

## Tailoring lignin polymer electrolytes for potassium-ion batteries

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The increasing demand for sustainable and cost-effective energy storage solutions has driven significant interest in potassium-ion batteries (KIBs) for stationary applications. Despite the obvious advantages as resource abundance and low costs, K-based systems face considerable challenges due to high reactivity and large ionic radius of K<sup>+</sup>. These factors result in the formation of a poor conductive and thick initial solid electrolyte interphase (SEI) and pronounced mechanical stress due to electrode deformation during cycling. AFM imaging of K-based cells at open circuit voltage reveals a highly irregular surface morphology, dominated by potassium oxides that promote dendrite growth. However, our findings indicate that through progressive decomposition of electrolyte and oxides, the passivation layer undergoes a remarkable transformation, decreasing its resistance by up to four orders of magnitude—ultimately achieving lower resistance than Li-based systems. This evolution necessitates the design of polymer electrolytes capable of adapting to both the chemical and mechanical changes at the electrode-electrolyte interface. Gel polymer electrolytes (GPEs) emerge as a promising solution, offering both mechanical robustness and flexibility. The smaller Stokes radius of K<sup>+</sup> allows for the use of thicker GPEs, which can provide higher Young’s modulus to suppress dendrite growth while preserving elasticity to accommodate dynamic interfacial changes. In this work, we present two innovative GPE formulations tailored for KIBs. The first GPE prioritizes mechanical strength, featuring a high storage modulus in the swollen state and thickness exceeding 200 μm to effectively mitigate dendrite formation. This system, based on pre-oxidized Kraft lignin crosslinked with PEGDGE1000 and plasticized with liquid electrolyte (KOx-PEGDGE), demonstrates ionic conductivity in line with literature-reported values and extends the half-cell lifespan beyond 2000 cycles without dendrite-related failure. The second GPE is a composite self-healing polymer electrolyte, comprising a UV-cured PCLDMA and UPyMA polymer network semi-interpenetrated by PEG, lignin-based fillers, and liquid electrolyte. The inclusion of lignin microparticles reduces crystallinity, yielding an exceptional ionic conductivity of 7×10<sup>3</sup> S cm<sup>-1</sup>—surpassing even liquid electrolytes. This GPE (G-PUL) exhibits an extended linear viscoelastic region, leading to improved plating and stripping profiles, lower overpotential, and significantly reduced charge transfer resistance over cycling. As a result, G-PUL delivers a high initial charge capacity of 125 mAh g<sup>-1</sup> with a C65-based cathode, maintaining 97.5% capacity retention over 1000 cycles.

These findings underscore the pivotal role of tailored polymer electrolytes in enhancing KIB performance. By strategically designing GPEs that balance mechanical strength, flexibility, and interfacial adaptability, we unlock new pathways for the advancement of K-based energy storage systems.

### References:

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