Controlling Electrochemical Lithiation/Delithiation Reaction Paths for Long-cycle Life Nanochain-structured FeS₂ Electrodes

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Abstract

Pyrite (FeS_2) stands out from other metal sulfides due to its abundance, low toxicity and high specific energy density. However, the low utilization and short cycle life of FeS_2 caused by irreversible side reactions and dramatic structural changes during charge/discharge process seriously hinders its practical application. In this work, we designed FeS_2 nanochains to address these issues because this unique structure not only shortens the ion/electron diffusion distance, increases the electrode/electrolyte contact area, but also accommodates the strain associated with Li ion intercalation and deintercalation reactions. Compared with nanoparticles, the FeS_2 nanochains synthesized by magnetic-field-assisted aerosol pyrolysis could cycle up to 800 cycles at 0.5 C, remained a discharge capacity of $342.4 \text{ mA h g}^{-1}$ in the voltage range of 1.0-2.6 V and achieved a remarkable capacity fading rate of 0.02% per cycle, which is among the best values demonstrated so far for Li/ FeS_2 cells. *In-situ* Raman experiments revealed that the irreversible side reactions could be suppressed through controlling the voltage window, which can be benefit for further improving the performance of Li/ FeS_2 cells.

Keywords: FeS_2 nanochains long-cycle life high capacity *in-situ* Raman lithium cells

1 Introduction

With the development of mobile devices and electric vehicles, high-energy-density and low cost lithium cells are urgently needed in energy storage device [1]. Pyrite (FeS_2) has attracted much attention due to its high specific energy density (1273 Wh kg^{-1}) and theoretical capacity (894 mA h g^{-1}) [2], abundance in natural mineral [3, 4], and low toxicity [5]. Additionally, the capability of overcharge protection enables FeS_2 to apply in large type cells [6]. However, the unsolved problems in FeS_2 cells, such as active material loss, severe volumetric change, agglomeration of Fe^0 nanoparticles, polysulfide dissolution and the subsequent redox shuttle [7, 8], seriously hinders its practical application [9, 10].

To address the above challenging issues in FeS_2 cathodes, researchers have devoted much effort on the device designs and materials synthesis. For example, Yersak *et al.* have developed a solid glass-ceramic electrolyte for suppressing the dissolution of polysulfides and agglomeration of Fe^0 particles into larger particles [8]. Son *et al.* have embedded cubic FeS_2 into a polyacrylonitrile matrix to confine the electroactive species (Fe^0 and polysulfides) of FeS_2 for improving the reversibility and efficiency [9]. Strauss *et al.* have modified cathode structure for improving the cycling performance by coating a very thin ion-conductive protective film on cathode [11]. However, achieving a high rate and a long cycle life performance for FeS_2 cathodes is still difficult. It is widely accepted that the morphology has a great influence on the electrochemical performance of electrode material [12, 13]. With the development of nanotechnology, FeS_2 with controllable microstructures, such as wires [14], rods [15], nanoplates [16], cubes [17], and nanocrystals [18], have been achieved. Unfortunately, the best reported performance for Li/FeS_2 cells was about 291 mA h g^{-1} after 500 cycles [19], which is far away

from the commercialization requirement. It is still a challenge to further improve the performance of Li/FeS₂ cells.

Among the controllable morphologies, one dimensional (1D) nanostructures are particularly attractive because their unique geometry not only accelerates the transport rate of electrons, but also creates more active sites for electrochemical reaction, and thus enhances the rate capability [14, 15]. In addition, the intertwined 1D structure can form a mechanically robust three dimensional (3D) network with porous structure, which can well endure the structural strain upon charging and discharging, and thus prolong the cycle life of electrodes [20]. Herein, we synthesized FeS₂ nanochains by magnetic-field-assisted aerosol pyrolysis. The as-prepared intertwined FeS₂ nanochain network with numerous nanometer-sized pores exhibits good mechanical properties, which is benefit not only for the rate capability but also for the cycle life. In addition, Raman spectrum as a sophisticated method in understanding the electrochemical reactions has been used in this paper. The results confirm that the control of voltage window is an effective method to reduce the shuttling effect of polysulfides upon cycling. When the voltage window was switched between 2.6 V and 1.0 V, the FeS₂ nanochain cathode could cycle up to 800 cycles and remained the capacity as high as 342.4 mA h g⁻¹, which is among the longest cycle life demonstrated so far for such a high capacity in FeS₂-based lithium cells.

2 Experimental

2.1 Materials

Iron pentacarbonyl (Fe(CO)₅, 99%, Sigma-Aldrich), Acetone (CH₃COCH₃, 99%, Sinopharm Chemical Reagent Company Limited), SP-type carbon black (TIMCAL, Graphite & Carbon Ltd), PVDF (polyvinylidene difluoride, Alfa Aesar), NMP (N-methyl-2-pyrrolidone,

Sigma-Aldrich), MWCNTs (Multi-walled carbon nanotubes, Sigma-Aldrich) were used as received. The electrolyte was 1 M bis (trifluoromethane)-sulfonamide lithium salt (LiTFSI, 99.95% trace metals basis, Sigma-Aldrich) dissolved in a mixture of 1,3-dioxolane (DOL) and dimethoxymethane (DME) (1:1 by volume).

2.2 FeS₂ nanochains preparation

As shown in Fig. 1, Fe(CO)₅ precursor solution (0.2 M) was firstly prepared by mixing 1 mL of Fe(CO)₅ with 35 mL of acetone, and then the precursor solution was pumped into a home-made ultrasonic atomizer (1.7 MHz). The setup of ultrasonic atomization and pyrolysis process can be found in Fig. S1. It should be noted that this ultrasonic atomization technology can generate very small droplets in range of an extremely narrow diameter compared with conventional pressure nozzles. The atomized aerosol (droplets with a diameter of about 5 μm) was carried by compressed air to a customized glassware, which was heated to 400 °C. As well known, Fe(CO)₅ can rapidly decompose into Fe and CO at 180 °C (refer to equation 1) [21]. In this work, the as-prepared Fe nanoparticles could easily react with air and form iron oxide at 400 °C (refer to equation 2). Finally, the Fe₂O₃ nanochain network reacted with H₂S gas at 300 °C for 30 min to obtain FeS₂ (refer to equation 3).

2.3 Electrochemical measurements

The cathode slurry was prepared by mixing the as-prepared FeS₂ materials, SP-type carbon black and PVDF binder with 80:15:5 (by weight) in NMP solution. Then the mixed slurry was spread evenly on an aluminum foil and roll-pressed to form cathode film after heating at 100 °C for 24 hours under vacuum. The thickness of cathode is around 25 μm. 2025 type coin cells were assembled in an argon filled glove box using lithium foil as the anode.

2.4 Material characterization

The microstructure of the as-fabricated FeS₂ nanochains were characterized by scanning electron microscopy (SEM, JEOL6700F) and transmission electron microscope (TEM, JEOL 2010). Raman measurement was conducted on a Lab RAM HR evolution Raman spectrometer. X-ray diffraction (XRD) was used to study the crystal structure of the samples with a Bruker D8 diffractometer. Cyclic voltammetry (CV, 1.0-3.0 V, 0.5 mV s⁻¹) measurements were performed on a VMP3 electrochemical working station. Galvanostatic charging/discharging was conducted on land cell program-control test system at rates of 0.05-2 C. X-ray photoelectron spectroscopy (XPS) characterization was performed to investigate information on the surface of FeS₂ nanochains on an Axis Ultra DLD XPS. The BET (Brunauere Emmette Teller) specific surface area of the sample was determined by an ASAP 2020 using the standard N₂ adsorption and desorption isotherm measurements at 77 K.

2.5 FeS₂ prototype cell for in situ Raman experiments

A 2025 type coin cells with a window (Fig. S2) was used to conduct *in-situ* Raman experiments. It should be noted that MWCNTs (Multi-walled carbon nanotubes) was used as current collector instead of Al foil for in situ observation. The cathode was prepared by mixing

MWCNTs and the FeS₂ nanochains with a weight ratio of 1:1, and then press the mixture into a disc. The following assembly procedure of coin cells is similar as the above.

2.6 FeS₂ prototype cell for ex situ XRD spectrum characterization

The electrochemical cells were disassembled after discharging to 2.1 V and 1.4 V. The structures of the electrode products after lithiation were investigated by powder X-ray diffraction.

3 Results and Discussion

As schematically illustrated in Fig. 1, FeS₂ nanochains were synthesized by three steps. Firstly, iron pentacarbonyl precursor from Fe(CO)₅ and acetone was decomposed into Fe nanoparticles through an ultrasonic process; then the as-prepared Fe nanoparticles reacted with air to form iron oxide at 400 °C, which was confirmed by XRD results (Fig. 2a). As shown in Fig. 2b, it can be found that the as-formed Fe₂O₃ shows porous network structure. TEM images (inset of Fig. 2b), reveal that this porous network is formed by these 1D chains intertwining together and the 1D chains are assembled by nanoparticles with size of 20-30 nm. Considering Fe nanoparticles could be assembled under magnetic-field, therefore, these Fe intermediates formed in equation 1 easily assemble into 1D nanochains with the assistance of magnetic field, and then these Fe nanochains are oxidized by air into 1D Fe₂O₃ nanochains. Finally, the as-formed Fe₂O₃ nanochains react with H₂S into FeS₂ nanochains. SEM and TEM results confirm that the as-obtained FeS₂ show 1D nanochain structure with a diameter range of 30-50 nm (Fig. 2c). Furthermore, the results in Fig. 2d well reveal that these nanochains are assembled with numerous nanoparticles with the size of 20-30 nm, which is similar to the structure of Fe₂O₃ (inset of Fig. 2b). A typical HRTEM image (inset of Fig. 2d) discloses the fine structure of the FeS₂ nanochains. The lattice fringes

with a spacing of 0.3 nm is consistent with the spacing of the (111) plane of FeS₂. Such a nanochain morphology is well preserved even after intense ultrasonic treatment in the TEM sample preparation or casting and roll-pressing step in the cathode preparation (shown in Fig. S3). These results indicate that these nanochains exhibit a good mechanism property, which may endure severe volumetric change of FeS₂ during cycling (about 162.2%, as we computed in Table S1). BET measurement results reveal that the surface area of the as-intertwined FeS₂ nanochains is around 37.3 m² g⁻¹ (Fig. S4), which is much larger than that of natural cubic FeS₂ (0.9431 m² g⁻¹) [22]. The total pore volume for this porous network is about 0.1494 mL g⁻¹ and the pore size is in the range of 5-40 nm.

The XRD pattern of the as-obtained nanochains cathode was shown in Fig. 3a, and it can be found that all the diffraction peaks match well with the standard pyrite FeS₂ peaks (JCPDS Card No. 42-1340). Besides, the crystal structure of nanoparticles was also studied by XRD. As shown in Fig. S5, it can be observed that all diffraction peaks are same for nanochains and nanoparticles, indicating that nanoparticles are also pyrite structure. The structure feature of the as-prepared nanochain was further investigated by Raman analysis. As shown in Fig. 3b, there are three peaks in the Raman spectrum, locating at 339, 376, and 429 cm⁻¹, corresponding to three different characteristic active modes of pyrite FeS₂, E_g, A_g, and T_g, respectively. The E_g mode (340-353 cm⁻¹) corresponds to the vibration of S atoms perpendicular to the dimer axes; whereas the A_g mode (377-383 cm⁻¹) corresponds to the S-S in phase stretching; and the T_g mode (427-466 cm⁻¹) assigns to mix coupled vibration and stretching mode [23-25]. No obvious vibrational peaks for Fe_xS or Fe_xO are detected in the range of 200-300 cm⁻¹ [26-29], indicating the high purity of the as-obtained FeS₂ nanochains. Moreover, XPS was also used to study the surface information of the as-prepared FeS₂ nanochains. As shown in Fig. 3c, the two peaks

locating at 162.3 eV and 163.5 eV can be attributed to S 2p_{3/2} and S 2p_{1/2}, respectively, which are also consistent with the previous reported sulfur binding energy of pyrite FeS₂ [30-33]. In Fe 2p spectrum (Fig. 3d), there are two peaks in Fe 2p core level XPS spectrum, locating at 707.3 and 720.1 eV, corresponding to Fe 2p_{3/2} and Fe 2p_{1/2} binding energies in FeS₂, respectively [34], indicating the formation of Fe²⁺. The peaks locating at 162.3 eV in Fig. 3c and 707.3 eV in Fig. 3d are attributed to pure pyrite [30, 31, 35]. These results again confirm that the as-prepared FeS₂ nanochains are of high purity.

To evaluate the superior electrochemical performance of the unique FeS₂ nanochains, the Nyquist plots of both FeS₂ nanochain and nanoparticle cathodes were obtained by applying a sine wave with amplitude of 5.0 mV over the frequency range from 200 kHz to 0.1 Hz. Compared with nanoparticle-based FeS₂ cathode shown in Fig. S6a and Fig. S6b, a smaller semicircle of the nanochain cathode suggests a lower electrochemical resistance. The fitted impedance parameters were listed in Table S2. The SEI film resistance R_f and charge-transfer resistance R_{ct} of the FeS₂ nanochain cathode are 67.1 Ω and 105.7 Ω , respectively, which are significantly lower than that of the nanoparticle cathode (97.8 Ω and 136.1 Ω). Furthermore, the cells performance based on nanochain cathode and nanoparticle cathode were compared. As shown in Fig. S7, the cell with the nanochain cathode delivers an initial discharge capacity of 598 mA h g⁻¹, much higher than 498 mA h g⁻¹ of the cell with nanoparticle cathode. After 100 cycles, the cell could retain the discharge capacity of 355 mA h g⁻¹ at 0.2 C, while the cell with the nanoparticle cathode just delivers around 179 mA h g⁻¹. The results well demonstrate that morphology has a significant effect on the electrochemical performance. This unique 3D nanochain structure not only benefits for improving the electron transfer kinetics, but also increasing the electrochemical active sites

with high porosity and large pore size through 1D chains, which thus improves the cell's discharge capacity and cycle stability.

Based on the above results, the electrochemical behavior of nanochain-based cell was carefully investigated. The CV (cyclic voltammetry) of nanochain cathode was conducted in half-cell configuration at room temperature, with a scan rate of 0.05 mV s^{-1} in the potential range of 1.0-3.0 V (vs Li/Li⁺). As shown in Fig. 4a, there are two main reduction peaks located around 2.1 and 1.4 V and two oxidation peaks located at 1.8, 2.4 V in the initial discharge/charge cycle. The involved reduction and oxidation reactions are summarized in Table S3 based on the previous reports [7-9]. During the initial discharging process, FeS₂ is reduced to Li₂FeS₂ at 2.1 V (equation S1) and further reduced to Li₂S and Fe⁰ at 1.4 V (equation S2); and then these products can be oxidized to Li₂FeS₂ (equation S3), and further to Li_{2-x}FeS₂ (equation S4), FeS₂, FeS_{8/7} and S (equation S5) [4, 36]. During 2nd discharging process, it can be found that a new peak located at 2.3 V appears, which can be attributed to the reduction of S into high-order Li₂S_x (equation S6). At 2.1 V, these high-order Li₂S_x can be further reduced to Li₂S (equation S7); while FeS₂ is reduced to Li₂FeS₂ (equation S1). The peak at 1.4 V, can be assigned to the formation of Li₂S and Fe⁰ (equation S2 and S8). Furthermore, the peak area shrunked obviously in the fifth cycle demonstrates the capacity fading of electrode upon cycling, which may be caused by the side-reactions from sulfur into high-order polysulfides.

The control of voltage window has long been proved to be an effective strategy for stabilizing cathode reactions in lithium ion cells, which has also been successfully applied in FeS₂ cathodes [37]. In this work, the effect of voltage window range was also investigated. By controlling the voltage in range of 1.0-2.6 V, only two reduction peaks appear at approximately 2.1 and 1.4 V in the initial discharging process in Fig. 4b, same as the results shown in Fig. 4a, which can also be

assigned to the transitions from FeS_2 to Li_2FeS_2 (equation S1) and then to the final discharge products, Li_2S and Fe^0 (equation S2), respectively. In the oxidation sweep, two peaks located at 1.8 V and 2.4 V can be observed. The peak at 1.8 V can be ascribed to the formation of Li_2FeS_2 from Li_2S and Fe^0 (equation S3). These Li_2FeS_2 can be further oxidized to FeS_2 , FeS and Li_2S_x at 2.4 V (equation S9 and S11). During the 2nd discharging process, there are only two peaks appearing at 2.1 V and 1.4 V, which are different from the results shown in Fig. 4a (three peaks: 2.3 V, 2.1 V, 1.4 V). The absent peak at 2.3 V well suggests almost no polysulfides forming under the window potential of 1.0-2.6 V. The peaks at 2.1 V can be ascribed to the reduction of FeS_2 and Li_2S_x described in equation S1 and S7. Then these reduction products can be further reduced to Li_2S and Fe^0 at 1.4 V, as described in equation S2 and S10.

The cycle performance comparisons between the cells with the regular voltage window (charging to 3.0 V) and the modified recharge setting (charging to 2.6 V) are shown in Fig. 4c. Stable electrochemical reactions can be accomplished as long as the voltage window is controlled to 2.6 V, it can deliver an initial discharge capacity of 598 mA h g^{-1} , remain about 432 mA h g^{-1} (0.58 mA h cm^{-2}) after 450 cycles at 0.2 C (101.6 mA cm^{-2}) and preserve stable cycling performance with 0.06% capacity decay per cycle. However, we found that when the cells were cycled under a regular voltage window (charging to 3.0 V), it delivers an initial capacity of 597 mA h g^{-1} , almost the same value obtained at 1.0-2.6 V (592 mA h g^{-1}), but the capacity rapidly reduces to 355 mA h g^{-1} after 100 cycles, which is much lower than the one operated at 1.0-2.6 V (541.4 mA h g^{-1} after 100 cycles). Correspondingly, the calculated capacity decay per cycle for the cell performed at 1.0-3.0 V is 2.42 mA h g^{-1} , much higher than the value of 0.36 mA h g^{-1} operated under the voltage range of 1.0-2.6 V. Moreover, the average Coulomb efficiency of the cell operated at 1.0-2.6 V is much better. To further study the

efficiency of the voltage window setting, the voltage profile operated at 1.0-2.6 V and 1.0-3.0 V were conducted. As shown in Fig. S8a, the initial discharge capacity of the FeS₂ nanochain electrode is 592 mA h g⁻¹ and it keeps at 530 mA h g⁻¹ for the 3rd cycle with a Coulombic efficiency of 99.8%. In contrast, although the same electrode operated at 1.0-3.0 V exhibits the almost same initial discharge capacity of 596 mA h g⁻¹, it sharply reduces to 468 mA h g⁻¹ in the 3rd cycle, as shown in Fig. S8b. These results again reveal that the potential cut-off to 2.6 V is an efficient way to suppress some side-reactions, can efficiently enhance the capacity of FeS₂ nanochain electrode, and thus significantly prolong the cycle life. These results again confirm that the voltage window setting can effectively suppress some side-reactions.

Based on the above results, the voltage window range of 1.0-2.6 V was selected to evaluate the following electrochemical behavior of FeS₂ nanochains. As shown in Fig. 4d, at a higher rate of 0.5 C (254 mA cm⁻²), the cell can deliver the highest specific capacity of 466 mA h g⁻¹ (0.54 mA h cm⁻²) at the 60th cycle, and the discharge capacity stabilizes at 421 mA h g⁻¹ (0.53 mA h cm⁻²) after 100 more cycles. The discharge capacity decreases to 380 mA h g⁻¹ (0.48 mA h cm⁻²) after 400 cycles and it can remain at 342.4 mA h g⁻¹ (0.44 mA h cm⁻²) after 800 cycles with a decay rate of 0.02% per cycle, which is better than the best performance reported so far for Li/FeS₂ cells (0.1% per cycle after 500 cycles) [11]. The average Coulomb efficiency of the cell is more than 95%. To further demonstrate the superior performance of our FeS₂ nanochain electrode, the specific capacity was also evaluated by the “total” weight (including active materials, super P additive, and binder). The cells can deliver discharge capacities of 352 mA h g_{total}⁻¹ after 450 cycles at 0.2 C and 255 mA h g_{total}⁻¹ after 800 cycles at 0.5 C based on the total weight, which exhibit a good performance in FeS₂-based cells. The advantage of these FeS₂ nanochains was further evaluated by the rate performance. As shown in Fig. S9, the discharge

capacity of electrode at discharge rates of 0.2 C, 0.5 C, 1 C and 2 C are 598, 503, 435, and 359 mA h g⁻¹, respectively. When the current rate changed back to 0.2 C, the discharge capacity recovered to 598 mA h g⁻¹.

To track the detailed electrochemical reaction processes upon cycling, an *in-situ* Raman experiment was conducted using a typical coin cell with an observation window as shown in Fig. S2. The fabrication details for this *in-situ* observation cell can be found in the experimental section. As shown in Fig. 5a, the Raman spectrum recorded at several special voltages upon cycling: (A) at open circuit 2.3 V, (B) initial discharged to 2.1 V, (C) initial discharged to 1.4 V, (D) initial charged to 1.8 V, (E) initial charged to 2.4 V, (F) 2nd discharged to 2.3 V, (G) 2nd discharged to 2.1 V, (H) 2nd discharged to 1.4 V. The corresponding Raman results were shown in Fig. 5(b-d). Table S4 and Table S5 summarized the Raman peaks at different voltages and their corresponding compounds. It can be found that the main compound at stage A was FeS₂ [38]. When discharged to 2.1 V, Li₂FeS₂ appeared at stage B, as evidenced by ex-situ XRD patterns (Fig. S10a). The peaks appeared at 14.1°, 26.4°, 28.8°, 42.8°, 46.8°, which well matched with the standard peaks of Li₂FeS₂ (JCPDS 36-1088). Other peaks located at 28.8°, 37.8°, 56.3°, 59.0° can be ascribed to FeS₂ (JCPDS 42-1340). When discharged to 1.4 V, the peaks of Li₂FeS₂ and Li₂S can be observed (Fig. S10b). These results are in good agreement with the Raman measurements and CV results described in the equations S1 and S2 of Table S3 [39, 40]. During charging process (Fig. 5c), it can be found that these Li₂S are oxidized to Li₂FeS₂ at 1.8 V (stage D), which is also proved by CV in Fig. 4b. At stage E, the appearance of FeS₂, FeS and Li₂S_x peaks, and the weaker signal from Li₂FeS₂ peaks well indicate the transformation from Li₂FeS₂ into FeS₂ and FeS. During the 2nd discharging process, it can be concluded that the main compounds are FeS₂ and FeS at 2.3 V, which again confirms the absence of the side-reaction

from sulfur into polysulfides under voltage window of 1.0-2.6 V. Further discharged to 2.1 V (stage G) and 1.4 V (stage H), the main reduced products are same as initial discharged products. Based on the *in-situ* Raman results, it can be concluded that the control of voltage window setting strategy is efficient to reduce some side-reactions, especially the one from sulfur into polysulfides. As well known, the polysulfides shuttle effect could cause serious capacity decay upon cycling, the absent of this side-reaction under 1.0-2.6 V may well explain why our cell can achieve a higher capacity and longer cycle life.

4 Conclusions

We have successfully synthesized FeS₂ nanochains through magnetic-field-assisted aerosol pyrolysis. This 3D FeS₂ nanochain network with porous structure exhibits a superior electrochemical performance. It can deliver a specific discharge capacity of 432.3 mA h g⁻¹ after 500 cycles at 0.2 C and 342.4 mA h g⁻¹ after 800 cycles at 0.5 C, which is among the best performance in the reported results, such as PAN-FeS₂ (470 mA h g⁻¹ 50 cycles) [9], FeS₂@C (495 mA h g⁻¹ 50 cycles) [41] and Li/FeS₂ film cell (291 mA h g⁻¹ 500 cycles) [19]. Our *in-situ* Raman experiments demonstrated that this remarkable performance is benefit from the successful suppression the side-reaction from sulfur into long-chain lithium polysulfides through controlling the voltage window. The simple, low cost synthesis approach enables the FeS₂ nanochain electrode to be more promising and competitive for the future applications in lithium cells.

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Fig. 1 Schematic diagram of the synthesis process of the FeS_2 nanochain network with the assistance of magnetic field.

Fig. 2 (a) XRD patterns, (b) SEM and TEM (inset) images of the as-prepared $\gamma\text{-Fe}_2\text{O}_3$ nanochain network; (c) Low-magnification and high-magnification (inset) SEM images of the as-prepared FeS_2 nanochain network; (d) TEM and HRTEM (inset) images of the as-prepared FeS_2 nanochain assembly.

Fig. 3 (a) XRD patterns, (b) Raman spectrum, (c) S core level and (d) high resolution Fe XPS spectra of FeS₂ nanochain.

Fig. 4 Cyclic voltammograms of the FeS₂ nanochain samples for the first, third and fifth cycles at 0.5 mV s⁻¹ in the voltage range of (a) 1.0-3.0 V vs. Li/Li⁺, (b) 1.0-2.6 V vs. Li/Li⁺. Cycling performance tests of FeS₂ nanochain under (c) 1.0-2.6 V and 1.0-3.0 V at 0.2 C. (d) 1.0-2.6 V at 0.5 C. Insets are their corresponding coulombic efficiencies.

Fig. 5 (a) Discharge/charge voltage profile of the FeS₂. Typical Raman spectrum of the electrode under specific voltages during (b) initial discharging process, (c) initial charging process, (d) 2nd discharging process.