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Covalently Interlinked Graphene Sheets with Sulfur-Chains Enable Superior Lithium Sulfur Battery Cathodes at Full-Mass Level

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Sulfur represents a low-cost, sustainable and high theoretical capacity cathode material for lithium-sulfur batteries, which could meet the growing demand in portable power sources. However, the shuttling-effect of the formed lithium polysulfides, as well as their low electric conductivity, compromise the electrochemical performance of lithium-sulfur cells.[1] To tackle this challenge, a so far unexplored cathode was developed, composed from sulfur covalently bonded directly on graphene. This was achieved by leveraging the nucleophilicity of polysulfide chains, which reacted readily with the electrophilic centers in fluorographene (Fig 1a). The reaction lead to the formation of carbon-sulfur covalent bonds and a particularly high sulfur content of 80 wt. %. Owing to these features, the developed cathode exhibited excellent performance with only 5 wt. % of conductive carbon additive, delivering very high full-cathode-mass capacities and rate capability, combined with superior cycling stability (Figure 1b,c). In combination with a fluorinated ether as electrolyte additive, the capacity persisted at ~700 mAh g^{-1} after 100 cycles at 0.1 C, and at ~644 mAh g^{-1} after 250 cycles at 0.2 C, keeping ~470 mAh g^{-1} even after 500 cycles.[2]

References

[1] A. Bhargav, J. He, A. Gupta, A. Manthiram, Joule 2020, 4, 285.

[2] I. Tantis, A. Bakandritsos, D. Zaoralová, M. Medveď, P. Jakubec, J. Havláková, R. Zbořil, M. Otyepka Adv. Funct. Mater. 2021, 2101326

Figures

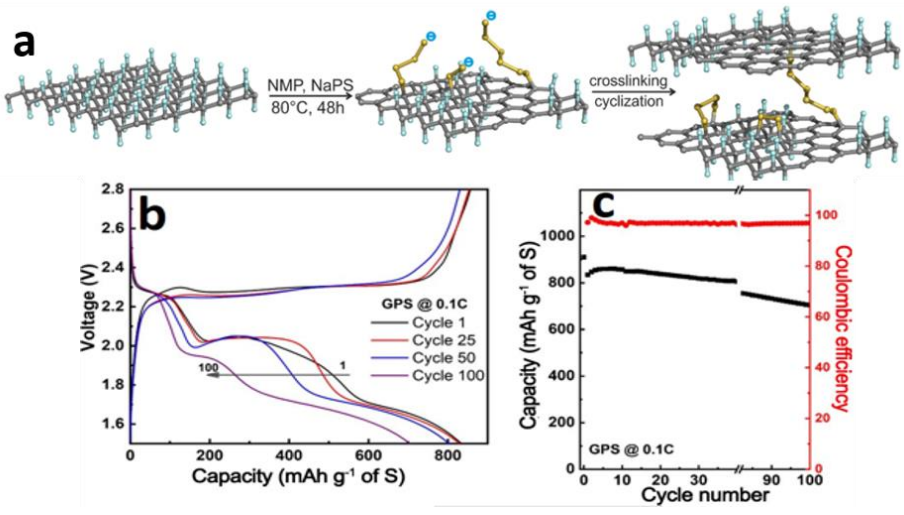

Figure 1: (a) Reaction scheme for covalent binding of sodium polysulfide (NaPS) to fluorographene (FG) leading to graphene-polysulfide (GPS), (b,c) Electrochemical performance of the prepared cathode against lithium in 1 M LiTFSI in 1:1 DOL:TTE electrolyte