

FREE ARTICLE

Understanding and Optimizing Aqueous Viologen Bromide Redox-Enhanced Electrochemical Capacitors

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Abstract

Research in electric double layer capacitors (EDLCs) and rechargeable batteries is converging, producing devices known as pseudocapacitors. Pseudocapacitors store energy electrochemically, utilizing fast and reversible faradaic redox reactions at the interface between high surface area electrodes and an electrolyte. The result is a technology that delivers a long cycle life, high specific power, and moderate specific energy. One frontier in pseudocapacitor research is redox-active electrolytes, which replace the traditional solid redox-active materials with soluble redox couples.

Here we have identified viologen bromide salts as promising aqueous redox-active electrolytes for redox-EDLCs.[i] During charging, Br^- is oxidized to Br_3^- at the positive electrode and the viologen dication (V^{2+}) is reduced to the stable monocation radical ($\text{V}^{+\bullet}$) at the negative electrode (Figure 1). The system shows unusually high coulombic efficiency and low self-discharge rates for an aqueous redox-supercapacitor. This was initially attributed to strong adsorption of the Br_3^- and $\text{V}^{+\bullet}$ to the activated carbon electrodes, but we now present a more detailed analysis that confirms the behavior is attributable to two electroprecipitation

mechanisms. In these mechanisms, each ion acts as a charge-storing redox couple at one electrode and as a complexing agent at the other electrode. The processes are highly reversible and cells show negligible capacity fade even after 20,000 cycles. The devices use conventional activated carbon electrodes, and because crossover is not a concern and self-discharge is suppressed, a simple cellulose separator is sufficient and ion-selective membranes are not required.

Heptyl viologen devices achieve specific energy densities of 11 Wh/kg (normalized to the mass of both electrodes and electrolyte), significantly higher than commercially available EDLCs (Figure 2). Using methyl viologen increased specific energy to 14 Wh/kg, but at the expense of cycling stability. By studying the operating mechanisms and synthesizing a large series of viologens with different alkyl substituents we have now identified viologens that deliver both the stability of heptyl viologen and the specific energy of methyl viologen. Finally, a simple electrochemical device model was developed to understand the behavior of the system and predict the maximum theoretically achievable performance (Figure 3). The results model experimental data well and suggest that significant performance improvement is possible, making this technology an exciting candidate for applications like automotive engine start-stop and grid-scale energy storage.

[i] S.-E. Chun, B. Evanko, X. Wang, D. Vonlanthen, X. Ji, G. D. Stucky, and S. W. Boettcher, "Design of aqueous redox-enhanced electrochemical capacitors with high specific energies and slow self-discharge," *Nat. Commun.*, vol. 6, p. 7818, 2015.

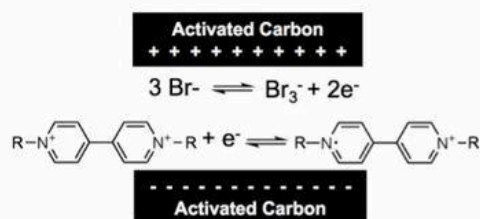


Figure 1. Simplified electrochemical reactions for energy storage in viologen bromide redox-enhanced electrochemical capacitor.

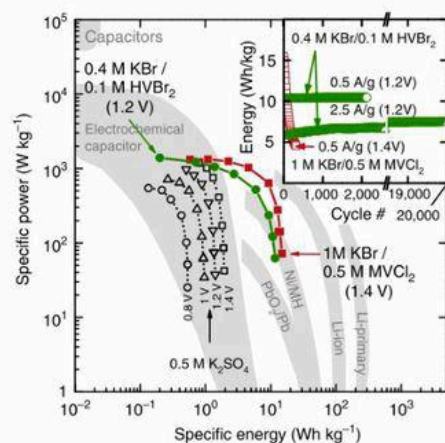


Figure 2. Ragone plot comparing the specific power and specific energy of viologen bromide cells with conventional EDLCs. Inset: Cycling stability of methyl viologen and heptyl viologen devices.

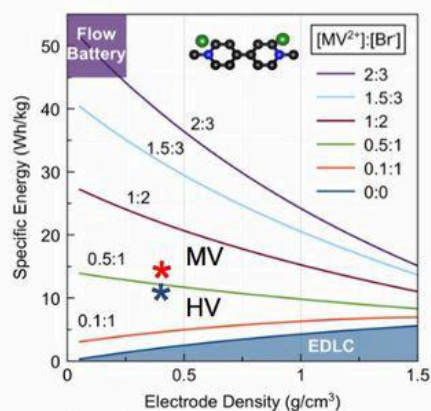


Figure 3. Electrochemical model predictions for maximum theoretical specific energy as a function of electrode density and redox couple concentration (in mol l^{-1}). Normalization is to combined mass of electrodes and electrolyte.

Figure 1

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