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Effect of Electrolyte Cation on the Charge Storage Mechanism of Manganese Dioxide for Electrochemical Capacitors

by

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Abstract

The performance of manganese dioxide ($\gamma\text{-MnO}_2$) as an electrode material in electrochemical capacitors is examined in aqueous electrolytes of 0.5 M Li_2SO_4 , Na_2SO_4 , K_2SO_4 or MgSO_4 . Performance and mechanistic analysis is conducted using cyclic voltammetry and step potential electrochemical spectroscopy. The choice of cation affects electrode performance, and this is explored in terms of the bare and hydrated ion size, and their interaction with the $\gamma\text{-MnO}_2$ surface and bulk structure. At fast sweep rates charge storage is dominated by double layer formation on the geometric (outer) electrode surface, in which case the smaller hydrated cations are best. At intermediate sweep rates charge storage occurs via double layer charge storage on the porous surface and diffusional (or pseudo-capacitive) processes, and is determined by the affinity of the cation to its hydration shell. Ions with a lower hydration shell affinity are more likely to undergo a solvation shell rearrangement or desolvation in order to access the smaller pores or undergo insertion into the bulk of the electrode. At slow sweep rates, charge storage is via diffusional capacitance. This mechanism is similarly dependent on the ability of an ion to undergo desolvation to be inserted into the bulk of the electrode.

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Keywords: electrochemical capacitors; pseudo-capacitance; manganese dioxide

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