Mesoporous Graphene Hosts for Dendrite-Free Lithium Metal Anode in Working Rechargeable Batteries

He Liu^{1,2} · Xinbing Cheng³ · Rui Zhang³ · Peng Shi³ · Xin Shen³ · Xiaoru Chen³ · Tao Li³ · Jiaqi Huang^{1,2} · Qiang Zhang³

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Abstract

Lithium (Li) metal anode has received extensive attentions due to its ultrahigh theoretical capacity and the most negative electrode potential. However, dendrite growth severely impedes the practical applications of the Li metal anode in rechargeable batteries. In this contribution, a mesoporous graphene with a high specific surface area was synthesized to host the Li metal anode. The mesoporous graphene host (MGH) has a high specific surface area (2090 m²/g), which affords free space and an interconnected conductive pathway for Li plating and stripping, thus alleviating the volume variation and reducing the generation of dead Li during repeated cycles. More importantly, the high specific surface area of MGH efficiently reduces the local current density of the electrode, which favors a uniform Li nucleation and plating behavior, rendering a dendrite-free deposition morphology at a low overpotential. These factors synergistically boost the Li utilization (90.1% vs. 70.1% for Cu foil) and life span (150 cycles vs. 100 cycles for Cu foil) with a low polarization of MGH electrode at an ultrahigh current of 15.0 mA/cm². The as-prepared MGH can provide fresh insights into the electrode design of the Li metal anode operating at high rates.

Keywords Lithium metal anode · Mesoporous graphene hosts · Dendrite-free plating behavior · Working rechargeable batteries · Composite electrode

Introduction

Lithium (Li) ion batteries (LIBs) have realized commercial applications in portable devices, including smart phones, computers, and electric vehicles [1]. Compared with conventional fuel vehicles, electric vehicles are ready to innovate nearly every aspect of transportation, such as potentially reducing costs and carbon emissions and even changing driving habits [2]. A question pertaining to electric vehicles

is how far they can go, which is mostly determined by the cell systems. Therefore, a high energy density battery is the primary demand for a long-range electric vehicle [3]. Compared with the graphite anode in commercial LIBs, the Li metal anode has a high theoretical capacity of 3860 mAh/g (372 mAh/g for the graphite anode) and the most negative potential of −3.04 V vs. SHE (usually 0.1 V vs. Li⁺/Li for the graphite anode) [4, 5]. Li metal batteries (LMBs) exhibit a high energy density and have the potential for practical applications in the top-end electronics, unmanned aerial vehicles, and electric vehicles [6–9].

The substitution of graphite anodes in commercial LIBs by Li metal anodes leads to a paradigm shift in the energy storage mechanism. In LIBs, Li ions can be intercalated into/deintercalated from the intercalated graphite compounds during repeated charge/discharge processes. By contrast, the “hostless” nature of conversion-type Li metal anode indicates that the electrode suffers from infinite relative volume change during each cycle [10, 11]. This “hostless” plating process induces considerable internal stress fluctuation of electrodes and deteriorates battery performance [12]. Another key issue of LMBs is the uncontrollable Li dendrite growth caused by

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✉ Qiang Zhang
zhang-qiang@mails.tsinghua.edu.cn

¹ School of Materials Science and Engineering, Beijing Institute of Technology, Beijing 100081, China

² Advanced Research Institute of Multidisciplinary Science, Beijing Institute of Technology, Beijing 100081, China

³ Beijing Key Laboratory of Green Chemical Reaction Engineering and Technology, Department of Chemical Engineering, Tsinghua University, Beijing 100084, China

the ultrahigh reactivity of Li metal, unstable solid–electrolyte interphase (SEI), and inhomogeneous current and Li-ion flux distribution on the anode surface [13, 14]. These issues collectively contribute to the short life span, reduced Coulombic efficiency (CE), low energy density, and poor safety performance of LMBs [15, 16].

Strategies are proposed to alleviate dendrite growth of the Li metal anode, such as constructing a stable SEI layer on the surface of Li metal electrode by regulating the components of electrolytes/additives and ex situ techniques [17–27], and utilizing solid-state electrolytes with high mechanical modulus [28–32]. The large volume variation in the “hostless” Li metal anode can only be remitted by providing a host framework to accommodate the deposited Li metal. Currently, 3D conductive metal/alloy current collectors [33, 34] and porous carbon-based frameworks [35, 36], as well as nonconductive 3D porous hosts [37], are proposed. Among them, porous carbon materials are regarded as superior hosts for Li deposition due to their high electronic conductivity, lightweight, superior electrochemical stability, high mechanical flexibility, and low cost [38]. Aside from providing space for Li deposition, 3D conductive host with a high specific surface area can also reduce the local current density of the electrode and delay the onset time of dendrite formation according to the classical Sand’s time model [39, 40]. Modification on the surface chemistry of carbon hosts can also regulate the nucleation and plating behaviors of Li metal, thus achieving a dendrite-free Li deposition morphology [41, 42]. Therefore, the use of efficient carbon hosts is highly expected to simultaneously address the large volume change and dendrite growth issues of working Li metal anodes.

In this contribution, a mesoporous graphene with a high specific surface area was prepared to host the Li metal anode. The mesoporous graphene host (MGH) can provide free space and an interconnected conductive pathway for Li plating and stripping, therefore alleviating the volume variation and reducing the generation of dead Li during repeated cycles. In addition, the higher specific surface area of MGH than that of flat Li foil anode efficiently reduces the local current density of the electrode, which favors a uniform Li nucleation and plating behavior, rendering a dendrite-free deposition morphology. These factors synergistically boost Li utilization, safety performance, and life spans of the Li metal anode at an ultrahigh current of 15.0 mA/cm². Therefore, the as-prepared MGH helps achieve a high-rate electrochemical performance of the Li metal anode, which is promising in practical applications.

Experimental Section

Synthesis of Mesoporous Graphene

The mesoporous graphene with an ultrahigh specific surface area was obtained via the chemical vapor deposition method by direct carbonization of CH₄ and MgO template [43]. The MgO template was obtained via the hydrothermal reaction, with commercial MgO powders as raw materials. The tube containing the as-obtained mesoporous MgO template was heated to 950 °C in a tube furnace at a heating rate of 10 °C/min with the protection of flowing Ar atmosphere. Then, the mixture of CH₄ and H₂ was simultaneously introduced into the fluidized bed reactor. CH₄ was decomposed into carbon and deposited onto the MgO template. Subsequently, the furnace was naturally cooled to room temperature with the protection of Ar and H₂. The as-prepared products were purified by 6.0 mol/L hydrochloric acid at 80 °C for 24.0 h to remove the MgO templates. Eventually, the mesoporous graphene was collected after washing, filtering, and freeze-drying.

Characterizations

The morphology and microstructures were investigated using a scanning electron microscope (SEM, JEOL JSM-7401F, Japan) operated at 3.0 kV and a transmission electron microscope (TEM, JEOL-2010, Japan) with an accelerating voltage of 120 kV. N₂ isotherms were obtained with an Autosorb-IQ₂-MPC system (Quantachrome, USA). The specific surface area was calculated by the multipoint Brunauer–Emmett–Teller (BET) method.

Electrochemical Measurements

Standard CR2025 coin cells with a Celgard 2400 separator were assembled in an argon-filled glove box with H₂O and O₂ contents both below 10^{−6}. All electrochemical measurements were conducted on a Neware multichannel battery cycler. For the half-cell test, the slurry of MGH powder and polyvinylidene fluoride binder (mass ratio of 85:15) in *N*-methyl-2-pyrrolidone was coated on a Cu foil and dried in vacuum. The as-obtained MGH-coated Cu foils were punched into Φ 13 mm disks as the working electrodes. Li metal foils with a thickness of 0.5 mm and a diameter of 16.0 mm (China Energy Lithium Co., Ltd.) were utilized as the counterelectrodes. Cu foils were used as the working electrodes in the control samples. The assembled Li||Cu half-cells were used to investigate the nucleation overpotential, plating/stripping behavior, and CEs of Li metal electrode with the electrolyte of 3.0 mol/L lithium bis(fluorosulfonyl) imide in 1,2-dimethoxyethane. The dosage of the electrolyte

in each cell was 60 μL . The concentrated electrolyte was selected to construct a stable SEI, excluding the effect of SEI instability on the function of MGH. In the long-term stability test, a pre-cycle at 0.5 mA/cm^2 was conducted to construct a stable SEI on the surface of the electrode. The impedance measurements were conducted on a Solartron 1470E electrochemical workstation (Solartron Analytical) over the frequency ranging from 100 kHz to 0.01 Hz.

Results and Discussion

Characterization of Mesoporous Graphene

The as-obtained mesoporous graphene exhibits a porous and hexagonal morphology (Fig. 1a, b). The lateral size

and thickness of graphene layers are approximately 2 μm and 50 nm, respectively (Fig. S1). The few-layered graphene flakes wrinkle into a honeycombed framework with in-plane nanocages in the size of 5–10 nm (Fig. 1c). Numerous defective sites are generated at the corners or edges of these nanocages, which favor the absorption of Li ions, and correspond to the nanocages in the TEM image. The BET surface area of 3D porous graphene reaches as high as 2090 m^2/g (Fig. 1d), which is expected to considerably reduce the local current density of the Li metal electrode and suppress dendrite growth at a high current rate.

Fig. 1 Characterizations of mesoporous graphene: **a** SEM image, **b** low-resolution and **c** high-resolution TEM images of MGH materials, **d** N_2 sorption isotherms of MGH materials and corresponding pore size distribution

Li Nucleation and Deposition Behavior on MGH Electrode

The Li nucleation overpotential on MGH and Cu foil electrodes at different current densities is compared to evaluate the Li nucleation and growth behavior. The overpotentials between these two electrodes during electrochemical deposition are the nucleation and mass transfer overpotentials. The nucleation overpotential is mainly influenced by the surface chemistry of the electrode, whereas the mass transfer overpotential depends on the applied current density and Li-ion migration properties. The stable platform voltage in the voltage–time profiles represents the mass transfer overpotential, and the value increases with large current densities (Fig. 2a, b). The nucleation overpotential is applied to evaluate the nucleation resistance and defined by the difference between mass transfer overpotential and sharp tip voltage [44]. Obviously, the Li nucleation overpotentials on MGH electrode are lower than those on Cu foil (Fig. 2). High nucleation overpotentials of 20–44 mV are observed on Cu foil even at low current rates ranging from 0.01 mA/cm² to 0.1 mA/cm². By contrast, a low nucleation overpotential of 10 mV is observed on MGH electrode at 0.1 mA/cm². The nucleation difference between these two electrodes can be attributed to their surface properties [45–48]. The ultrahigh specific surface area of MGH (2090 m²/g) compared with that of flat Cu foil effectively decreases the local current density of the electrode at the same bulk current density. The numerous defective sites generated at the corners or edges of graphene nanocages also favor the absorption and nucleation of Li ions, thus reducing the nucleation resistance of metallic Li. The Li nucleation overpotential on MGH is only 17 mV even at a current of 1.0 mA/cm²; however, this value increases to 79 mV on Cu foil. This finding further indicates that MGH with a high specific surface area can

efficiently regulate the nucleation behavior of the Li metal anode at high current densities.

SEM images are taken to compare the Li nucleation and deposition behavior on MGH and Cu foil electrodes visually. Striking differences in Li plating morphology can be observed between these two electrodes (Figs. 3 and S2). Dendritic Li deposition is observed on Cu foil even at a low capacity of 0.1 mAh/cm², and the deposits are distributed on Cu foil randomly (Figs. 3a and S2a). This nonuniform deposition behavior is induced by the inhomogeneous local current density on rough Cu foil surface. The nonuniform surface chemistry of Cu foil also leads to the inhomogeneity in Li-ion concentration distribution, resulting in the uncontrollable dendritic Li deposition on Cu foil electrode. The dendrites grow longer and thicker with the increase in capacity (Fig. 3b, c). The diameter of these dendrites ranges from several to a dozen micrometers at 2.0 mAh/cm², which leads to poor efficiency and high safety risk of LMBs.

By contrast, a uniform Li deposition behavior without any dendrite can be observed on MGH electrode (Fig. S2d–f). A porous framework is constructed by randomly stacking hexagonal graphene layers in MGH before Li plating (Fig. S2d). The deposited Li fills in the interspace between graphene layers at 0.5 mAh/cm² (Fig. S2e). Notably, a thin Li layer covers the MGH electrode uniformly with the increase in plating capacity to 2.0 mAh/cm², rather than forming Li dendrites (Figs. 3f and S2f). Further observation of morphological evolution under high magnification clearly depicts the deposition process (Fig. 3d–f). Li particles with dozens of nanometers in size uniformly distribute on the MGH surface at a low plating capacity of 0.1 mAh/cm² (Fig. 3d). The MGH is wrapped by more densely and uniformly distributed Li nuclei as the capacity is increased to 0.5 mAh/cm² (Fig. 3e). The uniform nucleation sites in MGH electrodes further contribute to the uniform deposition behavior, leading to the propagation and coalescence of these particles

Fig. 2 Comparison of the nucleation overpotentials between Cu foil and MGH electrodes. The voltage–time curves during Li nucleation at different current densities on **a** Cu foil and **b** MGH electrodes. **c**

The Li nucleation overpotentials on Cu foil surface and MGH electrodes at different current densities

Fig. 3 Li deposition morphology on MGH and Cu foil electrodes, SEM images of Li deposits on **a–c** Cu foil and **d–f** MGH electrodes after plating different capacities of Li at a current of 0.5 mA/cm^2

with the increasing lithiation capacity. Finally, a smooth Li depositing layer covers the MGH surface and fills the gaps (Fig. 3f). The contrast of Li deposition morphological evolution between MGH and Cu foil electrodes underscores the significance of the high specific surface area of MGH materials in reducing the local current density and regulating the nucleation and deposition behavior of Li ions.

Long-Term Stability of the MGH Electrode

The CE and long-term stability of the MGH and Cu foil electrodes were investigated. Obviously, the MGH electrode exhibits a higher CE and longer life span than the Cu foil electrode at different current densities at 2.0 mAh/cm^2 (Fig. 4a). At rates of 2.0 and 5.0 mA/cm^2 , the MGH electrode exhibits high CEs of 98.9% and 98.4% for 340 and 280 cycles, respectively, indicating highly reversible Li in the MGH electrode. By contrast, the life span of the Cu foil electrode under the same conditions is half that of the MGH electrode with lower CEs (97.0% and 96.0% at rates of 2.0 and 5.0 mA/cm^2 , respectively). Notably, high CEs of 97.7% and 90.1% can be retained in the MGH electrode at ultrahigh rates of 10.0 and 15.0 mA/cm^2 , respectively, for approximately 150 cycles. However, the Cu foil electrode exhibits a poor electrochemical performance with fluctuant CEs and sharply reduced life span at such high currents. These results can be attributed to the uncontrollable dendrite growth and fragile SEI layers on the Cu foil electrode. The higher CE and longer service time of the MGH electrode than the Cu foil electrode validate the superiority of the high specific

surface area in decreasing local current density and boosting Li utilization, particularly at high currents.

The MGH electrode exhibits low polarization during the entire cycling process (Fig. 4b, c). In Cu foil electrode, polarization increases from 150 to 230 mV after 150 cycles. This process is accompanied by the pulverization and volume expansion of the Li metal anode, as well as the accumulation of dead Li, leading to the increase in Li-ion transfer resistance. In comparison, the hysteresis of the MGH electrode increases from 100 to 150 mV during the initial 70 cycles to form a stable SEI layer on the Li metal electrode. Afterward, a stable plateau is maintained for 180 cycles, indicating the excellent long-term stability of the MGH electrode. In addition, the hysteresis of the MGH electrode during the fifth cycle is relatively lower than that of the Cu foil electrode at different rates (Fig. S3). In particular, a 50 mV lower hysteresis can be observed in the MGH electrode than in the Cu foil electrode at 15.0 mA/cm^2 . On the one hand, MGH can afford a free space for Li deposition and alleviate the volume variation during cycling. On the other hand, the conductive framework of MGH provides an interconnected pathway for electron transfer, alleviating the formation of dead Li. The considerably decreased local current density by the high specific surface area [40] of the MGH electrode also contributes to the stable cycling behavior.

Electrochemical impedance spectroscopy was conducted to investigate the Li-ion diffusion behavior in different electrodes. The MGH electrode exhibits a lower and more stable SEI resistance (semicircle at mid-frequency) than the Cu foil electrode (Fig. S4), indicating that a stable and ionically

Fig. 4 Long-term cycles of different electrodes, **a** CE of MGH and Cu foil electrodes at different rates with a capacity of 2.0 mAh/cm², **b** voltage profiles of MGH and Cu foil electrodes at 5.0 mA/cm², **c**

comparison of the polarization voltage between MGH and Cu foil electrodes at 5.0 mA/cm²

conductive SEI layer is constructed between the interfaces. These advantages render a stable and low-voltage plateau in the MGH electrode upon stabilization of the SEI layer.

The morphologies of the MGH and Cu foil electrodes after long cycles at different current densities were investigated. For the MGH electrode, the deposited Li metal uniformly covers the hexagonal graphene layers and dendrites do not grow in their gaps even after 200 cycles at high rates of 2.0 and 5.0 mA/cm² (Fig. S5a, b). The uniform deposition behavior can be retained for 100 cycles at ultrahigh currents of 10.0 and 15.0 mA/cm² (Fig. S5c, d). The conductive MGH with high specific surface area also favors a uniform

stripping behavior during long cycles, mitigating the formation of dead Li and facilitating the generation of superior SEI film (Fig. S6a–d). While for the Cu foil electrode, random and loose dendrites are observed during long cycles even at 2.0 mA/cm² (Fig. S5e–h). These loose structures are easily detached from the conductive Cu foil substrate during the stripping process, leading to the formation of dead Li (Fig. S6e–h). The accumulation of dead Li and ruptured SEI layers leads to the increase in ionic transfer resistance (Fig. S4) and polarization voltage (Fig. 4c). These results indicate that MGH can not only regulate the Li nucleation and deposition behavior at the initial stage but also maintain

the stability of the framework and work well during long repeated cycles.

Conclusions

Conductive MGH materials with a high specific surface area were designed to host the Li metal anode with a high-rate electrochemical performance. The following advantages can be provided by this unique host framework:

- (1) The high specific surface area of MGH can efficiently reduce the local current density of the electrode, rendering uniform nucleation, and deposition of Li ions without any dendrites.
- (2) MGH provides an interconnected conductive pathway for Li plating and stripping, thus alleviating the formation of dead Li.
- (3) MGH can maintain a long-term structure stability and ensure a uniform plating/stripping behavior during long cycles. All of these characteristics synergistically contribute to the high CE, low polarization, and long life span of the MGH electrode, particularly the outstanding performance at an ultrahigh rate of 15.0 mA/cm². Therefore, reducing the local current density is an effective strategy when designing a host framework for the Li metal anode.

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Qiang Zhang graduated from the Chemical Engineering Department, Tsinghua University, China, in 2004, where he continued doing research on mass production of carbon nanotubes and obtained his PhD in chemical engineering in 2009. After a short stay as a Research Associate in Case Western Reserve University, USA, in 2009, he joined the Fritz Haber Institute of the Max Planck Society, Germany, as a post-doctoral fellow. Prof. Zhang joined the faculty of Tsinghua University in 2011, and

now he is a full professor of chemical engineering. He was elected as Newton Advanced Fellowship from Royal Society, UK in 2015 and winner of the National Science Fund for Distinguished Young Scholars in 2018. Prof. Zhang is serving as an associate editor for *Transactions of Tianjin University*. His current research interests are advanced energy materials, including dendrite-free lithium metal anode, lithium sulfur batteries, and electrocatalysis, especially the structure design and full demonstration of advanced energy materials in working devices.