Exploiting solvated Mg²⁺ ions in layered transition metal sulfide for fast Mg storage

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Rechargeable magnesium batteries (RMBs) have attracted considerable attention owing to the high volumetric capacity, dendrite free and highly abundant metal Mg anodes. However, the divalent ions (Mg²⁺) need to overcome much larger energy barrier than the monovalent counterpart (Li⁺) during intercalation. Plus a sluggish desolvation process at the cathodeelectrolyte interfaces, the RMBs usually suffer from slow kinetics. Presently, we proposed to intercalate solvated ions ([Mg(DME)₃]²⁺) rather than the bare Mg²⁺ ions into the host, aiming at reducing the charge density of the multivalent ions and thus improving their mobility. Compared to other solvated Mg^{2+} ions intercalation been reported (e.g. $[Mg(H_2O)_x]^{2+}$ or MgCl⁻), the DME molecule is neither passivate to the anode nor corrosive to the cell which makes it more practical. The novel concept was verified using a layered MoS2 cathode. According to the thorough mechanism investigation, the intercalated cations into the host are exactly the same as that in the electrolyte, which means no extra solvation process needed during intercalation. With fast kinetics, the cell configuration exhibits improved RMB performances in terms of long-term cycling stability (>200 cycles) and high rate capability (47 mA h g⁻¹ at 0.5 A g⁻¹). In addition, we observed for the first time the reversible 2H to 1T phase transition during multivalent ions de-/intercalation, which also explains the charge storage mechanism of MoS₂ based cathode in RMBs. We believe that the idea to apply solvated Mg²⁺ ions have great potential to be extended to other cathode materials or other multivalent ions intercalation.

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