Enhancing Interfacial Stability in Sulfide-Based Solid-State Batteries: Amorphous LLZTO Coatings for Nickel-Rich High-Voltage Cathodes

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Nickel-rich high-voltage cathodes necessitate the use of a protective layer to suppress chemical reactions and enhance stability in sulfide-based solid-state batteries. One potential protective layer is garnet-type $Li_{6.75}La_3Zr_{1.75}Ta_{0.25}O_{12}$ (LLZTO), known for its high Li-ion conductivity and low electron transferability. However, crystallizing LLZTO on the cathode surface requires high-temperature treatment (>700 °C), which results in element interdiffusion and detrimental interface degradation. In this study, we propose the application of an amorphous LLZTO coating on $LiNi_{0.5}Co_{0.2}Mn_{0.3}O_2$ cathode particles and investigate the interface stability in a fully charged (4.55 V vs. Li/Li^+) state using an argyrodite sulfide-based solid-state battery. Our research aims to address the significant challenge of improving interfacial stability in ASSB studies by exploring the use of an amorphous Li-La-Zr-Ta-O coating on NCM523 cathode particles within an argyrodite sulfide-based solid-state battery. The durability of the cell was evaluated using a floating test sequence. The amorphous Li-La-Zr-Ta-O coating demonstrates exceptional protection against oxidative decomposition of the sulfide solid electrolyte near the interface, effectively preventing an increase in charge transfer resistance observed in previous studies. However, it should be noted that thermal treatment above 300 °C resulted in thermally induced degradation of the LLTO/cathode interface. These findings suggest that amorphous LLZTO coatings offer a promising strategy to enhance the performance and safety of ASSBs. However, further research is required to address the thermal degradation issue.

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