Titanium Sulfide-Iodine Hybrid Catalysts Promote Solid-Solid Redox Reactions in All-Solid-State Lithium-Sulfur Batteries

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All-solid-state lithium-sulfur batteries (ASSLSBs) have emerged as the next-generation energy storage systems due to their high energy density (~2600 Wh kg?¹) and safety, enabled by non-flammable solid electrolytes (SE). However, ASSLSBs are limited by (1) sluggish solid-state Li2S2-Li2S conversion due to sparse three-phase boundaries (Sulfur|SE|Carbon) and a high energy barrier, and (2) poor solid-solid interfacial contacts, which hinder ion/electron transport and cycling stability. Hence, we propose a TiS2 and I2 hybrid catalyst for the sulfur cathode to boost the sluggish reaction. I2 acts as a redox mediator that activates otherwise inactive two-phase boundaries (SE|Li2S), while TiS2 forms a mixed ion/electron conductive buffer layer that fills the interfacial gaps. Together, the number of active sites is increased. Furthermore, conductive carbon, sulfur loading, melt diffusion process, and high-energy ball milling (HEBM) rotational speed were systematically investigated. A composite cathode incorporating BP2000 with 70% sulfur, fabricated through a two-step sulfur-carbon melt diffusion process followed by HEBM at 500 rpm, delivered a high reversible capacity of 1400 mAh g?¹ over 300 cycles and retained 740 mAh g?¹ at a high rate of 4C. Overall, this study establishes an effective strategy to enhance sluggish reaction kinetics and interfacial contact in ASSLSBs, advancing their practical implementation.

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