A Comprehensive Cognition for the Capacity Fading Mechanism of FeS2 in Argyrodite-based All-solid-state Lithium Battery

Zhan Wu[1], Wenkui Zhang[1], Yang Xia[1], Hui Huang[1], Yongping Gan[1], Xinping He[1], Xin-Hui Xia[1], and Jun Zhang[1]

[1]Zhejiang University of Technology

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Abstract

Sulfide solid state electrolyte (SSE) possesses high ionic conductivity and great processability but suffers from narrow electrochemical window. Conversion sulfide cathode FeS2 has higher specific capacity and moderate redox potential, making it appropriate towards sulfide SSE. However, the complex reaction pathway and capacity fading mechanism in FeS2 are rarely studied, especially in all-solid-state lithium battery (ASSLB). Herein, argyrodite sulfide SSE is paired with FeS2 to investigate the electrochemical reaction pathways and the capacity fade mechanism. Instead of single conversion reaction, an anionic redox driven reaction of FeS2 is revealed. The oxidization of Li2S vanishes and large quantity of inactive Li2S accumulates to cause the interfacial deterioration, along with the stress concentration during cycling, which leads to the rapid capacity fade of FeS2. Finally, a simple strategy of slurry-coated composite electrode with highly conductive network is proposed to direct the uniform deposition of Li2S and alleviate the stress concentration.

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