Composite cathode for Li-sulfur system: Effect of inorganic additives on electrochemical performance

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Abstract

The rapid development of portable electronic devices together with new large-scale applications stimulate demand for battery systems providing high capacity and energy density. Since the classical Li-ion batteries based on intercalation approached their limits, researchers have moved their attention to a Li-sulfur battery with a theoretical capacity of 1675 Ah kg^{-1} and energy density of 2600 Wh kg^{-1}. Except for the non-conductivity of elemental S and large volume changes during redox reactions, the main problems are related to the dissolution of lithium polysulfides (PS) in the electrolyte. PS diffusion to the anode compartment of the Li-S battery causes severe redox shuttle between the cathode and Li anode and results in a low coulombic efficiency for charging/discharging and a fast self-discharge during storage[1]. Non-conductivity of sulfur requires the incorporation of electron-conductive additive and volume changes accompanying PS formation represent a challenge for cathode architecture. The carbonaceous materials were demonstrated to enhance the performance of sulfur-based cathode composites. In addition, the inorganic component provides a higher adsorption ability for PS. In our work, we studied the influence of two different activated carbons (Fluka or Penta) and five inorganic additives on the electrochemical performance of the sulfur composite cathode. The electrochemical behavior of the materials was evaluated by cyclic voltammetry at 0.1 mV s^{-1} and galvanostatic chronopotentiometry and correlated with their morphology. The inorganic additives (nano-TiO_2, TiO_xN_y-anatase, rutile/TiO_xN_y, P90, $Li_4Ti_5O_{12}$) in the cathode increase the capacity of the corresponding Li-S cell. This increase is more pronounced for Penta carbon with a larger external surface area of 345 m^2 g^{-1}. Commercial P90 titania in the Penta/sulfur composite cathode provides the highest voltammetric charge capacity of 731 mAh g^{-1}. This effect is ascribed to the excellent polysulfide adsorption ability of this disperse material. TiO_xN_y-anatase additive exhibits the second-highest capacity improvement due to surface-enhanced redox chemistry of its conductive and sulphiphilic surface. The electrochemical performance of all the Li-sulfur cells containing Penta/sulfur/inorganic additive cathode is boosted by the separator modification with P90. The best performance provides the cell containing Penta/sulfur/nano-TiO_2 cathode and P90-modified separator. The synergic effect of the Penta carbon large external surface area, the high surface area of nano-TiO_2 boosted by the presence of highly disperse P90 in the separator manifests itself by the voltammetric charge capacity of 881 mAh g^{-1}.

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References

[1] S.S. Zhang, Liquid electrolyte lithium/sulfur battery: Fundamental chemistry, problems, and solutions, J Power Sources, 231 (2013) 153-162.