

# Chelating Ligands as Electrolyte Additives to Modulate the Redox Potential of Organic Radical Batteries

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Poly(2,2,6,6-tetramethylpiperidin-1-oxy-4-yl methacrylate (PTMA) is one of the most promising cathode-active polymer materials in organic radical battery (ORB) with a redox potential of 3.6 V versus Li/Li<sup>+</sup>. To modulate the redox potential of ORB, modifications of polymer structure such as adding functional groups to the polymer chain or polymerizing with other conjugated polymer to form copolymers, are usually required. Herein, we report an electrolyte system using various concentrations of electrolyte salt and chelating ligand to regulate the redox potential of PTMA without changing the polymer structure. From the cyclic voltammetry of Li|0.1, 0.5, 1.0, and 2.0 M LiClO<sub>4</sub>-EC/DEC (1/1, v/v)|PTMA cells with 1, 2, and 4 equiv. of 12-crown-4 corresponded to LiClO<sub>4</sub>, respectively, the formal potential E<sub>1/2</sub> declines when increasing the LiClO<sub>4</sub> concentration in the electrolyte; however, a reverse change of E<sub>1/2</sub> is observed when increasing the equiv. of 12-crown-4 in the electrolyte. By applying different concentrations of LiClO<sub>4</sub> and 12-crown-4, the E<sub>1/2</sub> can be modulated between 3.557 to 3.855 V. The redox potentials calculated using density functional theory (DFT)/base set of B3LYP/6-31G in polarized continuum model (PCM) also shows the same trends in experimental results.

## References:

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