High-Performance Aqueous Batteries Utilizing Low-Cost and Sustainable Bisalt Electrolytes

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Lithium-free aqueous batteries (LFABs) offer a promising alternative to conventional lithium-ion batteries (LIBs) for large-scale energy storage, addressing safety, material scarcity, and geopolitical issues associated with LIBs. Utilizing water-based electrolytes, enhances safety, affordability, and environmental friendliness. Substituting Li with readily available alternatives, such as Na, K, Mg, and Ca, helps overcome material supply concerns. However, the narrow electrochemical stability window of water, as well as capacity degradation due to electrode dissolution restrict the energy density and cycle-life of these batteries. In many cases, these challenges are addressed by using extremely highly concentrated electrolytes like 13 m $NaClO_4$ [1], 22 m KCF_3SO_3 [2], or 21 m LiTFSI [3]. Whereas this approach mitigates water splitting and active material dissolution by significantly lowering the amount of free water within the electrolyte, it is also associated with high consumption of expensive and potentially hazardous salts.

In our recent work, we present a novel electrolyte design strategy that employs only cost-effective and safe salts at lower concentrations. Through a systematic investigation of cation species' influence on the stability and electrode potential of the copper hexacyanoferrate (CuHCF) cathode and the polyimide anode, we optimized the electrolyte composition for enhanced cell voltage and cycling stability.
Our proposed LFAB configuration consists of a $KCuFe(CN)_6$ cathode, a poly-(naphthalene four formyl ethylenediamine) (PNFE) anode, and a 1.8 m $MgCl_2$ + 1.8 m KCl aqueous electrolyte. This optimized LFAB exhibits an impressive energy density of 48 Wh kg^{-1} with an efficiency of 95 %. Furthermore, it demonstrates a remarkable capacity retention of 70 % at 50 C, making it suitable for supercapacitor applications. We believe that by utilizing readily available and inexpensive salts in a safer and environmentally benign electrolyte, our work represents a significant step toward practical applications of LFABs for large-scale energy storage[4].

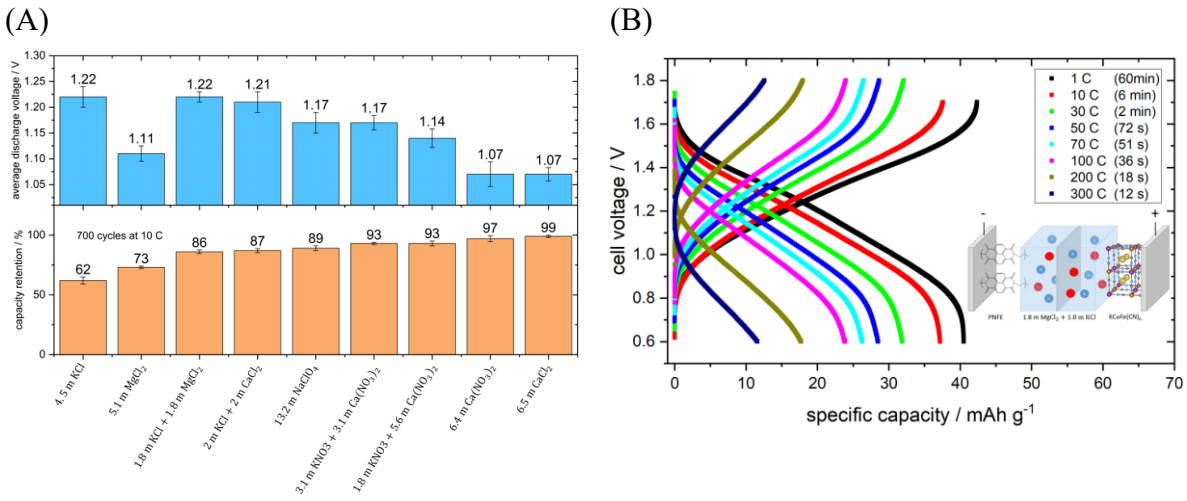

Figure 1: . (A) Comparison of the cell voltage and cycling stability of the CuHCF vs. PNFE full cell with different electrolytes. (B) galvanostatic charge-discharge curves of the CuHCF|1.8 m $MgCl_2$ + 1.8 m KCl|PNFE battery at several current densities.

References

[1] Bu et al., *Journal of Materials Chemistry A* 7.13 (2019): 7541-7547.
[2] Suo et al., *Science* 350.6263 (2015): 938-943.
[3] Jiang et al., *Nature Energy* 4.6 (2019): 495-503.
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