Oxygen Electrocatalysts Based on Ordered Mesoporous Manganese Oxides with Different Oxidation States and Crystal Phases

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Ordered mesoporous manganese oxides with different crystal structures and oxidation states (Meso-MnO_2, Mn_2O_3, Mn_3O_4, and MnO) were prepared by nanocasting KIT-6 mesoporous silica. Oxygen electrocatalytic activity of the Meso-MnO_x was investigated in 0.1 M KOH. Meso-MnO_2 and Mn_2O_3 showed higher oxygen reduction reaction (ORR) and oxygen evolution reaction (OER) activities than meso-Mn_3O_4 and MnO. Contrary to the previous report that revealed low ORR and OER activities for β-MnO_2, relatively higher oxygen electrocatalytic activity was observed with Meso-MnO_2, which can be attributed to its slightly lower oxidation state below 4 (~3.9), and unique disordered (or mixed) crystal phase. Comparing the Meso-MnO and Mn_3O_4, the former showed better ORR activity than the latter, demonstrating the advantage of the mixed phase in Meso-MnO for facilitating oxygen electrocatalytic activity. *In-situ* XAS showed noticeable potential-dependent change in oxidation state for Meso-MnO_x catalysts, suggesting the oxygen electrocatalytic reactions invoke *in situ* phase transformation of the catalyst surface.