Constitution, microstructure and electrochemical behaviour of magnetron sputtered Li-Ni-Mn-Co-O thin film cathodes for lithium-ion batteries as a function of working gas and annealing pressure

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Compared to the commercial $LiCoO_2$ cathode material for lithium-ion batteries $Li(Ni_{1/3}Mn_{1/3}Co_{1/3})O_2$ has a good thermal stability, high reversible capacity (290 $mAhg^{-1}$), good rate capability and is environmentally friendlier. In this work Li-Ni-Mn-Co-O thin film cathodes have been deposited onto Si and stainless steel substrates by non-reactive r.f. magnetron sputtering from a ceramic $Li_{1.25}(Ni_{0.42}Mn_{0.21}Co_{0.37})O_2$ target at various argon working gas pressures from 0.2 Pa to 20 Pa. Coating thickness is about 1 µm. Composition and microstructure were investigated comprehensively. It was found that the elemental composition varies with argon working gas pressure which was determined by inductively coupled plasma optical emission spectroscopy (ICP-OES) in combination with carrier gas hot extraction (CGHE). The as-deposited films are nanocrystalline and show their highest grade of crystallinity in the range between 0.2 Pa to 0.5 Pa and at 7 Pa argon working gas pressure. The crystallinity of the films deposited at 0.5 and 7 Pa argon working gas pressure is increased by annealing in a furnace. The samples were annealed at different pressures from 10 mPa to 80 kPa for one hour at 600 °C in a argon/oxygen atmosphere ($Ar:O_2$ = 80:20). The microstructure of the films varies with annealing gas pressure and was characterized by X-ray diffraction (XRD) and by unpolarized micro-Raman spectroscopy at room temperature. Electrochemical characterizations of as deposited and annealed films were carried out by cyclic voltammetry and galvanostatic cycling in Li-Ni-Mn-Co-O half cells against metallic lithium. Correlations between process parameters, constitution, microstructure and electrochemical behaviour are discussed in detail. These films are promising candidates for manufacturing of all solid state thin film batteries.