Fast Intercalation and Diffusion Kinetics of Magnesium Monochloride Cations in Interlayer-Expanded Titanium Disulfide

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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, 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 progress, 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 magnesium monochloride cations in expanded titanium disulfide [1]. Combined theoretical modeling, spectroscopic analysis, and electrochemical study reveal fast diffusion kinetics of magnesium monochloride cations without scission of magnesium─chloride bond. The battery demonstrates the reversible intercalation of 1.0 and 1.7 magnesium monochloride cations per titanium at 25 and 60 ℃, respectively, corresponding to ~400 mAh g^{-1} 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.

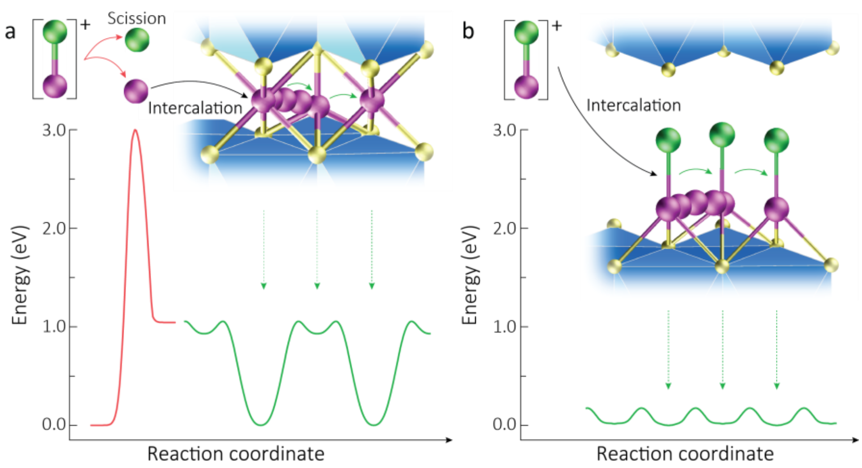

Figure 1. Energy diagrams for the intercalation and diffusion of Mg^{2+} and $MgCl^{+}$. (a) Typical intercalation of Mg^{2+}, which requires substantial activation energy of 3 eV and 1.06 eV for the intercalation and diffusion, respectively. (b) Intercalation of $MgCl^{+}$ that requires fairly low migration energy barrier of 0.18 eV. Mg and Cl atoms are shown as purple and green spheres, respectively.

References:

[1] H.D. Yoo, Y. Li, J. Lin, X. Qian, Y. Liang, Y.-S. Liu, J. Guo, Q. Ru, Y. Jing, Q. An, H. Wang, W. Zhou, S. Pantelides, Y. Yao, Nat. Commun. 8 (2017) 339.