Title:

Sol-Gel Derived Pb(Zr,Ti)O_3 Multilayer Thin Films: Residual Stress, Orientation, and Electrical Properties

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Abstract

Pb(Zr,Ti)O_3 compositions have been under investigation as potential integrated ferroelectric, piezoelectric and capacitor thin films for some time. Sol-gel synthesis and spin-coating are popular routes to the formation of high-quality, dense, crack-free, insulating films. However, electrical properties measured in thin film form are often inferior to those observed in bulk specimens of the same composition. Pb(Zr,Ti)O_3 films with a nominal composition at the MPB (53/47) were deposited from a 2-methoxyethanol based sol-gel system onto Pt/Ti/SiO_2//Si substrates via spin-casting. Multiple layers were sequentially deposited and heat-treated to 650ºC with the use of a PbO overcoat to ensure complete formation of the perovskite phase. Films with thicknesses varying from <0.2µm to >0.5µm were fabricated and studied by analytical techniques, including, electron microscopy, XRD, wafer curvature and electrical measurements to relate functional properties with the film's macroscopic state (phase, stress, orientation). Electron microscopy allowed for the study of phase assemblage and morphology in thin films of various thicknesses. X-ray diffraction studies were used to interrogate not only phase purity in the film, but also film orientation with respect to the substrate. *Ex-situ* wafer curvature measurements allowed for the determination of residual stresses in the Pb(Zr,Ti)O_3 films and the Pt/Ti/SiO_2//Si substrates. Final properties of interest such as dielectric constant, dielectric loss, remanent polarization, and coercive field were measured for films of various thicknesses and residual stress states. These measurements were employed as a whole in an attempt to separate the potential causes of property variations in a model Pb(Zr,Ti)O_3 thin film system when compared with their ceramic bulk counterparts. While, dielectric constant values of ~1200, loss tangents of ~2%, remanent polarizations of ~18$\mu C/cm^2$ and coercive field strengths on the order of 4 MV/m were measured for the thickest films, significant variations were observed as film thickness decreased (and the measured stress increased).