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Substrate effects on the growth of epitaxial $Pb(Mg_{1/3},Ta_{2/3})O_3$ thin films using chemical solution deposition

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Abstract

The effect of various substrates on the formation of epitaxial $Pb(Mg_{1/3},Ta_{2/3})O_3$ (PMT) thin films has been investigated. $Pb(Mg_{1/3},Ta_{2/3})O_3$ thin films were prepared on $SrTiO_3$ (STO), $LaAlO_3$ (LAO), and MgO substrates by the chemical solution deposition (CSD) method. Microstructural evolution of PMT thin films as a function of annealing temperatures (650–750 °C/1 h) has been studied using the transmission electron microscopy, the scanning electron microscopy, and the X-ray diffraction (XRD). Epitaxial PMT thin films could be grown on STO and LAO substrates with an epitaxial orientation relationship of $[100](001)_{films}||[100](001)_{substrates}$. However, pyrochlore phase was mainly observed and no epitaxy nature was observed in PMT thin films on MgO substrates. The difference in the epitaxy nature is explained in terms of the difference in the lattice mismatch and the crystal structure.

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1. Introduction

Lead-based relaxor thin films have been focused because they have numerous potential device applications by utilizing the unique dieletric [1], electromechanical [2], and electro-optic [3] properties. There have been many efforts to grow highly oriented or epitaxial relaxor thin films for the purpose of using their superior physical properties compared to non-oriented polycrystalline thin films that suffer from the presence of high-angle grain boundaries that are detrimental to device performance. Epitaxial $Pb(Mg_{1/3},Nb_{2/3})O_3$ [4], $Pb(Mg_{1/3},Nb_{2/3})O_3–PbTiO_3$ [5,6], $Pb(Sc_{1/2},Nb_{1/2})O_3–PbTiO_3$ [1], and $(Pb_{1-x},La_x)(Zr_{0.65},Ti_{0.35})O_3$ [7] thin films have been prepared on single crystal oxide substrates using the pulsed laser deposition and the chemical solution deposition (CSD) methods.

Lead magnesium tantalate $Pb(Mg_{1/3},Ta_{2/3})O_3$ (PMT) is one of the relaxor perovskite compounds. The maximum dielectric constant and dielectric maximum temperature of PMT are 8500–8700 at −88 to −85 °C [8–10] which shows its suitability in applications ranging from multi-layer ceramic capacitors, transducers, and electro-optic devices. Recently, there have been a few reports on the preparation of PMT thin films. Polycrystalline PMT thin films on Si substrates were prepared by Lee et al. using the sputtering process [11] and epitaxial PMT thin films on $SrTiO_3$ (STO) substrates were prepared by our group using the CSD method [12,13].

The CSD method is of particular interest because of its simplicity and stoichiometry compared with other methods. Therefore, epitaxial ferroelectric thin films such as $PbTiO_3/SrTiO_3$ [14], $Pb(Zr_{0.5},Ti_{0.5})O_3/LaAlO_3$ [15], and $SrBi_2Ta_2O_9/SrTiO_3$ [16] have been prepared using the CSD method. The CSD method requires a greater understanding of the crystallization behavior to optimize the quality of epitaxial films. In our previous work, it was reported that CSD-derived epitaxial PMT thin films on STO substrates

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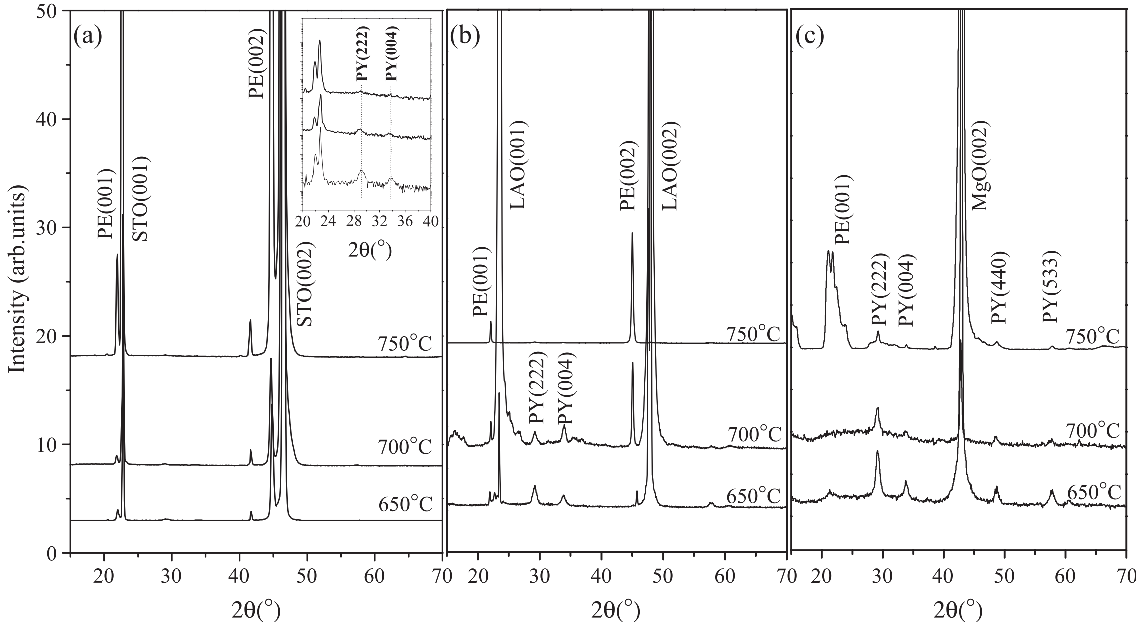

Fig. 1. X-ray diffraction $\theta-2\theta$ scans of $Pb(Mg_{1/3},Ta_{2/3})O_3$ thin films on (a) $SrTiO_3(001)$, (b) $LaAlO_3(001)$, and (c) MgO(001) substrates prepared from the double alkoxide precursor solution and annealed at various temperatures (650–750 °C) for 1 h in air. PE: perovskite, PY: pyrochlore.

are crystallized indirectly by the formation of a metastable fluorite phase first [13] as observed in $Pb(Zr,Ti)O_3/LaAlO_3$ system [15]. Epitaxial perovskite $Pb(Mg_{1/3},Ta_{2/3})O_3$ crystallites nucleate at the film/substrate interface and these nuclei grew across the interface and through the film toward surface by consuming the metastable nanocrystalline fluorite grains. However, details in the substrate effects such as the lattice mismatch and the crystal structure on the crystallization behavior and the epitaxy nature of PMT thin films have not been studied previously.

The purpose of the present work is to report the details on the preparation and characterization of epitaxial PMT thin films by the CSD method with attention to the effect of various substrates such as STO, LAO, and MgO on the phase evolution and the epitaxy mechanism.

2. Experimental procedure

$Pb(Mg_{1/3},Ta_{2/3})O_3$ thin films were prepared by spin coating on (001)-oriented STO, LAO, and MgO substrates (Materials and Technology, El Cerrito, CA) with a PMT alkoxide precursor prepared by a method reported previously [12], where Mg–Ta double-ethoxide solution was

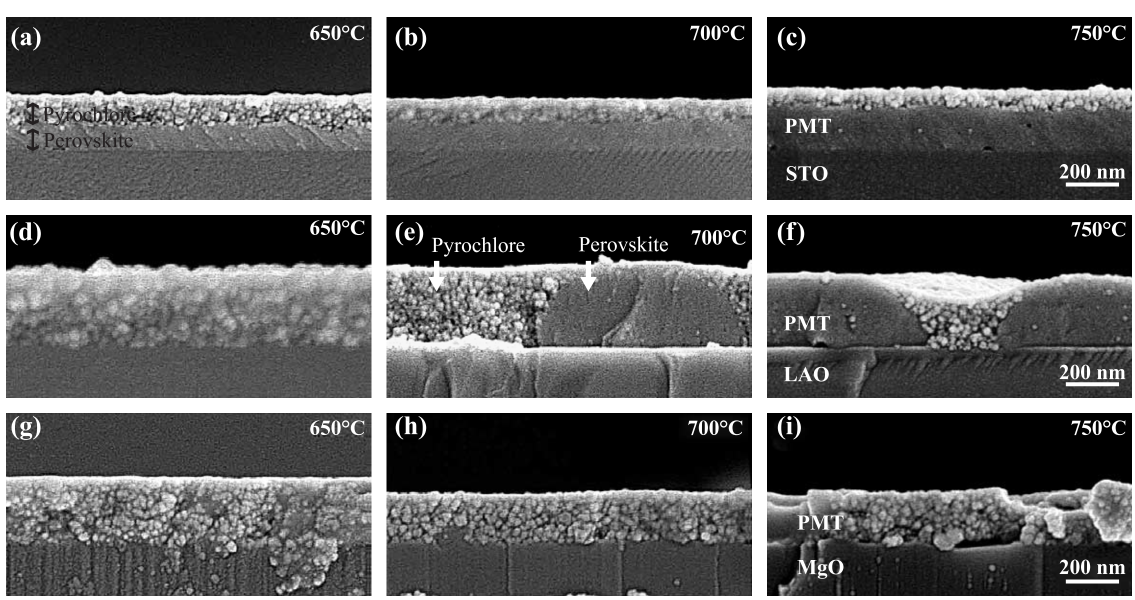

Fig. 2. Cross-sectional scanning electron micrographs showing the microstructural evolution of $Pb(Mg_{1/3},Ta_{2/3})O_3$ thin films on $SrTiO_3$, $LaAlO_3$, and MgO substrates as the annealing temperature changes from 650 to 750 °C.