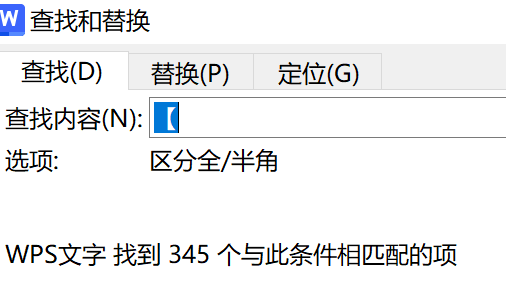
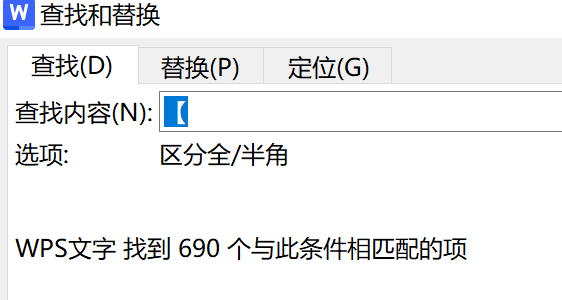
1



2.1



[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

【锂电池优点、应用】

## 【锂电池优点、应用】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

Because of their high energy and power density, lithium ion batteries (LIBs) are currently the most promising energy storage technology for mobile devices, electric vehicles, and large-scale energy storage. 【为提升锂电池能量密度，发展新电极材料和配方】

## 【为提升锂电池能量密度，发展新电极材料和配方】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

To further increase the energy density of LIBs, relentless research efforts have been invested in the development of new electrode materials and optimizing the electrode formulation.1−5 【商业锂电基于石墨，容量数据】

## 【商业锂电基于石墨，容量数据】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

Commercial LIB anodes typically rely on Li-ion intercalation in graphite (372 mAh/g) or lithium titanate (175 mAh/g).5 【然而，合金化或转化机理可获得更高的能量密度】

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However, much higher gravimetric energy densities have been achieved using anodes that react with lithium by alloying or conversion mechanisms (e.g., Si, Sn, GeO2, SnO2,Fe3O4, etc.).6−8 【高容量材料的缺点】

## 【高容量材料的缺点】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

Unfortunately, these high capacity materials have disadvantages including poor cycling stability, large volume change during Li+ insertion/extraction, high voltage hysteresis, poor rate performance, poor Coulombic efficiencies (CE), and low electrical conductivity.9−12 【提升电子/离子传输，缓解应力，前人的纳米化工作】

## 【提升电子/离子传输，缓解应力，前人的纳米化工作】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

To improve the ion and electron transport, as well as to alleviate mechanical stress, previous researchers have looked into nanostructuring these active materials.【纳米化的缺点】

## 【纳米化的缺点】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

However, nanostructuring often introduces problems of its own such as low tapped and electrode density as well as irreversible reactions taking place on the high surface area of the nanostructured materials. 【目前研究正在发展策略以解决问题】

## 【目前研究正在发展策略以解决问题】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

Ongoing research is developing strategies to alleviate some of these challenges.13−15 【吸引人的策略，改善导电性，碳包覆。文献举例，缓冲体积膨胀，但降低振实密度】

## 【吸引人的策略，改善导电性，碳包覆。文献举例，缓冲体积膨胀，但降低振实密度】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

In particular, attractive strategies have been proposed to address the poor conductivity of high capacity materials by advanced carbon coating processes. For instance, a number of promising carbon cages have been proposed to buffer the volume change of active materials,16−18 but these further reduce the tapped density of the electrodes. 【石墨烯和碳纳米管被研究。存在的问题。产生问题的原因及后果】

## 【石墨烯和碳纳米管被研究。存在的问题。产生问题的原因及后果】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

Alternatively, flexible high-aspect conductive carbons, such as graphene and carbon nanotubes (CNTs), have been studied.19−21 However, these materials tend to phase segregate after slurry mixing. This is because most carbon additives with high conductivity disperse well in NMP, whereas metal or metal oxides materials tend to disperse water well, which makes it difficult to codisperse them with battery binders such as PVDF in NMP or CMC/SBR in water.22,23 This phase segregation during the electrode coating and drying compromises electron conduction and results in poor material utilization and high ohmic losses. The phase segregation is particularly pronounced with high areal loading electrodes pursued in industry, which dry slower.24 Further, several researchers have shown that the interface between carbon additives and the active material can degrade over time, which further accentuates the above problems.25,26 【工业和学术界，减少碳添加物团聚的方法。学术界方法的问题】

## 【工业和学术界，减少碳添加物团聚的方法。学术界方法的问题】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

Industrially, time-consuming kneading and high intensity mixing processes are used to reduce the agglomeration of carbon additives,24 and academically, methods are developed to anchor active battery materials on the surface of carbon additives (e.g., by synthesizing the active material in the presence of CNTs or graphene).27,28 However, to nucleate the active material on the carbon additives, the carbon surface typically requires oxidation or other chemical modifications, which in turn decreases their electronic conductivity and is poorly scalable.

【本文，综合以上研究原则，硅/铁硅化物被碳纳米管包覆。纳米颗粒形成二次微米颗粒，表面生长碳纳米管】

## 【本文，综合以上研究原则，硅/铁硅化物被碳纳米管包覆。纳米颗粒形成二次微米颗粒，表面生长碳纳米管】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

In this work, we combine several of the above principles to fabricate advanced silicon/iron silicide anodes coated with CNTs. In particular, we focus on a continuous fabrication process that structures nanoparticles into secondary micrometer-sized particles on which CNTs are synthesized.【硅的优点】

## 【硅的优点】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

Si is a promising active material because of its high theoretical capacity (>3500 mAh/g), low reaction potential (∼0.35 V), and low cost.29−32 【硅已在石墨电极中混合少量，作为商业电动车应用】

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Because of these properties, Si is already mixed in very small amounts with graphite electrodes for commercial automotive applications.5【本文，体系选取理由】

## 【本文，体系选取理由】

[1] C. Jo, A.S. Groombridge, J. De La Verpilliere, J.T. Lee, Y. Son, H.-L. Liang, A.M. Boies, M. De Volder, Continuous-Flow Synthesis of Carbon-Coated Silicon/Iron Silicide Secondary Particles for Li-Ion Batteries, ACS NANO, 14 (2020) 698-707.

Here, we start from nanosized silicon particles (Si NPs) and react them with iron precursors to form iron silicide for three reasons: (i) to decrease the electrical resistance, which is 2.6 × 101 Ω cm in FexSiy compared to 6.0 × 103 Ω cm in Si33 and (ii) to reduce the volume expansion during cycling. This comes at the cost of a lower capacity (iron silicides <100 mAh/g),33−35 but it should be noted that because of limitations in the capacity of current cathode materials (approximately 200 mAh/g), anode capacities in excess of 550 mAh/g do not substantially improve the overall battery performance.36 Here, we first optimize Si/ FeSi cores with a capacity of 1200 mAh/g and then blend them with graphite to obtain 550 mAh/g anodes. (iii) CNTs can be synthesized from iron/iron silicide domains on Si nanoparticles.37,38 The latter results in a strong anchoring of the carbon additive on the active material and fosters a better electric network in the electrode. This approach inverts the classic approach where carbon additives such as CNTs or graphene are first oxidized followed by active material synthesis from surface defects, which compromises the electrical conductivity of the conductive material. These three properties address electron-transport challenges as well as problems related to phase segregation during the electrode mixing but do not solve problems related to the poor tapped density and poor electrode coating properties of nanomaterials. In commercial battery electrodes, small primary battery particles are often aggregated in larger secondary particles to alleviate electrodecoating challenges and increase the volumetric density.39,40 Here, we follow a similar strategy using spray drying to pack our Si nanoparticles into micrometer-sized spheres. The electrical resistance of micrometer-sized pure Si secondary particles would be too high for good battery operation, but this work shows that CNTs can be synthesized inside the crevices of the secondary particles hereby providing a good internal short-range electrical network. In addition, CNTs extending from the surface of the secondary particles can help with the interparticle long-range conductivity, which is particularly important as industry shifts to thicker electrode coatings.24

Impressive results were achieved in previous reports where CNTs grown on the surface of Si particles for battery applications, however in these reports, CNTs were not structured in secondary particles.41−43 As a result, these batteries were only tested in low loading levels and show poor CE. In contrast, our CVD process on secondary particles reduces the BET surface area and therefore improves the CEs of the material.44 Further, we have performed blending experiments of our electrodes with commercial graphite anodes to balance the electrode capacity, density and areal loading. Finally, we demonstrate a continuous flow synthesis method that allows to continuously synthesize these complex materials from simple precursors. As summarized in Figure 1, our approach enables a hierarchical design for battery electrodes where at the nanoscale the chemical composition of the primary particles and their interface with carbon additives is controlled while at the microscale the secondary particle size and carbon additives are controlling the ion and electron transport, and finally, flow synthesis allows for continuous material production.【】【】

RESULTS

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

1. Introduction

【迫切需要储能器件；目前锂电池可满足】

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To meet the surging demand for portable electronic devices and electric vehicles, efficient energy storage devices with high energy and power densities are urgently required. Lithium-ion batteries (LIBs) currently fill this need. 【锂电池容量和循环性尚不满意】

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[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

However, despite decades of progress, the capacity and cycle life of LIBs remain unsatisfactory. [1,2] 【锂电池通常使用石墨】

## 【锂电池通常使用石墨】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

LIBs commonly use a conventional carbonaceous anode (e.g., graphite). [3]【石墨容量限制，无法满足逐步提升的应用需求】

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However, a graphite anode, whose theoretical capacity is limited to 372 mAh g −1 ,cannot meet the escalating application requirements.【为提高锂电池性能，研究高容量负极】

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To improve LIB performance, several high-capacity anodes such as, those made of transition metal oxides,Si, Sn, Sb, Al, Mg, Bi, and their alloys have been studied. [4] 【硅的优点】

## 【硅的优点】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

Among them, Si is a promising anode material due to its high gravimetric capacity (theoretically 3579 mAh g −1 ), earth abundance, and low toxicity. [5] Moreover, its relatively high lithiation/delithiation potential compared to that of graphite makes it safer to operate(i.e., lower risk of Li metal plating). [6]Thus, Si-based anodes have the potential for application in high-performance, ecofriendly, and low-cost LIBs. 【硅的缺点】

## 【硅的缺点】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

Nevertheless, the practical implementation of Si anodes is hindered by its fast capacity decay. [7] The severe volume change (i.e., ≈400%) of Si during lithiation/delithiation causes the mechanical degradation and pulverization of Si particles. Repeated volume expansion/contraction also results in an unstable solid electrolyte interphase (SEI). Low Coulombic efficiency (CE) and SEI accumulation gradually increase the interfacial resistance, leading to electrode performance decay. [8]

【为缓解硅的快速劣化，提出的一些策略】

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To prevent the fast degradation of Si anode, several strategies have been investigated, including size and morphology control, [9] nano-architecture design, [10] application of composites/hybrids, [11] and binder optimization. [12]【硅表面包覆碳的作用】

## 【硅表面包覆碳的作用】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

A thin, uniform, and conformal carbon layer on the Si surface is highly desirable to boost Si conductivity and stabilize the interface.【但是，湿法碳包覆方法无法形成高质量表面涂层，原因】

## 【但是，湿法碳包覆方法无法形成高质量表面涂层，原因】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

However, a conventional wet-chemical carbon coating method cannot form a high-quality surface covering layer. [13] Because of the high surface tension of the liquid reaction medium, Si aggregates are not easily dispersed, and thus carbon precursors are not effectively delivered between Si nanoparticles. [14]【因此，气相碳包覆过程被应用，举例】

## 【因此，气相碳包覆过程被应用，举例】

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More advanced gas-phase carbon coating processes have thus been employed for Si particles. Magnetron sputtering deposition, [15] atomic/molecular layer deposition, [16] and pulsed laser deposition [17,18] have been applied to improve coating quality. 【气相碳包覆技术的缺点】

## 【气相碳包覆技术的缺点】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

However, these techniques require sophisticated facilities, complicate procedures, and high cost, which hinder their practical applications. 【需要一种怎么样的碳包覆方法】

## 【需要一种怎么样的碳包覆方法】

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A more facile, cost-effective, eco-friendly, and easily scalable carbon coating process is required. 【SiOx包覆的作用】

## 【SiOx包覆的作用】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

A SiOx coating can improve Si anode cycling stability. [19] SiOx shows a smaller volume change during lithiation/delithiation compared to that of Si. Moreover, the Li2O and lithium silicates in situ generated during the first lithiation buffer the volume variation and thus improve cyclability. [20] 【然而，构建连续和均匀的SiOx涂层有挑战，因此通常需要额外的合成步骤】

## 【然而，构建连续和均匀的SiOx涂层有挑战，因此通常需要额外的合成步骤】

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Nevertheless, creating a continuous and homogeneous SiOx coating layer is challenging and usually needs extra syn- thesis procedures. 【因此迫切需要一步法获得SiOx/碳多层涂层】

## 【因此迫切需要一步法获得SiOx/碳多层涂层】

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The formation of a SiOx /carbon multi-layer coating in one step, the goal of the present work, is thus highly desirable.

【我们认为超临界二氧化碳合成技术可克服以上局限】

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In this context, we believe that supercritical carbon dioxide (SCCO2 ) synthesis is an appealing method for overcoming the aforementioned constraints. 【超临界二氧化碳合成技术的优点，可控制涂层】

## 【超临界二氧化碳合成技术的优点，可控制涂层】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

SCCO2 fluid is characterized by its extremely low viscosity, gas-like diffusivity, and excellent mass transfer properties. [21] The near-zero surface tension of SCCO 2 allows excellent penetration between nanoparticles and the transport of reaction precursors throughout a sample. [22] The physicochemical properties of SCCO2 , such as, density, viscosity, and the dielectric constant, can be tuned via temperature and pressure. [23] The control of coating layers can thus be achieved. Moreover, SCCO2 is stable, nonflammable, non-toxic, and inexpensive; thus, the synthesis process is green and readily scalable.【超临界二氧化碳合成技术在其他材料合成中的举例】

## 【超临界二氧化碳合成技术在其他材料合成中的举例】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

SCCO2 has been used to exfoliate graphite, MoS2 , and MoSe 2 to produce two-dimensional nanosheets for various applications. [24,25] 【超临界二氧化碳合成可分散纳米硅颗粒】

## 【超临界二氧化碳合成可分散纳米硅颗粒】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

It was also reported that SCCO 2 fluid could help disperse Si nanoparticles between graphene nanosheets. [26] The resulting silicon/graphene composite showed promising rate capability and cycling stability as a LIB anode.【超临界二氧化碳合成可作为前驱体载体用于在不同纳米结构碳材料上锚定不同纳米材料，举例】

## 【超临界二氧化碳合成可作为前驱体载体用于在不同纳米结构碳材料上锚定不同纳米材料，举例】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

In addition, SCCO 2 fluid can act as a precursor carrier for the fabrication of various nanomaterials. For example, nano-size Ni, [27] Fe, [28] Pd, [22] Pt, [29] Au, [30] MnO 2 , [31] SnO 2 , [32] V 2 O 5 , [33] Fe 3 O 4 , [34] Co 3 O 4 [35] have been synthesized and anchored on various nanostructured carbon materials. 【作者前期工作，多孔CoCO3/石墨烯复合材料，用于储Li和Na】

## 【作者前期工作，多孔CoCO3/石墨烯复合材料，用于储Li和Na】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

In addition, our team has demonstrated that SCCO2 itself can serve as the precursor in the synthesis of a highly porous CoCO3/graphene nanocomposite anode for Li + and Na + storage. [36] The charge–discharge properties were superior to those of the conventionally synthesized CoCO3 counterpart. 【硅颗粒表面用超临界二氧化碳合成涂层没被尝试过。值得研究】

## 【硅颗粒表面用超临界二氧化碳合成涂层没被尝试过。值得研究】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

To the best of our knowledge, using SCCO2 to create coating layers on Si particles has not been previously attempted. We believe that this approach is worthy of investigation.

【本文，三种不同碳前驱体，超临界二氧化碳辅助制备碳涂层】

## 【本文，三种不同碳前驱体，超临界二氧化碳辅助制备碳涂层】

[2] R.F.H. Hernandha, P.C. Rath, B. Umesh, J. Patra, C.-Y. Huang, W.-W. Wu, Q.-F. Dong, J. Li, J.-K. Chang, Supercritical CO2-Assisted SiOx/Carbon Multi-Layer Coating on Si Anode for Lithium-Ion Batteries, ADVANCED FUNCTIONAL MATERIALS, 31 (2021).

In the present work, three kinds of carbon precursor, namely, glucose, sucrose, and citric acid, are used in the SCCO 2 -assisted deposition process to produce carbon coating layers. SCCO 2 fluid can oxidize Si, producing a SiO x shell around the Si core. A SiO x /carbon multi-layer coating on Si is thus formed in a one-pot and single-step process, which is facile and easily scaled up for mass production. A control sample synthesized using a conventional wet-chemical route without SCCO 2 [37] is examined for comparison. The microstructure, crystallinity, chemical composition, and lithiation–delithiation behavior of various electrodes are systematically investigated. In addition, the effects of carbon precursors on the material and electrochemical characteristics are explored. The proposed SCCO 2 synthesis approach can fabricate high-capacity, high-power, and long-life Si@SiO x @C composite anodes for next-generation LIBs.【】【】

2. Results and Discussion

[3] H. Su, A.A. Barragan, L. Geng, D. Long, L. Ling, K.N. Bozhilov, L. Mangolini, J. Guo, Colloidal Synthesis of Silicon–Carbon Composite Material for Lithium-Ion Batteries, Angewandte Chemie - International Edition, 56 (2017) 10780-10785.

【硅被认为是有前途的负极材料，可替代石墨】

## 【硅被认为是有前途的负极材料，可替代石墨】

[3] H. Su, A.A. Barragan, L. Geng, D. Long, L. Ling, K.N. Bozhilov, L. Mangolini, J. Guo, Colloidal Synthesis of Silicon–Carbon Composite Material for Lithium-Ion Batteries, Angewandte Chemie - International Edition, 56 (2017) 10780-10785.

Silicon is recognized as the most promising anode material to replace or complement graphite in lithium-ion (Li-ion) batteries.[1]【硅的缺点】

## 【硅的缺点】

[3] H. Su, A.A. Barragan, L. Geng, D. Long, L. Ling, K.N. Bozhilov, L. Mangolini, J. Guo, Colloidal Synthesis of Silicon–Carbon Composite Material for Lithium-Ion Batteries, Angewandte Chemie - International Edition, 56 (2017) 10780-10785.

However, the large volume change associated with lithiation/delithiation of Si causes significant challenges to practical Si-based anodes.[2] 【虽然采用纳米化硅后，粉化可能不再是主要问题，但是频繁的体积变化会带来副作用】

## 【虽然采用纳米化硅后，粉化可能不再是主要问题，但是频繁的体积变化会带来副作用】

[3] H. Su, A.A. Barragan, L. Geng, D. Long, L. Ling, K.N. Bozhilov, L. Mangolini, J. Guo, Colloidal Synthesis of Silicon–Carbon Composite Material for Lithium-Ion Batteries, Angewandte Chemie - International Edition, 56 (2017) 10780-10785.

Although particle pulverization may no longer be the primary problem since the use of Si of nanosize or with nanofeatures,[3] repeated volume change can degrade the electrical contact and destabilize the solid delithiation.[2a]

【硅碳复合材料，举例，被报道有良好的性能】

## 【硅碳复合材料，举例，被报道有良好的性能】

[3] H. Su, A.A. Barragan, L. Geng, D. Long, L. Ling, K.N. Bozhilov, L. Mangolini, J. Guo, Colloidal Synthesis of Silicon–Carbon Composite Material for Lithium-Ion Batteries, Angewandte Chemie - International Edition, 56 (2017) 10780-10785.

To date, Si–C composites with various nanostructures including yolk–shell Si–C,[2a, 8] sandwich-structured Si–graphene oxide,[9] pomegranate-like Si–C,[6c] graphene or carbon coated silicon nanoparticles or nanowires[10] have demonstrated excellent electrochemical performance【将硅融入碳基体中可提高性能】

## 【将硅融入碳基体中可提高性能】

[3] H. Su, A.A. Barragan, L. Geng, D. Long, L. Ling, K.N. Bozhilov, L. Mangolini, J. Guo, Colloidal Synthesis of Silicon–Carbon Composite Material for Lithium-Ion Batteries, Angewandte Chemie - International Edition, 56 (2017) 10780-10785.

. Clearly, incorporating Si into a carbon matrix does indeed improve the structural and electrical integrity of the Si–C composites.[11] 【大多数合成硅碳复合材料方法面临的挑战，均匀分散困难，原因】

## 【大多数合成硅碳复合材料方法面临的挑战，均匀分散困难，原因】

[3] H. Su, A.A. Barragan, L. Geng, D. Long, L. Ling, K.N. Bozhilov, L. Mangolini, J. Guo, Colloidal Synthesis of Silicon–Carbon Composite Material for Lithium-Ion Batteries, Angewandte Chemie - International Edition, 56 (2017) 10780-10785.

However, most of the synthetic methods in use still face challenges in the uniform dispersal of the primary Si particles due to their poor solubility in any solvent.

【本文，溶胶凝胶方法合成碳包硅复合材料，有机溶剂中溶解硅纳米颗粒】

## 【本文，溶胶凝胶方法合成碳包硅复合材料，有机溶剂中溶解硅纳米颗粒】

[3] H. Su, A.A. Barragan, L. Geng, D. Long, L. Ling, K.N. Bozhilov, L. Mangolini, J. Guo, Colloidal Synthesis of Silicon–Carbon Composite Material for Lithium-Ion Batteries, Angewandte Chemie - International Edition, 56 (2017) 10780-10785.

Herein we report a unique colloidal method to synthesize Si@C composites using Si nanoparticles (SiNPs) that are soluble in organic solvents. Using as calable non-thermal plasma synthetic method developed by the Mangolini group,[12] we synthesized SiNPs with hydrogen-terminated surface and an average particle size of approximately 5nm with narrow size distribution (Figure S1 in the Supporting Information). Enabled by the surface H, dodecyl chains (C12H25)were tethered onto the SiNP surface upon reaction with dodecane. The obtained C12H25-SiNPs became readily soluble in many organic solvents. The Si content in C12H25-SiNPs is 86.5 wt% as measured with thermogravimetric analysis (TGA, Figure S2). To synthesize the SiNPs@C composite, C12H25-SiNPs were first dissolved in styrene, and the obtained solution was dispersed in the aqueous solution of hexadecyltrimethylammonium bromide by sonication to form astable emulsion. Free radical emulsion polymerization was then initiated to produce acolloid of SiNPs@polystyrene (SiNPs@PS) particles dispersed in water. The SiNPs@PS particles were subsequently coated with a layer of resorcinol–formaldehyde resin (RF) via a seeded sol–gel synthesis followed by carbonization to produce the SiNPs@C composite denoted as SiNPs@CPS.【】【】

The transmission electron microscopic (TEM) image, high-angle annular dark-field (HAADF) image, and the energy-dispersive X-ray spectroscopy (EDX) elemental mapping of the SiNPs@CPS are shown in Figure 1.

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

【发展高能量密度和长循环寿命的锂电池，应用】

## 【发展高能量密度和长循环寿命的锂电池，应用】

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

There is considerable demand to develop rechargeable lithium-ion batteries (LIBs) with high energy capacity and long cycle life for applications in portable electronic devices and electrical/hybrid vehicles. [1,2] 【硅被认为有前途，高容量】

## 【硅被认为有前途，高容量】

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

Si is recognized as one of the most promising anode candidates because of its high theoretical lithium-storage capacity (4200 mAhg @1 for Li 22 Si 5 ) and moderate Li-uptake potential (0.4 V vs. Li/Li + ). [3–6] 【硅的缺点】

## 【硅的缺点】

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

However, the serious volume variation (ca. 400%) of Si electrodes, coupled with poor conductivity, often cause electrode pulverization and active material loss, resulting in overall deterioration of battery performance. [7–11] 【将硅融入碳基体被认为是一个有效方法提升性能】

## 【将硅融入碳基体被认为是一个有效方法提升性能】

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

Incorporating Si into a carbon matrix is recognized as an effective method by which to improve structural and electrical integrity of Si- based electrodes for enhanced LIB performance. [12–14] 【然而，物理混合硅碳复合物的缺点】

## 【然而，物理混合硅碳复合物的缺点】

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

However, the Si/C composites prepared by physical mixing of the two components [15–18] generally results in weak interfacial interactions and uneven dispersion of the two components, which is adverse to electron transfer at interfaces and cycling stability of composite anodes.

【化学气相沉积方法描述，用于制备硅碳复合物，方法的局限】

## 【化学气相沉积方法描述，用于制备硅碳复合物，方法的局限】

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

Chemical vapor deposition (CVD) based on metal catalysts, where catalyst nanoparticles are dispersed on substrates as catalytic sites for in situ growth of Si nanowires with a vapor–liquid–solid (VLS) mechanism, [8,10,19–22] is a successful method by which Si can be uniformly anchored into the carbon matrix of Si/C composites. However, the introduction of metal catalysts to a carbon matrix is primarily achieved by physical sputtering or chemical deposition, and such methods involve complicated processes and fail to distribute metal catalysts uniformly. 【生长的硅没有约束，带来的问题】

## 【生长的硅没有约束，带来的问题】

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

Moreover, the catalytic growth of Si is usually carried out in an unconfined space where Si can be supplied without restriction, leading to poorly controlled growth of Si and the formation of long Si nano-wires (hundreds of nanometers–micrometers).【这一方法会产生大直径的硅纳米线，问题】

## 【这一方法会产生大直径的硅纳米线，问题】

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

In most cases, the catalysts employed are in the form of nanoparticles with relatively big diameters, which, according to the catalytic growth mechanism, [23] results in the generation of Si nano-wires with large diameters (>50 nm). In principle, Si nano-wires that are long and large in diameter are not conducive to electron transfer from Si to the current collector and also suffer from large volume changes upon cycling. 【因此，需要发展合适方法，构建具有合适结构的硅碳复合物】

## 【因此，需要发展合适方法，构建具有合适结构的硅碳复合物】

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Therefore, the development of a simple and effective metal catalytic strategy to construct Si/C composites with favorable structures for high-performance LIBs is highly desirable. [24,25]

【本文，硅纳米点嵌入碳网络】

## 【本文，硅纳米点嵌入碳网络】

[4] B. Chen, L. Zu, Y. Liu, R. Meng, Y. Feng, C. Peng, F. Zhu, T. Hao, J. Ru, Y. Wang, J. Yang, Space-Confined Atomic Clusters Catalyze Superassembly of Silicon Nanodots within Carbon Frameworks for Use in Lithium-Ion Batteries, ANGEWANDTE CHEMIE-INTERNATIONAL EDITION, 59 (2020) 3137-3142.

Herein, we propose a novel strategy for the superassembly of a framework bearing Si nanodots (Si NDs) embedded in a carbon (Si NDs%C) framework for application in high-performance LIBs. The strategy was realized by copyrolysis of triphenyltin hydride (TPT) and diphenylsilane (DPS), during which Sn atomic clusters were produced from TPT pyrolysis and served as the catalyst for the pyrolysis of DPS and the catalytic growth of Si. The strategy breaks the Si @ C bonds of DPS and permits graphitization of the carbon framework at lower temperatures, thereby avoiding the generation of SiC. Moreover, the employment of Sn atom clusters can reduce the dimensions of Si product to form Si NDs as a consequence of the reported size-dependent correlation between the catalysts and the Si product. [26] The prepared Si%C composite made of Si NDs dispersed uniformly in a hybrid a/g carbon framework with a hierarchical pore structure (micropores of ca. 1.6 nm in diameter, large mesopores of 12.0–50.0 nm in diameter, and macropores over 50 nm in diameter is beneficial for stable, fast, and high-capacity Li storage. Significantly, the Si NDs%C framework demonstrates low lithiation stress and an outstanding mechanical stability, revealed by finite element calculations. [27–30] As a result, the Si%C framework anode exhibited high reversible capacity, long cycling stability, and superior rate capability. Moreover, a series of full batteries containing Si%C framework anodes and commercial cathodes exhibited remarkable performance compared to commercial full cells.【】【】

The strategy for preparation of the Si NDs%C framework is simple and straightforward, governed by stepwise elevation of temperature from 25 to 800℃ (Figures 1a,b).

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

1. Introduction

【锂电池是最快发展的储能器件，应用】

## 【锂电池是最快发展的储能器件，应用】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

Nowadays, lithium-ion batteries (LIB) are the fastest developing energy storage systems, and have widespread application in mobile phones, laptops, electric vehicles, etc. [1–3]. 【石墨材料的问题】

## 【石墨材料的问题】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

However, the commercial graphite anode material has a limited theoretical capacity of 372 mAh g−1, which cannot meet the energy density requirements in certain applications [4,5]. 【寻找高能量密度的材料】

## 【寻找高能量密度的材料】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

Various studies have been carried out to find new high energy density materials, such as Sn- [6,7] or Si-based anodes [8–10], and transition metal oxides [11,12]. 【硅被认为有前途，高容量】

## 【硅被认为有前途，高容量】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

Among these, Si-based material is considered as a promising candidate owing to its high theoretical capacity of 4200 mAh g−1 [13,14].【硅的挑战，问题】

## 【硅的挑战，问题】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

At present, the main challenge is the enormous volumetric change of the Si during the cycle process, leading to structural failure and pulverization [15,16], and the low electronic conductivity of silicon also induces severe irreversible capacity loss.

【目前工作聚焦在发展新的硅基结构解决体积变化问题，举例】

## 【目前工作聚焦在发展新的硅基结构解决体积变化问题，举例】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

At present, research has mainly focused on finding new Si-based structures to solve the problems of the volumetric changes. The utility of nanoparticles, nanosheets, nanohollows, nanowires, and nano-core-shells has been studied as well [17–21]. 【硅碳复合物被认为是有效结构，硅和碳所起的作用，对SEI的作用】

## 【硅碳复合物被认为是有效结构，硅和碳所起的作用，对SEI的作用】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

Among them, silicon/carbon composites are regarded as effective structures to alleviate the volumetric changes of the silicon. In them, the silicon acts as the active material to provide lithium storage capacity, and the carbon material acts as a buffer to resist the volumetric changes of silicon during the cycle process. In addition, the drastic capacity fading of the Si-based anode corresponds to the solid electrolyte interphase (SEI) films repeatedly formed during the cycles. It has been proved that the surrounding carbon material avoids direct contact between the silicon and the electrolyte, preventing repeated SEI formation [22–24].

【文献，纳米硅被碳包裹，固定在石墨烯上。三明治结构纳米硅/空心石墨纤维/碳涂层】

## 【文献，纳米硅被碳包裹，固定在石墨烯上。三明治结构纳米硅/空心石墨纤维/碳涂层】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

To develop a high-performance anode material, Park et al. [25] synthesized a Si/carbon/graphite composite, and the Si nanoparticles were encapsulated by the carbonized coal-tar pitch and attached to the surface of graphite; the amorphous carbon layer avoided the direct contact between the Si nanoparticles and electrolyte and alleviated the volumetric changes of the Si nanoparticles during the cycles. The specific capacity of the composite was 712 mAh g−1 at a current density of 130 mA g−1, with a capacity retention of 80% after 100 cycles [25]. Wang et al. [26] proposed a sandwich-structured silicon nanoparticle/hollow graphite fiber/carbon-coated composite. In this work, the volume expansion of the Si nanoparticles was effectively controlled by the new sandwich-structure, and the specific capacity of the composite was maintained at 910.1 mA h g−1 after the 50th cycle.

【大部分硅碳复合材料是嵌入结构，通过高温热解制备】

## 【大部分硅碳复合材料是嵌入结构，通过高温热解制备】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

Most silicon-carbon nanocomposites are embedded structures and are prepared by high-temperature pyrolysis of carbon materials, in which the Si nanoparticles are encapsulated by amorphous carbon layers to resists the volumetric change and enhance electrode conductivity. 【然而，碳涂层的厚度不均匀，硅纳米颗粒严重团聚，影响性能进一步提升】

## 【然而，碳涂层的厚度不均匀，硅纳米颗粒严重团聚，影响性能进一步提升】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

However, the further improvement of the electrochemical performance is limited by the non-uniform thickness of carbon coating and serious agglomeration of Si nanoparticles [27,28]. 【方法缺点，难以工业化】

## 【方法缺点，难以工业化】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

Moreover, the complex process, reagent pollution, and high cost make it difficult for industrialization.

【等离子技术被用来制备硅碳复合材料】

## 【等离子技术被用来制备硅碳复合材料】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

Recently, plasma technology has been introduced to prepare silicon-carbon nanocomposites due to the rapid synthesis process without expensive materials or toxic reagents.【传统等离子技术缺点，无法大量制备】

## 【传统等离子技术缺点，无法大量制备】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

However, the traditional plasma technology for the preparation of silicon-carbon nanocomposite is inadequate to achieve mass production. 【本文，等离子技术中引入磁场】

## 【本文，等离子技术中引入磁场】

[5] F. Wang, M. Gao, R. Hong, X. Lu, Magnetoelectric Plasma Preparation of Silicon-Carbon Nanocomposite as Anode Material for Lithium Ion Batteries, APPLIED SCIENCES-BASEL, 10 (2020).

Herein we proposed a novel plasma process with a magnetron plasma generator, in which the addition of magnetic force increases the volume and density of the plasma reaction region and reduces energy consumption. Thus, it lays a good foundation for mass production. The silicon-carbon nanocomposite was prepared by one-step magnetoelectric plasma pyrolysis of the mixture of methane, silane, and hydrogen, in which methane acts as the carbon source, silane as the silicon source, hydrogen as the reducing gas, and argon as the carrier gas. Finally, a new composite structure was obtained in which the evenly dispersed Si nanoparticles were adsorbed on the surface of tremelliform carbon nanosheets, effectively preventing the aggregation and volume expansion of the Si nanoparticles and showing excellent electrochemical performance.【】

【】2. Experimental

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

1．Introduction

【锂电池优点，占据主导】

## 【锂电池优点，占据主导】

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

Lithium-ion batteries (LIBs) have long cycling life, high energy density, and high power capability, thus dominating the current energy storage market.[1,2]【下一代电池，满足快速发展的储能需求，石墨容量有限】

## 【下一代电池，满足快速发展的储能需求，石墨容量有限】

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

Significant research efforts have been devoted to development of next-generation rechargeable batteries to meet the demand of the rapidly growing energy storage market,[3–7]as the graphite anodes in current LIBs have a rather limited theoretical capacity of 372 mAh/g.【硅的优点】

## 【硅的优点】

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

Silicon materials, with a nearly 10-fold higher theoretical capacity of 3579 mAh/g, relatively low discharge potential (<0.5 V vs. Li/Li+ considered as promising anode materials for LIBs.[8–10]), and abundant resource, have been 怕【硅的缺点】

## 【硅的缺点】

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

However, a major obstacle for silicon anodes is the dramatic volume change of silicon that occurs during lithiation and de-lithiation, which undermines the electrode’s integrity and disrupts the solid electrolyte interface (SEI). The breakdown of the SEI layers during cycling results in a range of problems, such as low Coulombic efficiency (CE), quick capacity fading and poor cycling stability.[11,12]

【为克服硅的问题，提出许多策略，举例】

## 【为克服硅的问题，提出许多策略，举例】

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

To address these challenges, many strategies have been proposed, including nanostructured silicon particles, coating of buffer layers, conductive binders, etc. 【设计出各类纳米硅，可缓解体积膨胀，但还是存在其他缺点】

## 【设计出各类纳米硅，可缓解体积膨胀，但还是存在其他缺点】

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

Recently, various delicate nanostructures have been designed and fabricated, such as nanotubes,[13,14] structures.[17–19] hollow sphere,[15,16] and yolk-shell. These void-containing nanostructures can effectively alleviate the large volume change and improve the cycling performance, but they also suffer from additional demerits such as low tap density, large surface area, complicated synthesis process, and relatively poor electrical properties due to the high inter-particle resistance.[20]

【导电聚合物和碳涂层对提升硅性能有利，文献举例】

## 【导电聚合物和碳涂层对提升硅性能有利，文献举例】

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

Conductive polymer and carbon coating have proven to be a practical approach to enhance the electrochemical performance of the silicon materials for LIBs.[21] layers not only can improve the electrical conductivity, but also can serve as an electrolyte layer on the surface of silicon to alleviate the side reactions and minimize the volume changes. For example, Wu et al.[22] Conductive coating reported a stable Si/C anode material by in-situ polymerization of polyaniline (PANi) to conformally coat silicon nanoparticles with conducting polymer, delivering a capacity of 550 mAh/g after 5000 cycles at 6 A/g with a mass loading of 0.2–0.3 mg/cm2. The average CE of the Si/C electrode from the 2nd to 5,000th cycle was 99.8%. Lee et al.[23] investigated the pyrolysis of polyacrylonitrile (PAN) at the temperature of 300–500°C to attain the cyclization of PAN, which was achieved without carbonization while maintaining PAN’s polymeric properties. This Si/C material with homogeneous coating layer on the surface of Si nanoparticles could provide superior performance of ~1500 mAh/g after 150 cycles with a CE approaching 100% at a current rate of 0.1 C. Choi et al.[18] used sucrose as a precursor via spray drying process to attain Si/C composite particles in which silicon nanoparticles (~ 70 nm) were embedded in porous carbon particles. The composite electrodes exhibited excellent electrochemical performance of 1243 mAh/g after 150 cycles at 4 A/g with a mass loading of 1 mg/cm2. In the cycling range from first cycle to fifth cycle, the average CE was 98.5%. 【然而，需要更高的库伦效率和循环稳定性】

## 【然而，需要更高的库伦效率和循环稳定性】

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

However, higher Coulombic efficiency and long-term cycling stability are still needed for these silicon-based anodes for practical applications.

【本文，规则的碳涂层包裹硅纳米颗粒】

## 【本文，规则的碳涂层包裹硅纳米颗粒】

[6] S. Zhou, C. Fang, X. Song, G. Liu, Highly Ordered Carbon Coating Prepared with Polyvinylidene Chloride Precursor for High-Performance Silicon Anodes in Lithium-Ion Batteries, BATTERIES & SUPERCAPS, 4 (2021) 240-247.

Here, we introduce polyvinylidene chloride (PVDC), a polymer containing stoichiometric ratio of sacrificial H and Cl elements with the formula of (C2H2Cl2 )n , as precursor to attain ordered carbon coatings via a simple yet efficient sintering strategy to encapsulate the silicon nanoparticles. The Si/C composites with ordered carbon coatings show excellent electrochemical performance. The composition and preparation conditions of Si/C composites are examined systematically to identify the optimum protocols. The benefit of ordered and compact carbon coatings for silicon anode materials are demonstrated.【】

【】2.Results and Discussion

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

1 INTRODUCTION

【锂电池主导市场，优点】

## 【锂电池主导市场，优点】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Lithium‐ion batteries (LIBs) have been dominant in the market for powering the portable electronic devices since they were first commercialized, due to their desirable energy and power densities. 【需要提高锂电池能量密度】

## 【需要提高锂电池能量密度】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

To meet the market demand for lighter batteries with longer service life, the energy densities of LIBs have to be continually increased. The recent development of electrical vehicles and their widespread use also call for LIBs with high energy. Therefore, there is always constant pressure on the relevant academic and industry communities to improve the energy density of LIBs.1-3 【然而，传统使用碳负极的锂电池接近理论能量密度】

## 【然而，传统使用碳负极的锂电池接近理论能量密度】

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Nevertheless, the conventional LIBs using carbonaceous anode and lithium transition‐metal oxide cathode are approaching their theoretical energy density.

【锂电池的能量密度与电极材料的容量有关】

## 【锂电池的能量密度与电极材料的容量有关】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

The energy density of LIBs is closely related to the electrode materials and their specific capacity. 【相对于正极，负极材料提升空间更大，举例】

## 【相对于正极，负极材料提升空间更大，举例】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Compared to the cathode materials, there is more room in anode materials to increase the capacity. For example, the gravimetric capacity of conventional LiCoO2 cathode is around 165 mAh/g, which is 0.2 to 1 times lower than that of Ni‐rich or Li‐rich cathodes,4-6 while the gravimetric capacity of Si anode is 10 times higher than that of conventional graphite anode (372 mAh/g). 【使用硅可限制提升能量密度】

## 【使用硅可限制提升能量密度】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

With the same cathode material, replacing graphite with Si can significantly improve the energy density of LIBs. 【因此，硅被认为最有前途的，优点】

## 【因此，硅被认为最有前途的，优点】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Therefore, Si has been considered as one of the most‐promising next‐generation anodes towards high‐energy LIBs because of its high theoretical capacity (3572 mAh/g), low working voltage (~0.2 V vs Li/Li+), and abundance in the earth’s crust.7【硅的缺点】

## 【硅的缺点】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

However, unlike the intercalation‐type anodes (eg, graphite), the alloying/dealloying reaction of Si with Li induces huge volume changes (>300%). Such huge volume changes during electrochemical cycling will lead to repeated cracking and pulverization of Si, and hence the disintegration and fracturing of the Si electrode, accompanied by electrical isolation. The repeated cracking and pulverization will also lead to the continual breaking up of the solid electrolyte interphase (SEI) layer and the explosion of new surface, which will quickly consume the electrolyte and Li ions.【单独使用硅负极的缺陷】

## 【单独使用硅负极的缺陷】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Therefore, the use of sole Si anode suffers from extremely fast capacity decay and low coulombic efficiency (CE) as a result of the severe volume changes and unstable SEI films. 【使用纳米材料，发展复合物以提升循环性能】

## 【使用纳米材料，发展复合物以提升循环性能】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Design strategies for advanced materials, such as employing unique nanostructures (nanowire, nanotube, core/shell, yolk shell, nanoporous materials, etc) and forming composites with electrochemically inactive/less active materials, such as carbon, conductive polymer, and so forth, have been applied as academic approaches to significantly improve the cycle life.8-10 【然而，体积能量密度和负载量太低，无法满足工业需求】

## 【然而，体积能量密度和负载量太低，无法满足工业需求】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Nevertheless, the volumetric energy density of these materials and the areal mass loading on electrodes are generally too low for industrial implementation. 【高性能负极的商业指标，容量，循环性，库伦效率，负载量，膨胀率】

## 【高性能负极的商业指标，容量，循环性，库伦效率，负载量，膨胀率】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

The commercial goal of achieving high‐performance anodes to replace the existing commercial graphite materials in the near future, involves reaching a specific capacity of 500 mAh/g or higher with a capacity retention of 80% after 500 cycles, while the initial CE and average CE should exceed 90% and 99.8%, respectively (Figure 1). Accordingly, the pressing density should reach ~1.65 g/cm3, and electrode swelling should be restricted to ~10%.11

【硅和石墨一起使用】

## 【硅和石墨一起使用】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Recently, the co‐utilization of Si and graphite has emerged as the most practical anode for high‐energy LIBs. 【石墨的优点】

## 【石墨的优点】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Graphite is a commercial anode with low cost, high CE, excellent cycle life, good mechanical flexibility, only small volume change, and high electrical conductivity.【硅中加石墨的优势】

## 【硅中加石墨的优势】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

The addition of graphite into Si can buffer the volume change, increase the electric conductivity, and achieve high specific, areal and volumetric capacities at the same time. 【硅和石墨一起使用在工业生产方面的优势】

## 【硅和石墨一起使用在工业生产方面的优势】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Moreover, the co‐utilization of Si and graphite can use the same commercial production lines, translating into high manufacturability and minimal investment. Therefore, the co‐utilization hybridizes two distinct anodes on the materials level into a single composite, retaining the advantages while circumventing the disadvantages of both, and can secure its success in the anode market. 【但是，将硅和石墨整合还是有困难，原因】

## 【但是，将硅和石墨整合还是有困难，原因】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

However, it is still a challenge to integrate the Si and graphite into a single system or composite to obtain the desired performance, since both of these materials are significantly different in terms of their physical and chemical properties. 【研究发展硅石墨碳复合物】

## 【研究发展硅石墨碳复合物】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

Growing research efforts have been devoted to improving and achieving high electrochemical performance with the design and development of Si/G/C composite.【碳添加剂起到整合硅和石墨的作用，优点】

## 【碳添加剂起到整合硅和石墨的作用，优点】

[7] J. Wu, Y. Cao, H. Zhao, J. Mao, Z. Guo, The critical role of carbon in marrying silicon and graphite anodes for high-energy lithium-ion batteries, CARBON ENERGY, 1 (2019) 57-76.

The carbon additives play important roles in integrating the Si and graphite from many aspects, such as increasing the interface bonding between the graphite and Si, alleviating the volume variations of Si, and improving the interfacial chemistry, conductivity, and mechanical integration. 【】【】This review attempts to highlight the critical role of carbon additives, as well as the associated synthesis methods and structures for achieving high‐performance Si/G/C composites for developing high energy‐density LIBs, and aims to provide possible insights for the future development of practical Si/G/C anodes.

2． CHALLENGE OF THE INTEGRATION OF SILICON AND GRAPHITE ANODES FOR HIGH‐ENERGY LITHIUM‐ION BATTERIES

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

1. Introduction

【硅的优点。有吸引力的锂电池负极】

## 【硅的优点。有吸引力的锂电池负极】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

It has been known Silicon (Si) has many advantages such as high theoretical capacity (4200 mAh/g),non-toxic, low cost, and abundant reserve [1–6]. Therefore, Si is the one of the most attractive materials as an anode for lithium ion battery. 【硅的问题】

## 【硅的问题】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

Two main problems should be solved for Si to be used as an anode commercially, which are low electrical conductivity and significant volume expansion (~300%) during Li insertion/extraction [7–12].【为提升导电性，硅表面包覆碳材料】

## 【为提升导电性，硅表面包覆碳材料】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

In order to enhance electronic conductivity, many researchers have been studied on several of Si-based electrodes. Carbon materials, including artificial graphite, natural graphite, and hard carbon are widely applied for Si coating and the mixture of high carbon Si/carbon composite was synthesized [13–15]. 【循环性有限。原因。】

## 【循环性有限。原因。】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

However, it was shown a limitation in the number of times the cycle test. It is because of the lack of enough change in volume of the internal volume of Si. 【核壳结构制备方法，可提升性能。文献。】

## 【核壳结构制备方法，可提升性能。文献。】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

Core–shell structural Si/C composites prepared by the coating of polymer delivered outstanding electrochemical performance, such as good rate capability and a long cycling life, as anode for LIBs [6,13–15]. Yi’s group has synthesized a C-Si and C-Si@G and shown the performance as an anode for LIB. It was clearly seen that the Si/C could only deliver ~617 mAh/g at 0.5 A/g after 100 cycles. The charge specific capacity of C-Si@G at ~1987 mAh/g under the same rates. Other researchers have reported that the specific capacity for porous Si/C anode is 600~750 mAh/g for LIB after 100 cycles [2,4,13,14].【碳可提升硅碳复合物导电性】

## 【碳可提升硅碳复合物导电性】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

It is confirmed that carbon materials in the silicon–carbon composite can improve the electronic conductivity of composite. 【但是，碳没法缓冲巨大的体积膨胀】

## 【但是，碳没法缓冲巨大的体积膨胀】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

However, it is not enough for carbon to play as a role of buffer the huge volume expansion caused by the processes of intercalating lithium ions in silicon.

【探索新的碳，提升循环稳定性和电化学性能。文献举例。】

## 【探索新的碳，提升循环稳定性和电化学性能。文献举例。】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

New types of carbon have been explored to improve the cycle ability and the electrochemical performance due to their high surface area and good electrical conductivity. Many researchers have studied carbon nanomaterials, such as carbon nanofiber (CNF), a novel one-dimensional (1D) carbon nanomaterial, graphene, and carbon nanotubes (CNTs) [16,17].【尤其是碳纳米纤维】

## 【尤其是碳纳米纤维】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

Especially, several types of CNF have been manufactured fromdifferent sources, which also demonstrated good electrochemical performance [18,19]. 【多数碳纳米纤维来自非可再生的，限制大规模应用】

## 【多数碳纳米纤维来自非可再生的，限制大规模应用】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

However, most of the CNFs are from non-renewable precursors consequently limiting their large-scale application. 【因此需要寻找低成本，可再生资源去生产碳纳米纤维】

## 【因此需要寻找低成本，可再生资源去生产碳纳米纤维】

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Therefore, finding low-cost and renewable resources to manufacturing high performance CNF materials is a pressing problem for every researcher. 【需要考虑降低化石资源消耗。生物质材料。优点。】

## 【需要考虑降低化石资源消耗。生物质材料。优点。】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

Also, reducing the consumption of fossil fuel resources should be considered. Recently, biomass has become more attractive in preparation of useful carbonaceous materials as a bountiful renewable resource in nature. Biomass sources such as catkin, lignin, alginate, and wood sawdust have demonstrated excellent electrochemical performance as electrode materials for energy applications [20–25]. Some biomass materials such as pollen, lignin, chitin, cellulose, and catkin have been studied on batteries and supercapacitors [26–30]. They have many advantages of inexpensive, abundant natural resources, renewable, and huge specific surface area. Besides, they could directly serve as both a template with porous structure for LIB and a precursor for the preparation of carbon microfibers by simple carbonization [31,32]. 【生物质没有被广泛应用于锂电池负极。相关文献】

## 【生物质没有被广泛应用于锂电池负极。相关文献】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

Biomass is not widely applied as an anode for LIB. These day, Chen’s group has reported that HPC@Si@GS shows high rate capability that the electrode delivers a high and stable capacity of 435 mAh/g at a high current density of 1.6 A/g. When the electrode is measured at original current density of 0.1 A/g, it can deliver a capacity of 1003 mAh/g after the subsequent 100 cycles [1]. Zhu’s group has investigated that the Si@C@CNT electrode exhibits a specific capacity of 620 mAh/g at extremely high charging rate of 7.50 A/g. Furthermore, the Si@C@CNT-G electrode exhibits the capacity of 420 mAh/g at the rate of 0.2 C after 100 cycles [2]. MW is the one of the biomass sources and biomass has been studied as a supercapacitor. Recently, there are research papers that biomass has good conductivity, and there are also research publications applied to sulfur batteries. However, biomass is not yet widely used in LIB.

【本文，采用MW生物质制备硅碳复合材料】

## 【本文，采用MW生物质制备硅碳复合材料】

[8] E.H. Chung, J.P. Kim, H.G. Kim, J.-M. Chung, S.-J. Lee, J.-S. Bae, E.D. Jeong, The Synthesis and Electrochemical Performance of Si Composite with Hollow Carbon Microtubes by the Carbonization of Milkweed from Nature as Anode Template for Lithium Ion Batteries, ENERGIES, 13 (2020).

In this study, we used MW as a template with carbon porous structure and a precursor to prepare HCMT by simple carbonization. HCMT can provide large surface area to load Si particles and buffer space to cushion the enough volume expansion for Si. Therefore, Si was accommodated to MW and calcinated with low-cost carbon sources such as Chitosan (CTS), Agarose, and polyvinylidene fluoride (PVDF) in N2 flow. Three different Si composites, HCMT@Si@C1, HCMT@Si@C1@C2 and HCMT@Si@C1@C3, were synthesized and discussed about electrochemical properties. Characterization for samples used after 100 cycle is in progress. In the future research, it will be the subject of future publication.

2. Materials and Methods

[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

1. Introduction

【锂电池被广泛使用，但需要进一步提升能量和功率密度】

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Lithium-ion batteries (LIBs) are widely used in many applications but there is an increasing demand for higher energy and power density.【能量和功率密度取决于用什么样的正极负极材料】

## 【能量和功率密度取决于用什么样的正极负极材料】

[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

These characteristics are mainly determined by which cathode and anode materials are used. 【硅的优点】

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[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

Among various anode materials, silicon has been investigated as one of the most promising alternatives to conventional graphite anodes because of its high theoretical specific capacity (about 4200 mAh/g) [1].

【硅的缺点】

## 【硅的缺点】

[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

However, the practical application of Si as an anode material is currently impeded by its large volume change (~300%) during lithiation and delithiation, resulting in capacity fading and a limited lifecycle. Moreover, the rate capability of Si electrodes is limited by the low electronic conductivity of Si and low lithium diffusion rates in Si [ 2 – 4 ]. 【大量工作提升结构稳定性和电学性能】

## 【大量工作提升结构稳定性和电学性能】

[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

Great efforts have been devoted to achieving improved structural stability and electrical performance of Si based anodes [ 5 – 10 ].【纳米硅碳复合物有优势】

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[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

Silicon-carbon composites using nanoscale Si have been recognized as a favorable approach to solve the abovementioned problems. 【硅的缺点可通过降低硅的尺寸实现】

## 【硅的缺点可通过降低硅的尺寸实现】

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Numerous studies have demonstrated that the drawbacks of Si as an anode material could be mitigated by reducing the size of the Si particle.【碳基体中分散纳米硅的优势】

## 【碳基体中分散纳米硅的优势】

[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

The dispersion of nanoscale Si in a carbon matrix has been a favorable approach in synthesizing Si-based anode materials for LIBs because it improves the performance of Si through avoiding the direct contact with electrolytes [ 11 , 12 ]. On the one hand, the dispersion of nanosized Si particles in a carbon matrix is known to be challenging since these nanosized particles are prone to forming agglomerations [ 13 , 14 ]. On the other hand, it is anticipated that the problem in dispersion of nanosized Si particles will be overcome by synthesizing a Si-carbon black (CB) carbon composite [15].

【石墨烯有前景，用于硅碳复合物，原因】

## 【石墨烯有前景，用于硅碳复合物，原因】

[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

Graphene(or graphite nanosheet) is regarded as a promising carbonaceous material for silicon–carbon composites due to its unique properties, in which graphene can provide an electrically conductive network and structural support for accommodating the mechanical stress, thus enhance the electrochemical performance of Si-based composites [ 16 , 17 ]. It also appears that the graphene could act as a protective barrier, inhibiting the Si particles from being directly exposed to the electrolyte [18].

【出于什么原因，包含一定比例硅的复合电极被研究】

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[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

To improve energy densities and simultaneously alleviate mechanical instabilities of electrodes, composite electrodes containing a small portion of Si have been increasingly researched [ 19 , 20 ].【然而很少研究关注提升倍率性能】

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However, little attention has been paid to an approach for improving their rate performance.

【本文，组合了以上提到的几方面，核壳结构硬碳/硅碳复合物】

## 【本文，组合了以上提到的几方面，核壳结构硬碳/硅碳复合物】

[9] J.-C. Kim, K.-J. Kim, S.-M. Lee, Preparation and Characterization of Core-Shell Structure Hard Carbon/Si-Carbon Composites with Multiple Shell Structures as Anode Materials for Lithium-Ion Batteries, ENERGIES, 14 (2021).

In this study, by combining several of the abovementioned aspects, a unique core-shell structure hard carbon/Si-carbon composites with multiple shell structures were synthesized to be applied as anode materials for LIBs. The concept was to improve the power density and energy efficiency by adopting a hard carbon with a high-rate capability as a core material and distributing Si nanoparticles in the shell layer. Thereby, it was expected that the utilization of active electrode materials would increase. In particular, we investigated the effect of the shell structure in the composite material on its electrochemical performance as an anode material for LIBs, in which we employed three types of shell structure: Si-carbon, Si-carbon black-carbon and Si-carbon black-carbon/graphite nanosheet. The resultant composite materials are denoted as Hard carbon/Si-carbon composite, Hard carbon/Si-carbon black-carbon composite and Hard carbon/Si-carbon black-carbon/graphite nano-sheet composite, respectively.【】【】

[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

1. Introduction

【锂电池是一些领域电气化的关键】

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Lithium-ion batteries (LIBs) are the key to underpinning the electrification of modern transportation and using intermittent renewable energies such as solar and wind. [1–3] 【需要提高锂电池的能量密度】

## 【需要提高锂电池的能量密度】

[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

To fulfill the requirements of batteries for electric vehicles and grid energy storage, it is necessary to increase the energy densities of LIBs. [4,5] 【使用高容量电极材料是有效方法】

## 【使用高容量电极材料是有效方法】

[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

The use of high-capacity electrode materials is one of the most effective ways to improve the energy densities of LIBs. 【硅的优点】

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[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

Si is an earth-abundant element and of a theoretical gravimetrical capacity of 3579 mAh g-1 ,which is ten-fold of the commercially used graphite anode (372 mAh g-1 ). [6] 【硅的缺点】

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[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

This means that one Si atom can accommodate around four Li + to form Li 15 Si 4 at a fully charged state, resulting in a vast volume expansion (~300%) of Si. [7] During discharge, Li + ions will be extracted out from the Si and subsequently incur a significant volume shrinkage thus causing the collapse of Si framework. [8] After repeated charge and discharge cycles, pulverization of the Si anode becomes severe along with the continual formation of solid electrolyte interface (SEI) layers, so that the active electrode materials gradually lose contact from current collectors and consequently the capacity of the electrode decays. [9]【因此，高容量硅负极发展的焦点是降低体积变化引起的性能衰减】

## 【因此，高容量硅负极发展的焦点是降低体积变化引起的性能衰减】

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Therefore, the central focus of developing the high-capacity Si anode is to minimize the capacity fading caused by the huge volume change.

【许多策略被提出提升硅负极寿命，举例】

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To increase the life span of the Si anode, various approaches have been employed such as

designing nanostructured Si materials, [10–12] preparing Si-C hybrid materials, [13–18] using

buffering matrixes, [19,20] and engineering electrolytes and binders. [21–23] 【纳米硅降低应力，碳层的作用】

## 【纳米硅降低应力，碳层的作用】

[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

It has been proven that the nanostructured Si can reduce the mechanical stress caused by the volume expansion, and a carbon layer on Si could remarkably stabilize the SEI layers and avoid the disconnection of the Si from the current collector. [24,25] 【传统制备纳米硅的方法举例。制备方法的问题。】

## 【传统制备纳米硅的方法举例。制备方法的问题。】

[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

The conventional approaches preparing nanostructured Si include physical vapor deposition (PVD), [26] chemical vapor deposition (CVD), [27] metallothermic reduction, [14,28,29] ball-milling, [30] and chemical etching. [31] In these methods, Si feedstocks are mostly organic Si compounds and Si halides, which require stringent synthesis conditions resulting in high energy consumption and cost. 【硅的储量，SiO2是最常见的硅形式。因此，直接将SiO2转化为纳米硅的优势】

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As we know, Si is the second most abundant element in the earth’s crust (26.4% by mass), and silica (SiO 2 ) is the most common natural Si resource. Thus, direct converting silica to nano-Si is of great interests due to the short process, low energy consumption, low cost, and sustainable raw materials.

【除了镁热还原，熔盐中直接电化学还原SiO2的优势。文献举例。问题。针对这一问题，文献报道的解决方案】

## 【除了镁热还原，熔盐中直接电化学还原SiO2的优势。文献举例。问题。针对这一问题，文献报道的解决方案】

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In addition to the magnesiothermic reduction of solid SiO2, [29] direct electrochemical reduction of solid oxides in molten salts offers a straightforward way to extract metals/alloys because the molten salts possess high ionic conductivity and wide electrochemical windows. [32–34] Si has been successfully prepared by electrochemical reduction of solid silica or soluble silicate in molten CaCl 2 -based molten salts. [35–39]However, the use of CaCl 2 -based molten salts could generate CO 3 2? At the graphite anode and then the CO 3 2? diffuses to the cathode and is then reduced to C, which decreases the coulombic efficiency of the electrolysis. [40–43] Jin et al. reported that the use of MgCl 2 -based molten salt can solve the parasite reactions caused by CO 3 2? in molten CaCl 2 - based molten salts. [44,45] Mai et al. [46] reported that Si nanowires can be prepared by electrode position in molten NaCl-KCl-MgCl 2 -CaSiO 3 .Yin et al. [47] tested the Li storage properties of the electrolytic Si film obtained in molten CaCl 2 -nano SiO 2 , and the capacity retention of the Si film is only 50% after 40 cycles. Recently, Xiao et al. Electrodeposited Si nanowires on a carbon cloth and revealed that the surface oxide layer significantly plagued the performance of the electrolytic Si. [48] 【很少研究报道固体二氧化硅中电还原硅，用于锂电池。原因以及因此带来的问题。】

## 【很少研究报道固体二氧化硅中电还原硅，用于锂电池。原因以及因此带来的问题。】

[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

However, there are few studies reporting the use of the electrolytic Si from the electrochemical reduction of solid SiO2 for the anode in LIBs. Since the properties of the Si are highly related to its structures such as morphologies, particle size, and porosities, [46,49,50] the electrochemical reduction of solid SiO2 involves both solid reduction and dissolution mechanism, [51] so that the morphologies of the electrolytic Si are difficult to control.

【二氧化硅包裹碳层是制备含孔洞的硅碳复合物的有效策略，原因】

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Precoating a carbon layer on SiO 2 (SiO 2 @C) could be an effective strategy to prepare the Si@viod@C because the voids can be created by the volume shrinkage from converting SiO 2 to Si. 【但是用熔盐电解制备硅碳的问题。文献解决方案】

## 【但是用熔盐电解制备硅碳的问题。文献解决方案】

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Unfortunately, the electrolysis of SiO 2 @C in molten CaCl 2 generates SiC but not the Si@C. [47,52–56] Likewise, the magnesiothermic reduction of SiO 2 @C generates SiC or Si@SiC but not the Si@C. [57,58] Xiao et al. designed a Si@C@Si structure by the electrolysis of SiO 2 @ polydopamine (PDA) in molten CaCl 2 -NaCl at 800 °C and revealed that the SiC could be retarded by the generated gas of thermal decomposition of the PDA. [48] The obtained Si@C@Si delivered a capacity of 904 mAh g ?1 at 500 mA g ?1 after 100 cycles. Yoo et al. [59] reported that the interface of C/SiO2 played a determining role in manipulating the products of the magnesiothermic reduction of the mixture of SiO 2 and C. Therefore, elimination of SiC by engineering the interface of SiO 2 /C in the process of the (electro-) reduction of SiO 2 @C is the key to preparing Si@C (Figure 1).

【相比镁热还原，电化学还原的优势。】

## 【相比镁热还原，电化学还原的优势。】

[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

Note that Si@C could be prepared by magnesiothermic reduction of SiO 2 @void@C, but the reduction of the SiO 2 @C will concomitantly generate SiC. [59,60] Unlike the magnesiothermic reduction, the electrochemical reduction process could be engineered by alternating the electrochemical potentials and electrolytes. To suppress the formation of SiC by the electrolysis of SiO 2 @C, the MgCl 2 -based mol-ten salts were applied. 【本文，SiO 2 @C 】

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[10] X. Zhou, H. Xie, X. He, Z. Zhao, Q. Ma, M. Cai, H. Yin, Annihilating the Formation of Silicon Carbide: Molten Salt Electrolysis of Carbon–Silica Composite to Prepare the Carbon–Silicon Hybrid for Lithium-Ion Battery Anode, Energy and Environmental Materials, 3 (2020) 166-176.

In this work, the SiO 2 @C was prepared by pyrolyzing the mixture of silica and sucrose. Then, the Si@C was prepared by the electrochemical reduction of SiO 2 @C without generation of SiC in the Mg-based molten salts, and the electrochemical performances of the electrolytic C-Si composite were studied as the anode for LIBs.【】【】

2. Results and Discussion

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

1. Introduction

【硅在锂电池中有前途】

## 【硅在锂电池中有前途】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

Silicon (Si)-based anode materials as a glaring star among the all currently reported anode materials, and it has attracted more and more attention in the industry of LIBs. 【石墨的问题】

## 【石墨的问题】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

Especially, nowadays commercialized graphite anodes are hard to meet the ever-increasing need of higher energy and power densities for LIBs, which will restrict the further development of consumer electronics and electric vehicles [1,2]. 【硅的优点】

## 【硅的优点】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

Generally speaking, the Si-based anode materials in LIBs have many merits, such as the great rich in the earth, theoretical specific capacity（3580 mA h g1）is ten times higher than that of graphite, and the relatively low potential of 0.3 V vs. Liþ/Li, which can avoid the formation of lithium dendrites [3–5]. 【硅的缺点】

## 【硅的缺点】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

However, Si-based anode materials undergo the large volume expansion (300%) in lithiation/delithiation process, which will lead to the collapsing and pulverization of the active material, corresponded to the active materials break away from the current collectors [6,7]. Besides, the repeated fracture and formation of solid electrolyte interface (SEI) results in the irreversible consumption of Liþ and electrolyte [8,9], and it causes the low coulombic efficiency and large irreversible capacity loss [6]. In addition, the performance of Sibased anode materials is restricted by sluggish electrochemical kinetics due to the inherent low conductivity [10].

【提出很多策略解决硅的问题。举例，纳米化。】

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[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

To overcome these thorny problems, many improvement strategies have been adopted. For instance, a variety of nanosized Si particles with different morphology and structures (nanoparticles [11,12], nanowires [13,14], nanosheets [15,16], hollow nanoparticles [17,18], and nanoporous networks [19,20]) have been designed and prepared. 【硅纳米化的优势】

## 【硅纳米化的优势】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

These solutions can buffer the huge volume change and enhance the mechanical integrity of Si electrodes to some degree. 【另一方面，提升寿命，硅基复合物】

## 【另一方面，提升寿命，硅基复合物】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

On the other hand, in order to improve the long cycling stability of Sibased anodes, the Si-based composites with different composite structures have also been designed, such as Si/carbon [21,22], Si/metallic oxide [23,24], and Si/carbon/metal oxide [25,26].【和其他改性方法相比，核壳蛋黄结构硅碳纳米结构复合物的优点】

## 【和其他改性方法相比，核壳蛋黄结构硅碳纳米结构复合物的优点】

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Compared with other modifications, core–shell Si/C and yolk–shell Si/C nanostructures are regarded as easier and higher efficiency strategies in the preparation of Si/C composite. In the Si/C composite structures, the carbon layer not only acts as a buffering shell to relieve the volume changes, but also decreases the contact resistance between Si particles with the current collector [27,28].

【小于150nm的硅，可以首次嵌锂过程中，缓解体积变化】

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[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

It is generally considered that the volume expansion of Si anodes can be limited in the initial lithiation process, when the diameter of particles is less than 150 nm [9]. 【但是，进一步降低硅颗粒尺寸的问题】

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[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

However, with the decrease of the particles size, the price of commercial nanosized Si increases obviously, and the manufacturing technique of commercial nanosized Si particles is complex.【硅的选择影响了生产硅的成本】

## 【硅的选择影响了生产硅的成本】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

Therefore, the selection of Si will directly affect the production cost of manufacturing Si-based anode materials.【硅可通过镁和二氧化硅的反应生产，可制备纳米硅。】

## 【硅可通过镁和二氧化硅的反应生产，可制备纳米硅。】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

Fortunately, Si also can be obtained by making use of magnesium (Mg) to scavenge oxygen from silica (SiO2) at high temperature (500 C–900 C), and the microscale morphologies of the original SiO2 can be maintained, so magnesiothermic reduction have been used to prepare the nanosized Si particles [29–31]. 【镁热反应方程】

## 【镁热反应方程】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

The magnesiothermic reduction process can be summarized as follows: Eq. (1) and (2) [32].

【镁热反应。硅的结构完整性和SiO2纳米颗粒的尺寸有关。文献，Si在MgO基体中。】

## 【镁热反应。硅的结构完整性和SiO2纳米颗粒的尺寸有关。文献，Si在MgO基体中。】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

The magnesiothermic reduction reaction is an exothermic process, and the obtained Si is melted and aggregated at high temperature. Therefore, the structural integrality of the reduced Si particles is related to the size of SiO2 nanoparticles. Besides, Yoo et al. reported that the Si nanoparticles (10 nm) were uniformly dispersed in MgO matrix via magnesiothermic reduction reaction, and Mg2Si still exists in the inner of product [32]. Moreover, the MgO matrix will contribute to form the ordered mesoporous structure, which will facilitate to enhance the electrochemical stability of Si.

【本文，介孔Si，来自SiO2镁热反应】

## 【本文，介孔Si，来自SiO2镁热反应】

[11] Z. Wu, J. Luo, J. Peng, H. Liu, B. Chang, X. Wang, Rational architecture design of yolk/double-shells Si-based anode material with double buffering carbon layers for high performance lithium-ion battery, GREEN ENERGY & ENVIRONMENT, 6 (2021) 517-527.

In this work, the mesoporous Si spheres were obtained from SiO2 (St€ober method) particles via the magnesiothermic reduction reaction. The yolk/double-shells (YDS) Si/C anode material with double buffering resin-based carbon layers was fabricated via making use of HF to dissolve the SiO2 in intermediate layer. The unique architecture with double buffering layers can provide good electron/ion pathways and alleviate volume change during charge/discharge process. And the costing of phenolic resin-based carbon is much lower than other organic carbon sources, e.g., polydopamine-based carbon, polyaniline-based carbon and polypyrrole-based carbon. As expected, the new designed yolk/double-shells Si-based anode material with double buffering carbon layers demonstrated many advantages, such as higher initial coulombic efficiency, improved reversible discharge specific capacity, enhanced capacity retention, and good rate capability as well as low cost.【】【】

2. Experimental

[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

1. Introduction

【锂电池使用碳电极的问题】

## 【锂电池使用碳电极的问题】

[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

Conventional lithium ion batteries (LIBs) based on carbon anodes, such as commercial graphite microspheres and mesophase carbon microbeads (MCMBs), with a theoretical capacity ofabout 372 mAh g1, have now reached their limits in terms of meeting the need for high energy storage.1–4【为提高能量密度，需要高容量，低成本的电极】

## 【为提高能量密度，需要高容量，低成本的电极】

[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

To meet the increasing demand for energy storage capability, novel electrode materials with higher capacity, low cost, and the ability to be produced on a large scale are of great interest.5

【硅和氧化铁被认为有吸引力】

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[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

Silicon (Si) and hematite (a-Fe2O3) have long been regarded as appealing anode materials for LIBs because of their much higher theoretical capacities ($4200 and 1007 mA h g1, respectively) than those of the commercially used ones, as well as their nontoxicity and natural abundance.3,6–14【硅和氧化铁的缺点】

## 【硅和氧化铁的缺点】

[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

Despite all these advantages, the full utilization of silicon- or a-Fe2O3-based batteries to date has been hindered by a series of obstacles, including poor cycle life and rate performance, that resulted from their large volumetric expansion during cycling and low ionic/electronic conductivity.15–19【为改善硅和氧化铁缺点做了大量努力，举例】

## 【为改善硅和氧化铁缺点做了大量努力，举例】

[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

Hence, great efforts have been made to further improve their electrochemical performance by using various silicon-containing (silicon nanowires,10,20,21Fe2O3 nanotubes,26 Fe2O3 and graphene@Fe2O3 composite9) materials. silicon nanotubes,22 porous structured silicon,23 and carbon coated silicon24,25)or aFe2O3-containing (Fe2O3 nanorods,15 nanosheets,6【其中，多孔carbon@Si或孔carbon@a-Fe2O3有前途，原因。文献】

## 【其中，多孔carbon@Si或孔carbon@a-Fe2O3有前途，原因。文献】

[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

Among them, porous carbon@Si or carbon@a-Fe2O3 composites in which the active particles are coated or embedded in a porous conductive carbon skeleton are quite promising, because the void space allows the expansion of silicon and a-Fe2O3 during the lithiation/delithiation processes, thus maintaining the structural and electrical integrity.7a For instance, Cui et al. Demonstrated a carbon@void@Si (yolk–shell) composite system having a high capacity of $2800 mA h g1.27 a-Fe2O3/reduced graphene oxide (rGO) nanocomposites ($1100 mA h g-1) in which the a-Fe2O3 nanoparticles were uniformly anchored on the graphene nanosheets were fabricated by Zhang et al.28 through a facile microwave hydrothermal method. 【（之前是结合文献）因此，硅和氧化铁的最主要问题可如何被解决】

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[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

Therefore, the main problem for silicon and a-Fe2O3 can be addressed by combining porosity and the incorporation of carbon in one structure. 【一些相关报道，硅与氧化物或合金组合，可利用两者优点】

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[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

Until now, some studies have reported that combining silicon with metal oxides or alloys in one system is an effective way to improve the electrochemical performance of the synthesized electrodes by taking advantage of the superiorities of both silicon and the active additions, such as Co2O3@Si,29 Ge@Si.32SnO2@Si,30 Fe3C@Si,31 and It is found that, the specific capacities of the composites were increased due to the introduction of silicon, and the cycling stability was also improved due to the specific structure of the metal oxides or alloys.【已有研究问题，大多数复合物，碳中要么有硅，要么有Fe2O3，很少有同时包含2个。】

## 【已有研究问题，大多数复合物，碳中要么有硅，要么有Fe2O3，很少有同时包含2个。】

[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

However, to the best of our knowledge, most of the composite anodes explored so far contain either silicon or a-Fe2O3 along with carbon, and only a few reports are available in which both the materials are utilized in one system. 【少有的相关文献，以及文献问题】

## 【少有的相关文献，以及文献问题】

[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

Just as reported this year, David Zitoun synthesized Si@hollow gFe2O3 nanoparticles as anodes for LIBs.33As reported, theprepared Si with iron oxide grown on its surface showed a very high specific capacity (around 2600 mA h g1). However, the preparation process in this reported work is very complex and high cost. In addition, without the introduction of carbon or other kinds of conductive additions, the conductivity of this prepared composite is low. 【因此，预期锂电池技术会取得进展，如何可以使用xxx方法。】

## 【因此，预期锂电池技术会取得进展，如何可以使用xxx方法。】

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Therefore, it is envisioned that advances in LIB technology can be achieved by incorporating both silicon and a-Fe2O3 into one porous carbon skeleton.

【本文，一步法，C–Si–Fe2O3复合物】

## 【本文，一步法，C–Si–Fe2O3复合物】

[12] L. Zhang, H. Guo, R. Rajagopalan, X. Hu, Y. Huang, S.X. Dou, H.K. Liu, One-step synthesis of a silicon/hematite@carbon hybrid nanosheet/silicon sandwich-like composite as an anode material for Li-ion batteries, JOURNAL OF MATERIALS CHEMISTRY A, 4 (2016) 4056-4061.

Herein, we report a facile one-step synthesis of a carbon–silicon–hematite (C–Si–Fe2O3) composite. In our case, silicon nanoparticles (Si NPs) are sandwiched between a-Fe2O3embedded porous carbon sheets (C@Fe2O3). The electronic conductivity of both the silicon and the a-Fe2O3 NPs can be improved by using the conductive carbon layers.34,35

Meanwhile, the flexible carbon sheets are adaptable and can freely switch between the expansion and contraction of the active particles upon lithiation and delithiation.28,36 In addition, the space between the carbon layers allows the Si NPs to expand freely,37 and the C@Fe2O3 layers can prevent the Si and a-Fe2O3 nanoparticles from aggregating. These unique characteristics facilitate the collection and transport of electrons, resulting in high capacity and good cycling stability.38,39【】【】

2. Experimental section

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

1. Introduction

【为满足xxx，需要发展新的锂电池负极材料】

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[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

To meet the increasing demands of rapidly developing market from cell phone to electric vehicles for the Li-ion batteries (LIBs), new anode materials with higher capacity have attracted significant attention. 【石墨的问题】

## 【石墨的问题】

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

Graphite, the most commonly used commercial anode material, has low theoretical specific capacity (372mAhg−1) and poor rate capability. 【硅有吸引力，优点】

## 【硅有吸引力，优点】

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

Silicon is considered as one of the most attractive and promising alternative anode materials to replace graphite in LIBs in the coming decades in virtue of its fascinating performance, such as relatively low working potential (∼370mV versus Li/Li+), rich abundance in earth, environmental benignity, and especially the highest theoretical capacity of4212mAh g−1 among the existing anode materials [1]. 【硅的挑战和问题】

## 【硅的挑战和问题】

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

However, there still exist several challenges which restrict the commercialization of such silicon anodes. Firstly, the large volumetric expansion (∼400%) of silicon anodes upon the lithiation results in high internal stress, causes mechanical fracture and pulverization of electrode and subsequent losses of electrical contact between the active material and current collector, and leads to poor reversibility and rapid fading of capacity. Secondly, the huge and repeated volume change during the charge/discharge process prevents the formation of a layer of stable solid electrolyte interface (SEI). Hence it can easily continuously grow through the cracks till being too thick for Li-ions to diffuse through, and in turn results in a low Coulombic efficiency and a decrease in capacity. Moreover, silicon anodes possess poor electrical conductivity [2, 3]. Such drawbacks lead to serious capacity fade during cycles and thus hinder the practical applications of silicon anodes.

【为解决以上硅的问题，需要xxx】

## 【为解决以上硅的问题，需要xxx】

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

In order to address abovementioned challenges, it would be crucial to release the mechanical strains, as well as reducing the diffusion length of Li-ions in Si electrode materials during charge/discharge process. 【纳米硅是一个解决方案。举例，发展了许多纳米硅。】

## 【纳米硅是一个解决方案。举例，发展了许多纳米硅。】

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

Nanostructured Si would be a good solution. So far, many nanostructured silicon and Si-based composites with various morphologies, such as nanoparticles [4–10], porous nanoparticles [11, 12], nanofilm [13–18], nanotubes [19–21], nanofibers [2, 22–37], and core shell structures [3, 38–42] have been prepared and evaluated. 【其中，一维纳米硅，举例，优点】

## 【其中，一维纳米硅，举例，优点】

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

Among them, one-dimensional (1D) nanostructured Si or Si containing nanotubes, nanofibers, and core-shell structures showed very good electrochemical performance in virtue of the enhanced electric conductivity of the 1D nanostructures. Furthermore, they could easily form three-dimensional (3D) interconnected networks to buffer the huge volume changes of active Si, thereby preventing the degradation of the electrode integrity and the breakdown of electric conductive networks. 【但是，一维纳米硅通常如何制备的，其中的缺点】

## 【但是，一维纳米硅通常如何制备的，其中的缺点】

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

However, such 1D silicon nanostructures were usually synthesized by using toxic SiH4 as silicon precursor and costly Au particles as catalysts [22, 24, 39, 40]. Moreover, the synthesis of them seemed to be too elaborate and suffered from low yield. 【因此，直接和实际方案是在碳基体中使用硅纳米颗粒，举例。优点】

## 【因此，直接和实际方案是在碳基体中使用硅纳米颗粒，举例。优点】

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

Thus, the most straightforward and practical approach recently to overcome these shortcomings is to directly utilize the commercially available Si nanoparticles incorporated into carbon matrix, for example, carbon nanofibers via electrospinning method [26–29, 31– 34]. Such products are typical interconnected nonwoven nanofiber networks with good mechanical integrity. The as-prepared free-standing fabrics can be directly used as anodes for LIBs without adding any other polymeric binders or conductive additives. They could increase the effective electrode-electrolyte interface area, facilitate the high-speed electron transport, and hold great potential to enhance the electrochemical performance. 【但是，硅和碳直接接触的缺点，循环性不行】

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[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

Nevertheless, in fact, their cycle stability is still inferior because the direct contact of carbon with silicon has the limited buffer capability, which is difficult to cope with about 400% of volume expansion.

【为提升循环性，纳米颗粒封装在连续空心碳中，性能提升原因】

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More recently, to improve the cycling stability of silicon anode materials, some new nanostructures with Si nanoparticles encapsulated in continuous hollow carbon tube shave gained much attention due to the enhanced electrical conductivity and stable solid electrolyte interface (SEI). Moreover, particularly, the ample empty space inside the hollow tubes among the silicon nanoparticles allowed for silicon nanoparticles expansion freely during electrochemical cycling, supporting a stable cycling of the entire electrode as well as high charge and discharge rates [2, 3, 35–37, 42]. 【利用酚醛树脂制备的优势】

## 【利用酚醛树脂制备的优势】

[13] D. Nan, Z.-H. Huang, R. Lv, Y. Lin, L. Yang, X. Yu, L. Ye, W. Shen, H. Sun, F. Kang, Silicon-Encapsulated Hollow Carbon Nanofiber Networks as Binder-Free Anodes for Lithium Ion Battery, JOURNAL OF NANOMATERIALS, 2014 (2014).

In addition, it is known that the low-cost phenolic resin (i.e., resol) has high carbon yield (over 50%) during carbonization and good anodic performance for LIBs by exhibiting a high reversible capacity of 550mAh g−1 [43]. More importantly, the resol could be solidified by occurrence of polycondensation reaction thus keeping the original morphology bymeans of solidification process. It is very favorable to generate the tubular nanostructure.

【本文，进一步提升硅循环性，硅被封装在空心碳纳米纤维网络中】

## 【本文，进一步提升硅循环性，硅被封装在空心碳纳米纤维网络中】

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With a motivation to further increase the cycling stability of the silicon anode materials, in this work, we put forward a facile strategy to synthesize novel silicon-encapsulated hollow carbon nanofiber networks with ample space around the Si nanoparticles (hollow Si/C composites) by dip-coating resol on the surface of electrospinning Si/PVA nanofibers along with the subsequent solidification and carbonization. These freestanding membranous hollow Si/C composites can be used directly as anodes for LIBs without adding any carbon conductors or polymer binders. The as-prepared hollow Si/C composites display a high electrochemical performance with excellent cycle stability of ∼90% of discharge capacity retention after 50 cycles.【】【】

2. Experimental

[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

1. Introduction

【资源消耗、碳排放问题。需要发展电动车。电动车的问题，增加续航里程】

## 【资源消耗、碳排放问题。需要发展电动车。电动车的问题，增加续航里程】

[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

Potential problems associated with natural resources including their depletion and increasing cost and challenges, such as global warming due to the increase in carbon dioxide generated by the combustion of natural resources, have become pronounced in recent years. This has accelerated the expansion of markets for environmentally friendly vehicles such as hybrid electric vehicles, plug-in hybrid electric vehicles, and fuel-cell vehicles. In particular, vehicles are highly anticipated owing to their environmental advantages stemming from their lack of carbon dioxide emissions; however, their actual ranges are still low compared to those of gasoline-powered cars, and the advanced batteries with high energy densities as the power sources of such vehicles are strongly desired to increase their range. In this context, solid-state and metal-air batteries have become increasingly active research and development areas; however, their practical use has not yet been realized.

【本工作关注锂电池用硅电极，原因】

## 【本工作关注锂电池用硅电极，原因】

[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

In this study, we focused on silicon-based electrode materials for lithium-ion battery (LIB); theoretical capacity is approximately 10 times greater than that of graphite electrode of LIB. 【SiO负极的商业化情况，企业进展。】

## 【SiO负极的商业化情况，企业进展。】

[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

Hitachi Maxell commercialized SiO negative electrode which is composed of major component of graphite and SiO additives. Panasonic preliminarily released large capacity battery (4.0 Ah) with Si-based negative electrode, but not yet commercialized. 【但是，还没有利用大容量硅负极的实际应用】

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[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

Thus, no practical application of LIB, taking advantage of large capacity of Si, has been performed.【硅电极的最大挑战，寿命短，原因】

## 【硅电极的最大挑战，寿命短，原因】

[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

The greatest challenges that obstruct the practical use of silicon-based materials for LIB is their short cycle life that arises from the alloying of silicon and lithium during their charging and discharging processes and the change in their big volume through large expansion and contraction during the repeated desorption of lithium from the alloy. In previous studies, these effects have been well documented to adversely affect the cycle life of LIB.【电动车的寿命需要15年，接近燃油车】

## 【电动车的寿命需要15年，接近燃油车】

[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

In addition, vehicles require batteries that span the vehicles’ expected lifetime of as long as 15 years, which is similar to that of the automobiles based on gasoline internal-combustion engine.

【许多研究被开展以克服寿命问题，包括锂硅合金化；薄的硅电极；硅微粉化】

## 【许多研究被开展以克服寿命问题，包括锂硅合金化；薄的硅电极；硅微粉化】

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Numerous studies to overcome this lifetime problem have already been conducted, including some reports that a longer battery lifetime can be achieved by lowering the absolute value of the volume change that occurs when silicon and lithium are alloyed; in these cases, longer lifetimes were achieved through the fabrication of thinned silicon electrodes [1e3] and micronization of the silicon [4,5]. 【薄膜方法的问题，和电极浆料涂敷对比】

## 【薄膜方法的问题，和电极浆料涂敷对比】

[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

However, the thin-film method has several shortcomings in terms of production costs that hinder its industrial development; specifically, the required manufacturing equipment is expensive, and its production speed is low compared to that of the method of applying an electrode paste, which is commonly practiced to produce LIB. 【硅微粉化的问题】

## 【硅微粉化的问题】

[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

Furthermore, micronized silicon forms SiO2 via oxidation by oxygen when handled in ordinary atmosphere, which results in increased irreversible capacity during the initial charging of LIB prepared using this method. 【许多工作研究硅和石墨的结合；但是均一的硅石墨复合物难制备，硅的量难提高】

## 【许多工作研究硅和石墨的结合；但是均一的硅石墨复合物难制备，硅的量难提高】

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A large number of studies have been conducted on the conjugation of silicon and graphite [6e21]; however, uniform silicon graphite composites are difficult to prepare, while the amount of added silicon is difficult to increase.

【本文，碳包裹的硅微粉化分散在软碳基体中】

## 【本文，碳包裹的硅微粉化分散在软碳基体中】

[14] N. Kobayashi, Y. Inden, M. Endo, Silicon/soft-carbon nanohybrid material with low expansion for high capacity and long cycle life lithium-ion battery, JOURNAL OF POWER SOURCES, 326 (2016) 235-241.

This study developed a new nanohybrid material, composed of carbon coated micronized silicon dispersed in soft-carbon matrix. It exhibited a long cycle life and suppressed the electrode expansion. These behaviors were discussed in connection with abundant nanovoids (diameter of ca. 70 nm) and hard bulk skeletal structure.【】【】

2. Experimental method

[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

1. Introduction

【锂电池是消费电子中的主要能源，原因。以及电动车中的应用】

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Lithium-ion batteries have become the power source of choice for consumer electronic devices such as cell phones and laptop computers due to their high energy density and long cycle life. In addition, lithium-ion batteries are expected to be a major breakthrough in the hybrid vehicle field. 【尽快取得成功商业化应用，还需要进一步性能提升】

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Despite their successful commercial application, further performance improvement of the lithium-ion battery is still required.

【石墨被广泛使用】

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[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

Graphitic or graphitization carbons have been used extensively as negative electrode materials for lithium-ion batteries. 【石墨的问题】

## 【石墨的问题】

[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

However, the lithium storage capacity of graphite as associated with its maximum LiC 6 stage 1 is limited to 372 mAh/g. 【锂合金引起兴趣。举例】

## 【锂合金引起兴趣。举例】

[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

Lithium alloys are also attractive anode candidates because they can incorporate large amounts of lithium. For example, Li 4.4 Si has a theoretical capacity of 4200 mAh/g.【锂合金的问题】

## 【锂合金的问题】

[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

However, lithium alloys are brittle and hence easily pulverized by the large volume change in charge and discharge reactions. The resulting loss of connectivity with the conducting additive particles causes poor cyclability in practice. 2-5 【为解决锂合金的问题】

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[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

，研究硅碳复合物。文献】To circumvent this problem, several research efforts have been focused on composites consisting of silicon and carbonaceous materials. For instance, Wilson and Dahn 6 have synthesized nanodispersed silicon in carbon using chemical vapor deposition (CVD). Although the Si–C anodes demonstrated a reversible capacity of 500 mAh/g, it is difficult to control the morphology of Si and C when using the CVD approach. Yoshio et al. 7 prepared carbon-coated silicon by a thermal vapor deposition (TVD) method, and a better cycle life than with the respective silicon has been achieved. Composite produced by mechanically mixing silicon and graphite? or acetylene black? shows high capacity but its cycling is still poor. 8 More recently, research work by many groups has demonstrated that silicon/disordered carbon (DC) composite, obtained by mechanically milling and pyrolyzing organic precursors, such as polyparaphenylene (PPP) or polyvinyl chloride (PVC), in an inert gas atmosphere can reversibly insert Li atoms with a capacity (>500 mAh/g) greater than that of graphite (372 mAh/g). 9,10 【虽然和硅比，硅碳复合物性能提升很多，但还存在一些问题，举例】

## 【虽然和硅比，硅碳复合物性能提升很多，但还存在一些问题，举例】

[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

Although the electrochemical performance is significantly improved compared with the respective silicon electrodes, the silicon-disordered carbon systems still pose some problems, such as a relatively high capacity fading rate and a relatively low capacity contribution from the disordered carbon (386 mAh/g from PVC-based disordered carbon; 451.3 mAh/g from PPP-based disordered carbon). 【近年来，其他热解体系的一些发现】

## 【近年来，其他热解体系的一些发现】

[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

Recently, it was reported that the disordered carbon prepared by pyrolyzing hexa(phenyl)benzene (HPB) demonstrated a stable capacity on the order of 500 mAh/g. 11 Carbon from pyrolyzed sugar under optimal conditions showed a large reversible capacity of 650 mAh/g. 12【因此，优化硅-不规则碳体系有必要】

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[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

Therefore, the optimization of the silicon-disordered carbon system and further improvement of the silicon-disordered carbon concept are necessary.

【本文，球磨制备一系列硅-不规则碳样品，球磨硅-蔗糖和硅pva，随后热解】

## 【本文，球磨制备一系列硅-不规则碳样品，球磨硅-蔗糖和硅pva，随后热解】

[15] Z.P. Guo, E. Milin, J.Z. Wang, J. Chen, H.K. Liu, Silicon/disordered carbon nanocomposites for lithium-ion battery anodes, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 152 (2005) A2211-A2216.

In this paper, a series of Si–DC samples were synthesized by mechanical milling of Si–sucrose and Si–polyvinyl alcohol (PVA) followed by pyrolysis. Here sucrose was chosen because the DC obtained by sugar pyrolysis shows a large reversible capacity (650 mAh/g); 12 PVA was used because it gives the maximum yield of amorphous carbon compared to other polymers. 13 The disordered carbon content in Si–DC nanocomposites was determined by thermogravimetric analysis (TGA) analysis for the first time. The electrochemical properties of the obtained Si–DC nanocomposites as anodes in lithium-ion cells were systematically evaluated.【】【】

2. Experimental

[16] J. Ha, H. Park, M. Kim, Y.T. Kim, J. Choi, Liquefied-Natural-Gas-Derived Vertical Carbon Layer Deposited on SiO as Cost-Effective Anode for Li-Ion Batteries, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 169 (2022).

1. Introduction

【近年来，高能量密度的锂电池的需求提升】

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Recently, the demand for Li-ion batteries (LIBs) with high energy density has increased.【硅的优点，容量，和其他材料对比】

## 【硅的优点，容量，和其他材料对比】

[16] J. Ha, H. Park, M. Kim, Y.T. Kim, J. Choi, Liquefied-Natural-Gas-Derived Vertical Carbon Layer Deposited on SiO as Cost-Effective Anode for Li-Ion Batteries, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 169 (2022).

Si-based materials are the most promising candidates for LIB anode materials because of their high theoretical capacity (∼4200 mAh g−1 for Si, ∼2400 mAh g−1 for silicon monoxide (SiO), and ∼1680 mAh g−1 for silicon dioxide (SiO2)) compared to transition metal oxide1,2 and organic based electrode3 adequate lithium alloying–dealloying potential (∼0.5 V vs Li/Li+), and abundance in the Earth’s crust (15.1 wt%).4,5 【硅的缺点】

## 【硅的缺点】

[16] J. Ha, H. Park, M. Kim, Y.T. Kim, J. Choi, Liquefied-Natural-Gas-Derived Vertical Carbon Layer Deposited on SiO as Cost-Effective Anode for Li-Ion Batteries, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 169 (2022).

However, because of their low conductivity and high-volume expansion rate during cycling (∼400%) leading to loss of electric contact via pulverization and electrolyte decomposition, Si-based electrodes exhibit poor cycling properties and fast capacity fading.6,7【因此，为克服缺点，活性材料表面碳涂层，可解决什么问题】

## 【因此，为克服缺点，活性材料表面碳涂层，可解决什么问题】

[16] J. Ha, H. Park, M. Kim, Y.T. Kim, J. Choi, Liquefied-Natural-Gas-Derived Vertical Carbon Layer Deposited on SiO as Cost-Effective Anode for Li-Ion Batteries, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 169 (2022).

Therefore, to overcome these shortcomings, recently, researchers have increasingly focused on coating the active material surface with a carbon layer that increases electrical conductivity and suppresses volume expansion by inducing the formation of a stable solid electrolyte interface (SEI).8–10 【cvd是大规模在活性材料表面生长碳的最有效方法。文献】

## 【cvd是大规模在活性材料表面生长碳的最有效方法。文献】

[16] J. Ha, H. Park, M. Kim, Y.T. Kim, J. Choi, Liquefied-Natural-Gas-Derived Vertical Carbon Layer Deposited on SiO as Cost-Effective Anode for Li-Ion Batteries, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 169 (2022).

Typically, chemical vapor deposition (CVD) is the most effective method for large-scale manufacturing of carbon coating layers on active materials. Kang et al. fabricated a fibrous yolk–shell-structured Si–C freestanding electrode with a high areal capacity over 150 cycles via CVD using acetone.11 In addition, an amorphous carbon-coated porous Si–graphene–carbon composite anode with superior cycling performance compared with commercial Si–carbon materials were fabricated via CVD method using methane (CH4) under an Ar atmosphere.12 【但是，使用昂贵的高纯碳氢化合物的问题，增加成本】

## 【但是，使用昂贵的高纯碳氢化合物的问题，增加成本】

[16] J. Ha, H. Park, M. Kim, Y.T. Kim, J. Choi, Liquefied-Natural-Gas-Derived Vertical Carbon Layer Deposited on SiO as Cost-Effective Anode for Li-Ion Batteries, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 169 (2022).

However, the use of expensive hydrocarbon-based carbon sources with high purity, including CH4, ethane (C2H6), acetylene, and n-butane (C4H10), during CVD leads to an increase in the overall electrode manufacturing cost13【为降低cvd成本，用成本低的碳源，举例，修饰活性材料】

## 【为降低cvd成本，用成本低的碳源，举例，修饰活性材料】

[16] J. Ha, H. Park, M. Kim, Y.T. Kim, J. Choi, Liquefied-Natural-Gas-Derived Vertical Carbon Layer Deposited on SiO as Cost-Effective Anode for Li-Ion Batteries, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 169 (2022).

To minimize the total production cost of CVD using expensive and high-purity hydrocarbon-based carbon sources, several researchers have attempted to obtain diverse carbon structures for decorating active materials using inexpensive carbon sources, such as liquefied petroleum gas,13,14 natural gas15 and coal gas.16,17

【本文，SiO表面沉积直立碳，用液化天然气】

## 【本文，SiO表面沉积直立碳，用液化天然气】

[16] J. Ha, H. Park, M. Kim, Y.T. Kim, J. Choi, Liquefied-Natural-Gas-Derived Vertical Carbon Layer Deposited on SiO as Cost-Effective Anode for Li-Ion Batteries, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 169 (2022).

Herein, we deposited a vertical carbon layer on SiO as an anode material for LIBs via CVD using liquefied natural gas (LNG, 0.27 USD/L, based on the retail price in 2021, South Korea), which was 13 times less expensive than high-purity CH4 (3.52 USD/L, based on the retail price in 2021, South Korea). Furthermore, the physicochemical properties and electrochemical performance of the carboncoated SiO electrode prepared using inexpensive LNG (denoted as C-SiO-LNG) were compared with those of a carbon-coated SiO electrode prepared using high-purity CH4 (denoted as C-SiO-CH4).【】【】

2. Experimental

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

1. Introduction

【硅的优点，相对石墨，锂电池的候选负极】

## 【硅的优点，相对石墨，锂电池的候选负极】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

As the one with exceptionally high gravimetric and volumetric capacity, low discharge voltage as well as abundant resource, silicon (Si) is a promising anode candidate as potential alternative to current commercial graphite in lithium ion batteries (LIBs). 【硅被广泛探索，过去15年几百篇文章】

## 【硅被广泛探索，过去15年几百篇文章】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

The exploration of Si-based anodes has been extensively carried out, as evidenced by hundreds of scientific papers published in the last 15 years and having been reviewed from varied perspectives.1–7 【尽管取得进展，硅的问题】

## 【尽管取得进展，硅的问题】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

Despite the great potential, several issues of Si as anode severely hinder its practical use, including large volume expansion (more than 300% upon full lithiation) that irreversibly breaks the structural integrity, poor intrinsic electrical conductivity that causes sluggish electrochemical kinetics, as well as gradual pulverization of the anode along with lithiation/delithiation cycles.

【典型的解决以上问题的策略主要依赖于稳定硅纳米结构】

## 【典型的解决以上问题的策略主要依赖于稳定硅纳米结构】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

Typical strategies to tackle the above issues mainly rely on stabilizing the Si nanostructure while maintaining its morphology/structure integrity during lithiation/de-lithiation.【各类硅纳米结构。优点】

## 【各类硅纳米结构。优点】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

Firstly, introducing “nano-effect” by reducing at least one dimension of Si to normally less than 100 nm, such as Si nanoparticles/nanospheres/nanoflowers,8–10 nanocubes,11 mesoporous Si,12–14 Si nanowires,.15–18 The nanoscale dimension of the Si realizes faster and more complete alloying and de-alloying with lithium ions, and meanwhile suppresses the volume expansion during the process. 【硅复合物】

## 【硅复合物】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

Secondly, combining with structurally stable and electrically conductive components to produce Si-based composites. 【诸多候选物中，碳最有前途，原因】

## 【诸多候选物中，碳最有前途，原因】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

Among all the candidates (metals, polymers, etc.),19–22 carbon is the most promising one, acting as host/support of Si, that is capable of buffering/accommodating the volume change of Si during charging/discharging, as well as providing good electrical conduction to the composites. In addition, the carbon phase is also electrochemically active and contributes to the overall battery performance. 【碳在硅中的应用】

## 【碳在硅中的应用】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

So far, carbon nanofibers/carbon nanotubes,23–25 graphene/graphite,26–31 amorphous/doped carbon32–36 have been synergistically introduced into Si nanostructures. 【硅碳比和硅碳之间交互的有效性是决定性能的关键因素】

## 【硅碳比和硅碳之间交互的有效性是决定性能的关键因素】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

The ratio of Si/carbon in the composites and the effectiveness of the interaction between Si and carbon are the critical factors that determine the properties. 【通过优化胶粘剂等，提升制备过程】

## 【通过优化胶粘剂等，提升制备过程】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

Last but not the least, improving the fabrication/assembling process through, for instance, optimizing the binders,37–39 inkjet-printing of the anode,40 etc.【总体而言，发展合适的硅纳米复合物以有效利用硅的活性迫切需要】

## 【总体而言，发展合适的硅纳米复合物以有效利用硅的活性迫切需要】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

Overall, the design/development of proper Si nanocomposites that could efficiently utilize the electrochemical activity of Si is still highly desired.

【需要考虑可回收性】

## 【需要考虑可回收性】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

Nowadays, the recyclability of the used materials has been becoming a more and more important consideration in industry that favors the utilization efficiency of the materials and the environment protection.【然而，xxx很少被考虑】

## 【然而，xxx很少被考虑】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

Yet this has rarely been investigated in the area of batteries, which the authors think it is worthwhile to unveil to add additional value to the good energy storage performance.【本文，商业硅纳米颗粒和纤维碳网络结合，自支撑薄膜，不需要胶粘剂和碳黑】

## 【本文，商业硅纳米颗粒和纤维碳网络结合，自支撑薄膜，不需要胶粘剂和碳黑】

[17] J. Kong, Y. Wei, Silicon Nanoparticles Confined in Thin Carbon Network: The Free-Standing Anode of Lithium Ion Batteries with High Performance and Easy Recyclability, JOURNAL OF THE ELECTROCHEMICAL SOCIETY, 166 (2019) A2013-A2020.

In this work, we proposed a straightforward strategy to utilize Si via binding commercially available Si nanoparticles (NPs) within fibrous thin carbon network, achieving freestanding anode without the usage of any binder and carbon black. Interestingly, the Si NPs collapse into smaller ones instead of expanding in volume upon lithiation, leading to excellent electrochemical properties. Moreover, the used collapsed Si NPs can be easily recycled from the anode after battery test, offering opportunity for the further reuse of the Si material.【】【】

2. Experimental

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

1. Introduction

【石墨的缺点】

## 【石墨的缺点】

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

The capacity of the commercial graphite has reached its limit, which prevent lithium ion batteries to meet the increasing demand of higher energy density. Besides, the commercial or indevelopment cathode materials also display a low specific capacity [1–8]. 【因此需要寻找新的负极，替代商业石墨】

## 【因此需要寻找新的负极，替代商业石墨】

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

Therefore, new anode materials should be developed to substitute the current commercial electrode materials. 【硅有前途，优点】

## 【硅有前途，优点】

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

Silicon is a promising candidate for anode material because of its high theoretical capacity (about 4200 mAh g-1), low discharge voltage and natural abundance [9–14].【硅的问题】

## 【硅的问题】

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

But a significant volume expansion and unstable solid electrolyte interphase (SEI) films occurred during the electrode process and results in particle fracture and loss of capacity with cycling [15–20]. 【硅纳米颗粒包裹在碳材料中，是个有效策略】

## 【硅纳米颗粒包裹在碳材料中，是个有效策略】

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

Si nanoparticles (NPs) wrapped in carbon materials is an effective way to strengthen the electrochemical properties.【近年来，各种碳被用来缓冲硅纳米颗粒的体积变化，作用】

## 【近年来，各种碳被用来缓冲硅纳米颗粒的体积变化，作用】

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

Recently, porous carbon [17,21], grapheme [18,22], carbon nanofiber [23,24] etc. have been utilized to accommodate the volume change of the SiNPs, thus effectively avoiding the direct contact between encapsulated SiNPs and the electrolyte and enabling the interfacial and structural stabilization of encapsulated SiNPs during cycling. 【虽然这些碳材料有他们的自身优势，但纳米硅的优势发挥有限，因为依赖单独的涂层材料】

## 【虽然这些碳材料有他们的自身优势，但纳米硅的优势发挥有限，因为依赖单独的涂层材料】

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

Though all of these carbon materials have their own advantages, the enhanced electrochemical performance of SiNPs has been limited depended on single coating materials. 【然而，只有很少报道关于用不同涂层包裹硅纳米颗粒，这样做的优点】

## 【然而，只有很少报道关于用不同涂层包裹硅纳米颗粒，这样做的优点】

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

However, there are only a few of research referred to wrapping SiNPs in two different coating materials, in which different coating materials can make their respective advantages complementary to each other, and then improve the electrochemical properties of SiNPs dramatically.

【本文，Si@RGO@C纳米纤维】

## 【本文，Si@RGO@C纳米纤维】

[18] Z. He, X. Wu, Z. Yi, X. Wang, Y. Xiang, Silicon/graphene/carbon hierarchical structure nanofibers for high performance lithium ion batteries, MATERIALS LETTERS, 200 (2017) 128-131.

In this work, we fabricated Si@RGO@C NFs with a hierarchical structure by encapsulating graphene coated Si nanoparticles in the interconnected carbon nanofibers. Firstly, for the Si@RGO, Si NPs were encapsulated into graphene shells with sufficient elasticity to accommodate the strain associated with volume change and enhance the conductivity of lithium ions and electrons. And then these as prepared Si@RGO powders have been wrapped in carbon nanofibers based on an electrospinning technology. This type of composite nanofibers as anode material possesses the advantages of high capacity and long cycle life.【】【】

2. Experimental

[19] T. Azib, C. Thaury, C. Fariaut-Georges, T. Hezeque, F. Cuevas, C. Jordy, M. Latroche, Role of silicon and carbon on the structural and electrochemical properties of Si-Ni3.4Sn4-Al-C anodes for Li-ion batteries, MATERIALS TODAY COMMUNICATIONS, 23 (2020).

1. Introduction

【能源需求增长】

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[19] T. Azib, C. Thaury, C. Fariaut-Georges, T. Hezeque, F. Cuevas, C. Jordy, M. Latroche, Role of silicon and carbon on the structural and electrochemical properties of Si-Ni3.4Sn4-Al-C anodes for Li-ion batteries, MATERIALS TODAY COMMUNICATIONS, 23 (2020).

Energy storage demands for current and future portable electronic devices and electrical vehicles are ever growing. 【锂电池功率和能量密度在某些应用方面还不足】

## 【锂电池功率和能量密度在某些应用方面还不足】

[19] T. Azib, C. Thaury, C. Fariaut-Georges, T. Hezeque, F. Cuevas, C. Jordy, M. Latroche, Role of silicon and carbon on the structural and electrochemical properties of Si-Ni3.4Sn4-Al-C anodes for Li-ion batteries, MATERIALS TODAY COMMUNICATIONS, 23 (2020).

Performances of Li-ion batteries in terms of specific power and density surpass other rechargeable battery systems but are still unsatisfactory for many applications such as extended-range electric vehicles [1–3]. 【发展比石墨容量高的负极材料很有挑战】

## 【发展比石墨容量高的负极材料很有挑战】

[19] T. Azib, C. Thaury, C. Fariaut-Georges, T. Hezeque, F. Cuevas, C. Jordy, M. Latroche, Role of silicon and carbon on the structural and electrochemical properties of Si-Ni3.4Sn4-Al-C anodes for Li-ion batteries, MATERIALS TODAY COMMUNICATIONS, 23 (2020).

The development of advanced anode materials exhibiting higher capacity than current graphite electrodes (C = 370 mAhg-1) remains very challenging [4]. 【许多负极候选材料，基于转化和合金化反应。后者举例展开描述。Sn和Si的其他优点】

## 【许多负极候选材料，基于转化和合金化反应。后者举例展开描述。Sn和Si的其他优点】

[19] T. Azib, C. Thaury, C. Fariaut-Georges, T. Hezeque, F. Cuevas, C. Jordy, M. Latroche, Role of silicon and carbon on the structural and electrochemical properties of Si-Ni3.4Sn4-Al-C anodes for Li-ion batteries, MATERIALS TODAY COMMUNICATIONS, 23 (2020).

Presently a lot of materials are studied as alternatives for carbonaceous ones to enhance the energy density of the negative electrode using various strategies such as conversion or alloying reactions [5,6]. For the latter, a special interest is brought to p-bloc elements that form lithiated compounds [7]. For instance Sn and Si can form Li4.4Sn and Li3.75Si compounds to store respectively 994 and 3600 mAhg-1 in electrochemical units, leading to much higher capacities than that of graphite [8,9]. In addition to their high theoretical capacity, these elements exhibit low potential and environmental friendliness, both suitable as anode properties.

【p区元素的缺点】

## 【p区元素的缺点】

[19] T. Azib, C. Thaury, C. Fariaut-Georges, T. Hezeque, F. Cuevas, C. Jordy, M. Latroche, Role of silicon and carbon on the structural and electrochemical properties of Si-Ni3.4Sn4-Al-C anodes for Li-ion batteries, MATERIALS TODAY COMMUNICATIONS, 23 (2020).

However, electrodes made of pure p-bloc elements are considered inappropriate because of large capacity fading during electrochemical cycling [10]. The large volume expansion that accompanies Li insertion and extraction during cycle life results in electrode pulverization and loss of electrical contact [11,12]. 【为保持机械完整，将反应元素嵌入某个缓冲基体，作用。这一方法的具体操作。体系举例】

## 【为保持机械完整，将反应元素嵌入某个缓冲基体，作用。这一方法的具体操作。体系举例】

[19] T. Azib, C. Thaury, C. Fariaut-Georges, T. Hezeque, F. Cuevas, C. Jordy, M. Latroche, Role of silicon and carbon on the structural and electrochemical properties of Si-Ni3.4Sn4-Al-C anodes for Li-ion batteries, MATERIALS TODAY COMMUNICATIONS, 23 (2020).

To ensure mechanical stability, embedding capacitive elements into a buffering matrix that helps accommodating volume changes, while improving electronic conduction, has been proposed [13,14]. This concept can be implemented, for instance, using binary intermetallic compounds made of one element that reacts with lithium (e.g. Si or Sn) and another one that is inactive (e.g. Fe, Ni, Co or Cu). Some reported intermetallics to achieve this goal are Ni3Sn4 [15–17], Cu6Sn5 [18], CoSn2 [19,20], FeSn2 [20], FeSi2 [21] and NiSi2 [22]. This approach has been successfully extended to ternary silicides such as Ti4Ni4Si7 [23,24], TiFeSi2 [25] and Ti3Si2C2 [26]. Moreover, binary and ternary intermetallics have been associated to carbonaceous materials to form highly efficient multicomponent systems, such as Si/FeSi2/C [27] and Si/Cu3Si/C [28], for further improvement of both the mechanical cohesion of silicon and the electronic conductivity of the whole active material.

【本文，Si-Ni3.4Sn4-Al-C，机械化学方法合成】

## 【本文，Si-Ni3.4Sn4-Al-C，机械化学方法合成】

[19] T. Azib, C. Thaury, C. Fariaut-Georges, T. Hezeque, F. Cuevas, C. Jordy, M. Latroche, Role of silicon and carbon on the structural and electrochemical properties of Si-Ni3.4Sn4-Al-C anodes for Li-ion batteries, MATERIALS TODAY COMMUNICATIONS, 23 (2020).

In this context, an efficient composite Si-Ni3.4Sn4-Al-C has been already synthetized by mechanochemistry [29]. It consists of sub-micrometric Si particles embedded in a nanostructured and complex matrix mainly formed by Ni3.4Sn4 intermetallic compound and disordered carbon. Si and Sn are the capacitive elements whereas Ni and C are expected to accommodate the volume changes and to insure good electronic as well as ionic conductivity of the electrode material. Al addition is based on reported beneficial effects on the cycle-life of Si-Sn-Al amorphous electrodes [30], best results being obtained for the composition 30Si67Sn-3Al which contains 3 wt.% of Al. This composite exhibits high reversible capacity (~ 700 mAhg-1) and long cycle life (over 280 cycles) [31,32]. Here we investigate the influence of silicon and carbon contents on the microstructure of this composite material and its electrochemical properties. Silicon and carbon contents were varied from 10 to 30 wt.% Si, and from 0 to 20 wt.% C, respectively.【】【】

2. Experimental

[20] S. Song, J. Li, A. Zheng, Y. Yang, K. Yin, Facile Synthesis of Sponge-Like Porous Nano Carbon-Coated Silicon Anode with Tunable Pore Structure for High-Stability Lithium-Ion Batteries, MOLECULES, 26 (2021).

1. Introduction

【随着xxx，具有xxx特点的锂电池发展被越来越重视】

## 【随着xxx，具有xxx特点的锂电池发展被越来越重视】

[20] S. Song, J. Li, A. Zheng, Y. Yang, K. Yin, Facile Synthesis of Sponge-Like Porous Nano Carbon-Coated Silicon Anode with Tunable Pore Structure for High-Stability Lithium-Ion Batteries, MOLECULES, 26 (2021).

With the rapid growth of energy demand in mobile devices and electric vehicles, etc., the development of lithium-ion batteries with a high capacity, long cycle life, and low cost has been paid much more attention around the world [1]. 【硅可替代，碳，容量】

## 【硅可替代，碳，容量】

[20] S. Song, J. Li, A. Zheng, Y. Yang, K. Yin, Facile Synthesis of Sponge-Like Porous Nano Carbon-Coated Silicon Anode with Tunable Pore Structure for High-Stability Lithium-Ion Batteries, MOLECULES, 26 (2021).

Silicon has become one of the candidates with the most potential to replace commercial graphite anode (theoretical specific capacity is only 372 mAh−1 for LiC6) due to the high theoretical specific capacity of 4200 mAh g−1 for Li22Si5 [2–4].【硅的缺点】

## 【硅的缺点】

[20] S. Song, J. Li, A. Zheng, Y. Yang, K. Yin, Facile Synthesis of Sponge-Like Porous Nano Carbon-Coated Silicon Anode with Tunable Pore Structure for High-Stability Lithium-Ion Batteries, MOLECULES, 26 (2021).

However, the extremely large volume expansion of silicon anode as high as 280% (vs. Li15Si4)~417% (vs. Li22Si5) causes severe problems such as pulverization, unstable solid electrolyte interface (SEI) film, etc. [5,6]. These issues lead to the rapid irreversible capacity decay of silicon anodes during the cycling, and then result in poor cycling stability.

【结构设计是提高硅基负极稳定性的主要策略】

## 【结构设计是提高硅基负极稳定性的主要策略】

[20] S. Song, J. Li, A. Zheng, Y. Yang, K. Yin, Facile Synthesis of Sponge-Like Porous Nano Carbon-Coated Silicon Anode with Tunable Pore Structure for High-Stability Lithium-Ion Batteries, MOLECULES, 26 (2021).

Structure design is one of the main strategies to improve the cycling stability of siliconbased anodes. 【碳包硅的优点。文献】

## 【碳包硅的优点。文献】

[20] S. Song, J. Li, A. Zheng, Y. Yang, K. Yin, Facile Synthesis of Sponge-Like Porous Nano Carbon-Coated Silicon Anode with Tunable Pore Structure for High-Stability Lithium-Ion Batteries, MOLECULES, 26 (2021).

Carbon-coated silicon’s design exhibits improved structural stability and higher cycling stability due to the protection of silicon by the carbon coating layer [7–9]. For example, Liu et al. [10] proposed a hierarchical structure design inspired by the structure of pomegranate through a evaporation-driven self-assembly method combined with coating and etching processes. In this structure, the silicon nanoparticles are encapsulated by a conductive carbon layer that leaves enough room for expansion during lithiation. Owing to the more stable SEI film and relieved stress, this pomegranate-like silicon-based anode exhibited a superior cyclability (97% capacity retention after 1000 cycles). Ryu et.al. [11] introduced a two-dimensional silicon nanosheet coated with a thin carbon layer through chemical vapor deposition. This carbon-coated silicon nanosheet formed ripples upon delithiation, which effectively releases the induced stress, rendering the electrode much more stable and durable than the uncoated counterparts.【但是，虽然这些碳包硅设计展现高循环稳定性，但合成复杂、甚至结构设计不可控，限制实际应用】

## 【但是，虽然这些碳包硅设计展现高循环稳定性，但合成复杂、甚至结构设计不可控，限制实际应用】

[20] S. Song, J. Li, A. Zheng, Y. Yang, K. Yin, Facile Synthesis of Sponge-Like Porous Nano Carbon-Coated Silicon Anode with Tunable Pore Structure for High-Stability Lithium-Ion Batteries, MOLECULES, 26 (2021).

However, although these improved carbon-coated silicon designs show excellent cycling stability, the complex synthesis methods and even uncontrollable structure adjustment still limit the practical application of carbon-coated silicon anodes.

【本文，笼状纳米碳包硅】

## 【本文，笼状纳米碳包硅】

[20] S. Song, J. Li, A. Zheng, Y. Yang, K. Yin, Facile Synthesis of Sponge-Like Porous Nano Carbon-Coated Silicon Anode with Tunable Pore Structure for High-Stability Lithium-Ion Batteries, MOLECULES, 26 (2021).

Herein, we design a sponge-like porous nano carbon-coated silicon (sCCSi) anode with a tunable pore structure based on a facile synthesis method. It was found that the water adding strategy and the heating rate of pre-oxidation are the two key conditions for forming the sponge-like porous structure and maintaining the material integrity. Moreover, the sponge-like porous structure can be easily manipulated by changing the stirring temperature of precursor solution and the oscillation methods of precipitates. The sCCSi with normal and excessive silicon-to-carbon ratios, 1:5 and 1:2, both maintain high capacity retention up to 70.6% and 70.2% after 300 cycles at 500 mA g−1, respectively. Furthermore, the enhanced buffering effect on pressure brought from the sponge-like porous structure in sCCSi is revealed by mechanical simulation.【】【】

2. Results and Discussion

[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

1. Introduction

【纳米硅在锂电池中展现巨大潜力】

## 【纳米硅在锂电池中展现巨大潜力】

[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

Nanostructured silicon (Si) electrodes have shown great potential as lithium-ion battery (LIB) anodes. 【需要更高能量密度的锂电池】

## 【需要更高能量密度的锂电池】

[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

As the power demands in consumer electronics, transportation, and large-scale renewable energy storage continue to increase, LIBs with a greater power and a higher energy density are urgently needed [1–4].【硅可替代石墨；容量；因此被认为有前途】

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[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

Sianodes [5–9] offer an alternative to commercial graphite, with Li storage capacity ten times higher than that of the graphite (372 mAh g-1)usedincommercialLIBs; thus, Si has been considered as a promising candidate for an anode material for next-generation LIBs.【硅的缺点】

## 【硅的缺点】

[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

Unfortunately, practical applications of Si as an anode material are not yet a reality due to the huge volume change (4300%) during the lithiation/delithiation process, which leads to dramatic destruction of the initial particle morphology and the loss of electrical contact between active materials and the electrode framework. In addition, the low intrinsic electrical conductivity of Si leads to dramatic pulverization of the Si anode material, rapid capacity decay, and poor cyclability, and thus should be addressed before its practical application [10,11].

【许多努力改善硅的问题，方法举例】

## 【许多努力改善硅的问题，方法举例】

[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

Significant efforts have been made towards mitigating the adverse mechanical effects of Si anodes and improving their overall electrochemical performance through diverse strategies, including engineering active material, current collector, binder and/or their interfaces [12–14]. 【一个特别吸引人的策略是形成双功能共形涂层。体系展开描述】

## 【一个特别吸引人的策略是形成双功能共形涂层。体系展开描述】

[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

A particularly attractive strategy is the use of in-situ polymerization to form a bifunctional conformal coating that binds to the Si surface and also serves as a short diffusion pathway for electronic conduction and three-dimensional (3D) interconnectivity [9,15].The Si nanoparticles (NPs) are trapped within the interconnected narrow pores of the polymer matrix, which can buffer the pulverization of larger particles during lithiation/delithiation and maintain good electrical connectivity among the fractured particles. 【（引出第二种策略）混合纳米结构包含碳和不同的硅形貌。展开。】

## 【（引出第二种策略）混合纳米结构包含碳和不同的硅形貌。展开。】

[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

Alternatively, hybrid nanostructures with carbon and various morphologies of Si have been created to tailor the nanostructure and buffer the mechanical strain. Nanometersized Si materials combined with carbon in the form of nanoparticles [16,17], nanowires [18], nanotubes [19], and core–shell nanofibers [20–22] have shown a long cycling life as candidate anode materials because they enable rapid Li transport with free volume around nanostructures and thus facilitate strain relaxation.

【碳在锂电池中常用，优点】

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[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

Carbon is a commonly used conductive additive in lithium-ion electrode materials, which can enhance electrical conductivity and constrain the large volume change of Si to maintain structural integrity by coating the Si surface with carbon [23– 27]. 【从电极结构角度，我们选择的是将硅纳米颗粒嵌入石墨烯薄膜中。这一方法的优点和缺点。】

## 【从电极结构角度，我们选择的是将硅纳米颗粒嵌入石墨烯薄膜中。这一方法的优点和缺点。】

[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

In the electrode structure viewpoint, we adopted Sigraphene with alternating multilayered composite structures, in which Si NPs are embedded in the in-between graphene films using a dip-coating method [28]. Nanostructured Si with graphene coating has been proven to be one of the most suitable Si anode designs because the graphene can accommodate the volume expansion of Si through its void space while enhancing the electrical conductivity [29,30]; however, fabricating those anodes is complex and time-consuming because the adhesion between the active materials and current collectors is not strong enough for longer cycling.【尽管有以上缺点，我们发现了安全和更经济的解决办法】

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Despite these issues, we have found a safer and more economical rapid fabrication method to improve anode performance by carbonating polymer on Si NPs and anchoring active materials on current collectors by carbon nanotubes (CNTs). This, in turn, forms cellular carbon coating and enforces interfacial contact between active materials and substrates by CNTs.

【本文，硅纳米颗粒嵌入3D蜂窝状碳骨架，石墨烯作为电子增强物】

## 【本文，硅纳米颗粒嵌入3D蜂窝状碳骨架，石墨烯作为电子增强物】

[21] J. Chang, X. Huang, G. Zhou, S. Cui, S. Mao, J. Chen, Three-dimensional carbon-coated Si/rGO nanostructures anchored by nickel foam with carbon nanotubes for Li-ion battery applications, NANO ENERGY, 15 (2015) 679-687.

Here we report a novel anode composed of Si NPs trapped in a 3D cellular carbon framework with reduced graphene oxide (rGO) as the electronic fortifier. The polymer, in which Si NPs and GO sheets are well distributed, can be carbonized to become amorphous carbon on Si NPs loaded on nickel (Ni) foams; CNTs rooted from Ni forms, can help active materials stabilization during cycling of LIBs. Such an electrode offers several advantages as LIB anodes: (1) the approach opens a new pathway for scalable, low-cost, and binder-free fabrication of carbon-coated Si/rGO/CNT (SiGC) nanostructures using a viscous solution method, which can assist with the mass loading of active materials; (2) the carbonized polymer generates a dense cellular carbon on Si NPs/rGO surfaces and an Ni foam through the entire electrode [31], attributing to the enhanced electrode integrity; (3) carbon coating and rGO wrapping on Si NPs restrict volume expansion and improve electrical conductivity, thereby enhancing lithium ion transport and electrical contact with Si NPs during the charge/discharge processes; and (4) the catalyzed CNT growth on Ni foams using the CVD method results in a strong interface connection, thereby helping to maintain the stability of the Si anode and improve its cyclability. In addition, the fabrication method using a polymer solution process leads to the uniform distribution of Si NPs and GO sheets in the carbon framework and thus high performance in cycle stability and rate capability. More importantly, CNTs grown on the Ni foam, like nails, can stabilize the active materials on the Ni foam and result in desirable structure integration.【】【】

2.Experimental section

[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

1. Introduction

【需要高性能电池】

## 【需要高性能电池】

[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

The ever soaring market of portable electronic devices has triggered significant demand and unprecedented research interests for high performance batteries. 【锂电池主导市场】

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[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

Lithium ion batteries (LIBs) have been dominating the market for electronic devices due to its relatively higher working potential, higher specific power and energy densities, and better environmentally friendliness compared to previous battery technologies such as lead acid, nickel cadmium, and nickel metal hydride batteries [1,2]. 【锂电池发展落后于消费电子器件的发展，后者遵循摩尔定律】

## 【锂电池发展落后于消费电子器件的发展，后者遵循摩尔定律】

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However, the development of LIB technology has fallen far behind the pace of the advancement of electronic devices, which roughly follows Moore's Law [3]. 【电动车替代传统车的原因】

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[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

Electric vehicles have emerged as a potential replacement of traditional vehicles due to the limited supply of fossil fuels, long-term increasing oil prices, and the environmental benignity and sustainability of electricity generated from green energy technologies. 【为满足高能量密度需求的应用，需要发展高性能电极材料。因为提升电极容量可以降低电池的质量和体积】

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To meet the demands of increasingly sophisticated electronic devices and competitive electric vehicles, which require both high energy density and power density, tremendous efforts have been devoted to developing high-performance electrode materials in LIBs [4,5]. As increase of specific electrode capacity can greatly reduce a battery's mass and volume.

【硅被认为是下一代负极材料】

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Silicon (Si) has been regarded as a promising candidate anode material for next-generation LIBs due to its outstanding properties.【硅的优点】

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[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

The theoretical gravimetric capacity of Si is approximately 4200 mAh g-1 – more than ten times that of the commercial graphite (372 mAh g-1) [6]. In addition, Si's high natural abundance, environmental compatibility, low working potential (0–0.4 V vs. Li/Li+), and maturity in the Si semiconductor industry all make it highly desirable as an anode material for LIBs [7–9]. 【硅的缺点】

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[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

However, several intrinsic drawbacks of Si have prevented it from being widely commercialized as LIB anodes. One of the critical challenges is that the large volume expansion of Si upon lithiation (up to 400% of its original volume) can cause it to crack and pulverize, resulting in the loss of contact points between the active material and the current collector [10–12]. This large volume change also leads to the continuous formation of an unstable solid electrolyte interphase (SEI) which consumes a considerable amount of electrolyte and lithium in a battery [13]. Columbic efficiency and electrode capacity are hence lowered due to these continuous side reactions. Furthermore, the repetitive growth of the SEI may severely block the conductive pathway of electrons between Si and the current collector. These phenomena significantly decrease the electrode's integrity and electron conductivity and, ultimately, the battery's rate performance [14]. Although the pulverization of Si can be prevented by engineering nano-sized structures, the severe volume variation can still lead to the displacement and the detachment of Si from the electrode's original conductive network during cycling [15]. The above disadvantages consequently result in poor cycling and rate performance of Si electrodes. 【为解决以上硅的问题，学者提出一些策略】

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To circumvent the aforementioned problems with Si electrodes, several strategies have been proposed by researchers. 【一个有效策略是设计不同的纳米硅电极。纳米硅的优点。体系举例。】

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One of the potential solutions is to design various nanostructured Si electrodes. Nano-sized Si can better accommodate mechanical strain during lithiation and delithiation processes, and avoid cracking and fragmentation [16]. Improved cycling stability has been demonstrated with mechanically-milled nanostructured particles, [17] and chemically synthesized nanostructures including nanowires, [18] nanotubes, [19] nanoporous structures, [20] and core– shell nanostructures [21].【另一个有效提升硅的策略是构建导电网络，举例。】

## 【另一个有效提升硅的策略是构建导电网络，举例。】

[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

Another effective strategy to improve Si's performance in batteries is to build an efficient conductive network for Si. Si/C, [22] Si/Graphene, [23] Si/ CNT [24] and Si/conductive polymers [25] have been reported to exhibit enhanced performance compared to bare Si.【粘结材料】

## 【粘结材料】

[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

Recent reports of novel binding materials in electrode fabrication have also shown significant improvement in Si electrode's performance [26–28]. 【虽然这些方法展现了一些前途，合成方法上有问题。展开，举例。】

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Although these results have shown some promise, there are some significant issues regarding their synthesis. Many of these procedures are highly complex and expensive. For example, the synthesis of Si nanowires (Si NWs) usually requires complex procedures and dangerous precursors such as silane gas. Template growth of Si usually involves multi-step template synthesis and removal steps, adding to the cost of the final Si product. All these factors make it merely impossible for these materials to be widely commercialized [29].

【rGO在电化学领域被广泛研究，优点。】

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[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

rGO has been intensively studied in the electrochemical field because of its unique structure and superior physical properties including conductivity, mechanical strength and flexibility.【rGO 在锂电池中的应用。rGO 包裹碳的优点和缺点】

## 【rGO 在锂电池中的应用。rGO 包裹碳的优点和缺点】

[22] K. Feng, W. Ahn, G. Lui, H.W. Park, A.G. Kashkooli, G. Jiang, X. Wang, X. Xiao, Z. Chen, Implementing an in-situ carbon network in Si/reduced graphene oxide for high performance lithium-ion battery anodes, NANO ENERGY, 19 (2016) 187-197.

It has recently been combined with Si to form composite anode materials in LIBs [30]. Wrapping rGO around Si NPs significantly improves the electrochemical performance of Si NPs in terms of both cycle and rate performance. However, despite the good contact between rGO and Si, the composite still fails to maintain long-term stability. Reported Si–rGO composites generally suffered considerable degradation after certain cycles [31,32]. 【以上硅-rGO负极性能退化的原因归因于采用物理混合方法，没产生硅rGO 的化学键合。以及其他原因 ，rGO 无法均匀包裹硅】

## 【以上硅-rGO负极性能退化的原因归因于采用物理混合方法，没产生硅rGO 的化学键合。以及其他原因 ，rGO 无法均匀包裹硅】

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The degradation of the above Si–rGO anode can be explained by the fact that many of these composites are synthesized using physical mixing, which does not lead to strong chemical bonding between Si and rGO. In addition, the size of rGO sheets is usually much larger than that of Si NPs. These make it difficult to homogeneously wrap Si NPs with rGO. Poor wrapping of Si with rGO and migration of Si NPs from rGO during cycling result in most of the reported unsatisfactory performances.

【本文，Si–rGO–C复合物，Si纳米颗粒嵌入GO，随后引入碳。CVD和冷冻干燥】

## 【本文，Si–rGO–C复合物，Si纳米颗粒嵌入GO，随后引入碳。CVD和冷冻干燥】

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Herein, we present a highly efficient Si reduced graphene oxide carbon (Si–rGO–C) composite with good Si wrapping and thus improved electrochemical performance compared to commercial Si NPs and regular Si–rGO composite. In order to take advantage of both carbon coating and rGO wrapping, a facile freeze-drying method and a subsequent chemical vapor deposition (CVD) process were conducted using commercially available Si NPs. Si NPs were well embedded onto the GO layers, and the sponge-like composite served as the precursor of the next step carbon incorporation. The following advantages of the obtained Si–rGO–C composite were first proposed and proved based on the electrochemical performances presented in the context. First, a layer of carbon on exposed Si NPs provides enhanced electron conductivity between particles and other rGO flakes with wrapped Si NPs. Second, the carbon rods between the rGO flakes create an effective conductive network in the whole electrode, helping electrons transfer from one rGO sheet to another and eventually to the current collector. Moreover, the flexibility of rGO and the carbon network can also help restrain the electrode morphology change during cycling, avoiding significant detachment of the active material from the conductive network and helping to maintain contact between the electrode material and the current collector. Various physical and electrochemical measurement techniques were adopted to characterize the samples.【】

【】2. Experimental

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

1. Introduction

【发展具有xxx性能特点的锂电池很重要】

## 【发展具有xxx性能特点的锂电池很重要】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

Developing rechargeable lithium ion batteries (LIBs) with a long cycling life, high energy density, and excellent ratecapability is of critical importance for electric vehicles and renewable energy storage [1–4]. 【石墨理论容量低，无法满足这些需要高能量密度的应用】

## 【石墨理论容量低，无法满足这些需要高能量密度的应用】

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The traditional graphite anode with its limited theoretical capacity of $372 mA h g-1 is unable to meet the high energy needs of such applications. 【探索了不同的负极材料和方法，提升容量】

## 【探索了不同的负极材料和方法，提升容量】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

Various anode materials and strategies have been explored and proposed for increasing the specific capacity of LIBs [5–7]. 【硅的特点，被认为有前途】

## 【硅的特点，被认为有前途】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

Due to its high theoretical capacity (Li4.4Si=4200 mA h g-1) and low discharge potential ($0.5 V versus Li/Li+), silicon has been considered as an attractive and promising candidate for LIBs [8]. 【硅的缺点】

## 【硅的缺点】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

The practical application of Si-based anodes, however, is still hindered by two critical problems: the relatively low electric conductivity and the huge volume expansion ($400%) upon lithium insertion and extraction [9]. These often cause fracturing, pulverization, and loss of electrical contact, leading to rapid capacity fading and poor cycling performance [10].

【一个有效的策略，使用不同形貌的纳米硅。与块体硅相比的优点】

## 【一个有效的策略，使用不同形貌的纳米硅。与块体硅相比的优点】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

It is a common and effective strategy to adopt nanoscale silicon materials with various morphologies, including nanoparticles [11,12], nanowires [13], nanotubes [14,15], and hollow spheres [16,17]. Compared to bulk silicon, such nanostructured Si is able to accommodate elevated mechanical stress, resulting in prolonged cycling stability. 【纳米硅的缺点】

## 【纳米硅的缺点】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

Nanostructured silicon materials, however, still suffer from poor electric conduction [18].【优化纳米硅，将纳米硅和不同导电基体融合，举例】

## 【优化纳米硅，将纳米硅和不同导电基体融合，举例】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

Further optimization is achieved by incorporating nano-silicon materials with various conductive matrixes, such as graphene [19,20], carbon nanotubes [21], and carbon [22,23] to form core-shell and yolk-shell nanocomposites [24]. The most promising carbon coating strategy has been explored to promote the electrochemical performance [25–28]. The obvious advantage of carbon shells is intensive improving the overall electrical conductivity of the Si-based anodes. In addition, the introduction of such a carbon shell plays a key role in alleviating the agglomeration of nano-silicon particles [29,30]. 【但是，目前包碳方法哪些，存在什么问题。导致硅纳米颗粒的倍率不行】

## 【但是，目前包碳方法哪些，存在什么问题。导致硅纳米颗粒的倍率不行】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

However, the well-established carbon coating methods are based on the chemical vapor deposition, hydrothermal of carbohydrates and polymerization of phenolic resin from sol–gel process, all of them unfortunately result in the compact carbon shells without open and connecting mesopore channels for fast transport of Li+ ions between the electrolyte and silicon. This could be the reason that Si NPs covered with carbon layer matrix as an anode present unsatisfactory rate-capability [18,29,31]. 【进一步的，SEI的影响被忽略，SEI很难控制。展开描述。】

## 【进一步的，SEI的影响被忽略，SEI很难控制。展开描述。】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

Furthermore, the crucial impact of SEI is usually be neglected and the formation of SEI is hard to control. Importantly, if a stable and compact SEI is constructed, the electrolyte molecules would not penetrate through SEI layer into the active material for further growth, thereby avoiding continual lithium loss and ensuring higher Coulombic efficiency.

【本文，Si@介孔碳，包含大量孔洞】

## 【本文，Si@介孔碳，包含大量孔洞】

[23] J. Yang, Y.-X. Wang, S.-L. Chou, R. Zhang, Y. Xu, J. Fan, W.-x. Zhang, H.K. Liu, D. Zhao, S.X. Dou, Yolk-shell silicon-mesoporous carbon anode with compact solid electrolyte interphase film for superior lithium-ion batteries, NANO ENERGY, 18 (2015) 133-142.

Here, aiming at outstanding cycling stability and ratecapability, we report a novel large-void-containing Si@mesoporous carbon yolk-shell structure from commercial silicon NPs for LIB anodes. This unique design has multiple attractive advantages: (i) the special design of the void spaces (10 and 50 nm) between the silicon NPs and the mesoporous carbon shells highlights the superiority of the unique yolk-shell structure and allows us to optimize the cycling stability while maintaining a reasonable storage capacity; (ii) the open-ended mesoporous carbon shells with accessible channels are able to facilitate the fast diffusion of Li+ ions, and guarantee the full immersion of active materials in the electrolyte, thus contributing to excellent rate capability; (iii) the mesoporous carbon shells are beneficial for the formation of a homogeneous and compact SEI-layer film on the external surface, retaining the internal void space for silicon yolk expansion and rendering superior capacity retention at a high current density. With this design, such elegant mesoporous carbon-encapsulated Si yolk-shell NPs address the challenges of rapid capacity decay and unsatisfied rate capability for Si-anodes, and deliver a high reversible capacity of $1000 mA h g-1 with outstanding cyclability during 400 long-deep cycles, as well as superior rate-capability at a very high current density of 8.4 A g-1.【】

【】2. Experimental section

[24] J. Lu, D. Wang, J. Liu, G. Qian, Y. Chen, Z. Wang, Hollow double-layer carbon nanocage confined Si nanoparticles for high performance lithium-ion batteries, NANOSCALE ADVANCES, 2 (2020) 3222-3230.

1. Introduction

【锂电池是最重要的储能器件，优点】

## 【锂电池是最重要的储能器件，优点】

[24] J. Lu, D. Wang, J. Liu, G. Qian, Y. Chen, Z. Wang, Hollow double-layer carbon nanocage confined Si nanoparticles for high performance lithium-ion batteries, NANOSCALE ADVANCES, 2 (2020) 3222-3230.

Lithium-ion batteries (LIBs) have been considered as one of the most important energy storage devices owing to their high energy density and lack of the memory effect.1,2 【商业负极的问题，容量】

## 【商业负极的问题，容量】

[24] J. Lu, D. Wang, J. Liu, G. Qian, Y. Chen, Z. Wang, Hollow double-layer carbon nanocage confined Si nanoparticles for high performance lithium-ion batteries, NANOSCALE ADVANCES, 2 (2020) 3222-3230.

However, the commercial anode material, graphite, has got a low theoretical specific capacity (LiC6, 372 mA h g-1), and it is challenging to meet the ever-growing demands of portable electronics and electric vehicles.3,4 【需要发展替代商业负极的高容量电极材料】

## 【需要发展替代商业负极的高容量电极材料】

[24] J. Lu, D. Wang, J. Liu, G. Qian, Y. Chen, Z. Wang, Hollow double-layer carbon nanocage confined Si nanoparticles for high performance lithium-ion batteries, NANOSCALE ADVANCES, 2 (2020) 3222-3230.

Therefore, to improve the energy density of current LIBs, it is urgent to develop a high specific capacity electrode material instead of commercial graphite anodes. 【硅被认为有前途，优点】

## 【硅被认为有前途，优点】

[24] J. Lu, D. Wang, J. Liu, G. Qian, Y. Chen, Z. Wang, Hollow double-layer carbon nanocage confined Si nanoparticles for high performance lithium-ion batteries, NANOSCALE ADVANCES, 2 (2020) 3222-3230.

Among the emerging anode materials, silicon (Si) is a promising candidate for next-generation LIB anodes due to its high specific capacity (Li15Si4, 3579 mA h g-1), low Li-uptake voltage (-0.5 V vs. Li/Li+) and natural abundance (second largest resource).5–7 【硅的缺点】

## 【硅的缺点】

[24] J. Lu, D. Wang, J. Liu, G. Qian, Y. Chen, Z. Wang, Hollow double-layer carbon nanocage confined Si nanoparticles for high performance lithium-ion batteries, NANOSCALE ADVANCES, 2 (2020) 3222-3230.

Unfortunately, the application of Si-based anodes has been impeded because of its inferior intrinsic conductivity and large volume effect (~300%) during lithiation.8,9 The huge volume variation produces high mechanical stress that causes the powdering of the electrode materials and formation of an unstable solid electrolyte interphase (SEI), resulting in fast capacity fading and poor coulombic efficiency.10

【发展大量策略解决硅的问题】

## 【发展大量策略解决硅的问题】

[24] J. Lu, D. Wang, J. Liu, G. Qian, Y. Chen, Z. Wang, Hollow double-layer carbon nanocage confined Si nanoparticles for high performance lithium-ion batteries, NANOSCALE ADVANCES, 2 (2020) 3222-3230.

Researchers have developed various strategies to resolve simple and multi-strategic coupling methods to mitigate the volume change of Si and form a stable SEI layer. 【金属有机框架材料如何制备的，在储能中的应用潜力。MOFs 的文献报道。MOFs 的优点。因此，硅可以和MOFs组合】

## 【金属有机框架材料如何制备的，在储能中的应用潜力。MOFs 的文献报道。MOFs 的优点。因此，硅可以和MOFs组合】

[24] J. Lu, D. Wang, J. Liu, G. Qian, Y. Chen, Z. Wang, Hollow double-layer carbon nanocage confined Si nanoparticles for high performance lithium-ion batteries, NANOSCALE ADVANCES, 2 (2020) 3222-3230.

Metal–organic framework compounds are constructed using metal ions/ clusters and organic ligands through coordination bonding, showing great potential in energy storage.22 The application of MOFs as a derivatized carbon shell in the field of energy storage has been reported.23,24 MOFs not only heighten the mechanical integrity of materials, but also derivate the heteroatom doping and porous structure of the carbon shell, which increases the conductivity, allowing rapid transfer of lithium ions. Furthermore, MOFs can form hollow structures through heterogeneous shrinkage during high temperature heat treatment.25 Thus, it is foreseeable that the combination of Si and MOFs will be a suitable method for the preparation of multifunctional materials.

【本文，硅空心碳纳米笼，碳层通过ZIF制备】

## 【本文，硅空心碳纳米笼，碳层通过ZIF制备】

[24] J. Lu, D. Wang, J. Liu, G. Qian, Y. Chen, Z. Wang, Hollow double-layer carbon nanocage confined Si nanoparticles for high performance lithium-ion batteries, NANOSCALE ADVANCES, 2 (2020) 3222-3230.

Herein, a Si-hollow double-layer hybrid carbon nanocage (Si@H-NC@GC) has been successfully prepared based on electrostatic self-assembly, seed-induced growth and heterogeneous shrinkage. The double hybrid carbon shells are obtained during heat treatment where a N-doped microporous carbon (NC) inner shell is derived from ZIF-8 and a mesoporous graphitic carbon (GC) outer shell is derived from ZIF-67.26,27 At the same time, the material is heterogeneously contracted during the high-temperature treatment to form the internal hollow, denoted as Si@H-NC@GC. In the novel structure of Si@H-NC@GC, GC has the effect of stabilizing the SEI, good electrical conductivity and rapid transfer of lithium ions, while hollow NC effectively accommodates the volume expansion of Si nanoparticles and increases lithium storage active sites.28–30 Si@H-NC@GC displays a high reversible capacity of 1052 mA h g-1 (92.5% capacity retention) after 550 cycles at a high current density of 1 A g-1 when used in LIBs. With the successful synthesis of Si@H-NC@GC, the employment of MOFs to synthesize double-layer and even multi-layer anode materials from self-templates will pave a new way for the manufacture of various anodes with high capacity and superior stability for next-generation LIBs.【】【】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

1. Introduction

【储能器件背景】

## 【储能器件背景】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

Electrochemical energy storage has attracted much attention and received increasing research interest worldwide for a variety of applications such as portable electronic devices and electric vehicles.【锂电池背景】

## 【锂电池背景】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

The lithium-ion battery (LIB) has become one of the most promising energy storage devices because of its relatively high energy density and reasonable rate capability 1,2. 【石墨电极背景】

## 【石墨电极背景】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

Since the development of the first commercial LIB in the early 1990s by Sony Energytech 3, graphite has been widely used as the anode material. 【石墨的缺点】

## 【石墨的缺点】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

However, the specific capacity of graphite is relatively low due to its intercalation mechanism 4. 【为提高能量密度，需要高容量电极材料】

## 【为提高能量密度，需要高容量电极材料】

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To further increase the energy density of LIBs for more demanding applications, novel electrode materials with higher specific capacity are required. 【硅的容量优势，相比石墨】

## 【硅的容量优势，相比石墨】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

Silicon is an exciting and promising alternative anode material to replace graphite due to its highest theoretical specific capacity of 3579 mA h g -1 among all of the alloy type anodes, which is almost ten times that of graphite anode (372 mA h g -1) 5. 【硅的问题】

## 【硅的问题】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

However, two major scientific and technical challenges have hindered its practical applications. First, due to the alloying mechanism, a large number of lithium atoms are inserted into Si and chemical bonds between Si atoms are broken. The structural pulverization induced by the large volume change (>300% at room temperature) during Li insertion and extraction leads to the loss of electrical contact between the active material and the current collector, eventually resulting in capacity fading and short battery lifetime 6,7. Second, due to the volume expansion and shrink, a thick solid-electrolyte interphase (SEI) layer can be formed deriving from the irreversible side reactions with the organic electrolyte 8. This causes the degradation of battery performance due to the consumption of electrolyte and lithium ions, the electrically insulating nature and the rather long lithium diffusion length through the thick SEI 9. 【因此，需要解决硅的问题】

## 【因此，需要解决硅的问题】

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Thus, to make silicon a good anode candidate for LIBs, two major issues must be settled: minimizing the degradation of the mechanical integrity and maintaining the stability of the SEI.

【很多工作被开展以解决硅的问题】

## 【很多工作被开展以解决硅的问题】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

Numerous works have been conducted on the research of silicon anode to meet the above challenges. 【降低尺寸到纳米级别消除机械破裂。纳米结构举例。这些纳米结构优点。】

## 【降低尺寸到纳米级别消除机械破裂。纳米结构举例。这些纳米结构优点。】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

Reducing the size of active materials into nanoscale could be one efficient way to alleviate the mechanical fracture. Various structures synthesized by different methods have been proposed, including nanoparticles 10,11 , nanotubes 12,13, nanowires 14,15, nanospheres 16, core-shell nanofibers 17,18 and thin films 19. These nanostructures have demonstrated superior performance compared to bulk silicon because of their ability to release the strain. Moreover, nanostructured silicon has settled the issue of the active material detachment from the current collector, resulting in significant improvement of electrochemical cycling. 【硅的缺点，和SEI相关】

## 【硅的缺点，和SEI相关】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

In spite of this, the surface of silicon is still directly exposed to the electrolyte and therefore unstable SEI is still formed.

【为解决硅直接接触形成SEI这个问题，表面涂层】

## 【为解决硅直接接触形成SEI这个问题，表面涂层】

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To tackle this problem, conformal coatings on silicon anode have been explored. 【无定形碳的优点】

## 【无定形碳的优点】

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Amorphous carbon is a good candidate and often used as a coating layer because of the relatively simple and inexpensive fabrication process, and more importantly it is both electronically and ionically conducting 20. 【近年来，ALD制备Al2O3 涂层。ALD描述】

## 【近年来，ALD制备Al2O3 涂层。ALD描述】

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Recently Al2O3 was used as a coating material fabricated by atomic layer deposition (ALD) 21–23. ALD is a mature technique for conformal coatings. The process is quite easy and the thickness of the deposited materials can be well controlled. 【硅表面涂层的工作。涂层的作用】

## 【硅表面涂层的工作。涂层的作用】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

Metallic and SiOx coatings have also been studied 9,24. These coatings prevent silicon from being exposed directly to the electrolyte. As a result, a thin SEI layer can be formed. 【硅表面涂层存在的问题，开裂】

## 【硅表面涂层存在的问题，开裂】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

Nevertheless, in some cases, these coatings will fracture upon the volume expansion of silicon, and therefore the silicon surface will be exposed to the electrolyte again. 【硅和涂层之间需要空隙。很多此方面的报道。报道所达到的效果】

## 【硅和涂层之间需要空隙。很多此方面的报道。报道所达到的效果】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

To maintain a stable SEI layer on the surface of the electrodes, void space around silicon inside the coatings is necessary to be built. Recently, numerous relevant structures have been designed to fulfil this requirement 25–35. The free volume allows silicon to expand without rupturing the outer coating layers, and thus the stable SEI is possibly maintained.

【本文，空心碳纳米球/硅/Al2O3核壳结构】

## 【本文，空心碳纳米球/硅/Al2O3核壳结构】

[25] B. Li, F. Yao, J.J. Bae, J. Chang, M.R. Zamfir, L. Duc Toan, P. Duy Tho, H. Yue, Y.H. Lee, Hollow carbon nanospheres/silicon/alumina core-shell film as an anode for lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

In this report, hollow carbon nanospheres/silicon/alumina (CNS/Si/Al2O3) core-shell structure was fabricated to overcome the above issues. The CNS thin film substrate was fabricated by electrophoretic deposition technique, followed by the deposition of amorphous Si and Al2O3 thin layer. The CNS/Si/Al2O3 hollow structure not only provided a conductive film connecting a series of CNSs into 3D network, but also accommodated large silicon volume expansion due to the existence of void space provided by CNSs. Moreover, the electrochemical performance was further improved by the reduced SEI formation due to the coated Al2O3 layer and the efficient electron transport facilitated by the surface-to-surface contact mode.【】【】

2.Results

[26] L.Y. Yang, H.Z. Li, J. Liu, Z.Q. Sun, S.S. Tang, M. Lei, Dual yolk-shell structure of carbon and silica-coated silicon for high-performance lithium-ion batteries, SCIENTIFIC REPORTS, 5 (2015).

1. Introduction

【石油价格，全球变暖引出锂电池，应用】

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Recently, the rapidly rising price of petroleum and growing concerns about global warming have brought a great deal of attention to lithium-ion batteries with high capacity and energy density for future electric vehicles and portable electronics1–6. 【锂电池性能需要进一步提高】

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Nevertheless, the performance of currently commercialized lithium-ion batteries must be further improved to meet the increasing demand for high energy storage capacity. 【因此，需要发展高容量电极】

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Therefore, novel electrode materials with higher capacity and higher power density are urgently needed.

【硅的优点】

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Among the various anode materials, silicon is one of the most promising candidates due to its high theoretical capacity (~3580 mA h g−1, Li15Si4) among alloy type anode materials and relatively low discharge potential (~0.4 V vs. Li/Li+)4,5. 【硅的缺点】

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Despite these advantages, silicon anodes have two major disadvantages that have prevented their widespread use. First, the large volume changes (~300%) in silicon upon insertion and extraction of lithium-ions lead to severe electrode pulverization, which results in the loss of contact between the active materials and the current collector, leading to rapid capacity fading. Second, the continual pulverization of silicon during cycling causes the electrode surface to be cyclically exposed to the electrolyte. This generates continual formation of solid-electrolyte interphase (SEI) films, contributing to capacity fading and poor coulombic efficiency.

【为解决硅的这些问题，许多工作致力于发展硅纳米结构，举例，硅复合物】

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In an attempt to overcome these limitations of silicon, much attention has been devoted to the design and fabrication of silicon nanostructures, such as silicon nanowires6–8 and nanotubes9,10, three-dimensional (3D) porous silicon11–14, and silicon in composites with carbon or oxides15–20. 【取得了令人鼓舞的结果。举例，硅复合物。性能提升原因】

## 【取得了令人鼓舞的结果。举例，硅复合物。性能提升原因】

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Encouraging results have been achieved through these efforts. In particular, Si/SiOx composites21,22, Si/SiO2/C23–25, and yolk-shell structured Si/C26–28 have demonstrated excellent electrochemical performance. These performances can be ascribed to the C or SiOx shell on the outside of the silicon, which can offer a static surface for the formation of a thin and stable SEI, preserving the anode from irreversible reaction with the electrolyte29–31. Additionally, the existing hollow structures in the yolk-shell structure can provide extra space for the volume expansion of silicon, which guarantees the structural integrity of the electrode29–31. These distinguishing features provide enlightened guidance for nanostructured design of high-performance silicon batteries.

【本文，硅表面包裹SiO2，随后包裹碳层】

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In this paper, we uniformly coated silicon with a silica layer via the Stöber method and then coated Si/SiO2 composites with a carbon layer. Taking advantage of the inhomogeneous nature of silica shells prepared by the Stöber method32, proper etching conditions were chosen for selectively etching the SiO2 shell by means of a hydrofluoric acid (HF) treatment. By this process, a small portion of the outer layer and a large portion of the interior layer of the SiO2 shell were removed. The novel Si/void/SiO2/void/C structures were obtained. The advantages of dual yolk-shell silicon structures lie in the presence of internal void spaces and the mechanically strong SiO2 layer, which limits the degree of volume expansion of silicon during lithiation. In addition, the amorphous SiO2 and C intrinsically have advantages as shell materials due to their chemical inertness, porous structure, and size-selective permeability33–37. They can provide a double barrier to prevent the electrolyte from reaching the surface of the silicon nanoparticles and protect the anode from subsequent irreversible reaction with the electrolyte31. With the help of the dual yolk-shell structures, the capacity of the silicon half-cell could be stabilized at 956 mA h g−1 at 0.46 A g−1 after 430 cycles with capacity retention of 83%, while the capacity of the Si/C core-shell structures rapidly decreases in the first ten cycles under the same experimental conditions. Therefore, such dual yolk-shell structures can also be extended to other battery materials that undergo large volume changes.【】【】

2.Results