

Friday, 28-Jan-2022

Dear Yuxuan Zhang,

Your abstract (ID: 3724477) entitled:

"Enabling High-Rate Long-lifespan Lithium-Sulfur Batteries via Stereolithography Technique and Oxidative Chemical Vapor Deposition"

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Control ID # (3724477)

Title: "Enabling High-Rate Long-lifespan Lithium-Sulfur Batteries via Stereolithography Technique and Oxidative Chemical Vapor Deposition"

Abstract Body: Abstract Body

Enhancing battery energy storage capability and reducing the cost per average energy capacity is urgent to satisfy the increasing energy demand in modern society. The lithium-sulfur (Li-S) battery is especially attractive because of its high theoretical specific energy (around 2600 Wh kg^{-1}), low cost, and low toxicity.¹

Despite these advantages, the practical utilization of lithium-sulfur (Li-S) batteries to date has been hindered by a series of obstacles, including low active material loading, shuttle effects, and sluggish sulfur conversion kinetics.² The traditional 2D planar thick electrode is considered as a general approach to enhance the mass loading of the Li-S battery.³ However, the longer diffusion length of lithium ions, which resulted in high tortuosity in the compact stacking thick electrode, decreases the penetration ability of the electrolyte into the entire cathode.⁴ Although an effort to induce catalysts in the cathode was made to promote sulfur conversion kinetic conditions, catalysts based on transition metals suffered from the low electronic conductivity and some elements (i.e.: Co, Mn) may even absorb and restrict polysulfides for further reaction.⁵

To mitigate the issues listed above, herein we propose a novel sulfur cathode design strategy enabled by additive manufacturing and oxidative chemical vapor deposition (oCVD).^{6,7} Specifically, the cathode is designed to have a hierarchical hollow structure via a stereolithography technique to increase sulfur usage. Microchannels are constructed on the tailored sulfur cathode to further fortify the wettability of the electrolyte. The as-printed cathode is then sintered at 700°C in a N_2 atmosphere in order to generate a carbon skeleton (i.e.: carbonization of resin) with intrinsic carbon defects. The intrinsic carbon defects are expected to create favorable sulfur conversion conditions with sufficient electronic conductivity. In this study, the oCVD technique is leveraged to produce a conformal coating layer to eliminate shuttle effects. Identified by scanning electron microscopy and energy-dispersive X-ray spectroscopy mapping characterizations, the oCVD PEDOT is not only covered on the surface of the cathode but also the inner surface of the microchannels. High resolution x-ray photoelectron spectroscopy analyses (C 1s and S 2p orbitals) between pristine and modified sample demonstrate that the high concentration of the defects have been produced on the sulfur matrix after sintering and posttreatment. In-operando XRD diffractograms show that the Li_2S is generated in the oCVD

PEDOT-coated sample during the charge and discharge process even with a high current density, confirming an eminent sulfur conversion kinetic condition. In addition, ICP-OES results of lithium metal anode at different states of charge (SoC) verify that the shuttle effects are excellently restricted by oCVD PEDOT.

Overall, the high mass loading ($> 5 \text{ mg cm}^{-2}$) with an elevated sulfur utilization ratio, accelerated reaction kinetics, and stabilized electrochemical process have been achieved on the sulfur cathode by implementing this innovative cathode design strategy. The results of this study demonstrate significant promises of employing pure sulfur powder with high electrochemical performance and suggest a pathway to the higher energy and power density battery.

- 1 Chen, Y. Adv Mater 33, e2003666.
- 2 Bhargav, A. Joule 4, 285-291.
- 3 Liu, S. Nano Energy 63, 103894.
- 4 Chu, T. Carbon Energy 3.
- 5 Li, Y. Matter 4, 1142-1188.
- 6 John P. Lock. Macromolecules 39, 4 (2006).
- 7 Zekoll, S. Energy & Environmental Science 11, 185-201.

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