



Contribution ID: 65

Type: Poster

Operando XAS Studies on Aqueous Zn-MnO₂ Batteries

Thursday, 8 September 2022 19:35 (20 minutes)

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-Mn edge 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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Session Classification: List of posters presented during the conference