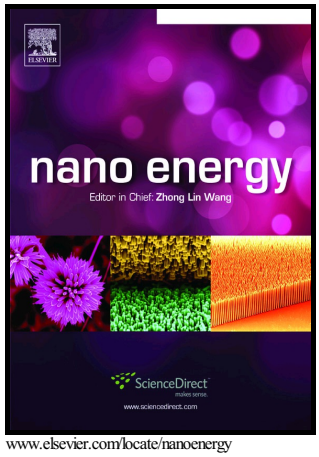

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Dynamic Observation of Reversible Lithium Storage Phenomena in Hybrid Supercapacitor Devices

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Abstract

Carbonaceous composites have attracted much attention as electrode materials for hybrid supercapacitors. Additionally, transition metal oxides, such as Co_3O_4, have high specific capacitance in the charging/discharging process. Here, we investigated the lithium storage mechanism of Co_3O_4/CNTs material via *in situ* transmission electron microscopy (TEM). Additionally, we analyzed the structure and composition of the anode material by high-resolution TEM, electron diffraction, energy dispersive spectroscopy (EDS) and electron energy loss spectroscopy (EELS). Using our unique *in situ* experimental setup that employs colloidal electrolyte, we elucidate two different mechanisms during operation, including the electrochemical reaction (battery-type) and ions intercalation (supercapacitor-type) of the electrode material. The cube-like Co_3O_4 nanoparticles were converted to Co nanograins dispersed in the Li_2O matrix after the first charging cycle. Subsequent cycles presented a reversible reaction between Co/Li_2O and CoO/Li_2O. Furthermore, the porous structure of the CNTs and conservation of the Li_2O matrix allow for the excellent ability to accommodate tremendous volume expansion, which enhances the life of hybrid supercapacitors. Our observations not only provide direct evidence of the