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Operando XAS Studies on Aqueous Zn-MnO2 Batteries

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Manganese-based (such as MnO2) cathodes have been attracting enormous attentions in aqueous Zn metal battery, thanks to its low-cost, suitable potential and considerable capacity. However, the electrochemical mechanism remains unclear, being ascribed to co-insertion/conversion with Zn2+, Mn2+, H+, or even precipitation of an intermediate products (Zn4SO4(OH)6·xH2O, ZHS), which is retrained by differing complicate structure of Mn in aqueous electrolyte and that might only be observable by real-time characterizations. Herein, operando XAS technique is used to probe the electronic and local structure of Mn and Zn during several consecutive cycles with the intention to shed some light on the underling the ambiguous mechanisms that govern the reactions. α-MnO2 are employed as the cathode in a 1M ZnSO4 + 0.2M MnSO4 electrolyte with a Zn metal anode. The XANES region of Mn K-edge shows a reversible shift of ca. 5 eV upon consecutive cycling, which is consistent with the formation of Mn (III) species upon reduction. Furthermore, the EXAFS indicates significant weakening of the nearest Mn-O shell at ca. 1.4 Å and strengthening of Mn-Mnegde in [Mn4+/3+O6] structures at ca. 2.6 Å during extended cycling. Meanwhile, Zn-K edge shows little change in all charging and discharging states. Hence, a mutual conversion reaction might be referred to between Mn (III) and Mn-ion (II) at the potential windows of 1.2-1.6 V.

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