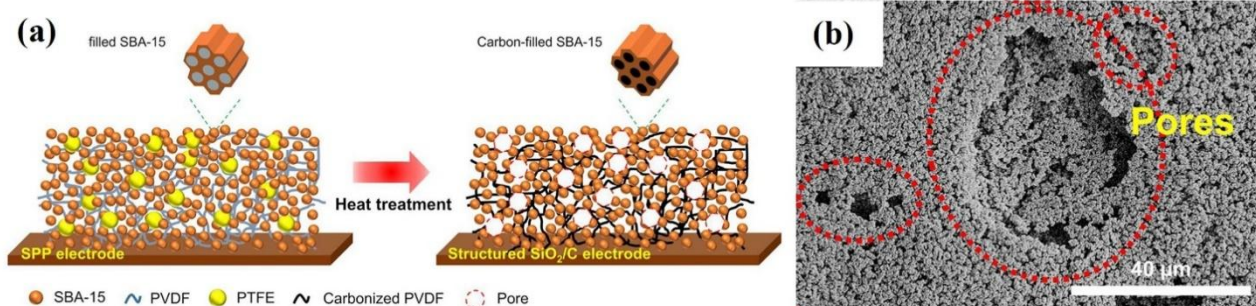


Figure 5 (a) Illustration of the formation of the SiO₂/C electrode structure, (b) SEM image of the SiO₂/C electrode (Reproduced with permission from Ref [65], Copyright 2022, Springer Nature Link).

Nanostructured SiO₂ anode

The electrochemical performance of SiO₂ anodes can also be improved by reducing the particle size to the nanoscale. Nanoparticles are materials that have a one-dimensional shape and a size of less than 100 nm. Small particle size can increase the contact area between the electrolyte and the electrode and reduce the transfer distance of lithium ions and electrons [67]. In addition, nanomaterials have low volumetric energy density and their structure can overcome excessive volume expansion and withstand stress during the charge-discharge process [68]. In general, particle size plays an important role in the electrochemical performance of lithium-ion batteries. Several studies have investigated the effect of SiO₂ particle size on battery performance. Gao *et al.* [24] in a previous study mentioned that commercial SiO₂ with a particle size of 7 nm can react with lithium ions at 0.0 to 1.0 V (versus Li⁺/Li) and produce a reversible capacity of 400 mAh/g. In a study related to SiO₂-based nanoparticle composites as lithium-ion battery anodes, Yao *et al.* [69] synthesized SiO₂/C nanoparticle composites with an average particle

size of 20 nm. The SiO₂/C composites showed a reversible capacity of 500 mAh/g after 50 cycles.

Tu *et al.* [31] also successfully synthesized SiO₂ nanospheres using the sol-gel method, which has good size uniformity. The SiO₂ anode exhibited a reversible capacity of 876 mAh/g at 1C for 500 cycles. The application of SiO₂ nanoparticles as battery anodes has also been studied by Lan *et al.* [70] who synthesized SiO₂ nanoparticles anchored on hollow porous carbon shells (HCP/SiO₂/C) through a carbonization process. The nano-scale SiO₂ particles are well distributed within the carbon layer, as shown in **Figure 6**. The composite has a low charge transfer resistance, indicating fast electron transfer kinetics. In addition, the semicircle formed after 500 cycles is also smaller than that of commercial SiO₂ anodes. This indicates that the nanoparticle structure of the composite contributes to the diffusion and transfer rate of lithium ions. The anode surface shows small cracks after 500 cycles, and the SiO₂ nanoparticle structure remains intact. This indicates the stability of the composite.

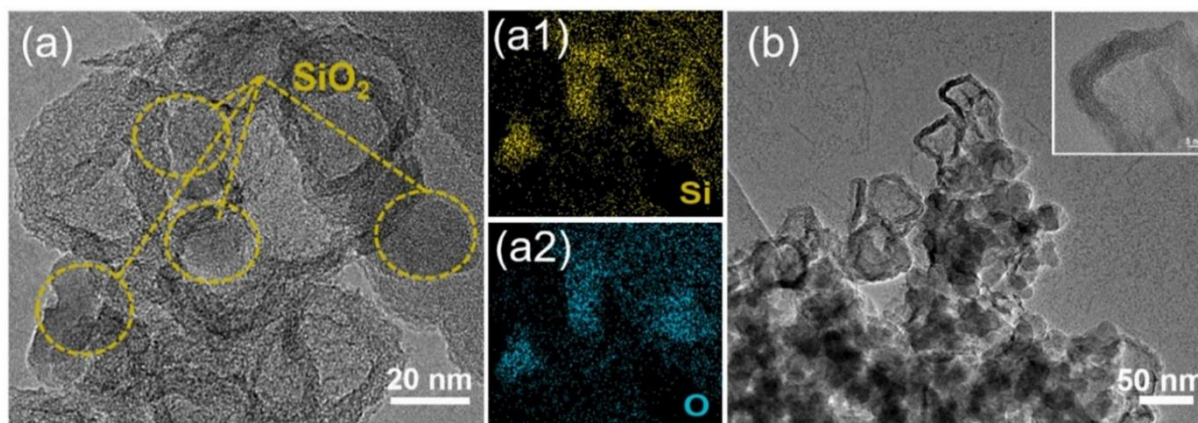


Figure 6 (a) and (b) TEM image of HPC@SiO₂/C; (a1,a2) elemental EDS image of HPC@SiO₂/C [70].

Silica-based composite for anodes

Various strategies have been employed to modify the electrochemical performance of lithium-ion battery anodes, such as the creation of composite materials, coatings, and modifications to nano-structures and porosity. SiO₂-based composite materials are currently being extensively researched as an effective means of improving the electrochemical performance of SiO₂-based anodes. Therefore, the strategies employed to overcome the challenges of SiO₂ anodes, particularly

modification as composite materials, will be discussed in this section.

SiO₂/C composite

In general, modifying the properties of SiO₂ using carbon materials as composites is an effective strategy for reducing volume expansion during the lithiation/delithiation process and improving the electrical conductivity of SiO₂. Various carbon materials such as activated carbon, graphene, and

carbon nanomaterials have been used to improve and enhance the performance of SiO₂ anodes. The carbon layer formed on the composite will form a protective shell that can reduce volume changes [71]. In addition, this layer can reduce the resistance between SiO₂ particles and the current collector. Carbon material has excellent electrical conductivity, which can improve charge transport and reduce internal resistance during the charge/discharge process [72].

SiO₂/C composites generally produce good electrochemical performance, as reported by Yao *et al.* [69], who synthesized carbon-coated SiO₂ nanoparticles through wet chemical reactions and heat treatment, producing a reversible capacity of 500 mAh/g over 50 cycles. The electrochemical impedance spectra (EIS) results show that carbon coating has a positive effect in reducing the interfacial resistance between the active material and the current collector. According to Lv *et al.* [1], who synthesized amorphous SiO₂/C composites

through a sol-gel process combined with mechanical milling and heat treatment, the carbon coating not only affects the electrical conductivity of silica but also accommodates the volume expansion that occurs during the charge-discharge process. The carbon layer on the SiO₂ surface can prevent particle aggregation and, together with the porosity of SiO₂, can support volume changes during the lithiation/delithiation process, thereby increasing cycle stability. Buga *et al.* [73] in their study on the synthesis of carbon-coated SiO₂ composites using the carbonization method produced good electrochemical performance. The composite showed a reversible capacity of 714.3 mAh/g with a CE of 98.9% during 200 cycles. These results are attributed to the carbon layer acting as a matrix that protects the material structure and keeps it stable. The electrochemical performance of SiO₂/C-based composite anodes over the past few years is shown in

Table 2.

Table 2 Electrochemical performance of various SiO₂/C-based composite anodes.

Anode sample	Method	Battery configuration	Electrochemical performance						Ref
			Current density A/g	Initial discharge capacity (mAh/g)	Initial CE (%)	Reversibel capacity (mAh/g)	CE (%)	Cycle	
SiO ₂ /C	Carbonization	Half cell	0.1	922.1	-	714.3	98.9	200	[73]
C/SiO ₂	Carbonization	Half cell	0.1	2,281.69	-	832.19	72.2	300	[74]
SiO ₂ /C	Hydrothermal, Carbonization	Half cell	0.1	1,462	59	888	-	100	[53]
SiO ₂ /C	Hydrothermal, Carbonization	Half cell	0.1	780	50	350	~100	500	[75]
SiO ₂ /C	Calcination	Half cell	1	1,385	~40%	534	~90	1,000	[76]
Porous C/SiO ₂	Carbonization	Half cell	0.1	-	-	1,105	99	360	[33]
3 Dimensional SiO ₂ -C	Carbonization	Half cell	-	~1,000	-	294	-	300	[65]
Porous Silica-carbon membrane	solid state photo-polymerization	Half cell	0.1	719	79.94	693	99	100	[77]
HPC@SiO ₂ /C	Polymerization	Half cell	0.1	1,321	46	804	98	500	[70]
SiO ₂ @C@graphene	Hydrothermal	Half cell	0.05	713.3	36.1	250	-	200	[78]
SiO ₂ /C/CNT	Chemical vapor deposition	Half cell	1	1,267.2	65.2	315.7	-	1,000	[8]
Carbon-SiO ₂ @SiO ₂ @CNT	Carbonization	Half cell	0.1	~1,400	-	644	-	200	[79]
Mesoporous SiO ₂ -CNFs	Vapor deposition	Half cell	-	2,420	-	2,420	86.4	30	[80]

FS-SiO ₂ /C-CNFMs	Electrospinning	Half cell	0.1	1,800	~50	754	100	200	[81]
Free-standing SiO ₂ /carbon nanofibers	Electrospinning	Half cell	0.5	-	-	405	~100	1,000	[82]
Silica@carbon nanofibers	Electrospinning	Half cell	0.1	~1,100	52.21	440	89	200	[83]

Biomass and organic materials are commonly used as carbon sources in SiO₂/C composites because they are efficient, environmentally friendly, and abundant. Rice husks have been widely used as a biomass source in the synthesis of SiO₂/C composites. Most of the rice husk content consists of lignin, organic cellulose, hemicellulose, and inorganic silicon compounds, making it a source of carbon and silica for battery anode synthesis. Silica naturally exists in the form of nanoparticles that are evenly deposited on the cell walls of rice husks as amorphous silica polymers [84]. The C/SiO₂ composite obtained through the heating process of rice husks produced an initial discharge capacity of 325 mAh/g, which increased to 485 mAh/g after 84 cycles [85]. Cui *et al.* [33] have successfully synthesized SiO₂/C composites through a carbonization process as shown in **Figure 7(b)**. The resulting SiO₂/C composites exhibit good cycle performance and rate capability as

seen in **Figure 7(a)**. The porous structure and large surface area of the composites provide more sites for Li ion storage and shorten the Li⁺ diffusion path. In addition, the carbon layer provides good electrical conductivity to SiO₂ and can withstand volume changes. Recent research conducted by Butcha *et al.* [75] utilized straw as a source of SiO₂/C composite with hydrothermal treatment. The composite showed good electrochemical performance with a reversible capacity of 350 mAh/g after 500 cycles. The temperature used in the carbonization process also effect the electrochemical performance of the composite. Chu *et al.* [86] synthesized SiO₂/C nanocomposites from rice husk through carbonization at various temperatures. The SiO₂/C produced by carbonization at 900 °C showed the highest capacity (a discharge capacity of 932.41 mAh/g with an initial coulombic efficiency of 61.93%) and good stability (650 mAh/g after 150 cycles).

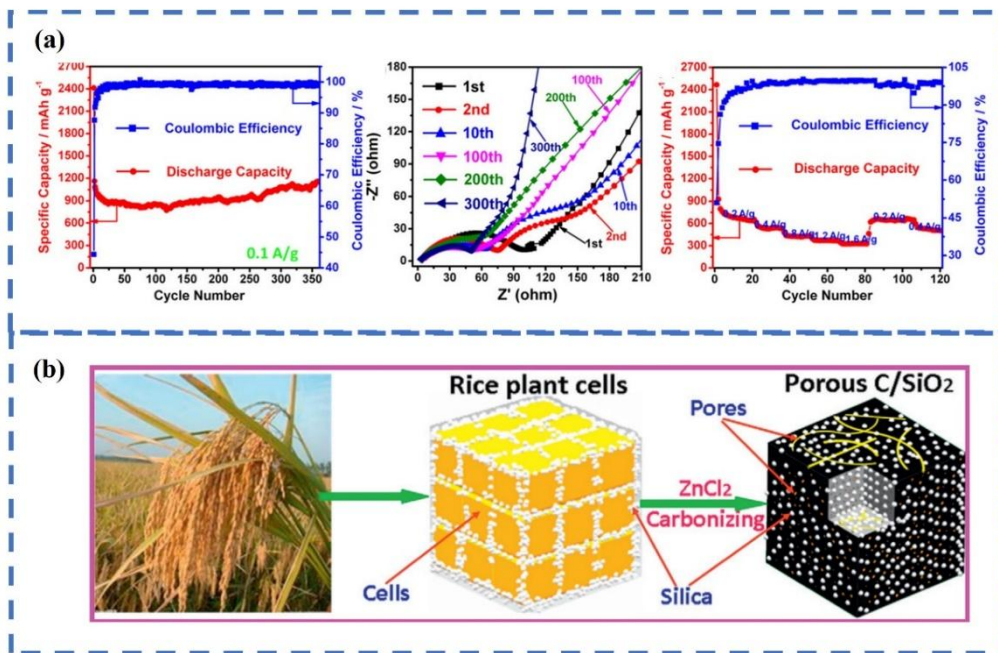


Figure 7 (a) Cycling performance, Nyquist plots under discharge conditions and rate performance of C/SiO₂ anodes, (b) C/SiO₂ composite formation process [33].

Some carbon materials in SiO₂/C composites have low crystallinity, limiting their ability to improve the electrical conductivity of SiO₂. Carbon nanotubes (CNTs) are a type of carbon material that has high crystallinity and good mechanical strength [87]. CNTs can also be used as a substrate to absorb mechanical stress from SiO₂ due to lithiation/delithiation reactions. The unique structure of CNTs is not only effective as a buffer for SiO₂ volume expansion, but also prevents direct contact between the active material and the electrolyte, thereby forming a stable SEI layer [88]. In addition, the carbon layer provides higher electrical conductivity. This structure can maintain the stability of the battery cycle [6].

Modification of SiO₂ with CNTs can not only improve conductivity but also support volume changes during the charge/discharge process. Wang *et al.* [8] synthesized SiO₂/C/CNTs composites through an in-situ chemical vapor deposition (CVD) method with a Co catalyst and produced good anode performance. The CV curve shows little difference over 100 cycles,

confirming that the composite has stable structural stability. The SiO₂/C/CNTs anode material has good rate capability and cycle performance, where the anode can maintain a capacity of 315.7 mAh/g for 1,000 cycles. CNTs with a one-dimensional structure can form a carbon conducting network that provides more electrical pathways between SiO₂/C composite particles, which can improve the electrochemical performance of the composite. Wang *et al.* [79] also synthesized a carbon-SiO₂@SiO₂@CNTs composite through a carbonization method and produced a porous material as shown in **Figure 8**. This pore structure not only provides pathways for Li ion transfer but is also effective in reducing volume expansion during cycling. CNTs as a support framework in the material not only provide space for volume expansion but also contribute to electrolyte and Li ion diffusion. In addition, CNTs also have high electrical conductivity, which accelerates electron transfer and produces good electrochemical performance.

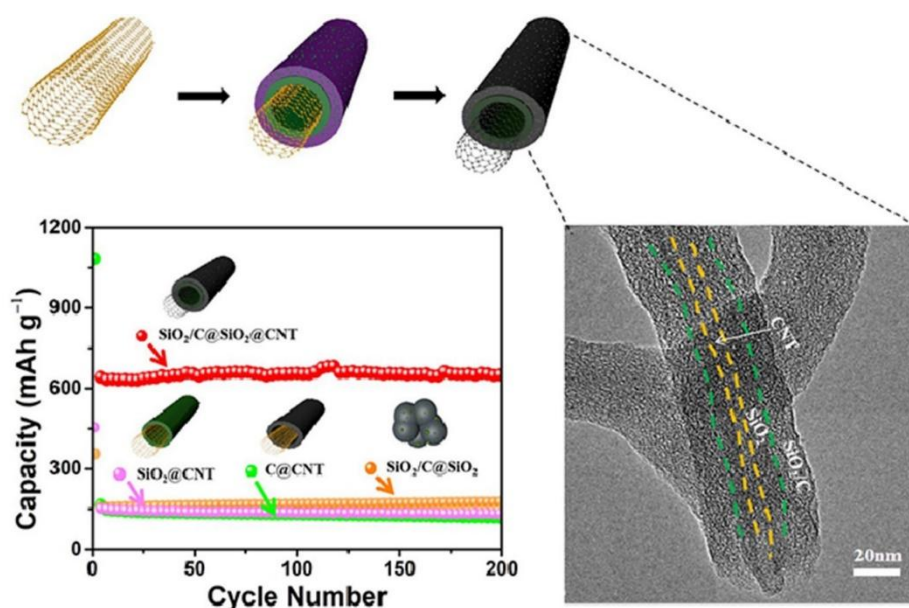


Figure 8 HRTEM image and performance of carbon-SiO₂@SiO₂@CNTs composite cycles [79].

Carbon materials with 1-dimensional architecture such as carbon nanofibers (CNFs), which have a large surface area, the ability to improve electrical conductivity, short diffusion distances, and strong resistance to pressure changes, have been considered as one of the supporting carbon materials for lithium-ion battery anode materials [89]. CNFs show great

application prospects due to their unique electronic characteristics. CNF materials can provide flexible space, thereby suppressing volume changes and SEI formation [82].

Hyun *et al.* [80]. synthesized SiO₂/CNFs composites using the vapor deposition method with Fe-Cu catalysts. The SiO₂/CNFs composites synthesized

without binders showed the highest charging and discharging capacities. The initial capacity of the composite (2,420 mAh/g) decreased to 2,092 mAh/g after 30 cycles at a retention rate of 86.4%. Free-standing SiO₂/C nanofibers composites were also successfully synthesized by belgibayeva *et al.* [81], using a 2-step heating electrospinning method. **Figure 9** shows the SEM results of the composites, revealing the morphology of SiO₂/C nanofibers under different heating treatments. The heating stage has a positive effect not only on the morphology of the material but also on the structure and electrochemical performance of the composite. SiO₂/C nanofiber composites with preoxidation treatment showed a better capacity of 470 mAh/g after 50 cycles. In addition, free-standing SiO₂/C nanofibers composites also showed better

electrochemical performance than SiO₂/C nanofibers electrodes prepared by coating on a current collector. The initial discharge and charge capacities of 1,800 and 984 mAh/g were due to shorter Li ion pathways. The diameter of the Nyquist EIS semicircle plot also decreased after continuous cycling compared to the fresh cell, indicating a decrease in charge transfer impedance. Recent research by Wang *et al.* [90], prepared a self-standing anode from SiO₂@Carbon Sphere/SiO₂-CNF using the electrospaying-electrospinning technique. The anode produced an initial capacity of 633 mAh/g and maintained a coulombic efficiency of 70% over 1,000 cycles. The SiO₂/carbon nanofibers composite anode is an attractive anode alternative due to its characteristics and structure.

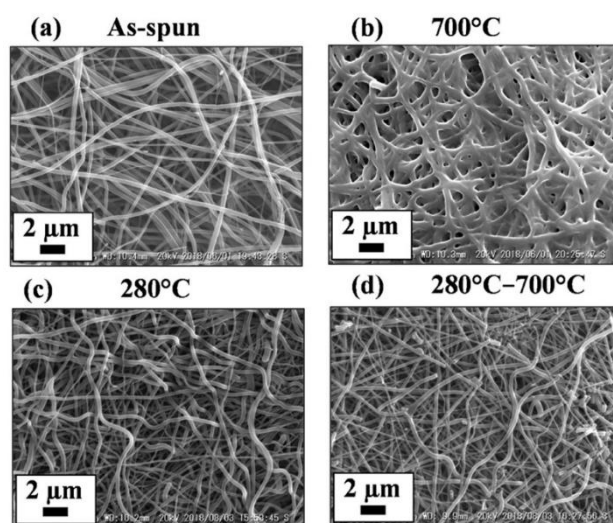


Figure 9 SEM results before and after heating treatment under different conditions [81].

SiO₂/metal composite

SiO₂ anodes have better cycle stability during the charge-discharge process compared to Si. The formation of Li₂O and Li₄SiO₄ phases during initial lithiation, which are inert, helps reduce volume expansion [91]. However, the formation of these irreversible phases in the first cycle results in low initial coulombic efficiency (ICE). In addition, SiO₂ anodes also have low electrical conductivity [92]. One strategy to overcome this challenge is to modify SiO₂ using support materials that have high charge conductivity, such as Ni, Cu, Sn, and Sb [93].

Wu *et al.* [54] in their latest research successfully synthesized silica gel combined with zinc nanoparticles (SG@Zn) through ball milling and heat-melting methods as shown in **Figure 10(a)**. The SG@Zn anode showed good cycle performance and rate capability. Zn has low volume expansion, good electrical conductivity, and large volume capacity, which can change the properties of the SiO₂ anode. **Figure 10(b)** illustrates that the addition of Zn can significantly increase the capacity and cycle stability of the anode. In addition, the presence of Zn contributes to the Li⁺ ion diffusion process and increases ionic conductivity.

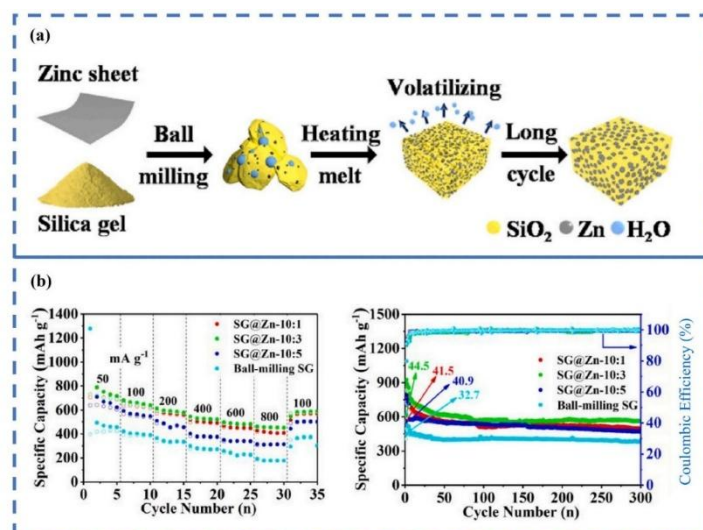


Figure 10 (a) Synthesis process scheme for SG@Zn anode material, (b) Rate performance and cycling performance of different SG@Zn anodes [54].

Modification of SiO_2 using metal-based materials was also successfully carried out by Tang *et al.* [92] who synthesized Ni/SiO_2 nanocomposites through a hydrothermal process to form nickel silicate and annealing. EIS results showed that Ni nanoparticles could increase the conductivity of the composite. In the synthesized composite, Ni nanoparticles were evenly distributed in the SiO_2 matrix, forming electron conduction pathways. The nanosheet form of the Ni/SiO_2 composite shortened the lithium ion diffusion pathway and increased the anode rate capability. Li *et al.* [94] also modified the properties of SiO_2 using Al as a composite through the melt-self-assembly method and produced good electrochemical performance. The unique structure of the SiO_2/Al composite produces good electrical conductivity and increases lithium ion diffusion capability. In addition, both materials have low discharge potential, which can increase the energy density of the anode.

Zhong *et al.* [95] synthesized $\text{SiO}_2/\text{Co@N}$ -doped carbon nanofibers composites using the electrostatic spinning method with high-temperature treatment. This method is simple but effective for distributing SiO_2 and Co particles evenly in the carbon matrix. The $\text{SiO}_2/\text{Co@N}$ -doped carbon nanofibers composite

showed good lithium storage performance, where the anode could maintain a discharge capacity of 552 mAh/g after 1,000 cycles at 1 A/g. Co nanoparticles are not electrochemically active but contribute to improving charge transfer and act as a catalyst that facilitates lithium release and provides energy for SEI and Li_2O decomposition. The addition of Co to the composite also successfully increased the conductivity of the material and reduced resistance. In addition, the anode can maintain its structure after cycling at 0.5 A/g, indicating that the addition of Co can improve the structural stability of the composite. In a recent study by Zhong *et al.* [96] metal-organic frameworks (MOFs) were used to form Co/N-C@SiO_2 composites. MOFs are known to have a large surface area, porous structure, and uniform component distribution, making them suitable for use as templates [97]. The hollow structure of the composite can help reduce the adverse effects caused by volume expansion. In addition, the presence of cobalt nanoparticles further enhances the conductivity and electrochemical performance of the Co/N-C@SiO_2 composite. The electrochemical performance of $\text{SiO}_2/\text{metals}$ -based composite anodes over the past few years is shown in **Table 3**.

Table 3 Electrochemical performance of various SiO₂/metal-based composite anodes.

Anode sample	Method	Battery configuration	Electrochemical performance						Ref
			Current density A/g	Initial discharge capacity (mAh/g)	Initial CE (%)	Reversibel capacity (mAh/g)	CE (%)	Cycle	
SG@Zn	Melt-self-assembly	Half cell	0.1	1,865.6	45	590	-	300	[54]
Cu-Sn/nano-SiO ₂	Electrodeposition	Half cell	0.1	1,470.8	~50.76	358.6	-	100	[98]
Ni/SiO ₂ hollow spheres	<i>In situ</i> reduction	Half cell	10	1,195	56.6	337	-	1,000	[92]
SiO ₂ /Co@N-doped CNF	Electrostatic spinning	Half cell	1	1,922	73	552	-	1,000	[95]
Co/N-C@SiO ₂	Carbonization	Half cell	5	~2,600	-	337	-	700	[96]
Mxene@SiO ₂	Electrostatic self-assembling	Half cell	0.1	430.7	88.3	380.2	~99	600	[99]
SiO ₂ /Al	Melt-self-assembly	Half cell	-	1,698.2	46.8	696	-	300	[94]
CNT/(Fe@Si@SiO ₂)	CVD, dip-coating	Half cell	1	1,787	53.8	804	83	500	[100]
SiO ₂ @SnO ₂ @C	Etching-free template	Half cell	0.2	1,822	51.6	923	99	340	[101]
NiS@SiO ₂ /graphene	Electrostatic self-assembling	Half cell	0.1	1,275	68	~750	~100	100	[102]
SiO ₂ /TiO ₂	Plasma electrolytic oxidation	Half cell	-	~550	88.7	400	-	500	[103]
SiO ₂ @a-TiO ₂ @Ag	Sol-gel	Half cell	0.1	1,320	58	712	99	300	[104]

SiO₂/other composite

Another strategy used to overcome the challenges of SiO₂ anode applications is the creation of composites with 2-dimensional materials that can improve mechanical stability, reduce volume expansion, and promote the formation of inorganic components in SEI. MXene is a type of 2-dimensional transition metal carbide/nitride material that has attracted widespread attention in recent years due to its high electrical conductivity ($6.76 \times 10^5 \text{ Sm}^{-1}$), good mechanical properties, and low resistance to lithium diffusion (0.05 eV) [105]. In addition, the high number of functional groups on its surface (-O, -OH, -F) can contribute to the formation of inorganic species in the SEI bond, thereby increasing the adhesive strength of the composite [106]. This material has a crystal structure, surface, and chemical composition that are suitable for energy storage. The use of MXene/metal oxide composites such as SiO₂ as an anode can improve

electrical conductivity, mechanical stability, and battery capacity [9]. However, research on the application of SiO₂/MXene composites has not been widely conducted.

Chen *et al.* [99] synthesized MXene@SiO₂ composites as lithium-ion battery anodes through a pre-lithiation process. The mesoporous structure of MXene@SiO₂ composites with uniform pore distribution at 4 - 25 nm can promote Li⁺ diffusion kinetics, while the presence of a conductive MXene layer will result in rapid charge transfer within the anode structure. After prelithiation for 1 day, the MXene@SiO₂ anode showed an increase in reversible capacity of 380.2 mAh/g over 600 cycles at 100 mA/g, which is a contribution of the MXene coating effect and the prelithiation process that promotes the long-term cycling capability of the battery. Based on XPS analysis, the illustration of the SEI structure on the SiO₂ and MXene@SiO₂ anodes after pre-lithiation is shown in

Figure 11(a). The ratio of inorganic Li_2CO_3 and LiF species formation continues to increase in the SEI layer, contributing to improved electrochemical performance

and cycle stability due to the high ionic conductivity and structural strength of the $\text{MXene}@/\text{SiO}_2$ composite.

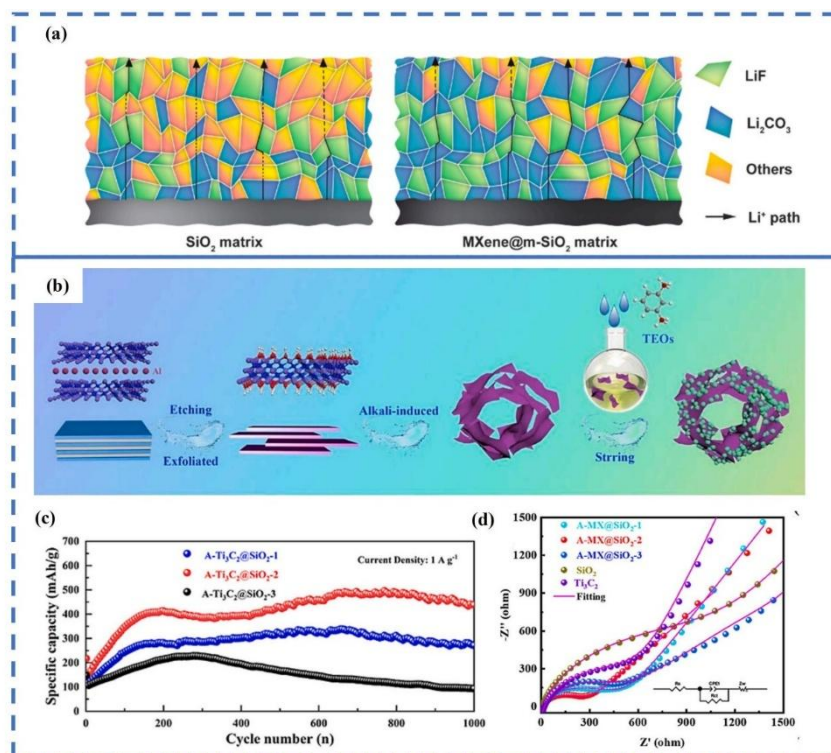


Figure 11 (a) Illustration of the SEI structure on SiO_2 and $\text{MXene}@/\text{SiO}_2$ composites after pre-lithiation [99], (b) Schematic illustration of the preparation process for $\text{Ti}_3\text{C}_2\text{T}_x@\text{SiO}_2$ composites, (c) Cycling performance, and (d) Nyquist plots of $\text{Ti}_3\text{C}_2\text{T}_x@\text{SiO}_2$ composites [107].

Research related to MXene composites was also conducted by Wang *et al.* [107] who synthesized $\text{Ti}_3\text{C}_2\text{T}_x@\text{SiO}_2$ composites as anodes using a low-temperature liquid phase method as shown in **Figure 11(b)** and provided good electrochemical performance. $\text{Ti}_3\text{C}_2\text{T}_x$ is one of the MXene materials that has a structure similar to graphene, but has higher electrical conductivity, faster lithium ion diffusion rate, and lower ion diffusion resistance (0.07 eV) [108]. The $\text{Ti}_3\text{C}_2\text{T}_x@\text{SiO}_2$ composite exhibits electrochemical performance with a specific capacity of 437.9 mAh/g at 1.0 A/g after 1000 cycles, as shown in **Figure 11(c)**. This cycling capability is due to the stable structure of the composite. Additionally, the composite also exhibits good rate capability due to the conductive $\text{Ti}_3\text{C}_2\text{T}_x$ network that facilitates electron transfer. Compared to the SiO_2 anode, the composite exhibits lower charge transfer resistance and a higher diffusion coefficient, as shown in **Figure 11(d)**. This is due to the addition of

$\text{Ti}_3\text{C}_2\text{T}_x$, which can alter the electrical conductivity of the composite and reduce the diffusion path of lithium ions due to its unique structural design. Mu *et al.* [91] stated that the $\text{SiO}_2/\text{MXene}$ composite forms a unique structure. The flexible MXene matrix can efficiently reduce the stress caused by the large volume expansion of SiO_2 . The MXene layer not only provides a conductive 3D network to accelerate electron transfer but also enhances lithium ion diffusion. Their research on $\text{SiO}_2/\text{MXene}$ composite anodes yielded good electrochemical performance with a reversible capacity of 635 mAh/g at 1 A/g over 200 cycles. This indicates that the application of $\text{SiO}_2/\text{MXene}$ composites as anodes holds promising prospects and warrants further development.

Conclusions

SiO₂-based materials are alternative candidates for high-capacity lithium-ion battery anodes. Low initial coulombic efficiency (ICE), insufficient electrical conductivity, and volume expansion during the charge-discharge process are challenges faced by SiO₂ anodes. Modifying SiO₂ into composites with carbon or metal materials is an effective strategy to improve the electrochemical performance of SiO₂ anodes. This article provides an overview of the electrochemical characteristics and lithium storage mechanisms of SiO₂-based anode materials and focuses on the application and development of SiO₂-based composites in improving the electrochemical performance of lithium-ion batteries to provide an overview of the challenges and prospects for the development of SiO₂ anodes in lithium-ion batteries. Carbon materials are ideal for use as a matrix in SiO₂/C composites due to their ability to reduce the volume expansion of the anode. Various types of carbon materials have been used to improve SiO₂ performance, such as carbon nanotubes and carbon nanofibers. Meanwhile, in SiO₂/metal composites, metals such as Ni, Al, and Co have excellent electrical conductivity and are effective in accelerating electron transfer due to the conductive network they form. In addition, the 2-dimensional structure of MXene can accommodate volume fluctuations during lithiation/delithiation and help maintain anode stability by reducing mechanical stress. The incorporation of carbon, metal, and MXene materials into the SiO₂ matrix appears to be equally effective in mitigating volume expansion, as well as enhancing electrical conductivity and cycling stability, particularly through the structural modification strategies employed. Nevertheless, challenges such as unstable SEI formation and low coulombic efficiency, which hinder the effective utilization of SiO₂-based materials, remain difficult to overcome. Although SiO₂-based anode materials show promising electrochemical performance and cycle stability through composite material modification, their practical application still faces several challenges, such as the overall system cost being higher than conventional graphite anodes due to the complexity of the production process.

Future work

Research on SiO₂-based composite materials is still in its early stages and further research is needed. Through proper structural design and modification, SiO₂-based composite anodes exhibit good electrochemical performance. So far, various modifications and structural designs of SiO₂-based anode materials have been attempted to improve structural stability and electrochemical performance. We hope that future research on SiO₂ anodes will contribute to the development of lithium-ion battery anodes and focus on the following aspects.

(1) Studies on the reaction mechanism between SiO₂ and lithium, as well as the formation of inactive phases (Li₂O and Li₄SiO₄) and the growth of the solid electrolyte interface (SEI), need to be fully investigated in order to create better materials.

(2) Efforts to increase the ICE value through structural design engineering and pre-lithiation procedures for SiO₂ anodes. Currently, SiO₂-based composite anodes with carbon or metals still exhibit low ICE values. The effect of pre-lithiation treatment on SiO₂ anodes on the resulting ICE needs to be studied further.

(3) Aspects such as conductive materials and innovative binders also need to be studied to improve the mechanical stability of anode materials during the charge/discharge process. In terms of binders, the impact of binders on SiO₂-based anodes is still relatively insignificant. Innovative binders such as conductive polymer binders or dual functional binders should ideally be adaptive, so that they can provide higher ICE and reversible capacity values compared to traditional binders.

(4) Low-cost alternative materials, simple synthesis processes, and environmentally friendly processes will be important in future production processes. Green synthesis-based approaches and the utilization of natural resources could be long-term solutions. In addition, a comprehensive evaluation of the environmental impact and economic feasibility of SiO₂-based anodes is needed. This study is important to ensure the sustainability of SiO₂-based anode applications.

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Declaration of Generative AI in Scientific Writing

The corresponding author, on behalf of all authors, declares that there is no generative AI.

CRediT Author Statement

Yayuk Astuti: Writing - original draft, Writing - review & editing, Validation, Resources, Formal analysis, Data curation, Conceptualization. **Yurike Candra Sefia:** Writing - review & editing, Writing - original draft, Formal analysis, Investigation, Visualization, Data curation. **Iis Nurhasanah:** Writing - review & editing. All authors have read and agreed to the published version of the manuscript.

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