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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 MnO<sub>2</sub> 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 MnO<sub>2</sub> 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 MnO<sub>2</sub> nanosheets can provide excellent capacity retention. The mass changes of the N-rGOae and MnO<sub>2</sub>-coated Au/TiO<sub>2</sub> quartz crystal EQCM electrodes during the charge process gradually increases to 8.15  $\mu\text{g cm}^{-2}$  and 10.34  $\mu\text{g cm}^{-2}$ , respectively. A finely tuned mass ratio of MnO<sub>2</sub> to N-rGOae is 1.75 providing the maximum charge storage performance. A single coin-cell asymmetric supercapacitor (CR2016) of MnO<sub>2</sub>//N-rGOae provides a maximum specific capacitance of ca. 467 F g<sup>-1</sup> at 1 A g<sup>-1</sup>, a maximum specific power of 39 kW kg<sup>-1</sup> and a specific energy of 40 Wh kg<sup>-1</sup> 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 MnO<sub>2</sub>//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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