ABSTRACT - ORAL

From Die-Pressed Prototypes to DLP-Printed Architectures: A Stepwise Route to All-Ceramic Solid-State Batteries

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All-solid-state batteries (ASSBs) represent a major breakthrough in electrochemical energy storage, offering superior safety, higher energy density, and extended cycle life compared to conventional liquid electrolyte systems [1]. Nevertheless, critical challenges persist at the electrode/electrolyte interface, where poor ionic contact and interfacial reactivity severely limit electrochemical performance [2].

Additive manufacturing technologies have opened transformative possibilities for fabricating complex, multi-material battery architectures with precise spatial control. In this work, we report a progressive strategy toward fully 3D-printed ASSBs based on a full ceramic system combining LTO ($Li_4Ti_5O_{12}$) as anode, LATP ($Li_{1.3}Al_{0.3}Ti_{1.7}(PO_4)_3$) as solid electrolyte, and NMC ($LiNi_{0.6}Mn_{0.2}Co_{0.2}O_2$) as cathode.

As a first milestone, we demonstrate the fabrication of a 3D-structured ASSB prototypes obtained by die-pressing, validating the processing and electrochemical viability of this LTO|LATP|Li^0 or Li^0|LATP|NMC combination. All powders were prepared/shaped via spray-drying to achieve optimized morphology and flowability. Rheological characterization using a Granudrum™ (measuring avalanche angle and cohesive index) confirmed powder suitability for dry deposition. Electrochemical impedance spectroscopy and cycling tests on die-pressed cells provide the first proof-of-concept for this innovative all-solid system.

Based on these results, we now develop a Digital Light Processing (DLP)-based printing approach for the full LTO|LATP|NMC combination. Ceramic suspensions are formulated into photopolymerizable resins with controlled viscosity, solid loading, and light penetration depth. This enables intricate 3D geometries inaccessible by conventional pressing, minimizing interfacial resistance through compositional gradients at each electrode/electrolyte interface [3].

This dual approach (from die-pressing to DLP) represents a scalable pathway toward next-generation 3D-printed ASSBs with enhanced interfacial engineering, architectural freedom and promising electrochemical performance.

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[3] Y. Liang et al., InfoMat 4 (2022) e12292

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