Fabrication of a Three-Dimensionally Networked MoO₃/PPy/rGO Composite for a High-Performance Symmetric Supercapacitor

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ABSTRACT: A three-dimensionally interconnected molybdenum trioxide (MoO₃)/polypyrrole (PPy)/reduced graphene oxide (rGO) composite was synthesized via an eco-friendly three-step method. The as-obtained electrode shows a high specific capacity of 412.3 F g⁻¹ at a current density of 0.5 A g⁻¹ and a good cycling stability (85.1% of the initial specific capacitance after 6000 cycles at 2 A g⁻¹ is retained), and these excellent electrochemical performances can be attributed to the unique structure, remarkable electrical conductivity, and the synergetic effects between MoO₃, PPy, and rGO. Furthermore, a symmetric supercapacitor based on a MoO₃/PPy/rGO electrode was assembled to investigate the practical application performance of this material. The results demonstrate a high energy density of 19.8 W h kg⁻¹ at a power density of 301 W kg⁻¹. These findings shine a light on the rational design of electrode materials with multicomponents for high-performance supercapacitors.

1. INTRODUCTION

Supercapacitors are attracting more and more interest due to their high power density and long-cycle life.¹,² However, the low energy density seriously restricts their further application in the field of energy storage. Therefore, the development of high energy density electrode materials for supercapacitors has become a key problem to be solved.³−⁵

MoO₃ is considered as an excellent electrode material because of its high specific capacitance, low cost, and environmentally benign properties. Despite these advantages, pure MoO₃ suffers from a low electrical conductivity and poor cycle stability, and this inevitably limits its further application as an electrode material for electrochemical energy storage.⁶,⁷

The combination of MoO₃ with a high-conductivity material is an effective approach to solve these problems.⁸ Yan et al. coupled MoO₃ with Ag nanoparticles to increase the conductivity of MoO₃-based electrodes. The as-prepared electrode showed a higher capacitance (225 F g⁻¹) compared with the MoO₃ electrode without Ag nanoparticles (69 F g⁻¹).⁹ Faraji and Abedini demonstrated the fabrication of the MoO₃/GO/MWCNTs/graphite composite. The composite exhibits an excellent conductivity and a remarkable areal capacitance of 103 mF cm⁻².¹⁰ However, although many efforts have been made to construct MoO₃-based composite electrodes, it remains a challenge to exploit composition rules for the design of high-performance composite electrodes.

In this work, polypyrrole (PPy) was applied to modify the conductivity of MoO₃ and provide additional pseudocapacitance for supercapacitors. In order to further improve the conductivity and cycle stability of the electrode material, reduced graphene oxide (rGO) was also used to construct a three-dimensionally (3D) interconnected conductive network via forming a high conductive coating layer on the surface of the MnO₃/PPy composite. Strong synergetic effects were observed between MoO₃, PPy, and rGO. Owing to the abovementioned advantages, the as-prepared composite exhibits a high specific capacitance (335 F g⁻¹, at 1 A g⁻¹) and a good cycling stability (remained 85.1%, at 2A g⁻¹, 6000 cycles). Moreover, the symmetric supercapacitor cell based on this composite shows a remarkable energy density (19.8 W h Kg⁻¹, at a power density of 301 W Kg⁻¹).

2. RESULTS AND DISCUSSION

The morphologies of the samples were characterized by scanning electron microscopy (SEM). As shown in Figure 1a, MoO₃ has a distinct wire-like structure, and the diameter of the MoO₃ nanowires (NWs) is about 100 nm. The rGO sheets in Figure 1b overlap each other to form a typical 3D network structure. Figure 1c is the SEM image of the MoO₃/PPy/rGO composite. The composite exhibits an excellent conductivity and a remarkable areal capacitance of 103 mF cm⁻². However, although many efforts have been made to construct MoO₃-based composite electrodes, it remains a challenge to exploit composition rules for the design of high-performance composite electrodes.

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to the surface of the MoO₃ NWs, and the composites shows a rough surface. Moreover, the average distance between two NWs in MoO₃/PPy has been increased obviously compared with that of pure MoO₃, indicating that PPy nanoparticles can efficiently prevent the agglomeration of MoO₃ NWs. This loose stacking structure can not only increase the electrode−electrolyte contact area but also provide more ion diffusion pathways. However, the large spacing between PPy nanoparticles may cause a low electrical conductivity. In Figure 1d, MoO₃/PPy is uniformly covered by rGO sheets to form an interconnected conductive network. Such a unique structure can efficiently increase the conductivity of the composite. Furthermore, the rGO coating can restrain the volume change in PPy nanoparticles and MoO₃ NWs during the charge and discharge process, leading to a good cyclic stability.

X-ray photoelectron spectroscopy (XPS) was carried out to investigate the chemical configuration of the MoO₃/PPy/rGO composites. In Figure 2a, the survey XPS spectrum indicates that the composite consists of four elements: molybdenum, carbon, nitrogen, and oxygen. The high-resolution Mo 3d spectrum in Figure 2b shows two peaks at 232.6 and 235.7 eV which are consistent with the Mo 3d₅/₂ and Mo 3d₃/₂, respectively. As shown in Figure 2c, the peaks at 398.3, 399.8, 400.9, and 402.2 eV are appointed to the $\equiv N−$, $−N=H$, $−N=H^+$, and $≡N−H^+$, correspondingly, originating from the synthesized PPy. In Figure 2d, the characteristic peaks at 530.4, 531.3, and 532.90 eV are corresponded to Mo−O, C−O−Mo, and C=O, respectively. The abovementioned results indicate that the MoO₃/PPy/rGO composite was successfully prepared.

Fourier transform infrared (FTIR) spectra were used to investigate the chemical structure of the samples. In Figure 3, the FTIR spectrum of pure PPy displays two peaks at 1466 and 1545 cm⁻¹, which correspond to the C−N and C≡C stretching vibration in the pyrrole ring, respectively. In the spectrum of the PPy/MoO₃ composite, the characteristic peaks of MoO₃ appear at 967 and 896 cm⁻¹, which is consistent with those in the literature. After the modification of rGO, a new peak is observed at 1623 cm⁻¹, which is assigned to the C≡C stretching vibration of the graphene sheet. Moreover, the characteristic peak of the O−H stretching at about 3500 cm⁻¹ did not appear, indicating that the GO was successfully reduced.

The electrochemical performances of MoO₃ NWs, rGO, PPy, MoO₃/rGO, MoO₃/PPy, and MoO₃/PPy/rGO were investigated in a three-electrode cell. Figure 4a shows the comparative cyclic voltammetry (CV) curves of these six electrodes at a scan rate of 10 mV s⁻¹. The MoO₃/PPy/rGO shows the largest enclosed area than those of the other electrodes, suggesting a superior specific capacitance. Moreover, except for the rGO electrode, the CV curves of the other electrodes are quasi-rectangles and have obvious redox peaks, originating from the pseudocapacitance behaviors of PPy and MoO₃. Figure 4b displays the CV curves of the MoO₃/PPy/rGO electrode at scan rates ranging from 10 to 100 mV s⁻¹. The shape of the CV curves is changed gradually with increasing scan rates. It is caused by the fact that the electrolyte ions do not have enough time to diffuse into the inner sites of the electrode with high scan rates. In order to further study the capacitive behavior of the samples, GCD tests were conducted at a current density of 1 A g⁻¹, and the results are presented in Figure 4c. As seen, the discharge times of these six electrodes increase in the order rGO < MoO₃ < MoO₃/rGO < PPy < MoO₃/PPy < MoO₃/PPy/rGO. This trend suggests that the specific capacitance of the MoO₃ can be improved via...

Figure 1. SEM images of (a) MoO₃ NWs, (b) rGO, (c) PPy/MoO₃, and (d) MoO₃/PPy/rGO.

Figure 2. (a) XPS spectra of MoO₃/PPy/rGO; (b) corresponding high-resolution Mo 3d peak, (c) N 1s peak, and (d) O 1s peak.

Figure 3. FTIR spectra of PPy, MoO₃/PPy, and MoO₃/PPy/rGO.
combining with PPy nanoparticles and rGO sheets. The relationships of the specific capacitances as a function of the current densities for these six electrodes are shown in Figure 4d. The highest specific capacitance of 412.3 F g\(^{-1}\) is obtained for the MoO\(_3\)/PPy/rGO electrode at a current density of 0.5 A g\(^{-1}\), compared with specific capacitances of 30.7, 57.5, 105, 116, and 208.1 F g\(^{-1}\) for rGO, MoO\(_3\), PPy, MoO\(_3\)/rGO, and MoO\(_3\)/PPy electrodes, respectively. Notably, the specific capacitance of the MoO\(_3\)/PPy/rGO electrode is evidently higher than the sum of the specific capacitances of the MoO\(_3\)/rGO electrode and MoO\(_3\)/PPy electrode. This may be due to the synergistic effect between MoO\(_3\), PPy nanoparticles, and rGO sheets. Moreover, when the current density is increased to 10 A g\(^{-1}\), the specific capacitance of the MoO\(_3\)/PPy/rGO electrode remains as 107.3 F g\(^{-1}\), still higher than those of the other electrodes at the same current density.

Electrochemical impedance spectroscopy (EIS) analysis was performed to further investigate the synergistic effect between

Figure 4. (a) CV curves of PPy, rGO, MoO\(_3\), MoO\(_3\)/PPy, MoO\(_3\)/rGO, and MoO\(_3\)/PPy/rGO electrodes at a scan rate of 10 mV s\(^{-1}\). (b) CV curves of the MoO\(_3\)/PPy/rGO electrode at scan rates from 10 to 100 mV s\(^{-1}\). (c) GCD curves of PPy, rGO, MoO\(_3\), MoO\(_3\)/PPy, MoO\(_3\)/rGO, and MoO\(_3\)/PPy/rGO electrodes at 1 A g\(^{-1}\). (d) Specific capacitances of PPy, rGO, MoO\(_3\), MoO\(_3\)/PPy, MoO\(_3\)/rGO, and MoO\(_3\)/PPy/rGO electrodes at current densities ranging from 0.5 to 10 A g\(^{-1}\).

Figure 5. (a) EIS curves of PPy, rGO, MoO\(_3\), MoO\(_3\)/PPy, MoO\(_3\)/rGO, and MoO\(_3\)/PPy/rGO electrodes. The inset of (a) is EIS curves of these electrodes in high-frequency regions and the equivalent circuit of the EIS spectra. (b) Cycling stability of PPy, rGO, MoO\(_3\), MoO\(_3\)/PPy, MoO\(_3\)/rGO, and MoO\(_3\)/PPy/rGO electrodes for 6000 charge/discharge cycles at a current density of 2 A g\(^{-1}\).
PPy nanoparticles and rGO sheets. The Nyquist plots of different samples are shown in Figure 5a. The inset is the equivalent circuit for the simulation of the EIS spectra. The internal resistance ($R_s$) values of MoO$_3$ NWs, rGO, PPy, MoO$_3$/rGO, MoO$_3$/PPy, and MoO$_3$/PPy/rGO electrodes are 2.6, 1.9, 1.3, 1.9, 2.2, and 1.3 Ω, respectively. The $R_s$ of PPy is evidently lower than that of MoO$_3$ and rGO. However, when PPy is combined with MoO$_3$, the $R_s$ increases from 1.3 to 2.2, which may be due to the granular PPy which is difficult to connect with each other to form a conductive network in the composites. On the other hand, the $R_s$ of rGO is 1.9 Ω, which is slightly higher than that of PPy. After being combined with MoO$_3$, the $R_s$ of MoO$_3$/rGO is still 1.9 Ω, which is the same as that of the pristine rGO. This may be due to the fact that the rGO sheets with a high specific surface area easily form an interconnected conductive network on the surface of the MoO$_3$ NWs. It is worth noting that the $R_s$ of the MoO$_3$/PPy/rGO electrode is as low as that of the pure PPy sample, which further proves that the synergistic effect between PPy nanoparticles and rGO sheets greatly enhances the conductivity of MoO$_3$/PPy/rGO.

Figure 6. (a) CV curves of the MoO$_3$/PPy/rGO//MoO$_3$/PPy/rGO symmetric supercapacitor at different voltage ranges at a scan rate of 10 mV s$^{-1}$. (b) CV curves of the symmetric supercapacitor at different scan rates from 10 to 100 mV s$^{-1}$. (c) Galvanostatic charge/discharge (GCD) curves of the symmetric supercapacitor at various current densities. (d) Specific capacitance of the symmetric supercapacitor at various current densities. (e) Cycle performance of the symmetric supercapacitor at a current density of 2 A g$^{-1}$. (f) Ragone plots of the MoO$_3$/PPy/rGO//MoO$_3$/PPy/rGO symmetric supercapacitor in comparison to other supercapacitors from literature studies.
conductive PPy nanoparticles acted as a conducting binder and promoted the electron transportation within the electrode. The slope of the line in the low frequency range is indicative of the capacitive behavior of the active material.

As seen, the MoO₃ electrode exhibits the lowest slope among these six electrodes. It may be due to the compact stacking structure of the MoO₃ NWs, which hinders the ions penetrating into the electrode pores. After being combined with PPy, the slope of the MoO₃/PPy composites increases obviously. It indicates that PPy nanoparticles can efficiently inhibit the aggregation of MoO₃ NWs. This result is also consistent with the SEM images mentioned above. Interestingly, the slope of the MoO₃/PPy/rGO electrode is slightly lower than that of MoO₃/PPy. This is because the coating layer formed by rGO sheets hindered the diffusion of ions. The cycling stability of MoO₃/PPy/rGO was also studied, and the result is shown in Figure 5b. After 6000 charge and discharge cycles at a current density of 2 A g⁻¹, the capacitance retention of MoO₃/PPy/rGO was 85.1%. This good cycling stability should be attributed to the rGO sheets acting as a protective layer to prevent the volume expansions of the MoO₃ and PPy during the charge/discharge process.

In order to further study the practical application performance of the MoO₃/PPy/rGO material, a symmetric supercapacitor made up of two MoO₃/PPy/rGO electrodes was assembled. Figure 6a displays the CV curves of the MoO₃/PPy/rGO/MoO₃/PPy/rGO supercapacitor at different voltage ranges at a scan rate of 10 mV s⁻¹. The curve keeps its rectangular shape even with a 1.2 V voltage range, suggesting a good cycle reversibility. According to the abovementioned test results, the electrochemical properties of the MoO₃/PPy/rGO/MoO₃/PPy/rGO symmetric supercapacitor were tested in the voltage window of 0–1.2 V. Figure 6b shows the CV curves of the symmetric supercapacitor at different scan rates. The curves at low scan rates show distorted rectangular shapes, suggesting the pseudocapacitance characteristics of the MoO₃ and PPy. The charge/discharge curves at various current densities are exhibited in Figure 6c. The curves present ideal linear behaviors even at a high current density of 10 A g⁻¹, indicating that the symmetric supercapacitor has excellent rate capability. The specific capacitances of the symmetric supercapacitor at various current densities are calculated from the charge/discharge curves and shown in Figure 6d. The specific capacity is 99 F g⁻¹ at a current density of 0.5 A g⁻¹. As the current density increases to 10 A g⁻¹, the specific capacity decreases from 99 to 42 F g⁻¹. The excellent practical performance is demonstrated by lighting an LED device, as shown in the Figure 6d inset. As shown in Figure 6e, the capacitance retention of the symmetric supercapacitor was 86.2% after 6000 cycles at a current density of 2 A g⁻¹, suggesting a good cycling stability. Ragone plots of the MoO₃/PPy/rGO/MoO₃/PPy/rGO symmetric supercapacitor are presented in Figure 6f. The symmetric supercapacitor exhibits a remarkable energy density of 19.8 W h kg⁻¹ at a power density of 301 W kg⁻¹, superior to those of other reported materials for supercapacitors, for instance, microrods architecture MoO₃ (7.33 W h kg⁻¹, 1200 W kg⁻¹), MoS₂/MoO₃/PPy (10 W h kg⁻¹, 203 W kg⁻¹), symmetric PPy/N-doped graphene (17 W h kg⁻¹ at 467 W kg⁻¹), PPy-rGO composites (15.8 W h kg⁻¹, 140 W kg⁻¹), PPy-GO composites (16.4 W h kg⁻¹, 80 W kg⁻¹), and PPy-rG (9.1 W h kg⁻¹, 375 W kg⁻¹).

3. CONCLUSIONS

In this work, we have successfully designed and synthesized a 3D networked MoO₃/PPy/rGO composite via a facile three-step method. The as-prepared electrode exhibits an excellent specific capacity of 412.3 F g⁻¹ at a current density of 0.5 A g⁻¹ and a good cycling stability (remained 85.1% after 6000 cycles at 2 A g⁻¹). The remarkable performances of the MoO₃/PPy/rGO electrode are attributed to the following reasons: (i) PPy nanoparticles can efficiently prevent the agglomeration of MoO₃ NWs, and this can not only provide more electrochemical active sites but also offer shortened pathways for ions; (ii) MoO₃ NWs are covered with a high conductive layer composed of PPy nanoparticles and rGO sheets, and this unique structure significantly reduces the Ri and Rs of the composite; (iii) the rGO coating can prevent the volume change in the MoO₃/PPy composite during charge/discharge cycles, leading to an excellent long-term cycle stability. Furthermore, the symmetric supercapacitor based on the MoO₃/PPy/rGO electrode shows a high energy density (19.8 W h Kg⁻¹) and good cycling stability of 86.2% capacitance retention after 6000 cycles. These results demonstrate that the MoO₃/PPy/rGO composite is a promising material for high-performance supercapacitor.

4. EXPERIMENTAL SECTION

4.1. Materials. Ammonium molybdate [(NH₄)₆MoO₂₄·4H₂O, AR], nitric acid (HNO₃, 65%), ammonium persulphate (NH₄)₃S₂O₸, AR] and the pyrrole monomer (AR grade) were purchased from Aladdin Reagent Co. Ltd. (China). rGO was purchased from XFNANO Materials Tech Co. Ltd. (Nanjing, China). The whole experiment process used deionized (DI) water. All of the reagents are analytically pure without further purification.

4.2. Material Characterization. Field-emission scanning electron microscopy was performed on a Hitachi S-4800 scanning electron microscope. XPS measurements were carried out on a Thermo Fisher X-ray photoelectron spectrometer equipped with Al radiation as the probe (Kα, radiation), with a chamber pressure of 5 × 10⁻⁷ Torr. The analysis spot size was 400 μm in diameter. The chemical structure of the samples was investigated using FTIR spectrometer (Nicolet6700, Thermo Fisher).

4.3. Electrochemical Tests. Whole electrochemical tests in this work were carried out on a CHI660E workstation (Shanghai Chenhua) in 1 mol L⁻¹ Na₂SO₄ aqueous solution. A three-electrode system was used to investigate the electrochemical performance of the samples. A saturated calomel electrode and Pt wire served as the reference and counter electrode, respectively. The working electrodes were fabricated by compressing the active materials onto nickel foams. A two-electrode system was used to study the electrochemical properties of the symmetric supercapacitor constructed by assembling two same PPy/GO/MoO₃ electrodes in 1 mol L⁻¹ Na₂SO₄ aqueous electrolyte. The symmetric supercapacitor is named PPy/GO/MnO₂/PPy/GO/MnO₂.

The specific capacitances (Cₑ) of the individual electrode and the two-electrode cell were calculated from the galvanostatic discharge curves using eq 1. The specific energy density (Eₑ) and specific power density (Pₑ) were calculated according to equation 2 and equation 3, respectively.

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where \( I \) is the discharge current, \( \Delta t \) is the discharge time, \( m \) is the mass of the active material in the three-electrode system and in the two-electrode cell, \( m \) is the total active mass of both electrodes, and \( \Delta U \) is the voltage window.

4.4. Synthesis of MoO₃ NWs. \((\text{NH}_4)_6\text{Mo}_7\text{O}_{24} \cdot 4\text{H}_2\text{O} (1.3 \text{ g})\) was dissolved fully in 50 mL of DI water and stirred. 65% HNO₃ was used to adjust the pH of the solution to 1.0. Then, the solution was transferred into a 150 mL Teflon-lined stainless-steel autoclave with heat at 185 °C for 15 h. Finally, the product was obtained by centrifuging and washing with DI water and then dried at 55 °C in vacuum.

4.5. Synthesis of the MoO₃/PPy Composite. In situ oxidative polymerization of pyrrole was carried out with the MoO₃ template to obtain the MoO₃/PPy composite. The \((\text{NH}_4)_2\text{S}_2\text{O}_8\) served as an oxidant. MoO₃ NWs (21 mg) were added in 90 mL of DI water and stirred. Then, 210 μL of pyrrole was added with an ice bath. Two hours later, 21 mL of \((\text{NH}_4)_2\text{S}_2\text{O}_8\) solution was added dropwise. The mole ratio of pyrrole and \((\text{NH}_4)_2\text{S}_2\text{O}_8\) was 2:1. After 10 h of ice bath reaction, the sediments were collected by centrifuging and washing with DI water and then dried at 55 °C in vacuum.

4.6. Preparation of the MoO₃/PPy/rGO Composite. MoO₃-PPy NWs (15 mg) were put into a round bottom flask and dispersed in 40 mL of DI water by sonication. Subsequently, 5 mg of rGO was added to the suspension and continued to sonicate for 40 min. The product was centrifuged and washed with DI water and absolute ethanol five times. Finally, the product was transferred into a vacuum drying oven and dried for 6 h at 65 °C. The schematic diagram of the composite material is shown in Scheme 1.
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