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Recently, the chemically disordered multi-cation oxide class of materials called High Entropy Oxides (HEOs) has been widely explored due to their tunable functional properties because of their higher configurational entropy and to understand their fundamental phase formation [1]. These HEOs have shown potential applications as thermoelectrics [2], ionic conductors [3], electrocaloric materials [4], etc. Since these systems have multiple aliovalent cations in a single lattice, it necessitates the understanding of elemental distribution and structure of these novel oxides. This would help us understand how the HEOs navigate the configurational space and enable us to establish a correlation between structure and property.

In this study, we are investigating the influence of oxygen partial pressure during pulsed laser deposition on the structure, and chemical environment on the HEO thin films. $(Mg_{0.2}Ni_{0.2}Co_{0.2}Cu_{0.2}Zn_{0.2})O$, commonly known as 'J14', is the prototype HEO that has been widely studied [5]. We are currently exploring the seven component HEO system with the composition: $(Mg_{0.167}Ni_{0.167}Co_{0.167}Cu_{0.167}Zn_{0.167}(Cr, Mn)_{0.167})O$, referred to as 'J14MnCr'. The goal of this study is to understand the influence of addition of Cr and Mn to J14 on the structure and how the growth conditions modulate the structure and tune magnetic properties. Due to the presence of seven cations with some cations that can adapt multiple valences, it is necessary to probe the structural nuances and chemical environment in these systems. In this study, Scanning/Transmission Electron Microscope is used to characterize the HEOs as S/TEM allows us to probe the structure, composition, and valence variation at nanoscale regime.

We have probed the J14MnCr thin film grown at 5mTorr and 50mTorr oxygen partial pressure. We have observed from Selected Area Electron Diffraction (SAED) and dark field TEM experiments (as shown in Fig.1) that there is a change in the crystal structure in the films grown at 5mTorr vs. 50mTorr. At 50mTorr, the SAED reveals the formation of rock salt structure with additional secondary phase. The increase in oxygen partial pressure during synthesis has led to the nucleation of a new phase with a different structure which is speculated to influence the magnetic property. We have further performed STEM experiments(as shown in Fig. 2) and will be implementing unsupervised machine learning to detect the local structural variation. STEM-EELS experiments have also been performed to investigate the local changes in structure and to correlate it to changes in the valences of the cations and chemical environment. These S/TEM experiments enable us to understand the influence of processing conditions on microstructures and chemical environment, informing us about the structural nuances at the nanoscale and allowing us to tune the structure for desired properties [6].

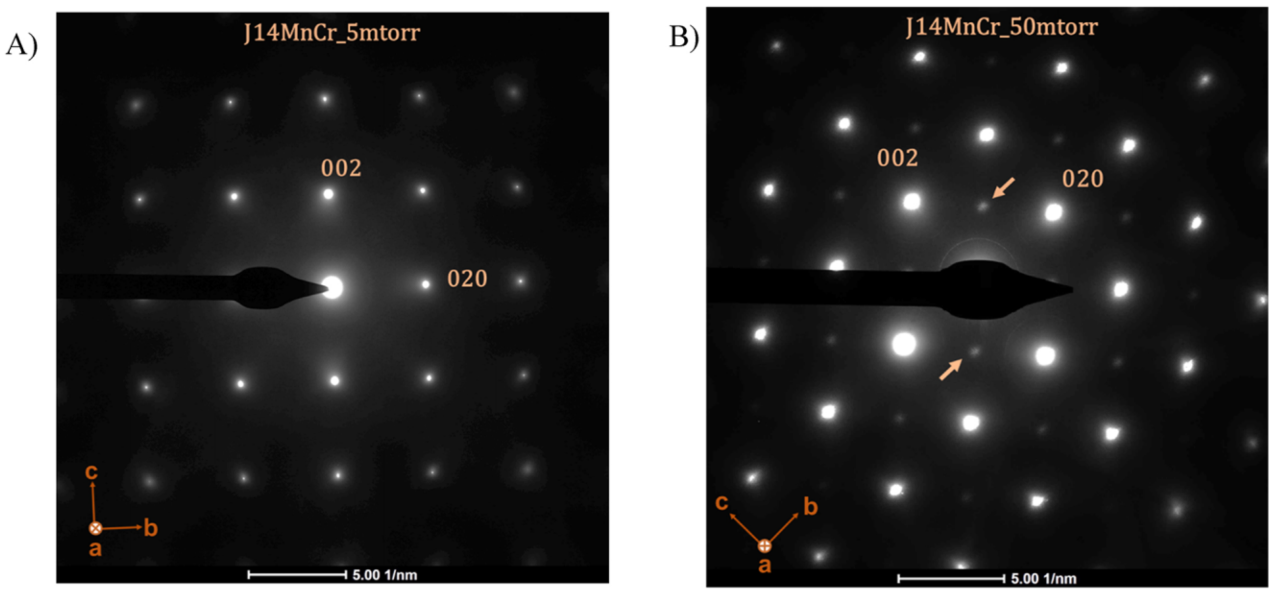

Fig. 1. Selected Area Electron Diffraction (SAED) pattern along [100] zone axis of J14MnCr thin film grown at A) 5mTorr B) 50mTorr oxygen partial pressure. In b) additional reflections marked by arrows can be observed apart from the reflections contributed by rock salt structure.

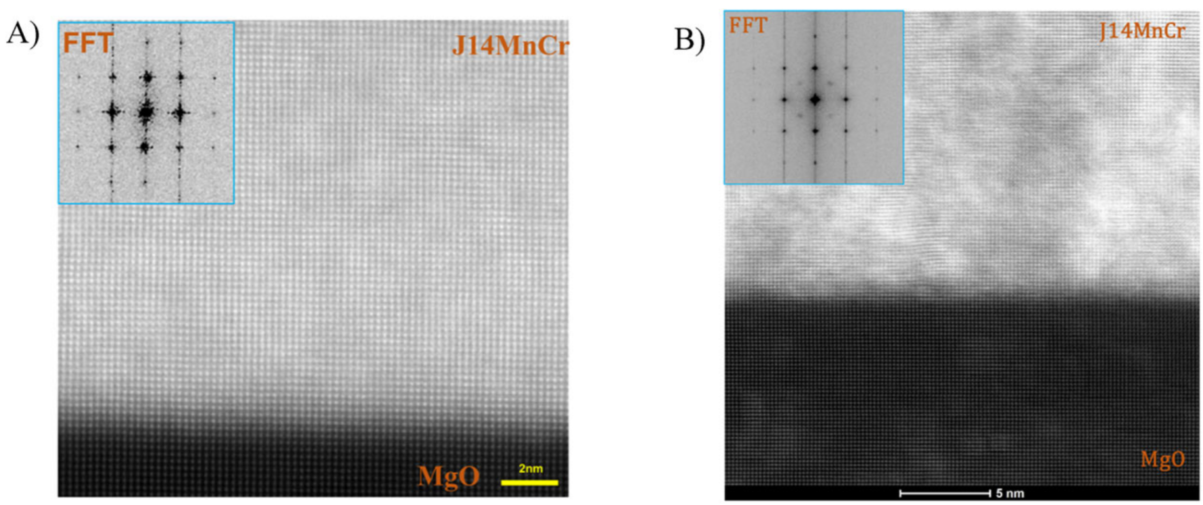

Fig. 2. ADF-STEM image along [100] zone axis of J14MnCr grown at a) 5mTorr b) 50mTorr oxygen partial pressure along with the FFT.

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