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In Situ Electrochemical Insight into Redox-active Macromolecular Radicals for Metal-free Batteries

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Abstract: Sociopolitical pressure towards elemental cobalt drives the demand for new energy storage materials. Specifically, Li-ion battery cathodes contain lithium cobalt oxide (LCO) or lithium nickel manganese cobalt oxides (LNCM), for which some cobalt is obtained from politically sensitive geographical regions or by means of child labor. One solution towards this challenge is the translation of organic or metal-free electrodes to replace of cobalt-containing cathodes. Here, macromolecular radicals as redox-active electrodes for metal-free batteries are presented. These polymers generally contain redox-active nitroxide radical groups that reversibly exchange electrons at rates much higher that of current metal oxide cathodes. This manifests as a higher power or a high charging rate. The current challenges for macromolecular radical batteries are to understand the redox mechanism, to increase the energy density in metal-free or aqueous conditions, and to consider a circular life cycle. Insight into the polymer's redox mechanism is provided using electrochemical quartz crystal microbalance with dissipation monitoring, in which mixed electron-ion-solvent transfer is quantified. This knowledge reveals why certain metal-free, aqueous electrolytes are well-suited to this polymer class. Lastly, an organic peptide battery that degrades on command into amino acids and byproducts provides a path forward toward recycling for a circular life cycle.