EIS and NMR Characterization of Storage & Degradation Mechanisms in Sodium Batteries

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Sodium batteries have emerged as a leading candidate to reduce dependence on lithium-based systems for sustainable energy storage. Their development in multiple configurations—such as metal oxide cathodes with hard carbon anodes, as well as anode-free architectures—has attracted growing interest due to sodium's abundance and cost advantages. Despite these benefits, large-scale commercialization remains constrained by limited specific capacity and poor cycle life. Overcoming these barriers requires a mechanistic understanding of sodium storage dynamics and the physicochemical origins of electrode degradation. Integrating electrochemical characterization with advanced structural probes is therefore essential for revealing the processes that govern performance.

In this work, we employ a combined electrochemical impedance spectroscopy (EIS) and ^{23}Na solid-state nuclear magnetic resonance (NMR) spectroscopy methodology to investigate sodium storage behavior and degradation pathways in sodium batteries. EIS enables quantification of interfacial resistances and charge-transfer kinetics [1], exemplified in Figure 1(a) for a representative hard carbon anode. Complementarily, NMR provides atomic-scale insights into the local coordination environments of sodium ions, as illustrated in Figure 1(b) for distinct sodium species in hard carbon [2].

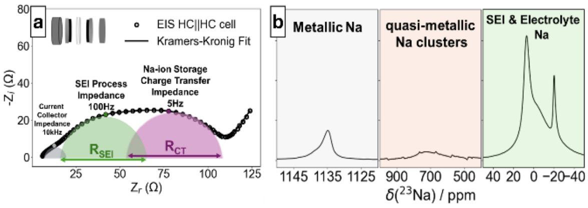

Figure 1. a) Nyquist plot of EIS response of hard carbon symmetric cell demonstrating SEI and charge transfer response, b) Solid state NMR spectra of different Na local environments in hard carbon.

We systematically examine the influence of electrolyte composition, electrode material, and state of charge on storage mechanisms and degradation pathways. EIS measurements performed under varying conditions—including temperature, cell geometry, and state of charge—enable precise evaluation of charge transfer processes and interfacial phenomena. Local structural changes during electrochemical cycling are monitored using a combination of in situ and ex situ NMR spectroscopy. Our results reveal critical correlations between structural evolution and electrochemical performance, providing design principles for improved material architectures.

In addition, we investigate interfacial behavior and plating/stripping characteristics of metallic sodium electrodes in anode-free cell configurations. Figure 2 presents the impedance response of a Cu||Na cell in different electrolyte environments. A two-order-of-magnitude decrease in impedance is observed when transitioning from carbonate-based to ether-based electrolytes, indicating the high reactivity of sodium and the associated formation of thick, heterogeneous SEI in carbonate systems. Ether-based electrolytes demonstrate better compatibility with sodium anodes, highlighting their potential for enabling stable and efficient Na anode-free designs.

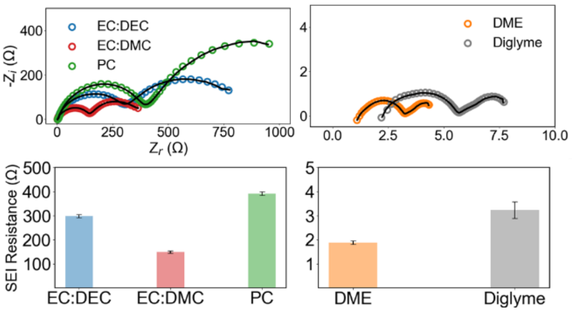

Figure 2. Nyquist plot of EIS response and fitted SEI resistance values of Na||Cu cell at different electrolyte envioirenments

These insights inform the design of optimized materials and cell architectures capable of delivering both high capacity and extended cycle life in sodium batteries.

Acknowledgment

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References

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[2] J. M. Stratford et al. J. Am. Chem. Soc., 143, 35, (2021) 14274–14286

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