Crosslinked disulfide materials as cathodes for stable cycling in Lithium–Sulfur batteries

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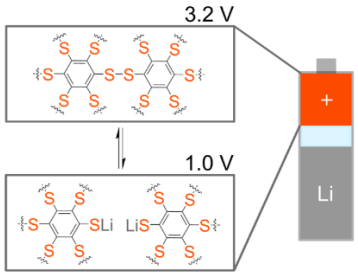
The main two advantages of Li-S is (i) the high theoretical capacity of elemental sulfur and (ii) the sustainability of sulfur-based materials over toxic, expensive heavy metals such as cobalt. Sulfur can theoretically achieve 1675 mAh g^{-1} , but due to the parasitic polysulfide shuttle problem, the capacity rapidly fades. We demonstrate a novel crosslinked disulfide system as a cathode material for Li–S cells that is designed with the two criteria of having only a single point of S–S scission and maximizing the ratio of S–S to the electrochemically inactive framework. The material therefore maximizes theoretical capacity while inhibiting the formation of polysulfide intermediates that lead to parasitic shuttle. The material we report contains a 1:1 ratio of S:C with a theoretical capacity of 609 mAh g^{-1}. The cell gains capacity through 100 cycles and has 98 % capacity retention thereafter through 200 cycles, demonstrating stable, long-term cycling. Raman spectroscopy confirms the proposed mechanism of disulfide bonds breaking to form a S–Li thiolate species upon discharge and reformation upon charge. Coulombic efficiencies near 100% for every cycle, suggesting the suppression of polysulfide shuttle through molecular design.



References:

[1] M.B. Preefer, B. Oschmann, C.J. Hawker, R. Seshadri, F. and Wudl. High Sulfur Content Material with Stable Cycling in Lithium-Sulfur Batteries. *Angew. Chem. Int. Ed.* 56 (2017) 15118-15122.