Process Optimization of Lead-Free Piezoelectric Thin Films Derived via Chemical Solution Deposition

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Abstract

Due to legislature such as the Restriction of Hazardous Substances (RoHS) being passed to eliminate the use of lead in consumer devices, there is a pressing need for environmentally-friendly alternative materials to be developed. In this work, $(Bi_{0.5}Na_{0.5})TiO_3$-$(Bi_{0.5}K_{0.5})TiO_3$ (BNT-BKT) was explored at the morphotropic phase boundary composition, 80BNT-20BKT, as a possible replacement for lead zirconate titanate (PZT) in applications such as transducers, sensors, and actuators due to its promising piezoelectric behavior. Here these materials are explored in thin film embodiments. Major issues in perovskite thin film systems can be the cation volatility and high processing temperatures that can alter system stoichiometry and create defects that have a large impact on material properties. This is exacerbated by the very large surface to volume ratios in thin films. There are numerous variables that can also affect the ultimate film performance. The effect of heating and aging of the precursor solution on the thin film properties were investigated as follows; Solutions were prepared using a modified inverted mixing order process and were spin cast onto platinized silicon substrates. Solutions were heated overnight at 45, 55, and 65 °C in addition to an unheated solution to find the optimal temperature to produce enhanced electrical and mechanical properties. Heating the solution to 55 °C resulted in the best properties overall and had the lowest dielectric loss at 0.03. The dielectric constant and $d_{33,f}$ remained approximately equal at 650 and 70 pC/N, respectively, between all four solutions. Atomic force microscopy surface scans showed a decrease in root mean square roughness upon solution heating. Cross sectional images of the films via scanning electron microscopy showed a decrease in interlayer porosity from 17 to 5 pores/μm from the room temperature to the 65 °C heated solution. Solution aging was also explored to determine the stability of solutions; films were spun ranging from same day to 21 days after the solution was prepared. Multiple time studies were completed, and data varied significantly between studies. In all studies, the dielectric constant and $d_{33,f}$ remained fairly constant until a second phase appeared. A 0.025 drop in loss between the films spun the same day as the solution was made and the 1day old solution suggests that solutions need time to adequately oligomerize and films should be spun next day at the earliest. The large inconsistencies between time studies indicate a more influential dependence on environmental factors such as humidity and temperature as well as minute differences in solution processing affecting the appearance of a second phase.

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