Reactivation of Dissolved Polysulfides in Li-S Batteries

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Abstract
This work demonstrates the effect of atomic layer deposited (ALD) \text{Al}_2\text{O}_3 on the reactivation of dissolved polysulfides in Li-S batteries. A layer of \text{Al}_2\text{O}_3 is coated onto highly porous carbon cloth (ACC) by ALD, and then assembled in a Li-S battery between the sulfur cathode and the anode to function as a reactivation component. The ALD-\text{Al}_2\text{O}_3 improves the initial specific capacity and stabilizes the cycle life remarkably. Scanning electron microscopy verified that the ALD-\text{Al}_2\text{O}_3-coated carbon cloth sorbs (adsorbs/absorbs) dissolved sulfur species from the electrolyte. The combination of an ultrathin ALD oxide coating with highly porous carbons presents a new strategy to improve the performance of Li-S batteries.

Background
- As an abundant byproduct, sulfur would offer an affordable material in lithium batteries.
- Polysulfides are produced from the electrochemical reaction of lithium and sulfur.
- Polysulfides easily dissolve between the anode and cathode, leading to the degradation of sulfur in Li-S batteries.
- This is the major reason of the decay of charging and discharging (cycling) in Li-S batteries.
- To slow this loss of polysulfides down, we added an additional activated layer, a circle of carbon cloth covered with \text{Al}_2\text{O}_3 using a process called atomic layer deposition, to adsorb and re-activate the dissolved polysulfides, making them return to the cathode.

Reactivation Idea

The reactivation idea is illustrated above. Instead of blocking polysulfide dissolution, the porous conductive ACC is used as a model material. The ALD-\text{Al}_2\text{O}_3 coating on ACC is found to further improve the reactivation effect by enhanced polysulfide sorption.

Step 1: Infiltration of Sulfur in ACC
1) Active Carbon Cloth is cut into disks with diameters of ¾ inch.
2) Samples are vacuum sealed in a glass tube with Sulfur Powder.
3) The samples are heated to 300 °C for 6 hours and cooled for 24 hours.

Step 2: Atomic Layer Deposition of \text{Al}_2\text{O}_3
- A plain carbon cloth disk is taken and placed in an atomic layer deposition system.
- This process deposits a one atom thick layer of \text{Al}_2\text{O}_3 onto the surface of the ACC.
- The purpose of this extra layer is to absorb the polysulfides that leave the cathode during discharging.
- To ensure coverage but maintain the cathode's conductivity, we performed atomic layer deposition five times on the ACC disk.

Step 3: Battery Assembly

Battery Performance

Below are comparisons that verify that ALD-\text{Al}_2\text{O}_3 is a more efficient battery structure than other Li-S systems.

When only using a sulfur cathode as in image (a) to the left, the specific capacity of the battery is limited. The charge/discharge tests are presented by the red (first cycle) and black (second cycle) lines. The drawings next to the graphs illustrate the corresponding cathode layouts. Figure (b) depicts a cathode that has an ACC reactivation layer. Part (c) shows the optimized charge/discharge cathode design with ALD-\text{Al}_2\text{O}_3 on ACC.

The picture below depicts the SEM micrographs for aluminum and sulfur distributions on the cross-section of a representative \text{Al}_2\text{O}_3-ACC fiber. The image in the middle illustrates that the aluminum distribution does not change during cycling, indicating the stable coating of ALD-\text{Al}_2\text{O}_3 on ACC. Most importantly, the image to the right displays that sulfur forms a distinguishable annulet with a 2 μm wall thickness (the boundary is marked with a dotted circle), coincident with the \text{Al}_2\text{O}_3 distribution in the fiber. The low density of sulfur interdispersed in the fiber center is a result of sulfur diffusion.

Conclusion

We have shown the viability of ultrathin ALD-\text{Al}_2\text{O}_3 for enhancing the collection (adsorption/absorption) and reactivation of dissolved polysulfides on ACC, improving the initial capacity and stabilizing the cycle life remarkably for Li-S batteries. We demonstrate this effect with a single ACC layer. Ultrathin ALD coatings not only maintain the high surface area of highly porous structures but also help the sorption of polysulfides.