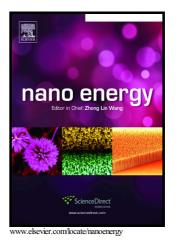
## Author's Accepted Manuscript

Dynamic Observation of Reversible Lithium Storage Phenomena in Hybrid Supercapacitor Devices

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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<sub>3</sub>O<sub>4</sub>, have high specific capacitance in the charging/discharging process. Here, we investigated the lithium storage mechanism of Co<sub>3</sub>O<sub>4</sub>/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<sub>3</sub>O<sub>4</sub> nanoparticles were converted to Co nanograins dispersed in the Li<sub>2</sub>O matrix after the first charging cycle. Subsequent cycles presented a reversible reaction between Co/Li<sub>2</sub>O and CoO/Li<sub>2</sub>O. Furthermore, the porous structure of the CNTs and conservation of the Li<sub>2</sub>O 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

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