An Electrosynthesized 3D Porous Molybdenum Sulfide/Graphene Film with Enhanced Electrochemical Performance for Lithium Storage

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Abstract
Molybdenum sulfide/graphene composites are promising anode materials for lithium-ion batteries (LIBs). In this work, MoS\textsubscript{x}/graphene composite film with an ideal 3D porous structure is developed via a facile and straightforward electrochemical route. The MoS\textsubscript{x} nanoparticles are uniformly anchored on the graphene nanosheets that are randomly arranged, resulting in MoS\textsubscript{x}/graphene composites with well-developed porous structure. Benefiting from such structure and the synergistic effect from two components, this material shows a high specific capacity over 1200 mA h g\textsuperscript{−1}, an excellent rate performance, and superior cycling stability. The dominating pseudocapacitive behavior in Li storage contributes to the outstanding rate capacity. Importantly, this kind of novel material can be easily produced as 3D microelectrodes for microscaled LIBs that are highly demanded for autonomous microelectronic systems.

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Keywords: molybdenum sulfide, graphene, 3D structure, electrochemical deposition, lithium ion battery

Abstract: Molybdenum sulfide/graphene composites are promising anode materials for lithium ion batteries (LIBs). In this work, MoSₓ/graphene composite film with an ideal three-
dimensional porous structure is developed via a facile and straightforward electrochemical route. The MoS$_x$ nanoparticles are uniformly anchored on the graphene nanosheets that are randomly arranged, resulting in MoS$_x$/graphene composites with well-developed porous structure. Benefiting from such structure and the synergistic effect from two components, this material shows a high specific capacity over 1200 mAh g$^{-1}$, an excellent rate performance and superior cycling stability. The dominating pseudocapacitive behavior in Li storage contributes to the outstanding rate capacity. Importantly, this kind of novel material can be easily produced as three-dimensional micro-electrodes for micro-scaled LIBs that are highly demanded for autonomous micro-electronic systems.

Molybdenum disulfide (MoS$_2$) is a promising anode material in lithium ion batteries (LIBs) due to its high theoretical lithium storage capacity (670 mAh g$^{-1}$) and low cost.$^{[1]}$ The incorporation with highly conductive and mechanically strong graphene to form MoS$_2$/graphene composites can further improve electrochemical properties,$^{[2]}$ owing to enhanced ionic and electronic conductivity. The strong interaction between sulfides and graphene prevents aggregation of sulfides stabilizing electrochemical performance.$^{[2c]}$ Such composites have demonstrated significant Li storage capabilities, such as 1100 mAh g$^{-1}$ reported for a sphere-like MoS$_2$/graphene$^{[2b]}$ and 1021.2 mAh g$^{-1}$ for MoS$_2$ on nitrogen-doped graphene.$^{[3]}$

The MoS$_2$/graphene composite is typically synthesized either by hydrothermal routes involving high pressure and temperature$^{[2b, 2c]}$ or by using the exfoliated MoS$_2$ nanosheets from time-consuming procedures with use of hazardous butyllithium.$^{[4]}$ Furthermore, the produced composites are commonly in the form of powders and used with added binders and conductive additives, limiting the application on a substrate with uneven surface. In contrast, electrodeposition can produce a conformal coating onto any shaped conductive surface with
controlled thickness at room temperature; even on those micro-scaled three dimensional (3D) architectures with an increased vertical length.[5]

MoS$_2$ can be electrochemically synthesized from (NH$_4$)$_2$MoS$_4$ in solution.[6] Electrodeposited MoS$_x$/graphene composite film has recently been used for hydrogen evolution.[7] We have previously demonstrated that graphene nanosheets act as a template and dopant in electrodeposited polypyrrole/graphene film.[8] Curiosity about the role of graphene in MoS$_x$/graphene composites and the electrochemical performance for Li storage have motivated this work. Excitingly, the MoS$_x$/graphene film produced demonstrates an ideal 3D porous structure created with randomly stacked graphene nanosheets decorated with MoS$_2$ nanoparticles delivering an impressive electrochemical performance.

The electrosynthesis of 3D porous MoS$_x$/graphene film was conducted in a solution containing (NH)$_4$MoS$_4$, graphene oxide (GO) and KCl. Anchoring of MoS$_4^{2-}$ ions on the GO nanosheets due to electrostatic attraction between the oxygen-containing functional groups on GO and the positively charged part in MoS$_4^{2-}$[4b] is expected, as illustrated in Figure 1a. These negatively charged nanosheets migrated and anchored onto the stainless steel mesh (SSM) substrate during cyclic voltammetry (CV), where the MoS$_4^{2-}$ ions were converted into particle-like molybdenum sulfide and GO was reduced to rGO. The continuously applied potential sweeps induced more nanosheets anchoring on the substrate and reduced; and the random stacking of these sheets created a porous 3D structure.

In this work, a potential range of -1.3 to 0.4 V (vs. Ag/AgCl) was applied, wherein the most efficient MoS$_x$ growth has been demonstrated.[9] Distinct peaks at 374.5, 406.6 and 455.7 cm$^{-1}$ were observed in the Raman spectra of MoS$_x$ and MoS$_x$/rGO (Figure S1, Supporting Information), which correspond to the respective E$_{2g}^1$, A$_{1g}$ and 2LA modes of hexagonal MoS$_2$ crystal[10] proving the existence of MoS$_x$. The D band and G band at 1329.1 and 1600.7 cm$^{-1}$ of graphene can also be found in MoS$_x$/rGO, evidence of the formation of composite. The cathodic peak at ~-0.8 V represents the formation of MoS$_2$ from the reduction of Mo$^{6b}$. 


The anodic peak at ~0.2 V corresponds to a process producing MoS$_3$ that involves oxidation of a sulfide ligand, followed by an intramolecular electron transfer to molybdenum metal and loss of sulfur$^{[11]}$ (Figure S2a and b, Supporting Information). The involved reactions are:

$$\text{MoS}_4^{2-} + 2e^- + 4H^+ \rightarrow \text{MoS}_2 + 2H_2S$$  \hspace{1cm} (1)

$$\text{MoS}_4^{2-} \rightarrow \text{MoS}_3 + \frac{1}{8}S_{8} + 2e^-$$  \hspace{1cm} (2)

Thus, the products are expected to be a mixture of MoS$_2$ and MoS$_3$ and denoted as MoS$_x$ in this work. The current responses increased with the proceeding scans indicative of materials growth. The additional cathodic peak at ~-1.1 V during the growth of MoS$_x$/rGO, is the reduction of GO.$^{[12]}$

The MoS$_x$ film with granular surface was uniformly coated on the SSM (Figure 1b, c). It is of a compacted nature with a thickness of ~354 nm (Figure 1d). The MoS$_x$/rGO film displayed a porous three-dimensional architecture created by the randomly stacked nanosheets (Figure 1e, f, g) with an average thickness of 1-2 µm by referring to the blank SSM (Figure S3, Supporting Information). The typical thickness of the nansheets is ~89 nm, and the sheet size several hundreds of nanometers is accordant with that of GO nanosheets (Figure S4, Supporting Information). The decorated nanoparticles in a size of 20 to 50 nm on the sheets formed a granular surface, which was MoS$_x$ as revealed by the energy dispersive X-ray spectroscopy (Figure S5, Supporting Information). This sheet-like structure with a granular surface can be further confirmed by the transmission electron microscopy (TEM) images (Figure 1h). The uncoated rGO sheet at the edge of the composite sheet (Figure 1i) evidences its role as a backbone or template for MoS$_x$ growth. The neat MoS$_x$ film consisted of bulky particles (Figure 1j). A typical lattice spacing of 0.66 nm corresponding to the (002) crystal plane of layered MoS$_2$, was shown for both MoS$_x$ and the composite (Figure 1j, k), yet in an amorphous state indicated by the absence of characteristic peaks in the XRD patterns (Figure S6, Supporting Information).
The chemical composition of these materials was characterized with X-ray photoelectron spectroscopy (XPS). Both MoS$_x$/rGO and MoS$_x$ showed strong Mo and S peaks in wide spectra (Figure 2a, d), the evidence of molybdenum sulfides. The intensity ratio of Mo:S for MoS$_x$ and MoS$_x$/rGO was 1:2.7 and 1:2.4, proving the existence of MoS$_3$. The ratio between MoS$_2$ and MoS$_3$ in MoS$_x$ and MoS$_x$/rGO was estimated to be 0.43 and 1.5, respectively. In the Mo 3d region, two major peaks at ~229.2 and 232.4 eV can be assigned to Mo 3d$_{5/2}$ and Mo 3d$_{3/2}$ binding energies (Figure 2b, e), typical for Mo$^{4+}$ in MoS$_2$. The peak at ~235.2 eV arises from Mo$^{6+}$. In the S 2p region (Figure 2c, f), two dominant peaks at the lower binding energy (~161.7 and 162.8 eV) represent S$^{2-}$ 2p$_{3/2}$ and S$^{2-}$ 2p$_{1/2}$, characteristic S 2p peaks of MoS$_2$ crystals. The other two peaks at ~163.0 and 164.3 eV are attributed to S$_2^{2-}$ 2p$_{3/2}$ and S$_2^{2-}$ 2p$_{1/2}$, suggesting the existence of bridging S$_2^{2-}$ ligands, which constitutes the MoS$_3$ in the form of Mo$^{IV}$(S$_2^{2-}$)(S$_2^{2-}$).$^{[13]}$ The peak representing C-O and C=O at ~289.2 eV and ~290.0 eV decreased dramatically in the C 1s spectra of MoS$_x$/rGO (Figure S7, Supporting Information). The C/O ratio in MoS$_x$/rGO increased to 4.0 from 1.9 for GO, which confirms the reduction of GO in this composite.

The electrochemical properties of these two materials in a LIB were investigated using cyclic voltammetry at 0.2 mV s$^{-1}$ and galvanostatic charge discharge at 0.1 A g$^{-1}$. They all exhibited similar characteristic peaks in the cyclic voltammograms (Figure 3a, b): a sharp cathodic peak at ~0.5 V in the first cycle corresponding to formation of solid electrolyte interphase (SEI), the main cause for the irreversible capacity in the charge/discharge profiles (Figure 3c, d).$^{[14]}$ In the following cycles, three cathodic peaks at approximately 1.83, 1.05 and 0.45 V were shown, which respectively corresponds to the formation of Li$_2$S and Li$_x$MoS$_2$, decomposition of lithiated MoS$_2$.$^{[15]}$ In the anodic process, two major peaks at ~1.52 and 2.31 V were presented which can be ascribed to the partial oxidation of Mo forming MoS$_2$, and formation of sulfur, respectively.$^{[15]}$ The plateaus at 2.0 and 1.2 V in the discharge process, and 2.1 and 1.3 V in the charge process (Figure 3c, d) agree well with the CV results.
The rate capabilities of MoS$_x$ and MoS$_x$/rGO were investigated (Figure 3e). The MoS$_x$/rGO film showed much higher specific discharge capacities at all the current applied (0.1-2 A g$^{-1}$). At 0.1 A g$^{-1}$, neat MoS$_x$ delivered an initial capacity of 588 mAh g$^{-1}$ and a reversible capacity of 562 mAh g$^{-1}$, whereas it was 1370 and 1214 mAh g$^{-1}$ for MoS$_x$/rGO. The MoS$_x$/rGO demonstrated an excellent rate capability: 1137, 1095, 1058, 1051 and 1016 mAh g$^{-1}$ at a current density of 0.2, 0.4, 0.8, 1 and 2 A g$^{-1}$. The lithium storage performance is better than or comparable to the reported MoS$_2$/graphene composite, including hydrothermally synthesized MoS$_2$ based$^{[2b, 14, 16]}$ and exfoliated MoS$_2$ nanosheets based-composites.$^{[17]}$ It is also superior than that of the MoS$_x$/carbon nanotubes composites.$^{[18]}$ The detailed comparison is listed in Table S1. Most importantly, this material can be readily electrodeposited onto the substrate with uneven surface and even those micro-scaled 3D type with an increased electrode height as a conformal coating.$^{[5b, 5c]}$ All these features prove that this MoS$_x$/rGO film is highly competitive as an anode in LIBs, especially for those micro-scaled LIBs which are highly demanded for autonomous micro-electronic systems such as medical implants, micro sensors, or microelectromechanical systems.$^{[5a]}$

The cycling performance of MoS$_x$ and MoS$_x$/rGO was studied at a current density of 0.1 A g$^{-1}$ (Figure 3f). The neat MoS$_x$ shows an ascending trend with a capacity of 709 mAh g$^{-1}$ delivered at the 100th cycle from the initial 585 mAh g$^{-1}$. The composite displayed an overall ascending trend, a capacity of 1504 mAh g$^{-1}$ at the 110th cycle compared to the initial 1299 mAh g$^{-1}$. The increasing capacity can be mainly due to the low crystallinity of MoS$_x$, in which the existence of MoS$_3$ can help stabilize the polysulfides generated from MoS$_2$ upon discharging, preventing them from diffusion loss.$^{[19]}$ This can be supported by the electrochemical impedance spectra (EIS, Figure 3g and h). The charge transfer resistance (R_{ct}) for MoS$_x$ and MoS$_x$/rGO became lower after the cycling test, demonstrating the enhanced kinetic performance upon cycling. These two materials displayed a Coulombic efficiency (CE) of 98.3% and 97% for the first cycle, and then it remained at ~100%, meeting the
requirement (>90%) of commercial graphite.\textsuperscript{[20]} The high CE at the first cycle might be ascribed to the easy access of electrolyte to MoS\textsubscript{x} film with nano-scale granular surface and MoS\textsubscript{x}/rGO film with a hierarchical porous structure. Absence of inactive materials in the electrode, free of surface oxide layer contamination may also contribute as previously reported.\textsuperscript{[21]} A high and stable CE is critical to achieve excellent cycling stability.\textsuperscript{[22]} Such excellent electrochemical Li storage performance afforded by MoS\textsubscript{x}/rGO may be ascribed to its unique structure: highly conductive rGO matrix acts as the pathway for electron transport benefiting the rate performance; the well-dispersed MoS\textsubscript{x} nanoparticles afford the fast redox reactions; the enlarged electrode/electrolyte interface from the 3D porous structure further facilitates the ion diffusion into the inner structure of the electrode materials.

The great rate performance of these MoS\textsubscript{x}-based electrodes inspired us to investigate the Li-storage mechanism. The overall capacity of a material is derived from the intercalation of Li ions and pseudocapacitive behaviors (e.g. faradaic reactions at the surface).\textsuperscript{[23]} This study is based on the CV data at different scan rates (0.2-1 mV s\textsuperscript{-1}) (Figure 4a, S8a, Supporting Information). Two oxidative and two reductive peaks are marked as O1, O2, R1 and R2, respectively. The pseudocapacitive contributions at various scan rates were estimated based on the peak current ($i$) and scan rate ($v$) (Figure 4c, S8c, Supporting Information) and shown in Figure 4d (see Supporting Information for detailed information). The slope of the linear line in the plot of log($i$) vs. log($v$) (Figure 4b, S8b, Supporting Information) is assigned as $b$-value. When $b$-value is 0.5, the electrochemical process is dominated by a diffusion-controlled intercalation, while it is mainly controlled by the pseudocapacitive reactions with a $b$-value of 1. The $b$-value of both samples is in a range of 0.79-1.00 (Table S2), inferring that the pseudocapacitive component takes the major part in the overall capacity. The charge storage of Li from faradaic reactions at the material surface is of great significance when the material is close to nanoscale,\textsuperscript{[24]} that can perform fast Li storage behaviors. It can hence elucidate the excellent rate capability of MoS\textsubscript{x} and MoS\textsubscript{x}/rGO.
In summary, the electrodeposited MoS$_x$ and MoS$_x$/rGO composite films exhibit Li storage behavior typical of molybdenum sulfide. The composite film demonstrates an exceptional performance including high capacity, excellent rate capability and superior cycling performance. Such performance can be attributed to its ideal structure: a 3D porous structure created with nanosheets that is composed of conductive graphene nanosheets decorated with nano-granuled MoS$_x$ particles. The large pseudocapacitive contribution to the overall capacity affords excellent rate performance. The combination of the outstanding electrochemical properties and the facile and straightforward fabrication method makes this highly integrated 3D MoS$_x$/rGO composite a promising material for LIBs and especially for micro-scaled LIBs.

**Experimental Section**

*Synthesis of graphene oxide (GO):* The GO was synthesized via a modified Hummer’s method.$^{[25]}$ Typically, graphite powder (2 g) and NaNO$_3$ (1 g) were added into the concentrated H$_2$SO$_4$ (75 mL) in an ice bath, followed by gradual addition of KMnO$_4$ (5 g) under stirring. This mixture was stirred at room temperature for 6 days, and then diluted with 5% H$_2$SO$_4$ (140 mL). It was stirred for another 2 h at 90 °C, followed by the addition of 30% H$_2$O$_2$ (5 mL). The resulting precipitate, graphite oxide, was rinsed with HCl aqueous solution (1:10) and water. Graphite oxide was sonicated into water to form graphene oxide dispersion, which was dialyzed for 1 week prior to use.

*Synthesis of MoS$_x$ and MoS$_x$/rGO:* The MoS$_x$ and MoS$_x$/rGO was synthesized using cyclic voltammetry (CV).$^{[9]}$ Briefly, the electrodeposition was performed over the range of -1.3 to 0.4 V (vs. Ag/AgCl) at a scan rate of 20 mV s$^{-1}$ for 30 cycles on a stainless steel mesh. The solution contained 5 mM (NH$_4$)$_2$MoS$_4$ and 0.1 M KCl with or without 0.25 mg mL$^{-1}$ GO.

*Material characterization:* The samples were characterized with field emission scanning electron microscopy (FE-SEM) (JEOL JSM-7500FA), and transmission electron microscopy (TEM) (JEOL JEM-2200FS). X-ray photoelectron spectroscopy (XPS) spectra were collected
by illuminating the samples with a non-monochromatic x-ray source (Omnivac) using Al Kα (1486.6 eV) radiation, and the photoemission collected by an SES2002 analyser (Scienta). The topographic data of GO sheets were collected by atomic force microscopy (AFM, Asylum Research, MFP-3D). The crystal structure was analysed by X-ray diffraction (XRD, GBC MMA diffractometer) with Cu Kα radiation at a scan rate of 4 degree min⁻¹.

**Electrochemical characterization:** The LR 2032 type coin cell was assembled with the electrode (0.8 × 0.8 cm) coupled with a lithium foil using 1 M LiPF₆ in 1:1 (v/v) ethylene carbonate/dimethyl carbonate as electrolyte. Cyclic voltammetry (CV) was performed on a Solartron SI 1287. The galvanostatic charge/discharge tests were performed on a LAND CT2001A battery test system. Electrochemical impedance spectroscopy (EIS) measurements were carried out on a Bio-logic workstation (VSP model) at an open circuit potential over the frequency range of 0.01 Hz to 100 kHz.

**Supporting Information**

Supporting Information is available from the Wiley Online Library or from the author.

**Acknowledgements**

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**Conflict of Interest**

The authors declare no conflict of interest.
References


**Figures**

**Figure 1.** (a) Schematic illustration of the formation of MoS$_x$/rGO; SEM images of MoS$_x$ (b-d) and MoS$_x$/rGO (e-g) at different magnifications; TEM images of MoS$_x$/rGO (h, i) and MoS$_x$ (j, k).
Figure 2. XPS survey spectra, Mo 3d spectra and S 2p spectra of MoS$_x$ (a-c) and MoS$_x$/rGO (d-f).
Figure 3 The first three cyclic voltammograms of MoS$_x$ (a) and MoS$_x$/rGO (b) over a potential range of 0.0-3.0 V vs. Li/Li$^+$ at 0.2 mV s$^{-1}$; The first three charge/discharge curves of MoS$_x$ (c) and MoS$_x$/rGO (d) at 0.1 A g$^{-1}$; Rate performance (e) and cycling behaviors at 0.1 A g$^{-1}$ (f); Nyquist plots of the fresh cells (g) and cells after 100 cycles (h) at 0.1 A g$^{-1}$ (solid line: fitted curves; Inset of (h): the equivalent circuit).
Figure 4. Cyclic voltammograms recorded at different scan rates (a), log $i$ vs. log $v$ plots for each peak (b), and relationship between the peak current and the scan rate (c) of MoS$_x$/rGO; Bar chart (d) displaying the pseudocapacitive contributions of MoS$_x$ and MoS$_x$/rGO.
A MoS$_2$/rGO composite film with an ideal three-dimensional porous electrode structure is realized using a facile and straightforward electrochemical method. The electrode produces an enhanced electrochemical performance as an anode in lithium ion battery.

**Keywords**
molybdenum sulfide, graphene, 3D structure, electrochemical deposition, lithium ion battery

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An electrosynthesized three-dimensional porous molybdenum sulfide/graphene film with enhanced electrochemical performance for lithium storage
Supporting Information

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Figure S1 Raman spectra of MoS$_x$, MoS$_x$/rGO and GO.
**Figure S2** Cyclic voltammograms during the growth of MoS$_x$ (a) and MoS$_x$/rGO composite (b).

**Figure S3** SEM images of the blank SSM (a), and with a coating of MoS$_x$ (b) and MoS$_x$/rGO (c).
Figure S4 AFM topology (a) and height profile (b) of GO nanosheets.

Figure S5 Energy dispersive X-ray spectroscopy (EDS) elemental mapping of Mo, S and C of MoS$_x$/rGO composite.
Computational details

The power-law relationship between peak current and scan rate is described in the following equations:\(^1\)

\[
i = av^b
\]

\[
\log(i) = b \log(v) + \log(a)
\]

where \(i\) is the peak current in mA, \(v\) is the scan rate in mV \(s^{-1}\), and \(a\) and \(b\) are adjustable parameters. The \(b\)-value can be obtained as the slope of the linear line in the plot of \(\log(i)\) vs. \(\log(v)\) (Figure 4b, S8b).
To quantify the contributions of capacitive and intercalation reactions, the following equation based on the dependence between peak current and scan rate was applied:[1]

\[ i = k_1 v + k_2 v^{1/2} \]  

(3)

where \( i \) is the peak current in mA, \( v \) is the scan rate in mV s\(^{-1}\), and \( k_1 \) and \( k_2 \) are constants. Herein, \( k_1 v \) and \( k_2 v^{1/2} \) are the capacitive and intercalation components, respectively. By rearranging the equation to \( \frac{i}{v^{1/2}} = k_1 v^{1/2} + k_2 \), the \( \frac{i}{v^{1/2}} \) vs. \( v^{1/2} \) plots were depicted in Figure 4c and S8c, in which constants \( k_1 \) and \( k_2 \) can be obtained as the slope and \( y \)-intercept of each line.

**Figure S8** Cyclic voltammograms of neat MoS\(_x\) recorded at different scan rates (a), log \( i \) vs. log \( v \) plots at each redox peak (b) and relationship between the peak current and the scan rate (c) of neat MoS\(_x\) electrodes.
Table S1 Comparison of capacitive performance of MoS$_x$/rGO composite in this work with the previously reported results.

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<th>Anode materials</th>
<th>Reversible capacity</th>
<th>Rate performance</th>
<th>Cycling stability</th>
<th>Ref.</th>
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<tr>
<td>MoS$_x$/rGO</td>
<td>1214 mAh g$^{-1}$ at 0.1 A g$^{-1}$</td>
<td>1016 mAh g$^{-1}$ at 2 A g$^{-1}$</td>
<td>1504 mAh g$^{-1}$ at 110 cycles at 0.1 A g$^{-1}$</td>
<td>This work</td>
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<td>~ 1100 mAh g$^{-1}$ at 0.1 A g$^{-1}$</td>
<td>~ 900 mAh g$^{-1}$ at 1 A g$^{-1}$</td>
<td>1187 mAh g$^{-1}$ at 100th cycle at 0.1 A g$^{-1}$</td>
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<td>1077 mAh g$^{-1}$ at 0.1 A g$^{-1}$</td>
<td>890 mAh g$^{-1}$ at 1 A g$^{-1}$</td>
<td>~ 900 mAh g$^{-1}$ over 400 cycles at 1 A g$^{-1}$</td>
<td>14</td>
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<td>Hydrothermally synthesized</td>
<td>~ 1300 mAh g$^{-1}$ at 0.1 A g$^{-1}$</td>
<td>1040 mAh g$^{-1}$ at 1 A g$^{-1}$</td>
<td>1290 mAh g$^{-1}$ over 50 cycles at 0.1 A g$^{-1}$</td>
<td>16a</td>
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<td>MoS$_2$/graphene composites</td>
<td>980 mAh g$^{-1}$ at 0.1 A g$^{-1}$</td>
<td>740 mAh g$^{-1}$ at 1 A g$^{-1}$</td>
<td>980 mAh g$^{-1}$ over 100 cycles at 0.1 A g$^{-1}$</td>
<td>16b</td>
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<td>912 mAh g$^{-1}$ at 0.1 A g$^{-1}$</td>
<td>571 mAh g$^{-1}$ at 1 A g$^{-1}$</td>
<td>808 mAh g$^{-1}$ over 100 cycles at 0.1 A g$^{-1}$</td>
<td>16c</td>
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<tr>
<td>Exfoliated MoS$_2$ nanosheets hybrid with graphene</td>
<td>1080 mAh g$^{-1}$ at 0.1 A g$^{-1}$</td>
<td>545 mAh g$^{-1}$ at 2 A g$^{-1}$</td>
<td>890 mAh g$^{-1}$ over 100 cycles at 0.1 A g$^{-1}$</td>
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<td>635 mAh g$^{-1}$ at 0.5 A g$^{-1}$</td>
<td>339 mAh g$^{-1}$ at 2 A g$^{-1}$</td>
<td>915 mAh g$^{-1}$ over 700 cycles at 0.5 A g$^{-1}$</td>
<td>17b</td>
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<td>MoS$_x$/CNT</td>
<td>&gt;1000 mAh g$^{-1}$</td>
<td>197 mAh g$^{-1}$ at 2 A g$^{-1}$</td>
<td>&gt;1000 mAh g$^{-1}$ over 45 cycles at 50 mA g$^{-1}$</td>
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Table S2 $b$-values at each redox peak of neat MoS$_x$ and MoS$_x$/rGO samples.

<table>
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<th>Samples</th>
<th>O1</th>
<th>O2</th>
<th>R1</th>
<th>R2</th>
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<tr>
<td>MoS$_x$</td>
<td>0.94</td>
<td>0.83</td>
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<td>0.79</td>
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<tr>
<td>MoS$_x$/rGO</td>
<td>0.85</td>
<td>0.86</td>
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