

Studies on Advanced Solid Electrolytes for Solid-State Metal Batteries

Submitted By: **Kuntal Ghosh**

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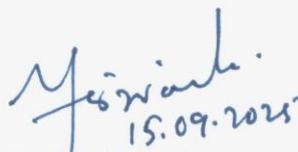
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Abstract

Deploying electric vehicles (EVs) is now becoming essential rather than a choice due to reducing environmental pollution and a smaller carbon footprint. EV batteries demand high energy and powder densities for long-range drive, high acceleration, quick charging operation, and better user safety. Conventional lithium-ion batteries are inadequate to fulfil these requirements due to the presence of flammable liquid electrolytes and poor compatibility with lithium metal. Solid-state lithium metal batteries (SSLMBs) emerged as promising candidates comprising solid electrolytes (SEs) with the potential to offer higher energy/power density and safety. Among various SEs, garnet-based $\text{Li}_7\text{La}_3\text{Zr}_2\text{O}_{12}$ (LLZO) has been the most researched SE across the globe due to its excellent ionic conductivity, wide electrochemical window, and chemical stability with Li metal. Yet, its practical deployment is restricted by the large grain boundary (GB) resistance, poor interfacial stability, and uncontrolled dendrite growth. My doctoral research addresses these challenges through a systematic and multi-scale engineering method. This includes developing a scalable synthesis, interface design, microstructural tailoring, dual-therapy introduction, and composite integration. An aqueous combustion synthesis route using alanine fuel was optimized to produce pristine cubic LLZO with a neck-fused structure. Interfacial instability was mitigated via surface fluorination (NH_4F) and a solvated ionic liquid infusion into the pristine LLZO matrix. This approach suppressed interfacial resistance, increased the critical current density up to $1.45 \text{ mA}\cdot\text{cm}^{-2}$ and delivered stable Li plating/stripping beyond 1200h with minimal overpotential (65 mV vs. 290 mV). Microstructural engineering via bio-templating derived plate-like and 3D interconnected LLZO architectures that reduced GB resistance, delayed dendrite growth and delivered excellent performance for more than 500 cycles in Li/LiMn₂O₄ full cell configuration. An innovative dual-therapy approach was introduced with NiO as a sintering aid (solid therapy) and SIL (liquid therapy) in doped LLZO. This synergistically enhanced densification up to 90%, minimized liquid uptake, and impeded the dendrite growth via forming a K_2NiF_4 -type lithiated impurity at the GBs. To offer the scalable integration of garnet electrolyte, LLZO fillers were incorporated into the cellulose matrix, dual-polymer composites, and hybrid paper-polymer scaffolds to improve conductivity, flexibility, and mechanical durability. Finally, exploratory extensions showed the adaptability of liquid therapy to $\text{Na}_3\text{Zr}_2\text{Si}_2\text{PO}_{12}$ (NZSP) solid electrolyte for sodium metal batteries and probed unconventional Al/S co-doping in LLZO for structural tuning. Together, the overall work develops a comprehensive roadmap for garnet electrolytes to address the various challenges and advance the practical realization of SSLMBs.

Kuntal Ghosh 15/09/2025

Kuntal Ghosh
CSIR-CGCRl, Kolkata 700032


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Dr. Mir Wasim Raja (Supervisor)

Date with Official Seal

डा. मीर वसीम राजा / Dr. Mir Wasim Raja

प्रमुख वैज्ञानिक / Principal Scientist

ऊर्जा सामग्री और उपकरण विभाग / Energy Materials & Devices Division

सीएसआईआर-केन्द्रीय काँच एवं सिरामिक अनुसंधान संस्थान

CSIR-Central Glass & Ceramic Research Institute

विज्ञान एवं प्रौद्योगिकी मंत्रालय, भारत सरकार

Ministry of Science & Technology, Govt. of India

196, राजा एस. सी. मुल्लिक रोड / 196, Raja S.C. Mullick Road

कोलकाता / Kolkata-700 032