Effect of Cathode Porosity on High Loading Li-S Battery Performance with Sparingly Polysulfide-Solubilizing Electrolyte

Yoshiharu Matsumae, Kenzo Obata, Ayumi Ando, Azusa Nakanishi, Masato Yanagi, Yutaro Kamei, Kazuhide Ueno, Kaoru Dokko, and Masayoshi Watanabe
Yokohama National University, Department of Chemistry & Biotechnology, 79-5 Tokiwadai, Hodagaya-ku, Yokohama, 240-8501, Yokohama, Japan

E-mail: matsumae-yoshiharu-mc@ynu.ac.jp

Lithium-sulfur (Li-S) batteries have attracted much attention due to their high theoretical capacity (1672 mA h g\(^{-1}\)), as well as the low cost and natural abundance of sulfur. One of the most serious problems for practical application of Li-S batteries is the dissolution of lithium polysulfides (PS), which causes poor charge-discharge cycle stability and low coulombic efficiency. Recently, sparingly PS solubilizing electrolytes, such as ionic liquid-based electrolytes\(^1\)\(^-\)\(^3\) and highly concentrated electrolytes\(^4\)\(^-\)\(^6\), have applied as new candidate of electrolytes for Li-S battery. We have reported that Li-S battery with the solvate ionic liquid (SIL) electrolyte, which is a type of highly concentrated electrolytes, showed good cycle stability and Coulombic efficiency\(^7\), however, increase of sulfur loading amount without lowering battery performance is still indispensable challenges. In this work, we achieved high sulfur loading using 3D current collector and elucidated how the cathode porosity affects the battery performance.

The sulfur / Ketjen black (KB) composites prepared by melt-diffusion strategy were dispersed in carboxymethyl cellulose (CMC) aqueous solution with sulfur : KB : CMC = 60 : 30 : 10 (wt %). The obtained slurry was infiltrated into Al foam as a 3D current collector. [Li(G4)][TFSA] was used as a SIL electrolyte. Cathode porosity calculated on the basis of physical properties of each composite material was controlled by changing press pressure of composite cathode. As shown in Fig. 1, the initial discharge capacity was greatly degraded in the cells with low porosity cathode (< 30%). Discharge curve with low porosity cathode degraded after the first voltage plateau with high overpotential. This capacity loss due to the overpotential was recovered by lowering discharge current densities, suggesting that the overpotential was caused by kinetic factors such as slow ionic transport in the composite cathode. We will also discuss the relationship between the electrolyte volume and cathode porosity.

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![Fig. 1 Initial discharge capacities (left) and curves (right) with different cathode porosities. Sulfur loadings were 1.2-5.7 mg cm\(^{-2}\).](image)