N-doped Graphene-CNT Cathode Composites for High-Performance Li-S Batteries

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Abstract

Reaching beyond the horizon of Li-ion batteries is a formidable challenge, as energy storage will be more important in the future than at any time in the past. This requires the exploration of new chemistry, especially electrochemistry, and new materials. Among the available choices, lithium-sulfur (Li-S) batteries are attractive because of their low cost, environmental friendliness, and superior energy density (theoretically 2500 Wh/kg) [1]. Despite their great promise, the commercialization of Li-S batteries is hindered by low cycle stability, fast capacity fading, and low efficiency; these problems are induced by the polysulfide shuttle reaction that occurs through dissolution into electrolyte, the loss of active sulfur material, and the electronically insulating nature of sulfur [2]. To tackle these issues, we synthesized a novel nanostructured composite consisting of Graphene and Carbon Nanotubes (CNT), each pre-doped with Nitrogen (N), as a host material for sulfur impregnation. In this unique composite, not only do CNTs act as the spacer units between the graphene sheets, avoiding losses in performance through restacking and agglomerate formation, but they also contribute to the material activity, overall surface area/nanoporosity, and the composite conductivity over the effective doping method strategy. In the next step, a new method of sulfur impregnating into the composite structure was implemented to effectively confine the sulfur into graphitic clusters, voids, and defects in N-doped graphene–CNT composite. The carbon nanostructured framework serves not only as a conductive layer for encapsulating sulfur and polysulfides without direct contact with electrolyte, but also as a nano-electrochemical reaction chamber. Cyclic voltammetry (CV) of the sulfur-impregnated N-doped graphene-CNT in high-dielectric solvent behaved similar to low-dielectric solvent, indicating that the material was not solvent dependent. Strong interaction between N-doped composite and sulfur or polysulfides enabled us to demonstrate Li–S cells with a high reversible capacity (1005-1592 mAh/g), outstanding cycling stability, and great Coulombic efficiency (~97.5%). The results indicate that the sulfur-impregnated N-doped graphene-CNT nanocomposite is a very promising cathode material for high-performance lithium–sulfur batteries.

References:

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