CNMS User Project Highlight

A Core-Shell Nanostructure for High Energy Lithium-Sulfur Batteries

Scientific Achievement

Core-shell-structured Li_2S nanoparticles, with Li_2S as the core and Li_3PS_4 as the shell, function as lithium superionic sulfide (LSS) cathode for long-lasting, energy-efficient lithium-sulfur (Li-S) batteries. The core-shell structure improves the ionic conductivity of Li_2S by 6 orders of magnitude, yielding an all-solid Li-S battery with excellent cycling performance.

Significance

The poor ionic and electronic conductivity of traditional Li-S cathodes limit their cyclability resulting in a short cycle life. The high lithium-ion conductivity of the new LSS imparts excellent cycling performance to all-solid Li-S batteries, which also promises safe cycling of high-energy batteries with metallic lithium anodes because of the replacement of flammable liquid electrolytes with solid electrolytes.

Research Details

- **CNMS Capability**: A core-shell nanostructure of Li_2S has been developed through a facile, solution-based, chemical synthesis approach.
- The nanostructure enhanced the ionic conductivity of Li_2S by 6 orders of magnitude, thus enabling efficient cycling of resulting all-solid-state Li-S batteries.
- The dissolution of sulfur species in liquid electrolytes of a conventional battery configuration limits the cycle life of Li-S batteries. The all-solid-state Li-S battery configuration eliminates this limitation and confers a long cycle life to Li-S batteries.
- All-solid-state Li-S batteries enable safe cycling of metallic lithium anodes.

Zhan Lin, Zengcai Liu, Nancy J. Dudney, and Chengdu Liang
ACS Nano. **DOI:** 10.1021/nn400391h

$$S + 2\,Li(CH_2CH_3)_3BH \rightarrow Li_2S\downarrow + 2\,(CH_2CH_3)_3B + H_2\uparrow$$

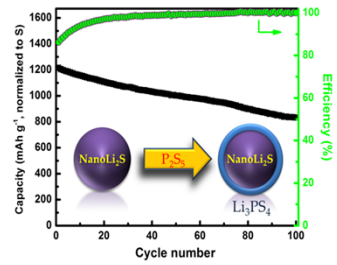

Figure 1 Electrochemical cycling and coulombic efficiency of nanostructured Li_2S in solid state.

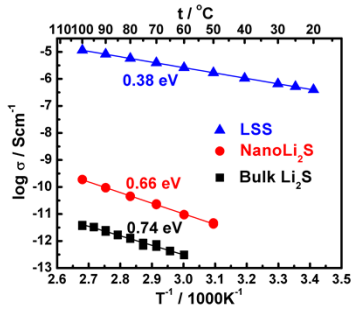

Figure 2 Temperature dependency of ionic conductivities of the Bulk Li_2S, NanoLi_2S, and LSS.

U.S. DEPARTMENT OF ENERGY | Office of Science

Center for Nanophase Materials Sciences

OAK RIDGE National Laboratory

Highlight Narrative Text (~175 words)

A facile synthesis approach of Li_2S nanoparticles was discovered by reacting elemental sulfur with lithium triethylborohydride in THF. Exposure of the surface of Li_2S nanoparticles with P_2S_5 results in a core-shell structure of $Li_2S@Li_3PS_4$, which is a superionic conductor, LSS. The high ionic conductivity and reduced particle size of LSS impart an excellent cyclability and rate capability to all-solid Li-S batteries. The ionic conductivity of cathode materials is identified as the key parameter for all-solid Li-S batteries. Although advances have been reported in systems using Li_2S in conventional Li-S batteries with liquid electrolytes, significant challenges still remain such as the migration of sulfur through polysulfide shuttle and the safety concerns of cycling metallic lithium in flammable liquid electrolytes. With a paradigm shift from a conventional battery configuration to all-solid batteries, the above challenges with liquid electrolytes are completely eliminated. However, the all-solid batteries face new challenges of low ionic conductivity and high interfacial resistance. This research introduces the approach of solving these problems through innovative nanostructured materials with enhanced ionic conductivity and reduced interfacial resistance.

Reference:

"A Lithium Superionic Sulfide Cathode for All-Solid Lithium-Sulfur Batteries"
Z. Lin (Materials Science and Technology Division, ORNL), Z. Liu (CNMS, ORNL), N. Dudney (MSTD, ORNL), C. Liang (CNMS, ORNL)
ACS Nano (2013).

Acknowledgment of Support:

This research was sponsored by U.S. Department of Energy (DOE)/Energy Efficiency and Renewable

Energy (EERE) through Vehicle Technologies Office. The investigation of the ionic conductivity of these new compounds and the preparation of solid electrolytes were supported by the Division of Materials Science and Engineering, Office of Basic Energy Sciences U.S. Department of Energy (DOE). The synthesis and characterization were conducted at the Center for Nanophase Materials Sciences, which is sponsored at Oak Ridge National Laboratory by the Division of Scientific User Facilities, U.S. DOE.