In-Operando Transmission X-ray Microscopy Study on Dynamics of Sulfur Dissolution And Re-Deposition in Li-S Battery

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Lithium-sulfur (Li-S) battery is a promising rechargeable battery system that has both high theoretical capacity (1675 mAh g^{-1}) and energy density (2600 Wh kg^{-1}).[1] Moreover, sulfur (S) is inexpensive and nontoxic, making Li-S suitable for large-scale energy storage applications. Sulfur (S_8) lithiation during the operation of Li-S battery is a multi-step electrochemical process that involves different lithium polysulfide (Li_2S_n, $1 \leqq n \leqq 8$) intermediates. The long-chain polysulfides are highly soluble in the aprotic organic electrolyte. The reported rapid capacity fading of Li-S batteries upon repeated cycling has often been linked to dissolution of the polysulfides.

Previous studies investigating the extent of sulfur dissolution during the operation of Li-S batteries are not without controversy. Previous transmission X-ray microscopy study showed rather minor change (reduction) in particle size, suggesting insignificant sulfur dissolution, while . In contrast, ex-situ scanning electron microcopy observation suggested extensive S dissolution upon lithiation along with formation of insoluble Li_2S. In none of the previous studies, however, the dynamics of the dissolution process has not been revealed.

We carry out in this work in-operando TXM study on the S particle electrodes of Li-S battery. S electrodes have been subjected to complex electrochemical protocols involving varied depths of charge/discharge. The object of this study is twofold: first, to resolve the conflicting reports on the extent of S dissolution; second, to study the dynamics of both dissolution and re-deposition of S within working Li-S batteries. Re-deposition of S may take place during de-lithaition, which has never been investigated in the literature but is believed to play equally important part as dissolution in affecting the cycle stability of S electrode. Better understanding of the dynamics of the morphological variation of the S particle electrode in response to the dissolution/re-deposition processes during the operation of the Li-S battery should provide valuable information to the development of countermeasures that might enhance the cycle performance of Li-S batteries.

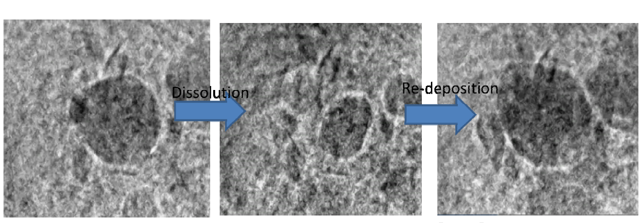

Figure 1. Transmission X-ray Micrographs showing dissolution and re-deposition of S particles