

## Density Functional Theory Study on Pyrd-PVA-CN-Based Metal-Ion-Chelating Organogel Electrolyte

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The transition-metal-based cathode materials severely suffer from the dissolution of transition metals during repeated oxidation and reduction, which causes structural deterioration of cathode and subsequent electrodeposition on anode resulting in capacity decay. In this study, Pyrd-PVA-CN-based (Pyrd = pyrrolidone) metal-ion chelating organogel electrolyte is presented to solve the problems triggered by metal dissolution from Mn-rich oxide cathode materials. The density functional theory calculations suggest that the  $Mn^{2+}$  dissolved to electrolyte can be captured by the pyrrolidone; the  $Mn^{2+}$  chelation by the pyrrolidone was thermodynamically more favorable than the complete solvation of  $Mn^{2+}$  in carbonate electrolyte. The most stable metal chelating configuration was found to be the  $Mn^{2+}$  double coordinated to O of ester groups in the bridge between pyrrolidone and vinyl backbone and N of pyrrolidone hetero-ring. Interestingly, while  $Mn^{2+}$  coordinated to N, the -NH of pyrrolidone was flipped in the direction of increasing electric dipole moment. This dipole enhancement, which strengthened ion-dipole interaction, contributed to the strong chelating ability of the pyrrolidone.