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Carbon Nanotube Based Sulfur Composite 3D Cathodes for Li-Sulfur Batteries

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Sulfur is one of the most promising cathode materials, with its theoretical specific capacity of 1675 mAh/g (the highest value for all known solid cathode materials), for the next generation of rechargeable batteries. However, the poor rechargeability and the fast capacity degradation owing to the insulating nature of sulfur and the dissolution of various soluble polysulfide intermediates formed during discharge process into the electrolyte are the major hurdles inherent in Li/S batteries that hindered their mass commercialization. The development of new battery architectures is essential to overcome these problems for Li/S batteries to succeed. In this study, the three-dimensional electrodes based on carbon nanotubes are utilized as electrode substrates to load sulfur inside their matrix. Two types of CNT based materials are examined in comparison to each other: MW-CNT buckypapers and vertically aligned carbon nanotubes (VACNT) are investigated as the 3D cathode materials. (The VACNTs are directly synthesized on stainless steel substrate by employing alumina catalyst support layer and a CVD process.) Carbon nanotubes, as a highly conducting form of carbon, can facilitate essential intimate contact of insulating sulfur to enable a reversible electrochemical reaction at high current rates. MW-CNT paper with large specific surface area and abundant micro- and meso-pores and VACNTs with its interconnected highly ordered structure, both exhibit a unique conductive matrix to confine sulfur in between their sole porous structures and deliver the ions and electrons efficiently to the sulfur. Moreover, they provide good structural stability of the cathode.

The CNT/Sulfur composites are prepared by a melt-infiltration and sublimation/condensation strategy with post heat treatments. The sulfur loaded CNT electrodes are further coated with conducting polymers. This external layer hinders the out-diffusion of the soluble polysulfides formed during discharge process into the electrolyte by encapsulating and adsorbing these intermediates in its unique highly torturous pore structure. These materials are tested as novel cathodes for Li/S batteries and the results demonstrate improved cyclability of the cells and the utilization of sulfur.