A Hybrid Electrolyte with 3D Bicontinuous Ordered Ceramic and Polymer Microchannels for All-Solid-State Batteries

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There are many challenges on the road to an all-solid-state battery with a lithium (Li) metal anode. One such challenge is identifying a solid electrolyte that combines sufficient ionic conductivity with suitable mechanical properties. It is essential that the solid electrolyte maintains contact with the solid electrodes while simultaneously inhibiting Li dendrites as well as be durable towards electrode volume changes and to external shock.

Significant attention has been paid to ceramic electrolytes due to their high Li-ion conductivity at room temperature. Nevertheless, finding a ceramic electrolyte that is stable with Li metal and inhibits the propagation of Li dendrites remains an active challenge. Additionally, the processing and manufacturing of ceramic electrolytes into sufficiently thin, highly dense, pinhole-free sheets, required for high specific energy and power devices is still in its early stages. The advantages of polymer electrolytes include good adhesion and contact to the electrodes. Nonetheless, their low conductivity at room temperature along with their limited capability of suppressing Li dendrite formation, are persistent drawbacks. [1] In recognition of this complementarity, efforts have been made to combine ceramic and polymer electrolytes to form composite electrolytes. [2-4]

We investigate a different approach, which gives full control of the ceramic-polymer ratio and 3D structure. In this work, we present a hybrid solid electrolyte, composed of 3D ordered bicontinuous ceramic and polymer microchannels, generated by 3D printing. The ceramic electrolyte endows the hybrid with high ionic conductivity at room temperature due to continuous pathways for ions through the ceramic phase, whereas the non-conductive polymer mitigates the brittleness of the ceramic, rendering the hybrid electrolyte more resilient to fracture. The conductivity of the hybrid is reduced by only the volume fraction of space that the non-conducting polymer occupies, demonstrating a well-sintered ceramic phase. Four different microarchitectures are investigated, with the gyroidal microarchitecture exhibiting the best properties, sustaining longer cycling in contact with Li metal electrodes than a dense ceramic disk. Electrochemical, mechanical, and microstructural characterisations of the hybrid are reported and a comparison analysis to a ceramic electrolyte disk is made.

References:

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