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Solid-state batteries: integration-ready separators, quantified interfaces and manufacturable microstructures

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All-solid-state batteries (ASSBs) are pursued to reduce flammability risk, widen operating-temperature windows, and enable cell designs that are difficult with liquid electrolytes, including lithium metal anodes and high-voltage cathodes.[1–3] The field has established multiple classes of fast ion conductors (sulfides, oxides, halides, polymers and hybrids).[1,3] The limiting factors are now predominantly system-level: (i) integrating solid electrolytes into separators and composite electrodes without losing transport,[4] (ii) controlling chemically and mechanically stable interfaces under electrochemical bias,[5] (iii) manufacturing microstructures that are reproducible at relevant thicknesses, loadings and stack pressures,[4,6] and (iv) developing predictive, experimentally anchored mechanistic understanding to enable transferable design rules across

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Nella M. Vargas-Barbosa was born and raised in the Caribbean Island of Puerto Rico, where she grew up and completed her Bachelor's in Chemistry at the Rio Piedras Campus of the University of Puerto Rico. After completing her PhD in Chemistry at Penn State University, she moved to pursue an academic career in Germany that has included stays at the University of Marburg, the Max Planck Institute for Solid State Research and Helmholtz Institute Münster, part of Research Center Jülich. Since August 2023, she has been Chair of Electrochemistry at the University of Bayreuth and the Bavarian Center for Battery Technology (BayBatt). There she leads the iPEC lab (interfacial photoelectrochemistry lab), which focuses on studying (photo)electrochemical systems that have applications for energy generation and energy storage. The goals of the group range from elucidating the fundamentals of charge-transport at heterogeneous interfaces in model systems to more application-oriented projects characterizing solid–solid interfaces and interphases in solid-state batteries.

chemistries and architectures.[7–10] This special collection in *Energy Advances* focuses on these constraints and on approaches that connect materials' properties to device-relevant performance.

The contributions in this special collection align around three themes that currently govern ASSB performance: separator and composite electrolyte design, surface and interphase chemistry, and processing and microstructure control.[1,4,5] Here we highlight three contributions, representing these themes, and collectively addressing the major failure modes of solid-state cells: percolation loss in composites,[4] poorly understood interfacial chemistry,[5] and uncontrolled microstructure.[4,6]

Attaining a fast-conducting, hybrid solid state separator for all solid-state batteries through a facile wet infiltration method (Heuer *et al.*) targets a practical problem in ASSBs: the gap between intrinsic conductivity and effective conductivity in a separator that must be thin, mechanically intact and manufacturable.[11]

Hybrid separators are often proposed to combine the conductivity of inorganic electrolytes with the compliance and processability of polymers. The challenge is that inorganic particles can form disconnected domains, and interfacial resistance between phases can dominate.[4] Wet infiltration approaches are technically attractive because they can improve phase connectivity and reduce void fractions relative to dry mixing or simple casting. This paper by Heuer *et al.* is therefore relevant not because it introduces a new electrolyte chemistry, but because it addresses microstructure formation as the controlling variable.

From a design standpoint, the critical metrics for such separators are: (i) ionic conductivity at the separator thickness used in cells; (ii) stability of conductivity after densification and cycling; (iii) interfacial impedance against both anode and cathode composites; and (iv) mechanical response under stack pressure.[4] Hybrid separators also need to suppress electronic leakage and mitigate local current hotspots that promote interphase growth or lithium penetration.[2,5] The contribution by Heuer *et al.* sits directly in this integration space: it demonstrates a route to embed a high-conductivity phase within a polymer matrix in a way that is compatible with scalable processing, and it frames microstructure as the enabling mechanism for transport rather than relying on bulk numbers alone.

Decoding the $AlPO_4$ and LATP surface with a combined NMR-DFT approach (Valenzuela Reina *et al.*) addresses one of the most persistent limitations in solid-state systems: interfaces are often treated empirically because surface structures and reaction products are not identified with sufficient confidence.[12]

Interfaces in ASSBs impose multiple, coupled resistances: (i) chemical reaction layers with low Li-ion conductivity; (ii) space-charge regions that modify local carrier concentrations; (iii) mechanical damage that reduces true contact area; and (iv) microstructural heterogeneity that creates non-uniform current distribution.[5,9,10] For oxide electrolytes and interlayers (including LATP and phosphate-based coatings), interpretation of spectroscopy can become 'under-constrained': similar spectral features can be assigned to multiple candidate local environments, and small changes in termination or hydration can shift signatures.[5]

The combined NMR–DFT workflow is important because it provides a controlled route from spectral features to specific local structures and terminations.[9,10] In practical terms, this matters for selecting and designing interlayers (*e.g.*, phosphates such as $AlPO_4$), defining processing conditions that stabilise desired terminations, and anticipating how surfaces evolve during contact formation and early cycling.[5,9,10] The work by Valenzuela Reina *et al.* also exemplifies how modelling is most useful in ASSBs: not as an abstract screening tool, but as a constraint that links measurement to mechanism, as also previously demonstrated for different battery materials systems.[7–10] As interface resistance is frequently the dominant impedance component, improving the certainty of surface assignments is directly actionable for materials selection and engineering.[7]

Microreactor assisted soft lithography of nanostructured antimony sulfide thin film patterns: nucleation, growth and application in solid state batteries (Chun *et al.*) focuses on manufacturing control, which is routinely underestimated in ASSB design.[13]

Thin-film and patterned architectures are not merely academic demonstrations. They provide controlled geometry for probing transport and interfacial processes and can be relevant to microbatteries or to engineered interlayers where thickness and morphology must be defined.[6,14] In solid-state systems, microstructure

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determines: (i) diffusion lengths; (ii) interfacial area (and therefore reaction kinetics and impedance); (iii) stress distribution during cycling; and (iv) contact continuity under pressure.[15] Uncontrolled nucleation and growth typically lead to roughness, porosity and discontinuities that dominate resistance and failure.

Microreactor-assisted soft lithography offers a method to control nucleation and growth kinetics while patterning nanostructured films. The contribution by Chun *et al.* is valuable because it treats pattern formation as a process with quantifiable steps (nucleation, growth and resulting morphology) and connects the fabricated structures to solid-state battery use cases. For the field, this supports a broader position: ASSB performance often hinges on whether the manufacturing route can consistently deliver the intended morphology and interfaces at scale.[15] Processing is therefore part of the functional design space, not an afterthought.

Taken together, contributions in this special collection reinforce several points that are increasingly supported across the ASSB literature:

1. Effective conductivity is microstructure limited. Composite and hybrid electrolytes require continuous percolation of the fast phase and minimisation of voids and high-resistance phase boundaries. Processing routes that reliably control connectivity can outperform nominally superior chemistries that cannot be integrated.

2. Interface characterisation must be mechanism-led and cross-validated. Electrochemical stability windows derived from bulk thermodynamics are necessary but insufficient; surfaces and interphases form under bias and mechanical constraint. Reliable interpretation requires constrained analysis (*e.g.*, NMR interpreted *via* DFT), otherwise interphase design remains empirical.

3. Manufacturability is a primary constraint. Reported performance depends strongly on stack pressure, thickness, areal loading, contact formation and microstructure uniformity. Thin-film and patterned methods provide useful control and can be directly relevant to engineered interlayers or microdevices.

4. Benchmarking needs discipline. Without consistent reporting of cell configuration, pressure history, interfacial preparation, and impedance fitting protocols, it is difficult to compare results or identify genuine improvements. This is particularly critical when claims depend on suppressing impedance growth rather than increasing initial conductivity.

Outlook

ASSBs will be judged by stability under realistic cycling conditions and practical formats. The near-term technical priorities are: (i) separators and composites that retain transport at manufacturable thickness, (ii) interfaces with predictable and low-resistance interphases, and (iii) processing routes that deliver consistent contact and microstructure across larger areas and higher areal loadings. The contributions in this collection address these issues directly by focusing on integration pathways, rigorous surface interpretation and controllable manufacturing. We thank the authors and reviewers for their work and the editorial office for coordinating this collection.

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