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Outline

This doctoral thesis entitled "Epitaxial thin film synthesis of high-entropy oxides using pulsed laser deposition" is composed of seven chapters in total. This work reports on a novel route to fabricate unexplored high-entropy oxides in an epitaxially stabilized thin film form using pulsed laser deposition (PLD).

In Chapter 1, the author reviews the high-entropy oxide materials and points out the limitation of conventional bulk synthesis to stabilize high-entropy oxides for new materials exploration. The author introduces why the author employs PLD as a non-equilibrium film growth method to fabricate high-entropy oxide epitaxial thin films for expanding the compositional and structural space of high-entropy oxides.

In Chapter 2, the author provides experimental details, including PLD techniques and thin film characterization methods.

In Chapter 3, the author focuses on increasing the upper-limit cation numbers (10) in high-entropy oxides. To incorporate as many as cations, the author designs a cubic perovskite (ABO_3: $(Ca,Sr,Ba)(Si,Ti,Cr,Mn,Fe,Co,Ni,Ge,Zr,Sn,Ce,Hf)O_3$) high-entropy oxide thin film containing 15 cations: 3 cations at the *A*-site and 12 cations in equiatomic ratios at the *B*-site. The author successfully grows the single-phase 15-cation-containing thin films with cubic perovskite structures on $SrTiO_3$(001) and LSAT(001) substrates. In addition, the author investigates the magnetic property of the thin film, suggesting spin/cluster-glass or canted antiferromagnetic states due to the complex oxidation states of six 3d transition metals (Ti, Mn, Cr, Fe, Co, Ni).

In Chapter 4, the author focuses on expanding high-entropy oxide films to anisotropic systems rather than cubic systems. The author prepares the epitaxial thin films of layered rock-salt ($LiMO_2$: $Li(Ni,Mn,Co,Fe,Cr,Cu)O_2$) high-entropy oxides containing six 3d transition metals. There have been no reports on high-entropy layered oxide materials in thin film form. The single-phase layered rock-salt $LiMO_2$ high-entropy oxide epitaxial thin films are successfully grown on Al_2O_3(0001) single-crystal substrates. In addition, the author demonstrates the electrochemical activity of the high-entropy oxide thin film as cathode materials for lithium batteries.

In Chapter 5, the author focuses on further expanding the structures of high-entropy oxide films into lower structural symmetry. The author expects to synthesize a monoclinic high-entropy oxide (Li_2MO_3: $Li_2(Ti,Mn,Fe,Co,Nb,Mo,Ru)O_3$) thin film by using PLD. However, instead of the expected monoclinic Li_2MO_3 structure, the author confirms that a spinel structure is formed for the high-entropy oxide film deposited on $SrTiO_3$(111) substrates. The author discusses the possible origins of spinel formation from the entropic points of view. For the future synthesis of monoclinic Li_2MO_3 films, the author proposes possible strategies according to configuration entropy.

In Chapter 6, the author states the importance of the non-equilibrium process and epitaxial thin-film form in high-entropy oxide synthesis. First, the author reveals that the non-equilibrium process suppresses the phase separation by comparing it to previous reports on the HEO bulk with the same structures. Second, the author discusses the epitaxial stabilization effect of single-phase high-entropy oxide structures by depositing perovskite, layered rocksalt, and spinel HEO films on glass substrates. As a result, each thin film is polycrystalline with impurity phases, suggesting that the epitaxial thin-film form contributes to stabilizing single-phase structures of high-entropy oxides and suppressing the phase separation.

In Chapter 7, the author summarizes and concludes this work, and describes the guidelines for future research on high-entropy oxide epitaxial thin films.

To sum up, this thesis demonstrates the epitaxial thin film growth through the non-equilibrium process as a powerful method to expand high-entropy oxide communities for new materials. The author deserves to receive a Doctor of Engineering from the Tokyo Institute of Technology.