CHAPTER 1

INTRODUCTION

Lead oxide based ferroelectric films have been investigated for a variety of device applications including electro-optic devices (Sayer et al. 1992), non-volatile Random Access Memories (RAMs) (Bondurant 1990 and Takayama et al. 1987), Dynamic Random Access Memories (DRAMs) (Scott and Araujo 1989), integrated capacitors on Si (Lee et al. 1993), non-linear optical elements, pyroelectric detectors (Takayama et al. 1987, Okada et al. 1990 and Baude et al. 1993), microelectromechanical systems (MEMS) (Polla and Schiller 1995, Chen et al. 1995 and Polla and Francis 1996), micromotors and actuators (Udayakumar et al. 1994 and Sayer et al. 1992).

Lead zirconate titanate (PZT) of the morphotropic phase boundary composition ($Pb(Zr_{0.52}Ti_{0.48})O_3$), PZT (52/48), is a candidate material for these applications because it exhibits superior dielectric, ferroelectric and piezoelectric properties. In the preparation of PZT thin film by solution deposition or sol-gel processing, metallo-organic starting reagents such as metal alkoxides are employed. Sol-gel processing offers several potential advantages for forming thin films and composite materials such as high purity, chemical homogeneity, low crystallization temperature and its ease of fabrication.

However, previous work by Kwok and Desu (1992), Griswold et al. (1995), and Aungkavattana (1996), demonstrated that it has been difficult to prepare phase-pure perovskite PZT thin films. Regarding to sol-gel derived PZT first crystallized upon heating to an intermediate phase (identified as either a fluorite-structured compound or a pyrochlore phase) at low temperature before transforming to the perovskite structure. Small volume percentages of residual intermediate phase can be especially detrimental for PZT thin film quality. The pyrochlore-like phase is not ferroelectric and has a much lower dielectric constant (k $\sim$ 50) than the perovskite phase. Therefore, device performance depends on the presence of this intermediate phase (Tuttle and Schwartz 1996).

Moreover, understanding the evolution of microstructure and transformation of the intermediate phase to the perovsite phase is very important for preparing a high quality film. Since the crystal structure of pyrochlore is considered a Pb-deficient structure, an addition of excess Pb in solution technique is used to reduce the pyrochlore formation problem. The excess Pb is added to compensate for lead-loss on heat treatment. This technique leads to crystallization of dense perovskite microstructure. Experimental parameters include not only the study of different precursor chemistry, but also the exploration of annealing conditions. However, film thickness in a conventional sol-gel system is generally limited to 50 to 500 nm because of the tendency of cracking as thickness approaches 1000 nm (Barrow 1998).

In recent years, a new coating technology was composite materials which were made by dispersing PZT particles into a PZT sol-gel matrix (Barrow et al 1995). The resulting solution can be spin-, dip- or spray deposited onto a substrate. Then, deposited film was fired and annealed in the same manner as conventional sol-gel. With this new technology, it is possible to fabricate sol-gel film thickness up to 60 μm on Pt-coated silicon, stainless steel or aluminum substrates. The screen printing process is a good deposition technique which is precise and efficient for the fabrication of composite materials having a specific image (Duccilli 1993).

In summary, the objectives of this thesis are the followings:

- to prepare good quality PZT ferroelectric thin film at the morphotropic phase boundary composition (52/48) by sol-gel processing;
- to form ferroelectric composite materials by screen printing method;
- to characterize ferroelectric properties, electrical properties, structure and microstructure evolution for PZT thin films.