

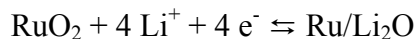
Fully reversible heterogeneous Li-storage in RuO₂ electrodes

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We present here the results of our investigation on the performance of RuO₂ as active electrode material for rechargeable lithium batteries. RuO₂ shows a high Li-storage capacity of 1120 mAh/g upon uptake of 5.6 Li, with 98% Columbic efficiency at the first discharge/charge cycle^[1].

Based on XRD, Raman spectroscopy and HRTEM measurements performed at various stages of discharge/charge processes, it is observed that full Li-uptake transforms polycrystalline RuO₂ into nanocrystalline composite of Ru and Li₂O with a grain size of 2-5 nm covered by a 5-10 nm surface solid/electrolyte interphase (SEI) layer. In the fully Li-extraction state, nanocrystalline RuO₂ formed again and the SEI layer disappeared completely.

The overall reaction can be written as:



This is analogous to the decomposition reaction observed in other transition metal oxides^[2] and fluorides^[3]. When the cycling is limited between 1.2–0.05 V, a reversible capacity of 150 mAh/g with a good cyclability is observed. We believe that this reversible Li-storage is caused by a heterogeneous interfacial storage process^[4].

Among the materials studied so far, it is worth mentioning that only RuO₂, allowed complete extraction of Li⁺ (nearly 100 % Coulombic efficiency) in the first cycle, while in other materials only 75 % or less Li⁺ can be extracted. We discuss in detail the cause for the complete extraction of Li⁺ in case of RuO₂ in comparison with few other materials such as TiF₃ and CoO in terms of the microstructure of M/LiX (X = O, F) nanocomposite and mass (e⁻, Li⁺ and O²⁻/F⁻) transport.

References

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3. H. Li et.al., *Adv. Mater.*, 15, 736 (2003).
4. J. Jamnik and J. Maier, *Phys. Chem. Chem. Phys.*, in press.