Superionic Cathode Materials for All-Solid-State Li-S Batteries

Chengdu Liang[*1], Adam J. Rondinone[1], and Nancy. J. Dudney[2]
1. Center for Nanophase Materials Sciences Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831
2. Materials Science and Technology Division, Oak Ridge National Laboratory, Oak Ridge, TN 37831

A sustainable energy future harvests intermittent renewable energies such as solar, wind, and tide energies into a stable supply of electricity on-demand. Battery technology is the central element to enable such a sustainable energy future. High energy, low cost, and enhanced safety are basic requirements for the development of the next generation of batteries that meet the needs of large scale storage of electricity. Among the known battery chemistries, lithium-sulfur (Li-S) holds the promise of high-energy batteries. However the development of Li-S batteries has been impeded by the intrinsic low electronic and ionic conductivities of the sulfur cathode and the poor cycleability of the metallic lithium anode. To enable the cycling of the sulfur cathode in a conventional Li-S battery, a liquid electrolyte that has a high solubility of polysulfides is required to overcome the poor ionic conductivities of solid sulfur species. As a result of the dissolution of polysulfides in the liquid electrolyte, the diffusion of polysulfides from the cathode to the anode during the battery cycling leads to intrinsically short cycle-life. In additional to the problems of the sulfur cathode, the dendritic growth of lithium metal in liquid electrolytes and the ever growing of solid electrolyte interphase on the anode surface shorten the cycle-life of Li-S batteries and also cause safety concerns.

In this work, we address the aforementioned challenges through an innovative all-solid-state configuration of Li-S batteries. Conventional liquid electrolytes have been replaced by a solid electrolyte with the advantages: (a) complete elimination of the polysulfide shuttle and (b) safe cycling of metallic lithium anode. The key challenges of all-solid Li-S batteries are to meet the following conditions: (1) good electrolytes that have high ionic conductivity and excellent electrochemical stability with both the anode and cathode; (2) a cell design that can handle the mechanical stress of large volume changes and prevent the penetration of lithium dendrites; and (3) sulfur cathode with high electronic and ionic conductivities. This presentation provides a detailed progress report on our research on thio-LISICON ceramic electrolytes[1], the cold-pressing fabrication of all-solid-state Li-S batteries, and novel cathode materials including a core-shell structured $Li_2S@Li_3PS_4$ nanoparticles[2] and lithium polysulfidophosphates[3]. The cycleability of the all-solid-state Li-S batteries is strongly correlated to the ionic conductivity of the cathode materials. This research gives insights on the design of all-solid-state Li-S batteries. Experimental details will be presented at the meeting.

Acknowledgment:
This work was sponsored by the Division of Materials Science and Engineering, Office of Basic Energy Sciences (synthesis and characterization of solid electrolytes), and the Assistant Secretary for Energy Efficiency and Renewable Energy, Vehicle Technologies Office (VTO) of the U.S. Department of Energy (investigation of all-solid-state Li-S batteries). The synthesis was 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. Department of Energy.

References:

1, Liu, Z.; Fu, W.; Payzant, E. A.; Yu, X.; Wu, Z.; Dudney, N. J.; Kiggans, J.; Hong, K.; Rondinone, A. J.; Liang, C.D., “Anomalous High Ionic Conductivity of Nanoporous β-Li_3PS_4,” *Journal of the American Chemical Society* **2013,** *135* (3), 975.
2, Lin, Z.; Liu, Z.; Dudney, N.J.; and Liang, C.D.; “A Lithium Superionic Sulfide Cathode for All-Solid Lithium-Sulfur Batteries,” *ACS Nano*, **2013** 7, 2829
3, Lin, Z.; Liu, Z.; Fu, W.; Dudney, N.J.; and Liang, C.D.; “Lithium Polysulfidophosphates: A Family of Lithium-Conducting Sulfur-Rich Compounds for Lithium-Sulfur Batteries,” *Angewandte Chemie-International Edition* **2013,** DOI: 10.1002/anie.201300680