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A Rechargeable $\text{Mg}^{2+}/\text{Li}^+$ Hybrid Battery Based on Sheet-like MoSe_2/C Nanocomposites Cathode

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Abstract

Sheet-like MoSe₂/C nanocomposites were synthesized and employed as the cathode of a rechargeable Mg²⁺/Li⁺ hybrid battery for the first time, which exhibited an excellent electrochemical performance. The structure transformation of the MoSe₂/C nanocomposite materials during the charge-discharge processes was identified by in situ XRD, and the electrochemical reactions taking place in the battery were evidenced.

Keywords: in situ XRD, sheet-like MoSe₂/C nanocomposites, Mg²⁺/Li⁺ hybrid battery

1 Introduction

With their many advantages, such as the high efficiency in energy conversion, good safety and reduced pollution, green electro-chemical energy storage devices have attracted ever increasing attention from both governments and industries, to be a key player as sustainable energy sources in the 21st century. So far the lithium-ion batteries (LIBs) are the state-of-art power sources in portable electronic and transport sections. However, with the fast reduction of lithium reserves, LIBs are unlikely to be continued as a sustainable choice for large energy storage applications.[1] In addition, when lithium metal is used as the anode, the formation of lithium dendrites during charge-discharge processes may cause the battery to be short-circuited. On the other hand, the rechargeable magnesium batteries have recently received more and more attentions.[2, 3] Furthermore, magnesium has a higher theoretical volumetric capacity than Li (3832 mAh cm⁻³) and a better safety profile because it has not suffered from the aforementioned dendritic growth problem during charge-discharge processes. [4] Besides, magnesium in the earth's crust is much more abundant than lithium, it is about 24 times cheaper than Li. These advantages have made rechargeable magnesium batteries become a highly promising choice for the electrochemical energy storage. Nevertheless, there are also significant disadvantages such as intrinsic strong coulombic interactions between bivalent Mg²⁺ ions and the host materials, which cause the sluggish kinetics for the diffusion of Mg²⁺ ions and result in a large polarization, and finally lead to low Mg²⁺ intercalation levels and rapid capacity decay. [5, 6] The absence of high capacity, high voltage and fast kinetics of Mg²⁺

intercalation has seriously hindered the development of magnesium batteries.

To solve these problems, a new type of $\text{Mg}^{2+}/\text{Li}^+$ hybrid battery has recently been proposed,[7-15] which has a Mg metal anode, a Li^+ ion intercalation cathode, and an electrolyte containing both Mg^{2+} and Li^+ . Such system takes the advantages of both the fast Li^+ intercalation in cathode and the high-safety of Mg anode. Recently, Cheng et al.[7] found that Mo_6S_8 can be used as cathode materials in the $\text{Mg}^{2+}/\text{Li}^+$ hybrid battery. The charge–discharge properties of Mo_6S_8 electrodes were significantly improved in the hybrid electrolyte (from $80 \text{ mAh}\cdot\text{g}^{-1}$ up to $120\text{mAh}\cdot\text{g}^{-1}$). The reaction mechanism of cathode for the hybrid battery is related to insertion and release of lithium, rather than magnesium involved in the reaction. However, the synthesis of Mo_6S_8 materials was extremely complicated and time consuming. The specific capacity of the Mo_6S_8 ($C_{\text{theoretical}} = 128 \text{ mAh g}^{-1}$) was low. Therefore, a more suitable material is required. Since the mechanism of lithium intercalation can be applied to the hybrid battery, the selectable range of the cathode material is greatly expanded. As a result, LiCoO_2 , $\text{LiNi}_x\text{Co}_y\text{Mn}_{1-x-y}\text{O}_2$ and a series of cathode materials for LIBs have been considered. Nevertheless, in these systems the electrolyte employed, all-phenyl-complex (APC), has a narrow electrochemical stability window, which in turn limits the overall operation potentials and requires the cathode materials to have a relatively low operation potential. [16]

Recently, MoS_2 , [10] TiS_2 , [8] TiO_2 , [12, 13] $\text{Li}_4\text{Ti}_5\text{O}_{12}$ [17] and Prussian blue analogues [14] have been employed to work with the Li^+ -containing APC electrolyte. In general, the cathode materials currently found to be suitable for the hybrid battery

are much less well documented than the mature lithium ion battery, so finding suitable materials for the hybrid system can contribute significantly to advancing the commercialization of the latter. Herein, we synthesized the sheet-like MoSe_2/C nanocomposites and applied them for the hybrid battery, for the first time. As one of the family members of transition metal selenides, MoSe_2 features a lamellar crystal structure which is comprised of Se-Mo-Se sandwich layers. These single layers are held together through weak van der Waals interactions to form the similar layered structure of graphite, which is in favor of the free insertion and removal of atoms between layers.[18] The carbonaceous conductive matrix would improve the electronic conductivity and structural stability of MoSe_2 . [19] The as prepared sheet-like MoSe_2/C nanocomposites were employed as cathode for the hybrid battery, which exhibited an excellent electrochemical performance. The structure transformation of the MoSe_2/C nanocomposite materials during the charge-discharge processes were studied for the first time by in situ XRD, and the in-situ XRD data obtained indicates that there was irreversible phase transformation of MoSe_2/C nanocomposites during the initial charge-discharge processes.

2 Experiments

2 mmol of $\text{Na}_2\text{MoO}_4 \cdot 2\text{H}_2\text{O}$ and 9.88 mmol of glucose were dissolved in 20 mL of deionized water and ultrasonically dispersed for 10 min. In a separate flask, 4 mmol of Se powder was added into the 10 mL of $\text{N}_2\text{H}_4 \cdot 2\text{H}_2\text{O}$ (85%) under constant stirring

about 0.5 h to form a red solution. After that, the solution ($\text{Se-N}_2\text{H}_4\cdot 2\text{H}_2\text{O}$) was then slowly added dropwise to the $\text{Na}_2\text{MoO}_4\cdot 2\text{H}_2\text{O}$ -glucose solution and stirred for 30 min. Then, the mixture was transferred to a Teflon-lined autoclave and hydrothermal treatment at 200 °C for 20 h. The precipitated product was washed thoroughly with deionized water and ethanol several times, and then dried in a vacuum for 10 h. Ultimately, the obtained samples (MoSe_2/C composite) was heat treatment at 600 °C in a high purity argon atmosphere for 2 h.

The electrode was prepared by mixing active material MoSe_2/C nanocomposites with 20 wt % acetylene black as conductive additive and 10 wt.% poly (vinylidene fluoride) in N-methyl-2-pyrrolidone solution as binder to make slurry. Then the slurry was coated onto Mo foil and dried at 90 °C overnight in the vacuum oven. CR2025 coin-type cells were assembled test. MoSe_2/C was tested as working electrode, metallic Mg foil and Celgard 2500 were used as the counter electrode and the separator, respectively. All-phenyl-complex (APC) electrolytes, containing 0.2 M aluminum chloride (AlCl_3), 0.4 M phenylmagnesium chloride (PhMgCl), and anhydrous tetrahydrofuran (THF) solvent, with 0.5 M LiCl concentrations were used. X-ray diffraction (XRD) patterns were recorded by a Bruker D8 Discover diffractometer detector with Cu $\text{K}\alpha$ radiation (40 kV and 40 mA). Morphologies and structures of the as-synthesized products were characterized by field emission scanning electron microscopy (HITACHI S-4800) and transmission electron microscopy (FEI Tecnai-F30 FEG). The coin-type cells were tested with a battery test system (LAND CT2011A). The cyclic voltammetry (CV) measurement of the

electrodes was carried out on a CHI660D electrochemical workstation.

3 Results and discussion

The crystalline structure and phase composition of the MoSe₂/C nanocomposite were analyzed by XRD. Figure 1 (a) shows the XRD patterns of MoSe₂/C nanocomposite, compared with the corresponding PDF card (JCPDS29-0914), the diffraction peaks can be well indexed to the 2H-MoSe₂ phase. The diffraction peaks at $2\theta = 25.2^\circ, 31.7^\circ, 38.3^\circ, 56.1^\circ, 65.8^\circ, 70.9^\circ$ correspond to the MoSe₂ (004), MoSe₂ (100), MoSe₂ (103), MoSe₂ (110), MoSe₂ (200) and MoSe₂ (203) planes, respectively. The SEM images of the as-synthesized MoSe₂/C nanocomposite are shown in Fig 1(b). The MoSe₂/C nanocomposite consists of several nanometer-thick curved nanosheets, which are bundled together to form a flower-like composite. The detailed microstructure of the MoSe₂/C nanocomposite was obtained by high-resolution TEM (HRTEM) as shown in Figs. 1c and d. As can be seen from the Fig. 1(d), this material exhibits 2D layered structure, which is wrapped in a thin amorphous carbon layer.

The galvanostatic discharge–charge curves of the Mg| Mg²⁺, Li⁺| MoSe₂/C cells are shown in Figure 2(a). During the first discharge process, the MoSe₂/C nanocomposite exhibited a well-defined plateau at approximately 0.90V. The discharge capacity was measured to be 204 mAh·g⁻¹, in the next process a capacity of 160 mAh·g⁻¹ was measured, showing an irreversible capacity loss, which is likely caused by irreversible capture of partial lithium ion and decomposition of electrolyte. Within the discharge process, the first cycle of the discharge plateau was different

from the next process, indicating that an irreversible structural transformation of MoSe₂/C nanocomposite might occur in the first cycle. Fig. 2(b) shows the discharge–charge curves of Mg| Mg²⁺| MoSe₂/C cell operated at the 100 mA·g⁻¹ current density. The sloped voltage curves in the cell were observed, which are similar to the behavior of electrochemical double-layer capacitors. The discharge capacity was only 13.9mAh·g⁻¹ in the first cycle, and decayed to 5.9 mAh·g⁻¹ and 4.8 mAh·g⁻¹ in the subsequent two cycles, respectively. The cyclic voltammetry (CV) curves of the Mg| Mg²⁺| MoSe₂ /C cell and Mg| Mg²⁺, Li⁺| MoSe₂ /C cells obtained from 0.05 V to 1.75 V potential region are shown in Fig. 2(c) and Fig. 2(d), respectively. The Mg| Mg²⁺| MoSe₂ /C cell showed a very weak current without any apparent reduction/oxidation peaks, only at a very low potential, a weak current appeared, indicating that its capacities were very small and resulted primarily from Mg²⁺ adsorption.[10] However, this situation changed with the addition of 0.5 M LiCl into the APC electrolyte. When the Mg²⁺/Li⁺ hybrid electrolyte was employed in the CV study, in the first cathodic process, two apparent reduction peaks appeared at approximately 0.87 V and 0.15 V. The first cathodic peak at 0.87 V corresponds to the intercalation of lithium ions into the MoSe₂ interlayers to form Li₂MoSe₂, along with the phase transformation from 2H to 1T structure of Li_xMoSe₂. [20, 21] The second peak at 0.15 V can be assigned to the reduction of Li_xMoSe₂ to form metal Mo and Li₂Se. In the first anodic process, a strong oxidation current peak appears at approximately 1.27 V, which is ascribed to the oxidation reaction of Li₂Se to Se.[22] In the subsequent cycle, the reduction current peak at 1.02V and oxidation current peak at 1.27 V are ascribed to the Li₂Se

associated with Mo, and the shape of the CV curves have been almost overlapped, illustrating highly reversible and stable discharge/charge cycling.[20, 22]

The cycling stability of the hybrid battery is shown in Fig 2(e). Here, the cycling stability of hybrid batteries was studied with a 100-cycle charge–discharge test at $200\text{mA}\cdot\text{g}^{-1}$. As shown in Fig 2(e), after 100 cycles, the capacity retention rate was 80% ($110\text{mAh}\cdot\text{g}^{-1}$ - $89\text{mAh}\cdot\text{g}^{-1}$ from the 3rd to 100th cycles) and the coulombic efficiencies of battery was $>99\%$. In order to evaluate the rate performance of $\text{Mg}|\text{Mg}^{2+}, \text{Li}^+|\text{MoSe}_2/\text{C}$ cells, the capacities at various current densities ($50\text{ mA}\cdot\text{g}^{-1}$, $100\text{ mA}\cdot\text{g}^{-1}$, $200\text{ mA}\cdot\text{g}^{-1}$, $400\text{ mA}\cdot\text{g}^{-1}$) were measured, and the results are shown in Fig 2(f). At a relatively low current density of $50\text{ mA}\cdot\text{g}^{-1}$, the cell exhibited a discharge capacity of approximately $175\text{ mAh}\cdot\text{g}^{-1}$. The discharge capacity at $100\text{ mA}\cdot\text{g}^{-1}$ and $200\text{mA}\cdot\text{g}^{-1}$ were about 150 and 115 $\text{mAh}\cdot\text{g}^{-1}$, respectively. It is worth noting that the capacity was still close to $100\text{ mAh}\cdot\text{g}^{-1}$ when the cell operated at a higher current density of $400\text{mA}\cdot\text{g}^{-1}$. The overall rate capability of MoSe_2/C nanocomposites cathode is significantly higher than the reported Mg-insertion materials prepared by much more complex methods. [23, 24] To get an insight into the stability of the cathode, in-situ XRD experiment was carried out to investigate the structure changes of the MoSe_2/C nanocomposites during the charge-discharge process. The main in-situ XRD results are shown in Figure 3, where the discharge and charge processes of the electrochemical cycling are shown with different colors. A variety of diffraction peaks were clearly seen, the most significant changes are the diffraction peaks at 31.7° and 56.1° which correspond to the $\text{MoSe}_2(100)$ and $\text{MoSe}_2(110)$ facets of the MoSe_2/C

nanocomposites materials. Both the (100) and (110) peaks changed as a function of potential, gradually disappeared in the discharge process but did not reappear in the charging process. The other peaks did not change during the discharging-charging cycle and were regarded as an internal standard features. Upon discharge, the aforementioned two peaks of the MoSe₂/C nanocomposites become weaker and weaker; at the end of discharge process, they disappeared completely. During the charging process, these peaks did not re-appear, indicating an irreversible reaction occurred in the first cycle. These observations are consistent with the results of the CV study discussed before. It is worth mentioning that because of the transformation reaction occurred between nanoparticles, it was difficult to obtain the obvious XRD signals of Li₂Se and Mo nanoparticles.

4 Conclusions

In summary, a highly reversible Mg²⁺/Li⁺ hybrid battery has been rationally designed and demonstrated, employing the sheet-like MoSe₂/C nanocomposite as the cathode and a metallic Mg foil as the anode, together with a dual-salt electrolyte containing both Li⁺ and Mg²⁺ ions. The hybrid battery showed a high capacity, good cycling performance and superior rate capability. The electrochemical reactions taking place in the hybrid battery were related to Mg stripping/plating at the anode side and Li⁺ intercalation at the cathode side, with a small contribution from Mg²⁺ adsorption. Thanks to the inherent safety and excellent electrochemical performance, the Mg²⁺/Li⁺ hybrid battery is evidenced to be very promising for practical applications,

especially for large-scale static energy storage. The design of such type of batteries also provides a new idea for the development of other magnesium-based batteries.

Conflict of interest

The authors declare no conflict of interests.

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Figure captions

Figure 1. XRD pattern of MoSe₂/C composite (a); SEM image of MoSe₂/C (b); TEM images of MoSe₂/C nanocomposites (c) and (d).

Figure 2. Discharge-charge profiles of Mg | Mg²⁺, Li⁺ | MoSe₂/C cells (a); discharge-charge profiles of Mg | Mg²⁺ | MoSe₂/C cells (b). CV curves of Mg | Mg²⁺, Li⁺ | MoSe₂/C cells and Mg | Mg²⁺ | MoSe₂/C cells (c) and (d), respectively; scan rate: 0.1 mV s⁻¹. (e) Cycling performances and (f) rate performances of Mg | Mg²⁺, Li⁺ | MoSe₂/C cells.

Figure 3. In-situ XRD patterns of the MoSe₂/C nanocomposite in the first discharging-charging cycle.

Figures

Fig. 1

Fig. 2

Fig. 3

Highlights

The sheet-like MoSe₂/C nanocomposites were firstly employed as cathode of Mg²⁺/Li⁺ hybrid battery.

The sheet-like MoSe₂/C nanocomposites exhibited an excellent electrochemical performance in the hybrid battery

In situ XRD reveals the structure transformation of materials.