Abstract

The development of catalysts with high thermal stability is receiving considerable attention. Here, we report manganese oxide octahedral molecular sieve (OMS-2) materials with remarkably high thermal stability, synthesized by a simple one-pot synthesis in a neutral medium. The high thermal stability was confirmed by the retention of the cryptomelane phase at 750 °C in air. Mechanistic studies were performed by X-ray absorption near-edge structure (XANES) spectroscopy and *ex situ* X-ray diffraction (XRD) to monitor the change in oxidation state and the phase evolution during the thermal transformation. These two techniques revealed the intermediate phases formed during the nucleation and growth of highly crystalline cryptomelane manganese oxide. Thermogravimetric analysis, Fourier transform infrared spectroscopy (FTIR), time-dependent studies of field emission scanning electron microscopy (FE-SEM), and high-resolution transmission electron microscopy (HR-TEM) techniques confirm the formation of these intermediates. The amorphous phase of manganese oxide with random nanocrystalline orientation undergoes destructive reformation to form a mixture of birnessite and hausmannite during its thermal transformation to pure crystalline OMS-2. The material still has a relatively high surface area (80 m^2/g) even after calcination to 750 °C. The surfactant was used as a capping agent to confine the growth of OMS-2 to form short nanorods. In the absence of the surfactant, the OMS-2 extends its growth in the *c* direction to form nanofibers. The particle sizes of OMS-2 can be controlled by the temperatures of calcination. The OMS-2 calcined at elevated temperatures (400–750 °C) shows high remarkable catalytic activity for oxygen reduction reaction (ORR) in aqueous alkaline solution that outperformed the activity of synthesized solvent-free OMS-2. The activity follows this order: OMS-$2_{500\ °C}$ > OMS-$2_{750\ °C}$ > OMS-$2_{400\ °C}$. The developed method reported here can be easily scaled up for synthesis of OMS-2 for use in high-temperature (400–750 °C) industrial applications, e.g., oxidative dehydrogenation of hydrocarbons and CO oxidation.