The emerging applications of electric vehicles (EV) and grid scale energy storage are pushing the limit of current energy storage technologies. To meet the US Department of Energy (DOE)'s targets for EV batteries and grid storage, battery chemistries beyond the lithium-ion systems are required. Among the many new chemistries studied, lithium-sulfur battery is extremely attractive and promising because of its high theoretical specific energy of ~2600 Wh/kg, six times higher than that of the current lithium-ion battery technology. However, successful practical applications of lithium-sulfur batteries have been impeded by challenges associated with both sulfur cathode and lithium metal anode. Here I will present my research on tackling the problems of lithium-sulfur batteries through multifaceted approaches at different length-scales, allowing unprecedented control over the electrode architecture from the nanoscale to macroscale. For the cathode side, I will present my work on fundamental understanding of the lithium-sulfur reaction and rational design and synthesis of unique sulfur nanostructures with multifunctional coatings to overcome the issues related to electrode volume expansion, loss of active materials, and insulating nature of sulfur. For the anode side, I will present that the growth of lithium dendrites, which is the cause for battery internal short-circuit, can be effectively suppressed by chemical modification of lithium metal anodes. These research findings provide new insights and open up exciting opportunities for the next generation of cost-effective and high-energy batteries.