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Low-Temperature Processing of PZT Thick Film by Seeding and High-Energy Ball Milling and Studies on Electrical Properties

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Lead zirconate titanate thick film with molecular formula $PbZr_{0.52}Ti_{0.42}O_3$ (PZT) was prepared by a modified conventional sol–gel method through seeding and high-energy ball milling, resulting in perovskite phase formation at lower temperatures. The ball-milling time was optimized by keeping the seed particle loading (5 wt.%) constant in the sol–gel solution. This methodology helped in reduction of the crystalline phase formation temperature to 300°C, which is much lower than that reported in the literature (450°C). The well-established perovskite phase was confirmed by x-ray diffraction (XRD) analysis. Scanning electron microscopy (SEM) of PZT films revealed uniform and crystalline microstructure. Film prepared by this methodology showed higher spontaneous polarization (2.22 $\mu C/cm^2$), higher capacitance (1.17 nF), and low leakage current density (18 $\mu A/cm^2$). The results obtained from ferroelectric characterization showed a strong correlation with the XRD and SEM results.

Key words: PZT thick film, seeding, high-energy ball milling, XRD, ferroelectric

INTRODUCTION

Lead zirconate titanate material has been extensively investigated by researchers for its good piezoelectric, ferroelectric, and optical properties.[1–4] In the late 1980s, use of bulk $PbZr_{0.52}Ti_{0.42}O_3$ (PZT) was increasingly supplemented by thin/thick films due to their integration into semiconductor chips with the advent of nonvolatile random-access memory (NVRAM) devices. PZT materials in thick film (10 μm to 20 μm) form have been used in high-frequency transducers and vibration control devices for their actuation properties.[5] Fabrication of thicker (i.e., 100 μm to 500 μm) films using methods like tape casting and screen printing suffers from drawbacks due to nonuniformity, the requirement for a high sintering temperature (~1200°C) for film densification, and the lack of reproducibility in their properties.[6,7] It is a very challenging task to fabricate thicker films at low temperature. Though the sol–gel-based chemical method has been widely used for fabrication of such films, it is not suitable for production of thick films higher than 3 μm due to the requirement of large deposition cycles. The present research aims at obtaining thicker (up to 100 μm) films at lower processing temperatures (< 400°C) to aid their smooth integration into semiconductor device compounds.[8]

Many methodologies have been tried for low-sintering phase formation by the aerosol deposition method,[9,10] laser annealing processes,[11] microwave irradiation using a magnetic field,[12] PZT ink,[13] etc. The effect of seeding on reduction of firing temperature (450°C to 650°C) for fabrication of PZT thick films has been reported earlier.[14–16] A novel approach to reduce the processing temperature of PZT thick films below 450°C using the combined effect of high-energy ball milling and seeding (i.e., a small percentage of crystalline PZT seeds) on the

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