Advanced Mesoporous Spinel Li$_4$Ti$_5$O$_{12}$/rGO Composites with Increased Surface Lithium Storage Capability for High-Power Lithium-Ion Batteries

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*Supporting Information

ABSTRACT: Spinel Li$_4$Ti$_5$O$_{12}$ (LTO) and reduced graphene oxide (rGO) are attractive anode materials for lithium-ion batteries (LIBs) because of their unique electrochemical properties. Herein, we report a facile one-step hydrothermal method in preparation of a nanocomposite anode consisting of well-dispersed mesoporous LTO particles onto rGO. An important reaction step involves glucose as a novel linker agent and reducing agent during the synthesis. It was found to prevent the aggregation of LTO particles, and to yield mesoporous structures in nanocomposites. Moreover, GO is reduced to rGO by the hydroxyl groups on glucose during the hydrothermal process. When compared to previously reported LTO/graphene electrodes, the newly prepared LTO/rGO nanocomposite has mesoporous characteristics and provides additional surface lithium storage capability, superior to traditional LTO-based materials for LIBs. These unique properties lead to markedly improved electrochemical performance. In particular, the nanocomposite anode delivers an ultrahigh reversible capacity of 193 mA h g$^{-1}$ at 0.5 C and superior rate performance capable of retaining a capacity of 168 mA h g$^{-1}$ at 30 C between 1.0 and 2.5 V. Therefore, the newly prepared mesoporous LTO/rGO nanocomposite with increased surface lithium storage capability will provide a new opportunity to develop high-power anode materials for LIBs.

KEYWORDS: lithium-ion batteries, anode, high power, spinel Li$_4$Ti$_5$O$_{12}$, reduced graphene oxide

1. INTRODUCTION

Spinel Li$_4$Ti$_5$O$_{12}$ (LTO) has been intensively investigated as a promising anode candidate for high-power lithium-ion batteries (LIBs) due to its rapid Li$^+$ diffusivity, remarkable structural stability, and excellent safety.1−4 However, its inherently low electrical conductivity significantly limits its high rate performance in LIBs.5−11 Many strategies have been explored to address the electrical conductivity issue for the promising LTO anodes. They include development of novel methods to prepare nanostructured LTO and the introduction of conductive additives.4,12,13 Among others, mesoporous nanostructures represent an ideal architecture for facilitating mass and electron transfer within battery electrodes. As a matter of fact, nanostructured mesoporous materials have attracted much attention reflecting a wide variety of applications including energy storage, catalysis, chemical sensors, and biomedical applications.14−23 A number of publications have already reported the synthesis of mesoporous structures for LTO by using modified templates.24−27 However, complex template methods have many technical issues, which make it very difficult to control synthesis of well-defined mesoporous structures.24−26 New methods based on facile
synthesis procedures are needed to prepare unique mesoporous electrodes for battery applications.

Meanwhile, reduced graphene oxide (rGO) has rapidly emerged as a potential electrode material for electrochemical energy storage due to its superior electron conductivity, high surface area, excellent chemical stability, and structural flexibility. It should be noted that LTO/rGO composites have previously been studied as electrode materials for LIBs. A few investigations on the synthesis and applications of LTO/rGO composites have also been reported, using various synthesis routes such as hydrothermal, solvothermal, and microwave methods. It can be concluded that the LTO/rGO composites prepared by all of these methods exhibit significantly improved rate capability and cycling performance. In this experiment, highly electrically conductive rGO was subsequently integrated into mesoporous LTO to overcome the poor electrical conductivity of LTO. More importantly, we report a simple strategy which uses glucose as a linker and reducing agent to prepare a mesoporous LTO/rGO composite with enhanced electrochemical properties. Different from other reported LTO/rGO composites, our newly prepared mesoporous LTO/rGO composite provides increased surface lithium storage capability and greatly improved electrochemical performances for LIBs. The detailed formation and improved mechanism of mesoporous structured LTO/rGO composites are systematically studied in this paper.

2. EXPERIMENTAL DETAILS

2.1. Preparations of Graphene Oxide and LTO/rGO Composites. Graphene oxide (GO) was synthesized from natural flake graphite powders (Sinopharm Chemical Reagent Co., Ltd., Shanghai, China) using the modified Hummer method. LTO/GO synthesized with the assistance of glucose was labeled as g-LTO/rGO. It should be noted that the results of Raman spectra indicate that GO could have been reduced to rGO by the hydroxyl groups of glucose. It was prepared via a hydrothermal process followed by a heat treatment. The Ti(SO4)2·H2O (2.87 g) was first dispersed in 10 mL of H2O. Then 0.02 g of GO, 0.1 g of glucose, and 8 mL of prepared Ti(SO4)2 solution were dissolved into 20 mL of H2O. The resulting solution was sonicated for 1 h. Next, the solution was added dropwise into a 23 mL solution that contained 0.380 g of LiOH·H2O. After the solution was stirred for 15 min, the resulting solution was transferred into a Teflon-lined autoclave (100 mL) and treated at 180 °C for 24 h. The collected precipitate was washed with ethyl alcohol and dried in a vacuum oven at 80 °C for 24 h. Finally, the obtained powder was calcined at 600 °C for 2 h in an Ar atmosphere to obtain g-LTO/rGO. For comparison, LTO/GO composites were prepared through an almost identical procedure that excluded the addition of glucose.

2.2. Materials Characterization. X-ray diffraction (XRD) was performed on a Rigaku D/MAX-RC X-ray diffractometer with Cu Kα (45 kV, 50 mA) monochromated radiation to determine crystalline phases and lattice parameters of prepared LTO composites. XRD patterns were collected from 5° to 80° with a step size of 0.02°. The particle morphologies of the samples were examined with transmission electron microscopy (TEM, H-800). Nitrogen adsorption–desorption measurements at 77 K were performed using a Micromeritics ASAP 2010 instrument to measure Brunauer–Emmett–Teller (BET) surface areas and porous structures. The pore size distribution was calculated from the adsorption branch of the isotherms using the Barrett–Joyner–Halenda (BJH) method. Raman measurements were conducted with a Renishaw 2000 system with a 514.5 nm Ar-ion laser and charge-coupled device detector. Thermogravimetry analyses (TGA) were performed on a TG-DSC instrument at a heating rate of 10 °C min−1 in air atmosphere from 30 to 700 °C (Mettler-Toledo, TGA/DSC 1).

2.3. Electrochemical Measurements and Battery Tests. Electrochemical measurements were carried out in a two-electrode system using a CR2025-type coin cell with lithium metal as the reference and counter electrode. The work electrodes were prepared by mixing active materials, conductive material (acetylene black), and binder (polytetrafluoroethylene) with a weight ratio of 8:1:1. They were then cut into disks (12 mm in diameter) and dried at 120 °C for 12 h in vacuum. The cell assembly was operated in a glovebox filled with pure argon. The electrolyte was 1.0 M LiPF6 dissolved into a mixed electrolyte containing ethylene carbonate, propylene carbonate, and diethyl carbonate with a volume ratio of 1:1:1. Cyclic voltammetry experiments were conducted using a CHI 660E electrochemical workstation at various scanning rates of 0.2, 0.4, 0.8, and 1.6 mV s−1 between 1.0 and 2.5 V.

Galvanostatic charge/discharge battery experiments were performed on an automatic galvanostatic charge–discharge unit (Land CT 2001A, Wuhan, China) at different current densities and a voltage range between 1.0 and 2.5 V at 25 °C. All experiments shown in this work were performed multiple (more than five) times. The specific capacity was calculated according to the total composite mass. The deviation of the calculated values between identical test conditions at five different rates was <1%.

3. RESULTS AND DISCUSSION

3.1. Structure and Morphology. Figure 1 presents XRD patterns of GO, LTO/GO, and g-LTO/rGO. The intense peak at 10.6° observed in Figure 1a corresponds to the layered structure of GO. In addition, other diffraction peaks of LTO/GO (Figure 1b) and g-LTO/rGO (Figure 1c) are consistent with a cubic spinel structure of Li4Ti5O12 (JCPDS No. 00-49-0207). There was no impurity-related peak observed for materials such as TiO2. These results indicate that the addition of glucose has no effect on the crystal structure of LTO, based on the identical XRD patterns between LTO/GO and g-LTO/rGO. There was no obvious difference in the patterns of LTO/GO and g-LTO/rGO. There was no obvious difference in the patterns of LTO/GO and g-LTO/rGO.
Figure 2. Raman spectra of GO (a), LTO/GO (b), and g-LTO/rGO (c).

Figure 4. Nitrogen adsorption—desorption isotherms and BJH pore-size distribution (insert) analysis for the as-obtained g-LTO/rGO.

LTO/rGO exhibits a more dominant G-band peak. Generally, G-peak intensities increased with the average size of the sp² domains upon reduction of the exfoliated GO. The prominent G peak in g-LTO/rGO could be explained by a higher number of new graphitic domains that were smaller than those in the GO before reduction. The results suggested that GO was reduced to rGO due to the addition of glucose. Previous works also revealed that the GO could be reduced to rGO by the hydroxyl groups of glucose.42,43 Therefore, the reduced GO (rGO) resulting from glucose was very unique in the synthesis procedure, which could provide improved electrical conductivity for the g-LTO/rGO nanocomposite.

The TEM images of LTO/GO and g-LTO/rGO are compared in Figure 3. Unlike the significant particle aggregation observed in glucose-free LTO/GO (Figure 3a and Figure S2), a well-dispersed LTO particle morphology was clearly determined in g-LTO/rGO (Figure 3b,c). Furthermore, a high-resolution TEM (HR-TEM) image (Figure 3d) clearly shows lattice fringes of 0.484 nm, corresponding to (111) interplanar spacing of LTO. In combination with XRD analysis, these results verify the formation of well-crystallized LTO in LTO/GO and g-LTO/rGO composites. Interestingly, unlike LTO/GO, a larger mean pore size of ~5 nm was determined in the g-LTO/rGO according to the HR-TEM image as exhibited in Figure 3d, showing a mesoporous structure. To further study the porous structure and pore size distribution of the g-LTO/rGO nanocomposite, BET measurements were performed. As displayed in Figure 4, a typical mesoporous structure was identified in g-LTO/rGO, as evidenced by its nitrogen adsorption—desorption isotherm. In good agreement with observation of its HR-TEM image (Figure 3d), its pore size was mainly distributed in the range of ~5 nm (insert of Figure 4). Consequently, addition of glucose during the synthesis could not only prevent LTO particle aggregation but also lead to mesoporous structures in the composite.

To elucidate the formation mechanism of the well-dispersed mesoporous LTO onto rGO supports, XRD, TEM, and Raman tests were employed to study the structure and morphology of the precursor of g-LTO/rGO before calcination treatment. As shown in Figure S1, the precursor was indexed as hydrous...
lithium titanate (JCPDS No. 47-0123). In addition, it exhibits a similar mesoporous microstructure to g-LTO/rGO (Figure S2). Moreover, precursor GO has already been reduced to rGO by the glucose hydroxyl groups, even before the high-temperature calcination (Figure S3). Glucose has been used before as a facet-controlling agent and a connection linker to prepare ultradispersed mesoporous TiO$_2$ particles on graphene aerogels surfaces, although the unique function of glucose has been well-addressed, the possible mechanism of the formation of mesoporous TiO$_2$ has not yet been provided. Herein, the function of glucose along with the formation process of well-dispersed mesoporous LTO nanocrystals on rGO is discussed.

During synthesis, Ti(SO$_4$)$_2$ was first dissolved in an aqueous solution as a Ti source to cultivate crystal seeds. Then glucose was used to fix the seeds onto GO, followed by the addition of stoichiometric amounts of LiOH (Li source). During the hydrothermal process, large amounts of glucose molecules are adsorbed onto the (001) surfaces of titanium oxide, resulting in nanosized seeds due to hydroxyl groups on glucose. This process can effectively hinder the aggregation of nanoparticles. Simultaneously, (1) a stoichiometric amount of lithium can spontaneously react with titanium and form hydrous lithium titanate while (2) GO is reduced to rGO by the hydroxyl groups of glucose. During the growth of lithium titanate, the hydroxyl groups at one end of glucose connect with rGO, and the hydroxyl groups at the other end attach to the lithium titanate facets. Notably, the repulsions between the free hydroxyl groups on glucose can not only effectively hinder the aggregation of lithium titanate particles but also produce a mesoporous structure. Thus, the unique hydroxyl groups on the glucose molecule are able to serve as a linker and promote the in situ growth of mesoporous lithium titanate nanocrystals onto the rGO surface. The obtained mesoporous lithium titanate/rGO is directly transformed into a mesoporous-structured LTO/rGO during subsequent calcination under an Ar atmosphere. Accordingly, a schematic illustration for the formation mechanism of the mesoporous-structured LTO/rGO is summarized in Figure 5. The resulting porous structure offers sufficient surface area, which allows electrolyte to effectively access active materials. These mesoporous nanocrystals supported on highly electrically conductive rGO hold great promise to facilitate Li$^+$ and electron transfer, thereby leading to high capacity and excellent rate performance LTO anodes for LIBs as discussed of late.

In this experiment, we calculated the capacity according to the total composite mass. However, it would also be helpful to determine the carbon content in these LTO-based nanocomposites. As shown in Figure 6, TGA tests indicate that the carbon content of g-LTO/GO and LTO/GO composites are estimated to be 4.9 and 2.8 wt %, respectively. The nominal ratio of LTO and GO is 95:5. The carbon content of 2.8 wt % in LTO/GO sample suggests that about 40 wt % GO is lost during the preparation process. In addition, the nominal LTO/C ratio should be 90:10 in g-LTO/rGO sample, assuming that all of the carbon atoms in glucose could be converted into graphitized carbon. In principle, the carbon content in the g-LTO/rGO is derived from GO and the carbonization of glucose. The loss of GO should be decreased in g-LTO/GO because of the linker function of glucose. Accordingly, the carbon content of 4.9 wt % determined in g-LTO/GO sample revealed that nearly 80 wt % glucose was lost during the preparation process. Excess amounts of glucose were used to provide sufficient linker agent and reducing agent for GO in this work. In particular, the carbon formed from the carbonization of glucose could increase the electrical conductivity of the final product, thereby improving the rate performance of nanocomposite anodes. Obviously, the optimum weight/reactant ratio of LTO:glucose would vary with the change of the weight/reactant ratio of LTO:GO. Therefore, although determination of optimal carbon content in LTO nanocomposites is extremely important to achieve maximum battery performance, it will be very challenging due to the variation of loss rates of GO and glucose. The relevant studies are ongoing and will be reported in the future.

**3.2. Electrochemical Properties and Battery Performance.** The prepared LTO/GO and g-LTO/GO composites were then studied as anodes in LIBs. Figure 7 displays typical charge−discharge curves for LTO/GO and g-LTO/rGO recorded at a potential range between 1.0 and 2.5 V as a function of current rates. Both electrodes exhibit a well-defined flat plateau at about 1.5 V, which is a typical characteristic of the insertion process of Li$^+$ in LTO. Although the capacity gradually decreased with an increase of applied current density, the recorded charge and discharge voltage plateaus of the LTO/GO and g-LTO/rGO anodes were still well-defined even at an extremely high charging−discharging current of 30 C. It should be noted that, relative to the LTO/GO, the g-LTO/rGO electrode demonstrated elongated charge and discharge plateaus, indicating a higher utilization efficiency and a lower polarization. In addition, the g-LTO/rGO anode displayed a high reversible current capacity of 193 mA h g$^{-1}$ at 0.5 C and superior rate performance of more than 168 mA h g$^{-1}$ at 30 C when voltages were set between 1.0 and 2.5 V vs Li$^+/\text{Li}$,
whereas these values of LTO/GO were only 166 and 125 mA h g$^{-1}$, respectively.

Furthermore, cycling stabilities of LTO/GO and g-LTO/rGO anodes were tested at different rates. Compared with LTO/GO, g-LTO/rGO possesses higher reversible capacity and much improved rate capability as given in Figure 8. When the current density was increased from 0.5 to 30 C, the capacity differences measured between LTO/GO and g-LTO/rGO became larger. Compared to the reversible capacity of 0.5 C, the retaining capacities of g-LTO/rGO were 98.4%, 96.4%, 93.8%, 90.7%, and 87% when charging−discharging current densities were increased from 0.5 to 1, 2, 5, 10, and 30 C, respectively. However, the corresponding values for LTO/GO anode were 96.5%, 92%, 87.1%, 82%, and 75.2%. Notably, even at 30 C, the reversible capacity of the g-LTO/rGO was higher than 168 mA h g$^{-1}$, whereas this value was only 125 mA h g$^{-1}$ for the LTO/GO. These comparisons suggest that the mesoporous LTO/rGO prepared via a glucose-assisted synthesis is superior to conventional LTO/GO in terms of reversible capacity and rate capacity as LIB anodes.

To understand the role of glucose in promoting electrochemical properties of g-LTO/GO, cyclic voltammetries were measured at scan rates ranging from 0.2 to 1.6 mV s$^{-1}$ in the potential window of 1.0−2.5 V. It is well-known that the relation between peak currents and scan rates is closely associated with different electrochemical reaction characteristics, including solid-phase diffusion-controlled or surface-confined charge-transfer processes.$^{1,39,41,52}$ As shown in Figure 9, unlike previously reported LTO/rGO composites, the newly prepared g-LTO/rGO possesses two couples of redox peaks at all scan rates. As displayed in the insets of Figure 9a,b, peaks of LTO/GO and one couple of peaks of g-LTO/rGO demonstrate a linear relationship between peak currents and the square root of the scan rates, suggesting a typical diffusion-limited reaction.$^{1,52}$ The diffusion coefficient calculated from the slope (K values)$^{53,54}$ of the charge−discharge process for the g-LTO/rGO was much larger than that for the LTO/GO, indicating a faster diffusion rate of Li$^+$ in g-LTO/rGO compared to that in LTO/GO. Furthermore, the dependence of peak currents on the scan rates as exhibited by another couple of peaks of the g-LTO/rGO analysis reveals a typical surface lithium storage process.$^{39,41}$ Accordingly, the electrode reaction of the g-LTO/rGO was controlled by a mixed process involving a surface lithium storage reaction and a diffusion-limited reaction. It is well-known that that surface lithium storage reaction is a quick lithium-ion insertion process, yielding extra capacity and improved rate performance.$^{1,52}$ Therefore, the surface lithium storage process is responsible for the remarkable improvement in reversible capacity and high-rate performance of the g-LTO/rGO anode. Our previous investigation elucidated that mesoporous LTO possessed extra surface lithium storage capability.$^{55}$ Accordingly, the mesoporous LTO in the g-LTO/rGO contributes to the detected extra lithium storage capacity. Table 1 compares the cycling and rate performances of g-LTO/rGO with other LTO-based anodes reported previously. According to the comparison, the newly prepared mesoporous LTO/rGO nanocomposite is
superior to those of the state-of-the-art LTO-based materials for LIBs in terms of enhanced reversible capacity and rate performance.

Figure 10 further provides correlations between the polarization of $\Delta E$ and the rates used for charging and discharging of LTO/GO and g-LTO/rGO anodes. The values of $\Delta E$ are defined here as the difference between the potentials of charge and discharge plateaus. These potential differences represent the degree of polarization of the electrode during the charging and discharging reactions. The $\Delta E$ values for the g-LTO/rGO were much smaller than those of the LTO/GO at all rates from 0.5 to 30 C, which suggested that g-LTO/rGO had lower polarization and better reaction kinetics. Therefore, these results further indicated that the mesoporous LTO deposited onto rGO in the g-LTO/rGO nanocomposite could effectively reduce the electrode polarization, thus resulting in increased reversible capacity and improved rate performance. Developments of mesoporous structured LTO-based composites with increased surface lithium storage capability may provide a new route to prepare a high-performance LTO anode for power LIBs.

Figure 11 displays the first and second charge-discharge curves for the electrode of LTO/GO and g-LTO/rGO recorded over a potential range between 2.5 and 1.0 V at 0.5 C. The initial capacity loss of g-LTO/rGO was greater than that of LTO/GO. This indicates that the higher reversible capacity of g-LTO/rGO might be used to form a solid electrolyte interface (SEI) film. Initially, LTO anodes are considered free from SEI films when cycled between 1 and 3 V. However, it was found that reactivity and the formation of SEI films were much affected by the morphology and surface area of the electrode. Also, previous investigations indicated that the formation of a smooth SEI film (as compared to a rough SEI film) could greatly improve the reversible capacity and rate performances of mesoporous electrode materials. Therefore, the well-dispersed mesoporous LTO/rGO morphology may facilitate the formation of smooth SEI film contributing to its ultrahigh reversible capacity and superior rate performance.

4. CONCLUSIONS

In summary, we prepared a novel mesoporous LTO/rGO nanocomposite through a facile hydrothermal process followed by a heating treatment. Compared to previously studied LTO-based anodes, our newly prepared composite provides an extra surface lithium storage capability for LIB anode applications. The utilization of glucose during the hydrothermal process not only inhibits aggregation of LTO nanocrystal particles but also leads to a nanostructured mesoporous composite. The mesoporous LTO in g-LTO/rGO nanocomposite was responsible for extra surface lithium storage reaction, which significantly improved its reversible capacity and rate performance. Even at an extremely high charging-discharging rate of 30 C, the mesoporous LTO/rGO anode was able to retain 87% capacity measured at 0.5 C with well-defined voltage plateau.

Table 1. Comparison of Cycling and Rate Performance of g-LTO/rGO with Other LTO-Based Anodes Reported Before

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<th>samples</th>
<th>capacity (mA h g$^{-1}$)</th>
<th>retention (%)</th>
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<tr>
<td></td>
<td>1 C</td>
<td>2 C</td>
</tr>
<tr>
<td>g-LTO/rGO$^\dagger$</td>
<td>190</td>
<td>187</td>
</tr>
<tr>
<td>LTO/GO$^\dagger$</td>
<td>160</td>
<td>153</td>
</tr>
<tr>
<td>n-LTO/GO$^{32}$</td>
<td>171</td>
<td>151</td>
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<tr>
<td>n-LTO/MWNT$^{33}$</td>
<td>158</td>
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<tr>
<td>LTO-NS$^{39}$</td>
<td>148</td>
<td>139</td>
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$^\dagger$Present work; n, nanosized; MWNT, multiwalled carbon nanotube; M, mesoporous; NS, nanosheets; MS, microspheres.
We believe that this novel mesoporous LTO/rGO composite can offer a new opportunity to develop advanced LTO-based anode materials for high-power LIBs with significantly improved performance. The synthetic strategy using glucose as an effective additive in this work could be further extended to preparations of other rGO-based composites to generate uniform mesoporous nanocrystal particles.

- **ASSOCIATED CONTENT**

  **Supporting Information**
  The Supporting Information is available free of charge on the ACS Publications website at DOI: 10.1021/acsami.6b01644.

  XRD pattern and TEM image for the precursor of g-LTO/rGO before calcination. HR-TEM image of the LTO/GO. Raman spectra of the GO, precursors of LTO/GO and g-LTO/rGO (PDF)

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  **Notes**
  The authors declare no competing financial interest.

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