Lithium Sulfide (Li_2S)/Graphene Oxide Nano-spheres with Conformal Carbon Coating as High-rate, Long-life Cathode for Li/S cells

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Introduction

Advantage of Li/S cell

- Large volumetric and gravimetric energy density
- Very low production cost of sulfur (~ $150/ton)
- High abundance of sulfur.
- Low environmental impact.

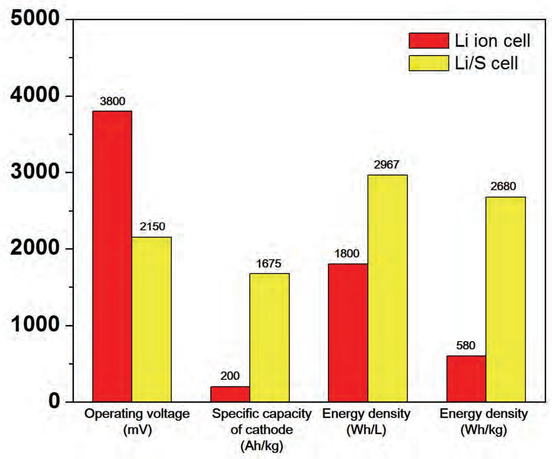


Figure 1. Comparison of theoretical electrochemical properties of Li/S and Li-ion cells.

Reaction mechanism of Li/S cell

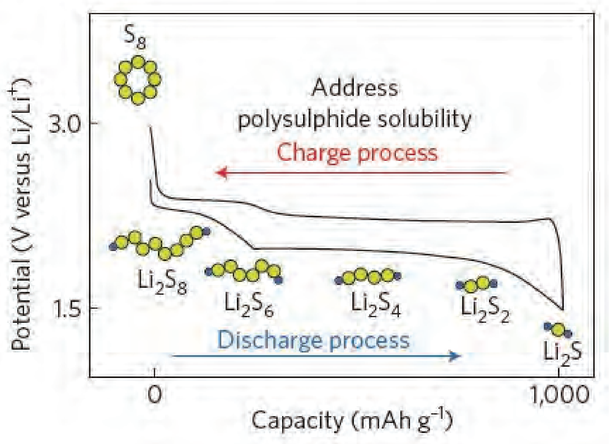


Figure 2. Schematic and voltage profiles of a Li–S cell. P. G. Bruce et al. *Nat. Mater.* **2012** , 11 , 19.

- Reaction chemistry between S and Li: $S_8 + 16Li^+ + 16e^- \leftrightarrow 8Li_2S$

Drawbacks of Li/S cell

- Insulating nature of S.
- Large volume change of S up to 76 % during lithiation/delithiation process.
- Dissolution of polysulfides into the organic liquid solvent and their shuttle effect.

Results and Discussion

Strategy of the Li_2S/GO@C nano-spheres

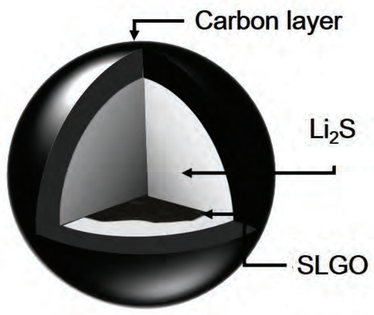


Figure 3. Schematic illustration of the Li_2S/GO@C nano-sphere

- Improving electrical conductivity.
- Suppressing polysulfide dissolution into organic liquid electrolyte.
- The carbon shell will not need to expand during cycling because Li_2S occupies the maximum volume relative to S.
- GO sheets embedded in the Li_2S@C act as sulfur immobilizer.

Rotating furnace

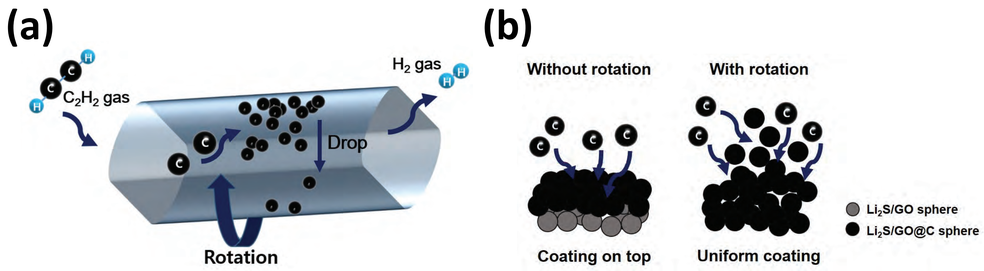
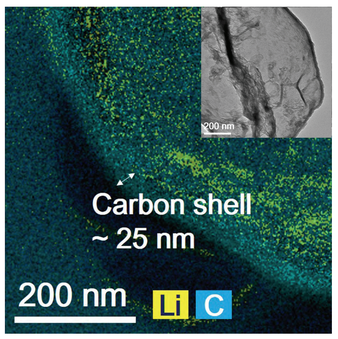


Figure 4. Schematic illustrations of (a) the rotating furnace, (b) carbon deposition process using the conventional CVD method and CVD using the rotating furnace.

- The continuous mixing in the rotating furnace through a "lifting and falling" process during the CVD coating provides a uniform carbon deposition on the Li_2S/GO nano-spheres.

Material characterization



- The core-shell structure of the Li_2S/GO@C nano-spheres and embedded GO in the Li_2S particles were successfully demonstrated by TEM with an EFTEM elemental mapping.

Figure 5. Elemental mapping of the Li_2S/GO@C nano-sphere by energy filtered TEM (inset: TEM image of hollow carbon nano-sphere including GO in its structure obtained by removal of Li_2S from the Li_2S/GO@C nano-sphere.)

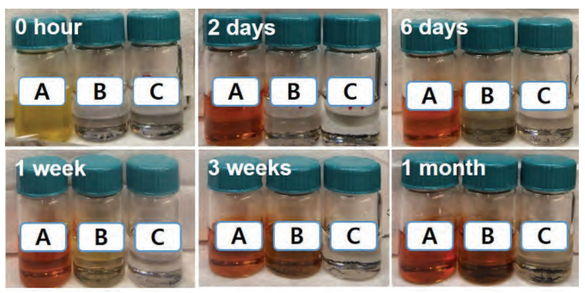


Figure 6. Polysulfide dissolution test results of Li_2S (sample A), Li_2S/GO@C-NR* (sample B) and Li_2S/GO@C (sample C).

- The conformal carbon shell of the Li_2S/GO@C successfully prevents the dissolution of Li_2S into the test solution.

*NR: Carbon coating using the conventional CVD method

Electrochemical Tests

Comparisons

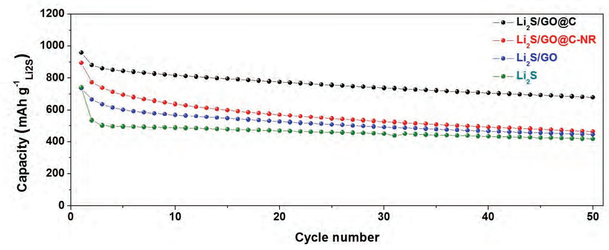


Figure 7. Comparison of cycling performances of the electrodes at 0.2 C

- The Li_2S/GO@C electrode showed much better cycleability compared to the other electrodes for 50 cycles.

Long-term cycling performance

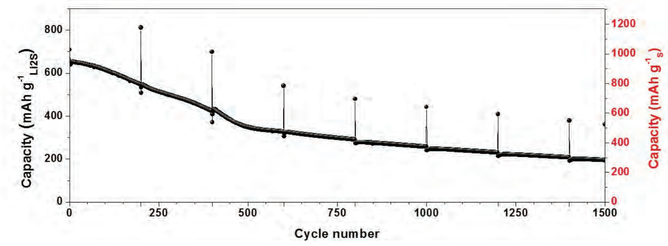


Figure 8. Long-term cycling performance of the electrode at 2.0 C discharge for 1500 cycles.

- During 1500 cycles, the Li_2S/GO@C electrode exhibited a very low capacity decay rate of 0.046 % per cycle.

Summary

- The Unique structure of the Li_2S/GO@C nano-spheres successfully improved the electrochemical performance of the Li/S cell.
- A conformal carbon coating was obtained using a lab-designed rotating furnace.
- The Li_2S/GO@C electrode exhibited a very high initial discharge capacity of 964 mAh/g of Li_2S at 0.2 C and a very low capacity decay rate of only 0.046 % per cycle at 2.0 C discharge for 1500 cycles.

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