Elucidating Redox Cycling Driven Transformation of Manganese Oxide Nanoparticles

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Mn (hydr)oxide nanoparticles (hereafter Mn oxides) play critical roles in contributing to electron transfer in energy and environmental systems, thus it is of great importance to understand the fate of Mn oxides during redox cycling. However, while Mn oxides show significant structural changes under redox cycles, the fundamental understanding of the structural transformation has remained puzzled. Here, using an electrochemical cyclic redox reaction, we demonstrate the direct transformation from layered Mn oxide (d-MnO_2) to tunneled Mn oxides. Both reduction and oxidation processes involved a two-step, one-electron transfer (i.e., Mn(Ⅳ) ↔ Mn(Ⅲ) and Mn(Ⅲ) ↔ Mn(Ⅱ)), and interlayer Mn(Ⅲ) was found to be critical in facilitating the structure transformation. We further revealed that the kinetics and electron flux of the cyclic redox reaction are key to the layer-to-tunnel structure transformation of Mn oxides, provided new insights for Mn redox reactions and explained the structural transformation of Mn oxides in nature and engineered systems.