INTERLAYER CONTROL OF 2D MATERIALS FOR FACILE INTERCALATION OF MAGNESIUM CATIONS

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Magnesium rechargeable batteries potentially offer high energy density, safety, and low cost due to the ability to employ divalent, dendrite-free, and earth-abundant magnesium metal as the negative electrode. In addition to the ultrahigh capacity of Mg metal anode, maximum two-electron transfer per site of cathode materials can effectively enhance the capacity of the cathode. As a result, the energy density of magnesium rechargeable batteries can meet the requirements for large-scale energy storage including electric vehicles, overcoming the theoretical limits in state-of-the-art Li-ion batteries. Despite recent progresses, further development remains stagnated mainly due to the sluggish scission of magnesium–chloride bond and slow diffusion of divalent magnesium cations in cathodes. Here we report a battery chemistry that utilizes interlayer control of 2D materials for facile intercalation of magnesium or magnesium monochloride cations. Metal dichalcogenides, namely TiS_2 and MoS_2 were utilized as model compounds for the expanded 2D materials. Combined theoretical modeling, spectroscopic analysis, and electrochemical study reveal fast diffusion kinetics of magnesium cations in the expanded interlayer. Even faster intercalation and diffusion kinetics were observed for magnesium monochloride ions because of the alleviated electrostatic force between S anion and Mg cation, circumventing the energy barrier due to the scission of magnesium–chloride bond. Such battery demonstrates the reversible intercalation of 1.0 and 1.7 magnesium monochloride cations per titanium at 25 and 60 ^{o}C, respectively, corresponding up to ~400 mAh/g capacity based on the mass of titanium disulfide. The large capacity accompanies with excellent rate and cycling performances even at room temperature, opening up possibilities for a variety of effective intercalation hosts for multivalent-ion batteries. Additionally, this presentation will address pros and cons as well as the prospects of the interlayer-expanded materials, based on theoretical studies and experimental observations.

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