Sulfurized-Carbonized Polyacrylonitrile Cathode with Modified Binder and Electrolyte Additive for High-Performance Lithium-Sulfur Batteries

Hee Min Kim [a], Doron Aurbach [b], Yang-Kook Sun [a,*]

[a] *Department of Energy Engineering, Hanyang University, Seoul 04763, South Korea*

[b] *Department of Chemistry and BINA (BIU Institute of Nano-technology and Advanced Materials), Bar-Ilan University, Ramat-Gan 5290002, Israel*

E-mail: *cssilver87@gmail.com*

Li−S batteries are among the more auspicious candidates as next-generation ESSs, owing to the abundance and inexpensive cost of raw sulfur, environmental benignity, theoretical capacity of 1675 mAh g^{-1} for sulfur cathodes, and theoretical energy density of 2500 Wh kg^{-1} for these ESSs.[1] Yet, in spite of their benefits, Li−S batteries have not been successfully commercialized to date because they suffer from several problems: (1) lithium sulfide dissolution and their shuttle reactions that avoid full oxidation of the lithium sulfide to sulfur in the cathodes, (2) volatility of the conventional ether-based electrolyte solutions (necessary to operate Li−S batteries properly), and (3) only a partial utilization of sulfur at the cathodes due to the low sulfur conductivity of 5×10^{-30} S cm^{-1}.[2] We prepared novel sulfurized polyacrylonitrile cathodes by melting sulfur into carbonized polyacrylonitrile (CPAN) polymer. As the charge−discharge mechanism of sulfurized CPAN (S-CPAN) cathodes is different from that of most sulfur cathode Li−S batteries, it has the benefit of bypassing the lithium sulfide dissolution phenomenon and the shuttle problem, thereby preventing active material loss. In addition, the lack of soluble lithium sulfide formation allows the use of carbonate-based electrolyte solutions (provided that the right selection of materials has been made), instead of the ether-based electrolyte solutions used in conventional Li−S batteries.[3] To take advantage of the unique charge−discharge mechanisms of S-CPAN electrodes at high sulfur loadings, we introduce a novel cell design through the application of poly(acrylic acid) (PAA) binder to S-CPAN electrodes using alkyl carbonate-based electrolyte solutions with fluoroethylene carbonate (FEC) as an additive. PAA-based binders were found to be superior over PVdF in a previous work, in terms of excellent adhesion and cohesion properties of composite electrodes. FEC is considered additive or cosolvent in Li salt electrolyte solutions, working with Li metal or Li−Si anodes and high-voltage Li insertion cathodes or S−C composite cathodes, as its surface reactions lead to polymerization and formation of effectively passivating surface film.[4] Here, we tested S-CPAN cathodes with high (fully practical) loading. The combination of these three components "S-CPAN, PAA, and FEC" resulted in outstanding electrochemical performance of practically loaded sulfur cathodes with stable cycle life. We demonstrated the scale-up pouch cells with four electrodes connected in parallel has cell capacity of 260 mAh.[5]

References:

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