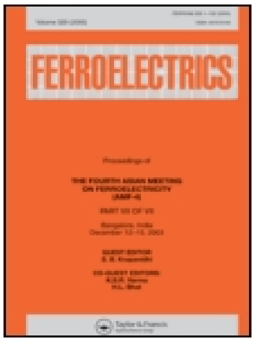

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PREPARATION AND DIELECTRIC PROPERTIES OF $Pb(Sc_{1/2}Nb_{1/2})O_3$-$PbTiO_3$ THIN FILMS NEAR MPB COMPOSITIONS

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The $(1\text{-}x)Pb(Sc_{1/2}Nb_{1/2})O_3\text{-}xPbTiO_3(x=0.4$, PSNT(60/40)) thin films near the morphotropic phase boundary(MPB) composition were successfully deposited via sol-gel method. Taking the strict controls of processing factors such as the stable and homogeneous precursor solutions, conditions of heat treatment and suitable substrates, it was possible to obtain the pyrochlore-free PSNT(60/40) thin films. The effect of substrate on the microstructure and crystallographic orientation is shown. Dielectric behaviors of sol-gel derived PSNT(60/40) thin films have been investigated.

Keywords thin film; PSN-PT; MPB; sol-gel; dielectric behavior

INTRODUCTION

Recently, relaxor ferroelectric and their solid solutions with $PbTiO_3$(PT) have attracted much attention due to their excellent dielectric and electromechanical properties[1][2]. The large piezoelectric responses combined with unique dielectric behaviors make them very attractive for application in micro-actuators, high frequency ultrasonic medical imaging transducers, sensors, various dielectric and microelectromechanical systems[2][3]. For those applications benefiting from device integration, thin film deposition of relaxor ferroelectrics-PT solid solutions are very much desired[4].

$Pb(Sc_{0.5}Nb_{0.5})O_3$(PSN) is a well-known relaxor ferroelectrics[5]. In bulk state, their solid solutions with PT near morphotropic phase boundary(MPB, x=0.42) show excellent piezoelectric coefficients and dielectric constants[6]. However, there are only a few reports published on preparation and properties of PSNT thin films because of the difficulty in preparation of the relaxor-PT thin films with low PT contents due to the unwanted formation of pyrochlore phases[7-9].

In this article, we report the preparation of pyrochlore-free PSNT(60/40) thin films by a sol-gel method. Optimized electromechanical properties are observed for compositions just on the rhombohedral side of the MPB. The effect of different substrates on the crystallization and microstructure of thin films are shown. Electrical and dielectric properties of sol-gel derived PSNT(60/40) films are firstly investigated.

EXPERIMENTAL PROCEDURES

The PSNT(60/40) thin films were processed from a modified alkoxide solution precursor. The solution was prepared under a controlled inert gas atmosphere. Lead acetate trihydrate [$Pb(CH_3COO)_2 3H_2O$], scandium acetate hydrate, niobium ethoxide [$Nb(C_2H_5O)_5$] and titanium isopropoxide [$Ti((CH_3)_2CHO)_4$] were used as precursor materials and 2-methoxyethanol as the solvent. To compensate for lead loss during thermal annealing, 5mol% excess lead was added to the precursor solution. The thin films were deposited via spin coating. After pyrolysis heat treatment at 380°C for 1 min, the films were annealed gust above 650°C. The orientation and microstructural analysis of the films were investigated by x-ray diffraction(XRD) and scanning electron microscopy(SEM). For electrical measurements, gold top electrodes were deposited by sputtering method at room temperature. The P-V(polarization-voltage) hysteresis loops were measured using a standardized RT66A ferroelectric test system operating in a Virtual-Ground measurement. To perform the dielectric measurements, an impedance analyzer and a furnace with controlled heating rates were used.

RESULTS AND DISCUSSION

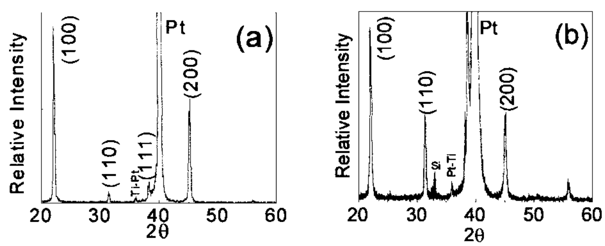

FIGURE 1 XRD patterns of PSNT(60/40) thin films annealed at 650°C for 20min on (a) S_1 and (b) S_2 substrates.

FIGURE 1 shows the XRD patterns of PSNT(60/40) thin films annealed at 650°C for 20min on two kinds of substrates. All the PSNT(60/40) thin films have the perovskite single phase without pyrochlore a phase precipitation. In FIGURE 1(a), the diffraction pattern of PSNT thin film deposited on a $Pt(111)/TiO_2/SiO_2/Si$ substrate(S_1) exhibits a high degree of (100) orientation while PSNT film in figure 1(b), formed on a $TiO_2/Pt/TiO_2/SiO_2/Si$ substrate(S_2) with 5nm-thick TiO_2 seed layer shows rather random orientation with a tendency toward the (111) orientation. The SEM micrographs of PSNT(60/40) films are illustrated in FIGURE 2. Microstructures of both films on S_1 and S_2 also exhibit that dense and homogeneous

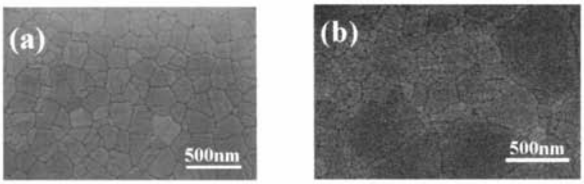

FIGURE 2 Plan view scanning electron micrographs of PSNT(60/40) thin films annealed at 650°C for 20min on (a) S_1 and (b) S_2 substrates.

films without thermal cracks are synthesized. The average grain sizes of (100)oriented PSNT(60/40) thin films on S_1 is about 300nm and seemingly more homogeneous while their microstructure of the specimen on S_2 show abnormal grain growth. The above evidences illustrate that the orientation and microstructure of the films are strongly influenced by the property of substrate.

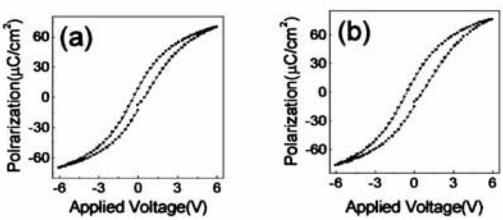

FIGURE 3 Polarization hysteresis loops vs. applied voltage for 220nm-thick PSNT(60/40) thin films measured at room temperature. (a) Au/PSNT(60/40)/S_1 and (b) Au/PSNT(60/40)/S_2

Figure 3 shows the P-V hysteresis loops for PSNT(60/40) films annealed at 650°C for 20min. Both hysteresis loops are almost symmetrical about the voltage axis. The remanant polarization(P_r) value of Au/PSNT(60/40)/S_2 capacitor is larger than that of Au/PSNT(60/40)/S_1. Remanant polarization(P_r) and coercive field(E_c) of Pt/PSNT(60/40)/S_2 capacitor are found to be 13μC/cm^2 and 29kV/cm at an applied voltage of 6V.(in case of Au/PSNT(60/40)/S_1, P_r = 9μC/cm^2 and E_c = 25kV/cm at an applied voltage of 6V) Although the tails of the hysteresis loop indicate high resistivity, the value of remanant polarization is smaller than expected. In general, these thin films resemble a relaxor in the vicinity of transition temperature(T_m). Evidences of a relaxor-like behavior and a reduced transition temperature value are observed in the temperature dependence of the dielectric constant at different frequencies(FIGURE 4). As can be seen in FIGURE 4, the maximum dielectric constant(ε_m) of 3400 and transition temperature(T_m) of approximately 150°C at 1kHz are shown. The lower ε_m and the T_m shift compared to those of the bulk ceramics may result from thin thickness of the film and clamping of the film by the substrate(strain effect)[4][10].

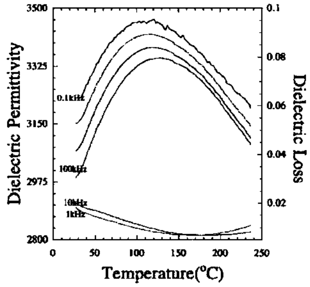

FIGURE 4 Temperature dependence of the relative permittivity and dielectric losses of 220nm-thick PSNT thin films on S_2 substrate at various frequencies. Measurements were made at 1kV/cm.

SUMMARY

The PSNT(60/40) thin films were firstly deposited via sol-gel method with special concern `for stable and homogeneous precursor solution, suitable substrate and narrow processing window. These films showed diffuse ε-T response, frequency dispersion and relaxor-like P-V hysteresis loops at R.T.

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