Formation and Evolution of Solid Interphase Layers in an Interface-Modified Solid-State Li-Sulfur Battery

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The Li⁺ transport kinetics at the solid-solid electrode|electrolyte interfaces is crucial for the stable and durable performance of solid-state batteries (SSBs). A poor interface due to mechanical problems and/or (electro-)chemical instabilities will curtail the performance of such batteries. Herein, we present a detailed study on the interfaces of a lithium-sulfur (Li–S) SSB with a Li anode, Li-garnet (LLZO) solid electrolyte (SE), and a sulfur-carbon composite as cathode. Interlayer gels based on ionic liquid were introduced to improve the interfacial properties of the system. For Li symmetric cells, the strategy resulted in a decrease in cell resistance by about a factor of 5 and stable voltage profiles with low overpotentials (~300 mV at 0.4 mA cm⁻² after 400 hours). Furthermore, the LLZO SE efficiently blocked the polysulfide shuttle to the Li anode. Due to the advantageous features of the design, an excellent electrochemical performance was obtained, where the Li–S SSB delivered an initial discharge capacity of ca. 1360 mAh g⁻¹ sulfur⁻¹ and ca. 600 mAh g⁻¹ sulfur⁻¹ after 100 cycles. The formation and evolution of the interphase layers was investigated through X-ray photoelectron spectroscopy (XPS), applying depth profiling techniques, and scanning transmission electron microscopy (STEM). The results revealed presence of amorphous interphase nanolayers with varying thickness on LLZO surface that contained organic and inorganic species.

Keywords: solid-state battery, solid-interphase layer, Li-sulfur, solid-electrolyte, interface characterization
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