Mesoporous Hydroxylated Carbon Nanofibers as an Effective Cathode Scaffold to Enhance the Lithiation-Delithiation Reversibility of Lithium-Sulfur Batteries

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Taking a reversible electrochemical reaction of elemental sulfur towards metallic lithium into consideration, it seems to be inappropriate to calculate Li^+ -ion diffusion coefficient (D_{Li}) within a solid-state cathode of carbon-sulfur composites using the concentration of lithium ions in electrolyte solution. In this paper, cross-linked non-porous carbon nanofibers (CNF) and mesoporous hydroxylated carbon nanofibers (m-HCNF) are prepared and comparatively utilized as sulfur-loading matrices to improve the lithiation-delithiation reversibility between molecular S_8 and metallic lithium. When model cells are assembled at a constant dosage of electrolyte (30 mL) per gram of sulfur, at open-circuit voltage the D_{Li} value of a lithium-sulfur (Li-S) battery cathode can be estimated according to the S_8 concentration therein: $3.51 \times 10^{-15} \text{ cm}^2 \text{ s}^{-1}$ for the low sulfur-loading cathode S/CNF (47.0 wt%); $4.21 \times 10^{-15} \text{ cm}^2 \text{ s}^{-1}$ for the high sulfur-loading electrode S/m-HCNF (~ 62.0 wt%). At 0.5 C the initial Coulombic efficiency of S/m-HCNF (96.9%) is higher than that of S/CNF (88.8%), and thereafter the better cycling stability and rate performance of the former coincide well with the varying D_{Li} values. In a word, these indicate an enhanced lithiation-delithiation reversibility of composite S/m-HCNF for high-performance Li-S batteries.

Keywords: Lithium-ion diffusion coefficient; Lithiation-delithiation reversibility; Mesoporous carbon nanofiber; Lithium-sulfur batteries.

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