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Phase selectivity in the synthesis of manganese oxides by plasma assisted pulsed laser deposition

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Manganese oxides have aroused considerable interest due to their technological applications in energy storage devices, catalysis, electrochemistry and supercapacitors. Manganese cations can exist in several oxidation states leading to the crystallization of a variety of oxides into different phases of which MnO, Mn_2O_3, Mn_3O_4, and MnO_2 are the most commonly known. These have different physico-chemical properties and one of the main challenges in the synthesis of such oxides is to have good phase control of the synthesized material in view of a specific technological application. In this work, we will be presenting results on the synthesis of pure manganese oxide phases, in thin film form, using plasma assisted pulsed laser deposition (PAPLD). We have determined the experimental conditions to grow one phase of manganese oxide as opposed to another, and we particularly emphasize the growth of highly crystalline manganese dioxide MnO_2 layers. After the oxygen plasma conditions are optimized, we show that operation at low pressure values (10 – 25 mTorr) with plasma activation of the oxygen gas induces a shift in the Mn-O phase diagram from Mn_2O_3 to MnO_2. X-ray diffraction, infra-red spectroscopy, raman scattering and atomic force microscopy analyses confirmed the synthesis of the rutile structure β-MnO_2 films having smooth surfaces with roughness values as low as 0.63 nm. While deposition at high pressure (250-500 mTorr) also promoted the formation of MnO_2, the structure of the resulting films consisted of an irregular intergrowth of two MnO_2 polymorphs and consequently poor crystalline quality as well as high surface roughness. The experimental conditions to synthesize Mn_2O_3 and Mn_3O_4 compounds have also been determined. A comparison of our results with those obtained using other film deposition techniques, such as atomic layer deposition, will be presented as well as perspectives of applications for our PAPLD films.

Keywords
MnO2
remote plasma
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XRD
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