

# LI-TETRAALKYLAMMONIUM DUAL-CATION BOROHYDRIDES AS MELT-INFILTRATIVE SOLID-STATE ELECTROLYTES FOR 3D LITHIUM-ION MICROBATTERIES

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Three-dimensional solid-state lithium microbatteries (3D-SSLMB) offer the ability to simultaneously enhance the energy and power density of microbatteries, the potential to attain higher energy density, and improved safety, than liquid electrolyte-based lithium microbatteries. However, the full integration of 3D-SSLMB still remain elusive due to the high processing temperature of solid electrolytes that may alter the 3D electrode geometry.<sup>1</sup> In this context, melt-infiltrative solid electrolytes, that is, solid electrolytes with low melting points ( $< 200$  °C) that can be infiltrated into 3D electrodes and the interelectrode spacings of 3D-SSLMB without altering the 3D geometry, which then solidify on cooling, have arisen as a viable solution for the integration of 3D-SSLMB.<sup>2</sup> In this work, we report on a family of Li-tetraalkylammonium dual-cation borohydrides, prepared by ball-milling lithium borohydride with the corresponding tetraalkylammonium borohydride. The properties of the resulting dual-cation borohydrides were found to depend on the molar ratios of the ball-milled borohydrides and length of the alkyl chain, resulting in solid electrolytes that melt as low as  $< 50$  °C, have an activation energy as low as 0.29 eV and a room-temperature conductivity as high as  $4.46 \times 10^{-2}$  mS cm<sup>-1</sup>. Our work lays the foundation for a new class of hydride materials that could enable 3D-SSLMB as well as conventional lithium batteries, and may find other applications in the energy storage field.

## References

1. Hur, J. I., Smith, L. C. & Dunn, B. High Areal Energy Density 3D Lithium-Ion Microbatteries. *Joule* **2**, 1187–1201 (2018).
2. Xiao, Y. *et al.* Electrolyte melt infiltration for scalable manufacturing of inorganic all-solid-state lithium-ion batteries. *Nature Materials* (2021) doi:10.1038/s41563-021-00943-2.



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