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Charge storage mechanisms of manganese oxide nanosheets and N-doped reduced graphene oxide aerogel for high-performance asymmetric supercapacitors

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Although manganese oxide and graphene supercapacitors have been widely studied, their charge storage mechanisms are not yet clear. In this work, we have investigated the charge storage mechanisms of MnO2 nanosheets and N-doped reduced graphene oxide aerogel (N-rGOae) using in situ X-ray absorption spectroscopy (XAS) and electrochemical quartz crystal microbalance (EQCM). The in situ XAS carried out together with a chronoamperometry indicates that the oxidation state of manganese in the MnO2 electrode being charged increases from +3.01 at 0.0 V vs. SCE to +3.12 at +0.8 V vs. SCE and then returns to +3.01 for the discharge process. This is an origin why the MnO2 nanosheets can provide excellent capacity retention. The mass changes of the N-rGOae and MnO2-coated Au/TiO2 quartz crystal EQCM electrodes during the charge process gradually increases to 8.15 μg cm-2 and 10.34 μg cm-2, respectively. A finely tuned mass ratio of MnO2 to N-rGOae is 1.75 providing the maximum charge storage performance. A single coin-cell asymmetric supercapacitor (CR2016) of MnO2//N-rGOae provides a maximum specific capacitance of ca. 467 F g-1 at 1 A g-1, a maximum specific power of 39 kW kg-1 and a specific energy of 40 Wh kg-1 with a wide working potential of 1.6 V at 93.2% capacity retention after 7,500 cycles. The coin-cell supercapacitor can practically supply electricity to a spinning motor with a nominal voltage of 3 V for 1.45 min. The enhancement in the specific energy and specific power of the MnO2//N-rGOae supercapacitors can compete with the batteries in many applications.

KEYWORDS

Charge storage mechanism; Asymmetric supercapacitors; Birnessite-type potassium manganese oxide; N-doped graphene aerogel; In situ X-ray absorption spectroscopy

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