

Magnesium solvate-based electrolytes for high voltage magnesium rechargeable batteries

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Magnesium rechargeable batteries (MRBs) are promising large-scale energy storage technologies, owing to the remarkable properties of magnesium metal. Many researchers have focused on and developed many different electrode and electrolyte materials, aiming at materialization of practical MRBs. Spinel-type transition metal oxide, MgM_2O_4 , are attracted much attention as high voltage cathode active materials; however recent theoretical and experimental studies suggested that these compounds require high operation temperature due to extremely large diffusion barrier of Mg^{2+} ions in the oxide-based lattice.^{1,2} We herein propose the highly stable electrolytes which are composed of magnesium-ether solvate and ionic liquids (ILs). The electrolyte solutions composed of typical TFSA based ILs and corresponding Mg salts are inactive electrochemically, probably due to strong association of Mg^{2+} and anions, making Mg-species presented in the IL-solutions neutral $\text{Mg}(\text{anion})_2$ and/or aggregate $[\text{Mg}(\text{anion})_n]$. This situation however can be solved simply by addition of ether into the solutions.³ In particular case, equimolar mixture of $\text{Mg}(\text{TFSA})_2$ and appropriate glymes dissolved in aliphatic ILs showed somewhat reversible Mg deposition/dissolution behavior, and this electrolyte achieved excellent anodic stability of > 4 V vs. Mg^{2+}/Mg even at 100 °C on Pt and Al electrodes. Such electrolyte is found to be compatible with high voltage MgM_2O_4 ($M = \text{Co}$ and Mn) while conventional $\text{Mg}(\text{TFSA})_2/\text{glyme}$ electrolyte solutions suffer from severe decomposition during charging. The effect of the ligand choice and chloride-based additive on the Mg deposition/dissolution behavior will also be presented.

References

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