Hierarchical Porous Carbon Spheres for High Performance Na–O_2 Batteries

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The ever-growing energy storage demands have generated a great deal of interest in exploring novel battery chemistries (Li–O_2, Li–S, Na–O_2, Na–S, etc.), with the aim of realizing practical energy storage devices having higher capacity and lower cost than today's lithium-ion batteries.[1, 5] Research interest in aprotic sodium-air (Na–O_2) batteries has surged recently because of their relatively high theoretical specific energy and particularly their uncompromised round-trip efficiency, in which carbon-based oxygen electrode plays a critical role.[6, 7] Towards unlocking the energy capabilities of Na–O_2 batteries, the major challenge is the rational design and fabrication of carbon-based oxygen electrode that can sustain the desired Na-O_2 electrochemistry (i.e., formation and decomposition of NaO_2) and maximize the capacity and rate capability.

In this work, we report that a hierarchical porous carbon sphere (PCS) electrode has outstanding properties to realize Na–O_2 batteries with excellent electrochemical performances, including high capacity and rate capability and long cycle life. The controlled porosity of the PCS electrode, with macropores formed between PCSs and nanopores inside each PCS, enables effective formation/decomposition of sodium superoxide (NaO_2) by facilitating the electrolyte impregnation and oxygen diffusion to the inner part of the oxygen electrode. In addition, the discharge product of NaO_2 was deposited on the surface of individual PCSs and had an unusual conformal film-like morphology, which can be more easily decomposed than the commonly observed micron-sized NaO_2 cubes in Na–O_2 batteries. Moreover, a combination of Coulometry, Fourier transform infrared spectroscopy (FTIR), X-ray diffraction (XRD) and *in situ* differential electrochemical mass spectrometry (DEMS) provided compelling evidence that the operation of the PCS-based Na–O_2 battery was underpinned by the formation and decomposition of NaO_2 with a high reversibility and less parasitic side reactions.

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