Correlation between constitution, microstructure and electrochemical behaviour of magnetron sputtered Li-Ni-Mn-Co-O thin film cathodes for lithium-ion batteries

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In comparison to well established cathode LiCoO₂ Li(Ni_{1/3}Mn_{1/3}Co_{1/3})O₂ shows a higher reversible theoretical capacity (290 mAhg⁻¹), better thermal stability and environmental The scientific goal improvement of electrochemical properties compatibility. of $Li(Ni_{1/3}Mn_{1/3}Co_{1/3})O_2$ can be achieved by the synthesis of nanocrystalline metastable phase thin films with a higher amount of Ni and Li to extend the operating voltage and capacity. Li-Ni-Mn-Co-O thin film cathodes have been deposited by non-reactive r.f. magnetron sputtering from a ceramic Li_{1.25}(Ni_{0.42}Mn_{0.21}Co_{0.37})O₂ target at various argon working gas pressures. The elemental composition was determined by inductively coupled plasma optical emission spectroscopy (ICP-OES) in combination with carrier gas hot extraction (CGHE). The microstructure of the films was characterized by X-ray diffraction (XRD) and by non polarized micro-Raman spectroscopy at room temperature. The electrochemical characterizations were carried out by cyclic voltammetry and galvanostatic cycling in Li-Ni-Mn-Co-O half cells against metallic lithium. The samples were annealed at different pressures from 10 mPa to 80 kPa for one hour at 600 °C in an argon/oxygen atmosphere (Ar: $O_2 =$ 80:20) allowing to adjust different microstructures. Correlations between process parameters, constitution, microstructure and electrochemical behaviour are discussed in detail. These films are promising candidates for the production of all solid state thin film batteries.