# **Reactivation of Dissolved Polysulfides in Li-S Batteries** Xiaogang Han, Raphael Elspas, Chen-yu Chen, Jiaqi Dai, Liangbing Hu\*

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## Abstract

This work demonstrates the effect of atomic layer deposited (ALD)  $Al_2O_3$  on the reactivation of dissolved polysulfides in Li–S batteries. A layer of Al<sub>2</sub>O<sub>3</sub> 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-Al<sub>2</sub>O<sub>3</sub> improves the initial specific capacity and stabilizes the cycle life remarkably. Scanning electron microscopy verified that the ALD-Al<sub>2</sub>O<sub>3</sub> 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 degeneration 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 Al<sub>2</sub>O<sub>3</sub> using a process called atomic layer deposition, to adsorb and reactivate the dissolved polysulfides, making them return to the cathode.

<b>Reactivation Idea</b>		
а	b	
+		
S cathode	ACC Polysulfides	s→
Reactivation Layer		
Separator	ACC Polysulfides	s
Li anode 	Al <sub>2</sub> O3-ACC	

The reactivation idea is illustrated above. Instead of blocking polysulfide dissolution, the porous conductive ACC is used as a model material. The ALD-Al<sub>2</sub>O<sub>3</sub> 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 ¼ 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.



Valve

#### **Device for Sulfur infiltration in ACC**

- Vacuum-sealed glass tube
- Vacuum connector
- ACC disks linked with wire
- Sulfur powder

## Step 2: Atomic Layer Deposition of Al<sub>2</sub>O<sub>3</sub>

- A plain carbon cloth disk is taken and placed in an atomic layer deposition system.
- This process deposits a one atom thick layer of  $Al_2O_3$ 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.





### ACC with $Al_2O_3$ Separator coating (Step 2 above) Polysulfides Lithium Anode Sulfur Cathode (Step 1 above)

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### **Battery Performance** Below are comparisons that verify that $ALD-Al_2O_3$ is a more efficient battery structure than other Li-S systems. S-ACC 400 600 800 1000 1200 S-ACC Reactivation shows Al<sub>2</sub>O<sub>3</sub>-ACC S-ACC Reactivation ACC. PRECURSOR Specific capacity (mAh/g vs. S)

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) the charge/discharge cathode design with  $ALD-Al_2O_3$  on

The picture below depicts the SEM micrographs for aluminum and sulfur distributions on the cross-section of a representative  $Al_2O_3$ -ACC fiber. The image in the middle illustrates that the aluminum distribution does not change during cycling, indicating the stable coating of ALD-Al<sub>2</sub>O<sub>3</sub> on ACC. Most importantly, The image to the right displays that sulfur forms a distinguishable annulet with a 2  $\mu$ m wall thickness (the boundary is marked with a dotted circle), coincident with the  $Al_2O_3$  distribution in the fiber. The low density of sulfur interspersed in the fiber center is a result of sulfur diffusion.



## Conclusion

We have shown the viability of ultrathin ALD-Al2O3 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.

