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Real-time observation of rapid densification and degradation in flash sintered multilayer systems for battery materials

Content

With increasing demand for advanced energy storage, cost-effective and efficient production of all-solid-state batteries (ASSBs) is critical for sustainable, low-waste energy solutions. ASSBs offer enhanced safety, higher energy density, and improved durability due to their solid electrolytes. However, conventional fabrication methods are costly, energy-intensive, and often yield poor interfacial contact, limiting commercial scalability. Advanced sintering techniques such as spark plasma sintering (SPS), cold sintering (CS), and flash sintering (FS) have emerged as promising alternatives. Flash sintering, in particular, significantly reduces sintering times from hours to seconds and greatly lowers energy consumption, aligning with circular economy principles.

In this study, we explore the fundamental mechanisms of electrochemical flash sintering (EFS) in model multilayer all-solid-state architectures composed of a $\text{Li}_{1.4}\text{Al}_{0.4}\text{Ti}_{1.6}(\text{PO}_4)_3$ (LATP) electrolyte and $\text{Li}_3\text{V}_2(\text{PO}_4)_3$ (LVP):LATP composite electrodes sandwiching an LATP layer.[1,2] Using total scattering (Bragg + Pair Distribution Function), we spatially mapped structural evolution to capture the formation of hotspots, localized regions of intense Joule heating that drive particle cracking and phase decomposition on sub-second timescales. In-situ total scattering experiments performed with the newly commissioned ARC detector at the i15-1 beamline (Diamond Light Source) achieved millisecond temporal resolution, allowing direct observation of both local and mesoscale structural dynamics. These measurements revealed the rapid formation of a transient amorphous intermediate that crystallizes within seconds, providing new insight into metastable states previously inaccessible to observation. Together, these findings elucidate how localized electrochemical and thermal processes drive densification and interfacial reactions in materials relevant to solid-state batteries, enabling assessment of whether EFS can effectively improve interfacial contact in future ASSSB architectures. Our use of ultra-fast total scattering thus establishes a framework for understanding reaction fronts and transient interfaces in electrochemically driven sintering across broader functional materials systems.

References :

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