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# 1. Introduction

Nowadays, lithium ion batteries (LIBs) and supercapacitors which can realize the conversion of electric energy and chemical energy are predominant power supplies with a wide range of applications. The energy storage mechanism in LIBs is a typical chemical process accompanied by Li<sup>+</sup> ion insertion/ extraction into/from electrode materials, while in supercapacitors it is mainly based on the physical adsorption of charges coupling with pseudocapacitive reactions on the surfaces of electrode materials. The different energy storage mechanisms impart high energy densities to LIBs,<sup>1</sup> while high power outputs to supercapacitors.<sup>2</sup> For both devices electrode materials are considered as the key components determining the overall performances. Therefore, developing high-perform-

# One-pot mass preparation of MoS<sub>2</sub>/C aerogels for high-performance supercapacitors and lithium-ion batteries†

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In this paper, we report the successful design and synthesis of a hierarchically porous  $MoS_2/C$  composite aerogel by simple one-pot mass preparation. The synthesis involves the *in situ* formation of  $MoS_2$ nanosheets on agarose molecular chains, the gelation of  $MoS_2$ -deposited agarose monomers to generate a composite hydrogel, and *in situ* transformation of the composite hydrogel into a  $MoS_2/C$  composite aerogel through carbonization. This composite aerogel can be used as a high-performance electrode material for supercapacitors and lithium-ion batteries. When tested as a supercapacitor electrode, it achieves a high specific capacitance of 712.6 F g<sup>-1</sup> at 1 A g<sup>-1</sup> and 97.3% capacity retention after 13 000 cycles at 6 A g<sup>-1</sup>. In addition, as a lithium-ion battery electrode, it exhibits a superior rate capability (653.2 mA h g<sup>-1</sup> at 0.1 A g<sup>-1</sup> and 334.5 mA h g<sup>-1</sup> at 5.0 A g<sup>-1</sup>) and an ultrahigh capacity retention of nearly 100% after 1000 cycles at 1 A g<sup>-1</sup>. These performances may be ascribed to the unique structure of the  $MoS_2/C$  composite aerogel, such as hierarchical pores, (002) plane-expanded  $MoS_2$  and interconnected carbon networks embedded uniformly with  $MoS_2$  nanosheets. This work may provide a general and simple approach for mass preparation of composite aerogel materials and pave the way for promising materials applied in both supercapacitors and lithium-ion batteries.

> ance electrode materials has been a significant strategy to address the ever-increase demand for practical energy applications. Nevertheless, subjected to the different working mechanisms in two devices, electrode materials were often exclusively designed and applied to either LIBs or supercapacitors, which limit the practical application scope of the developed electrode materials. It is highly desired to develop suitable materials as high-performance electrodes for both LIBs and supercapacitors.

> Molybdenum disulfide  $(MoS_2)$ , as a typical two-dimensional layered metal dichalcogenide with interconnected sulfurmolybdenum-sulfur bonds,<sup>3</sup> has drawn great attention due to its unique structure and attractive properties.<sup>4-6</sup> Similar to graphite, MoS<sub>2</sub> has a layered planar structure and the interlayered space conducive to the lithium intercalation.<sup>7,8</sup> In addition, the central Mo atoms have the valence ranging from +2 to  $+6^{9}$ showing great potential for electrochemical pseudocapacitance. Therefore, MoS<sub>2</sub> is considered as an ideal material for LIBs and supercapacitors. However, as a semiconducting material, MoS<sub>2</sub> shows poor electrical conductivity which is a drawback for LIBs and supercapacitors. To improve the conductivity of molybdenum disulfide, a variety of strategies have been developed to synthesize carbonaceous MoS2 composite materials such as graphene– $MoS_2^{10-12}$  and other carbon– $MoS_2$ composites<sup>13-19</sup> for LIBs and supercapacitors. However, few of



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 $MoS_2/C$  composites reported so far are suitable for both lithium-ion batteries and supercapacitors. Moreover, the  $MoS_2/C$  composites are produced on a gram scale, which is difficult to scale up.

Herein, we propose a simple method for the mass preparation of  $MoS_2$ /carbon composite aerogels which can be employed as excellent electrodes for both lithium-ion batteries and supercapacitors. The prepared  $MoS_2/C$  aerogel combines the advantages of 3D interconnected frameworks, hierarchical pores, interlayer-expanded  $MoS_2$  which are beneficial for electron transport, fast electrolyte/Li ion diffusion and efficient ion intercalation into  $MoS_2$ . When used as an electrode material for supercapacitors, the  $MoS_2/C$  aerogel exhibits high specific capacitance and electrochemical stability. In addition, the  $MoS_2/C$  aerogel also shows excellent lithium storage performance as an anode material, such as high rate performance and long-term cycle stability.

# 2. Experimental section

#### Chemicals

Sodium molybdate dihydrate  $(Na_2MoO_4 \cdot 2H_2O)$ , thioacetamide  $(C_2H_5NS)$  and agarose  $(C_{12}H_{18}O_9)$  were purchased from Aladdin Reagent Co. Ltd and used without further purification.

#### Synthesis of MoS<sub>2</sub>/agarose aerogel

In a typical experiment, a mixed solution was prepared by dissolving 0.3 g sodium molybdate dihydrate and 0.6 g thioacetamide in 20 mL deionized water. Then, 1.5 g agarose was added. The reaction solution was kept at 90 °C for 5 days with strong stirring. A dark brown hydrogel was formed when the reaction system was cooled down to 4 °C in a freezer. Subsequently, the gel was rinsed with deionized water and dialyzed for 3 days.

#### Synthesis of MoS<sub>2</sub>/C composite aerogel

The as-synthesized hydrogel was treated through the freezedrying process and then thermally decomposed in a tube furnace at 550 °C for 4 h and then 700 °C for 2 h under an Ar atmosphere with a heating rate of 2 °C min<sup>-1</sup>.

#### Characterization

The general morphologies were characterized by using a fieldemission scanning electron microscope (FE-SEM, Hitachi S4800, 3 kV) equipped with an X-ray energy dispersive spectroscopy (EDS) analysis system and a high-resolution transmission electron microscope (HR-TEM, JEM 2011, 200 kV). The crystal structure was characterized by X-ray diffraction (XRD) using a D/max2550VB3+/PC X-ray diffractometer with a Cu K $\alpha$  radiation source ( $\lambda = 0.15418$  nm). The Raman spectrum was recorded by using a spectrophotometer (inVia, Renishaw, Germany) with a 514 nm laser. The specific surface area was analyzed on a Quantachrome Nova 2200e and calculated *via* the Brunauer–Emmett–Teller (BET) model, and the pore size distribution was estimated by the Barret–Joyner–Halenda (BJH) method. The Mo element was detected by inductively coupled plasma atomic emission spectroscopy (ICP-AES, PerkinElemer Optima 8000).

#### Evaluation of the performance of supercapacitors

Measurements were done using a standard three-electrode cell, including an Ag/AgCl electrode as a reference electrode, a platinum wire as a counter electrode and a working electrode. The working electrode was a mixture of the as-synthesized sample, active carbon and polytetrafluoroethylene (PTFE) in ethanol with a weight ratio of 8:1:1. The slurry was pasted onto nickel foam after being treated by ultrasound for 1 h and dried in a vacuum oven at 110 °C for 24 h. The electrolyte was an aqueous solution containing 6 M KOH. Electrochemical measurements were conducted using a CHI660E electrochemical analyzer (CH Instruments, Inc., Shanghai).

#### Evaluation of the performance of lithium-ion batteries

The working electrode slurry was a mixture of the assynthesized sample, active carbon and polyvinylidene difluoride (PVDF) in *N*-methyl pyrrolidinone (NMP) with a weight ratio of 8:1:1. The slurry was pasted onto a copper foil after being stirred overnight and dried in a vacuum oven at 80 °C for 24 h. Cells were assembled in an Ar-filed glove box. Cyclic voltammetry (CV) was performed using a CHI660E electrochemical analyzer (CH Instruments, Inc., Shanghai). Galvanostatic charge– discharge experiments were conducted using a LAND CT2001A battery testing system (Wuhan, China) at 25 °C.

### 3. Results and discussion

Fig. 1 shows a schematic illustration of the formation of the  $MoS_2/C$  aerogel. Firstly,  $Na_2MoO_4 \cdot 2H_2O$  gives  $Na^+$  and  $MoO_4^{2-}$  ions in water and  $C_2H_5NS$  hydrolyzes (reaction (1)). As the temperature rises,  $H_2S$  released from thioacetamide reacts with  $MOO_4^{2-}$  ions slowly to produce  $MoS_2$  according to reaction (2).<sup>20</sup>

$$C_2H_5NS + H_2O \rightarrow CH_3CONH_2 + H_2S$$
 (1)

$$MoO_4{}^{2-} + 3H_2S \rightarrow MoS_2 + 3H_2 + SO_4{}^{2-}$$
 (2)

Agarose monomers dissolve in water acting as surfactants. The agarose monomer with four hydroxyls can absorb the reactant of  $MoO_4^{2^-}$  ions in solution and allow *in situ* nucleation and formation of  $MoS_2$  nanosheets on agarose molecular chains, which provide steric protection of  $MoS_2$  nanosheets from agglomeration. When the reaction system cools down, a network structure composed of agarose monomers, *i.e.* an agarose hydrogel is formed due to the strong interaction of hydrogen bonds, and  $MoS_2$  nanosheets are locked separately within the space of the network structure. Introducing the inorganic nanoparticle component into the structure is able to strengthen the toughness of the whole gel.<sup>21</sup> In the following carbonization step, the networks of agarose chains deposited



Fig. 1 Schematic illustration of the synthetic route to the formation of the  $MoS_2/C$  composite aerogel.

with  $MoS_2$  in situ transform to the  $MoS_2$  nanosheet/carbon skeleton aerogel. In this step, the carbon skeleton prevents  $MoS_2$  nanosheets from agglomeration, and in turn  $MoS_2$ nanosheets exert the same effect on agarose chains, conducive to forming a variety of pores in different sizes in the composite aerogel during carbonization.

Fig. 2a shows the optical photograph of the obtained  $MOS_2/C$  composite aerogel. It is clear that the diameter and height of the cylinder-like aerogel are approximately 3 cm and 1.7 cm, respectively. The density of the aerogel was calculated to be approximately 0.04 g cm<sup>-3</sup>, which makes the aerogel stand freely on a brush. In addition, the production of this aerogel can be easy to enlarge, depending on the preparation of the precursor. Fig. S1† shows the precursor formed on a larger scale with a size of 15.8 cm (diameter) and 4.7 cm (height). Fig. 2b and c display the SEM images of the obtained  $MOS_2/C$  composite aerogel at different magnifications. The hierarchically porous structure is shown and pore sizes range from less than one micrometer to several hundred micrometers. The

pores of different sizes in the carbon skeleton are formed due to the sublimation of water during the freeze-drying process,<sup>22</sup> the removal of the oxygen-containing group in the agarose monomers during thermal processing and the steric effect of MoS<sub>2</sub> nanosheets dispersed on the composite. It is found from the BET results (Fig. S2<sup>†</sup>) that the surface area of the obtained  $MoS_2/C$  composite aerogel is 212.0 m<sup>2</sup> g<sup>-1</sup> and the distribution of the pore diameter centers at approximately 3.8 nm. Also, the pores with different sizes are shown in Fig. S3,† which further confirms that the MoS<sub>2</sub>/C composite aerogel is a hierarchically porous structure comprising macro-, meso- and micropores. The element maps indicate the homogeneous distribution of the three elements C, Mo and S (Fig. 2d), in which the C element accounts for a greater proportion as the color standing for the C element is more intensive than others. Specifically, the atomic ratios of three elements in the composite aerogel are 86.3% (C), 4.6% (Mo) and 9.1% (S), respectively, indicating a mass percentage of 41.3% for MoS<sub>2</sub> in the composite aerogel. The mapping analysis matches the energy dispersive



Fig. 2 Optical photographs (a), SEM images (b and c), element maps (d) and TEM images (e, f) of the MoS<sub>2</sub>/C composite aerogel.

X-ray spectrometry (EDS) results (Fig. S4<sup>†</sup>), which furthermore proves that the MoS<sub>2</sub> composite aerogel is composed of the three elements. Fig. 2e and f show the typical TEM images of the  $MoS_2/C$  composite aerogel at a higher magnification. It is found that MoS<sub>2</sub> nanosheets are embedded uniformly in the carbon skeleton, showing a distorted morphology (Fig. 2e). The crystal lattices of MoS<sub>2</sub> nanosheets inside the composite aerogel are difficult to be clearly observed due to the interference of thick carbon skeletons. To have a clear observation, an area on the surface of the composite aerogel was selected for study, as shown in Fig. 2f. The TEM image in Fig. 2f displays the expanded (002) planes of MoS<sub>2</sub> (JCPDS card no.37-1492), with a d-spacing of approximately 7.20 Å, which may be caused by the insertion of Na<sup>+</sup> ions. It is reported that Na<sup>+</sup> ions are able to intercalate into MoS<sub>2</sub> layers, increasing the distance of the interplanar spacing of the (002) planes.<sup>23</sup> For example, Hu et al. found that the intercalation of Na<sup>+</sup> ions into the MoS<sub>2</sub> layers during the hydrothermal process led to the peak of the (002) plane shifting to a low angle.<sup>24</sup> Moreover, reported by Alla Zak et al., Na atoms could also intercalate into MoS<sub>2</sub> layers though a vapor transport technique, leading to the d-spacing increase of the (002) plane, which was proved by XPS and EDS analyses.25

Furthermore, the composition of the MoS<sub>2</sub>/C composite aerogel is confirmed by XRD and Raman analyses. Fig. 3a shows the XRD pattern of the aerogel and it is found that there are three diffraction peaks at  $2\theta = 32.7$ , 39.5 and 58.3°, which can be assigned to the (100), (103), (110) planes of the hexagonal-phase MoS<sub>2</sub> (JCPDS card no.37-1492). It is noted that the peak position (13.2°) corresponding to the (002) plane shifts to the left compared with that  $(14.4^{\circ})$  of the normal MoS<sub>2</sub>, indicating that the distance between the (002) planes increases, which is in good agreement with the interplanar spacing measured in the TEM image (Fig. 2f). Except for the (002) peak, other peaks have no apparent shifts because their corresponding interplanar spacings are too small to be intercalated by Na<sup>+</sup> ions. Fig. 3b displays the Raman spectrum of the  $MoS_2/$ C composite aerogel. It is found that the Raman spectrum from 350  $\text{cm}^{-1}$  to 420  $\text{cm}^{-1}$  exhibits two vibration peaks at 383 cm<sup>-1</sup> and 405 cm<sup>-1</sup> which respectively belong to the  $E_{2g}^1$ and A<sub>1g</sub> vibration modes of MoS<sub>2</sub>.<sup>26,27</sup> There are additional two

bands in the range of 1300–1700 cm<sup>-1</sup> matching the D band and G band of the carbon skeleton. The D band at approximately 1400 cm<sup>-1</sup> associates with the defect and lattice distortions in the carbon skeleton and the G band located at about 1580 cm<sup>-1</sup> corresponds to the relative motion of sp<sup>2</sup> carbon atoms.<sup>28</sup>

Fig. 4 displays the electrochemical performance of the  $MoS_2/C$  composite aerogel when used as an electrode material in supercapacitors. Fig. 4a shows the CV curves at different scan rates, ranging from 5 to 800 mV s<sup>-1</sup>. All the CV curves display a pair of redox peaks, proving the presence of a pseudo-capacitive behavior and reversible Faradaic reactions. As the scan rate increases, the current response increases accordingly and the peak position changes, but the shape of CV curves retains, which indicates a good electrochemical stability and rate performance of the MoS<sub>2</sub>/C aerogel. The galvanostatic charging-discharging curves of the MoS<sub>2</sub>/C composite aerogel are measured at different current densities, ranging from 1 A g<sup>-1</sup> to 20 A g<sup>-1</sup>, as displayed in Fig. 4b. The specific capacitances at the corresponding current densities are plotted in Fig. 4c. The specific capacitance is calculated based on eqn (1):<sup>29</sup>

$$C = \frac{2i_{\rm m} \int V \mathrm{d}t}{\frac{V^2 |_{V_i}^{V_f}}{V_i}} \tag{1}$$

where C (F g<sup>-1</sup>) is the specific capacitance, and  $i_m = I/m$  $(A g^{-1})$  is the current density, where I represents the current and *m* represents the active mass of the electrode.  $\int V dt$  represents the integral current area and V represents the potential with the initial and final values of  $V_i$  and  $V_f$ , respectively. Among various tested conditions, the capacitances are calculated with eqn (1) using the integral current areas of the discharge curves in Fig. 4b, as shown in Fig. 4c. The MoS<sub>2</sub>/C aerogel electrode exhibits the specific capacitances of 712.6, 669.6, 640.1, 615.2, 592.8, 578.2, 543.3, 513.6, 464.3, 447.5 and 415.1 F g<sup>-1</sup> at current densities of 1, 2, 3, 4, 5, 6, 8, 10, 14, 16 and 20 A  $g^{-1}$ , respectively. The specific capacitance achieves a maximum of 712.6 F  $g^{-1}$  at a low current density of 1 A  $g^{-1}$ , which is comparable to or higher than previously reported supercapacitors based on MoS<sub>2</sub> materials. The high capacitance may be attributed to the pseudocapacitance caused by the central Mo atoms



Fig. 3 XRD pattern (a), and Raman spectrum (b) of the MoS<sub>2</sub>/C composite aerogel.

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with the valence ranging from +2 to +6 in the discharging/ charging processes. Even if measured at a high current density of 20 A  $g^{-1}$ , the specific capacitance still maintains 415.1 F  $g^{-1}$ , which is 58.2% of the maximum at 1 A  $g^{-1}$ . The good rate performance may be attributed to the hierarchically porous structure of the composite aerogel, which may provide shorter pathways for fast and efficient ion transport. Fig. 4d shows the long-term cycle stability of the aerogel at 6 A  $g^{-1}$ , which is another critical factor for practical applications especially at a relatively high current density. The specific capacitance is 578.2 F  $g^{-1}$  at the initial cycle and can still maintain 562.6  $F g^{-1}$  after 13 000 cycles, giving a high capacitance retention of ~97.3%. This excellent cyclability may be owing to the fact that the carbon skeleton inhibits the aggregation of MoS<sub>2</sub> layers which enhances the stability of the composite. Table S1<sup>†</sup> summarizes some of the results that have been reported about MoS<sub>2</sub> as an electrode material used in supercapacitors.<sup>30-34</sup> Obviously, the performance of the MoS<sub>2</sub>/C composite aerogel is among the best level for MoS<sub>2</sub> material-based supercapacitors.

Fig. 5 displays the electrochemical lithium storage performance of the MoS<sub>2</sub>/C composite aerogel when used as an anode in LIBs. Fig. 5a shows the initial three CV curves in the potential range of 0.05–3.0 V at a scan rate of 5 mV s<sup>-1</sup>. In the first cathodic sweep, there are two peaks at approximately 0.7 V and 0.4 V in the first discharge. The 0.7 V peak is attributed to the formation of  $Li_xMoS_2$  (reaction (3)) that results in a phase transition from 2H to 1T,<sup>4,11</sup> which indicates the intercalation of Li<sup>+</sup> into MoS<sub>2</sub> layers.<sup>35</sup> The peak at 0.4 V corresponds to the conversion reaction of Li<sub>x</sub>MoS<sub>2</sub> to Li<sub>2</sub>S and Mo

(reaction (4)),<sup>4</sup> followed by the formation of a SEI layer irreversibly.36

$$MoS_2 + xLi^+ + xe^- \rightarrow Li_xMoS_2$$
 (3)

$$Li_xMoS_2 + (4 - x) Li^+ + (4 - x)e^- \rightarrow 2Li_2S + Mo$$
 (4)

6 A/g

8 A/g 10 A/g

14 A/g

16 A/a

250

300

200

In the first anodic scan, a weak peak at 1.4 V and a main peak at 2.3 V are found, which belong to the conversion reactions of Mo to  $MoS_2$  (reaction (5)) and  $Li_2S$  to S (reaction (6)) respectively.15,37

1

$$Mo + 2Li_2S \rightarrow MoS_2 + 4Li^+ + 4e^-$$
(5)

$$\text{Li}_2 S \rightarrow 2 \text{Li}^+ + S + 2 e^-$$
 (6)

In subsequent reduction cycles, a peak at 1.0 V and a new peak at 1.7 V are observed, which are associated with the formation of  $Li_x MoS_2$  (reaction (3)) and  $Li_2S$  (reaction (7)), respectively.7,36,38

$$S + 2Li^+ + 2e^- \rightarrow Li_2S$$
 (7)

In subsequent oxidation cycles, there are two peaks at approximately 2.3 V and 1.6 V, which belong to the dissociation oxidation of Li<sub>2</sub>S (reaction (6)) and delithiation of  $Li_xMoS_2$  (reaction (8)) produced in the cathodic process, respectively.38,39

$$\text{Li}_x \text{MoS}_2 \rightarrow x \text{Li}^+ + \text{MoS}_2 + x \text{e}^-$$
 (8)

Fig. 5b displays the typical capacity-voltage curves of the MoS<sub>2</sub>/C composite aerogel for charging and discharging at a current density of 0.1 A  $g^{-1}$  in the potential range of 0.05–3.0



**Fig. 5** Initial three CV curves at a scan rate of 5 mV s<sup>-1</sup> (a), charging–discharging curves of the initial three cycles (b), capacities at different current densities (c) and cycling performance at the current density of 1A  $g^{-1}$  (d) of the MoS<sub>2</sub>/C composite aerogel used as an electrode material in lithium-ion batteries.

V. In the first discharge, the plateaus appear at approximately 0.7 V and 0.4 V, which correspond to the formation of  $Li_xMoS_2$ and the subsequent conversion of Li<sub>x</sub>MoS<sub>2</sub> to Li<sub>2</sub>S and Mo, respectively.4 The peak at 1.7 V appearing in the next two discharges indicates the formation of Li<sub>2</sub>S.<sup>38</sup> During the first cycle, the MoS<sub>2</sub>/C aerogel electrode shows a discharge capacity of 1054.2 mA h  $g^{-1}$  and a charge capacity of 663.7 mA h  $g^{-1}$ . The irreversible capacity loss is likely attributed to the decomposition of the electrolyte and the formation of the SEI layer.<sup>40</sup> Fig. 5c shows the capacities of the MoS<sub>2</sub>/C electrode at different current densities, ranging from 0.1 A  $g^{-1}$  to 5 A  $g^{-1}$ . The capacities are 653.2, 630.2, 552.7, 512.9, 439.7 and 334.5 mA h  $g^{-1}$  at current densities of 0.1, 0.2, 0.5, 1.0, 2.0 and 5.0 A  $g^{-1}$ , respectively. As the current density returns to 0.1 A  $g^{-1}$  after 60 cycles, the capacity still maintains 653.5 mA h  $g^{-1}$ , which is even higher than the previous capacity at  $0.1 \text{ A g}^{-1}$ . Fig. 5d displays the excellent cycling performance of the  $MoS_2/C$  composite aerogel at the current density of 1 A g<sup>-1</sup>. The capacity shows an upward trend upon cycling. The high capacity of 604.3 mA h g<sup>-1</sup> is achieved after 1000 cycles, which is higher than the initial several cycles, giving a capacity retention of nearly 100%. The excellent lithium storage performance may be ascribed to the structural advantages of the MoS<sub>2</sub>/C composite aerogel such as hierarchical pores, interconnected conductive carbon networks, (002) plane-expanded MoS<sub>2</sub> embedded stably in the composite structure, which facilitate Li<sup>+</sup> diffusion and intercalation, electron transport, and electrode pulverization.

Furthermore, the influence of the reactant ratio on the structure and lithium-ion performance of the end products

has been investigated. Fig. 6 shows the comparison of the obtained MoS<sub>2</sub> composite materials at different reactant ratios. Fig. 6a shows the SEM image of the product without sodium molybdate dihydrate and thioacetamide, recorded as MC-0. Fig. 6b, c and d show the SEM images of MoS<sub>2</sub>/C composite materials prepared at different reactant ratios of  $w(Na_2MoO_4 \cdot 2H_2O) : w(C_2H_5NS) : w(C_{12}H_{18}O_9) = 1 : 2 : 15,$ 1:2:5, 1:2:2.5, recorded as MC-1, MC-2 and MC-3, respectively, where MC-2 is the typical aerogel product that has been discussed above. By ICP analyses, the contents of MoS<sub>2</sub> formed in the composite aerogels increase with increasing amount of the reactants Na<sub>2</sub>MoO<sub>4</sub>·2H<sub>2</sub>O and C<sub>2</sub>H<sub>5</sub>NS (Table S2<sup>†</sup>). It is found that the amount of reactant precursors of MoS<sub>2</sub> has a significant influence on the final structure of aerogels and the electrochemical performance. It seems that fewer MoS<sub>2</sub> reactants lead to a low cross-linking degree and an undeveloped pore structure (Fig. 6b), while more reactants lead to a higher degree of cross-linking with obvious pore structures (Fig. 6d). When too many reactants are added, the ion concentration is so high that the reaction for producing MoS<sub>2</sub> is out of control, which tends to form large MoS<sub>2</sub> aggregates (Fig. S5<sup>†</sup>). The agglomeration of MoS<sub>2</sub> may have adverse effects on the electrochemical performance. Fig. 6e displays the comparison of the cycle performance of MC-1, MC-2 and MC-3 as electrode materials in LIBs at a current density of 1 A  $g^{-1}$ . It is clear that MC-1 has good cycle performance, but its specific capacity is the lowest among the three possibly due to the fewest MoS<sub>2</sub> content with insufficient active sites for lithiation in the obtained MoS<sub>2</sub>/C composite aerogel. It is also found that MC-3 has the highest initial specific capacity but poorer cycle



**Fig. 6** SEM images of MC-0 (a), MC-1 (b), MC-2 (c) and MC-3 (d); (e) cycling performance of MC-1, MC-2 and MC-3 electrodes in lithium-ion batteries at a current density of 1 A  $g^{-1}$ ; (f) galvanostatic charging–discharging curves of MC-1, MC-2 and MC-3 electrodes in supercapacitors at a current density of 1 A  $g^{-1}$ .

stability than the other two, which may be owing to the high content of  $MoS_2$  with serious aggregation in the composite aerogel. Fig. 6f displays the galvanostatic charging–discharging curves of MC-1, MC-2 and MC-3 electrodes in supercapacitors at a current density of 1 A g<sup>-1</sup>. It is quite evident that MC-2 has the highest capacitance by comparing the integral current areas of the discharge curves. To sum up, MC-2 shows the best comprehensive performance.

# 4. Conclusion

In summary, a hierarchically porous MoS<sub>2</sub>/C composite aerogel is successfully designed and synthesized by simple one-pot mass preparation. The strategy involving the in situ formation of MoS<sub>2</sub> nanosheets on agarose molecular chains followed by the gelation of agarose monomers allows the largescale generation of the composite hydrogel and composite aerogel upon carbonization. This composite aerogel can be used as an excellent electrode material in lithium-ion batteries and supercapacitors, even though the mechanisms are totally different in these two devices. This MoS<sub>2</sub>/C composite aerogel shows high capacitance/capacity, excellent rate capability and long cyclability in both devices, which may be attributed to the unique structure of the MoS<sub>2</sub>/C composite aerogel, such as hierarchical pores, (002) plane-expanded MoS<sub>2</sub> and interconnected carbon networks embedded uniformly with MoS<sub>2</sub> nanosheets. This work may provide a general and simple

approach for the mass preparation of composite aerogel materials that are promising for practical applications in both lithium-ion batteries and supercapacitors.

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