

# Synthesis of Perovskite-type Oxide Thin Films by Hydrothermal Electrodeposition and Their Structural Characterization

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Recently, concerning energy saving, "Soft Solution Processing" has been focused in thin film synthesis. We have proposed a novel solution process for thin film synthesis, hydrothermal electrodeposition. Hydrothermal electrodeposition is a combined process of hydrothermal method with electrodeposition, and enable us to synthesize the oxide thin film at relatively low temperature. In this thesis, the functional ceramic, perovskite-type oxide thin films have been epitaxially grown on (100) oriented strontium titanate single crystal substrate.

We tried to synthesize thin film of perovskite-type oxides: lead titanate ( $\text{PbTiO}_3$ ), lead zirconate titanate ( $\text{PbZr}_x\text{Ti}_{1-x}\text{O}_3$ , PZT), potassium niobate ( $\text{KNbO}_3$ ), potassium tantalate ( $\text{KTaO}_3$ ), potassium niobate tantalate ( $\text{KTa}_{1-x}\text{Nb}_x\text{O}_3$ , KTN), and elucidate the optimum synthesis conditions of reaction temperature and KOH concentration of the reaction solution in binary system of  $\text{PbTiO}_3$  and  $\text{KTaO}_3$ , and control the film compositions in ternary system of PZT and KTN. Namely, the  $\text{PbTiO}_3$  and PZT thin films could be synthesized at  $90\sim 110\text{ }^\circ\text{C}$  in  $4\sim 10\text{ M}$  and  $6\sim 10\text{ M}$  KOH solution, respectively. The  $\text{KTaO}_3$  and KTN thin films also could be synthesized at  $90\sim 110\text{ }^\circ\text{C}$  in  $18.5\text{ M}$  KOH solution and the  $\text{KNbO}_3$  thin film could be synthesized at only  $110\text{ }^\circ\text{C}$  in  $18.5\text{ M}$  KOH solution. Among soft solution processes reported up to date, the hydrothermal electrodeposition enables us to synthesize perovskite-typed oxide film at the lowest temperature. Further, the crystalline structures and surface morphologies in the film were changed with the preparation conditions. For example, the PZT film prepared in lower KOH concentration consisted of the large and flat crystallite ( $\sim 500\text{ nm}$ ), while high KOH concentration brought about the small crystallite in size ( $100\text{ nm}$ ). Moreover, from the TEM observation of PZT thin film, we confirmed that PZT thin film consisted of the fine grains of  $2\sim 15\text{ nm}$  in size, suggesting that the growth mechanism of thin film synthesized by hydrothermal electrodeposition was the electrophoretical deposition of colloidal nuclei generated in the reaction solution.