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Fabrication and characterization of ceramic thin films by RF magnetron sputtering using Zn-enriched $(Ba_{0.3}Sr_{0.7})(Zn_{1/3}Nb_{2/3})O_3$ as target

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ABSTRACT

Dielectric ceramic thin film was fabricated on SiO₂ (110) substrates by radio frequency (RF) magnetron sputtering method using a Zn-enriched ($Ba_{0.3}Sr_{0.7}$)($Zn_{1/3}Nb_{2/3}$)O₃ target. The microstructure, components, and morphological properties of the thin films were characterized thoroughly. The results reveal that the main phases of the thin films are $Ba_xSr_{1-x}Nb_2O_6$, which are of different compositions from that of the ceramic target due to Zn loss. The thin films are polycrystalline and of dense structure with uniform grain sizes and well-defined grain boundaries.

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1. Introduction

Recently, dielectric ceramic thin films have been intensively studied for the potential applications in microelectronic and microwave integration circuits [1,2]. The researches of these ceramic thin films, such as $Ba(Zr_{0.3}Ti_{0.7})O_3$ [3], $BaTi_4O_9$ [4], and $Ba(Zn_{1/3}Ta_{2/3})O_3$ [2] have been reported by different methods such as Sol-Gel [5], PLD [2], and so on. In the past years, our group has studied ceramic thin films fabricated by radio frequency (RF) magnetron sputtering method using stoichiometric $(Ba_{0.3}Sr_{0.7})(Zn_{1/3}Nb_{2/3})O_3$ ceramic as target [6,7]. However, there exist many problems for these thin films, such as the composition discrepancy between the target and the thin films (i.e., serious Znloss) and oxygen-vacancies not only in PLD method [2] but also in magnetron sputtering deposition [3,4,6,7]. Taking into account that a major barrier in the widespread use of these films in a number of high-frequency applications is their high leakage currents determined by structural defects as grain boundaries and oxygen vacancies [8], it is significant for the research on the microstructure and the composition of the thin films. To obtain a dense thin film with fewer oxygen vacancies and less Zn loss, in this article, we adjusted the target component to fabricate ceramic thin films using

magnetron sputtering deposition, which differs from those targets reported previously [6,7], i.e., 1 mol excess ZnO was incorporate in this stoichiometric ($Ba_{0.3}Sr_{0.7}$)($Zn_{1/3}Nb_{2/3}$)O₃ target so as to compensate the ZnO volatilization during the sputtering and annealing process.

2. Experimental procedure

Ceramic thin films were deposited on SiO₂ (110) substrates by RF magnetron sputtering, using a Zn-enriched target comprised of a homogeneous mixture of 1 mol (Ba_{0.3}Sr_{0.7})(Zn_{1/3}Nb_{2/3})O₃ and 1 mol ZnO, which were synthesized by a conventional solid-state sintering technique. The monocrystal SiO₂ (110) substrates were placed on the substrate holder, which can rotate around a central axis to improve the homogeneity of the thin films. The thin films were deposited using an Ar–O₂ gas mixture in a JGP450 RF magnetron sputtering pressure, and 610 °C of the substrate temperature. The Ar gas flow rate was fixed at 16 ml/min and the oxygen flow rate was fixed at 3.2 ml/min, i.e., the O₂/Ar flow ratio of 0.2:1 was used as working atmosphere. The distance between the substrates and the targets was 11 cm, the sputtering time was 180 min, and the base pressure of the chamber was 1.0×10^{-3} Pa. As-deposited films were annealed in flowing oxygen (99.999%) at 1150 °C for 30 min in a tube furnace.

A Rigaku D/max-rB X-ray diffraction (XRD) meter with Cu K_{α} -line, X-ray photoelectron spectroscope (XPS, ESCALAB 250) with Al and Mg K_{α} -lines, a Hitachi S-450 scanning electron microscope (SEM), a Park Autoprobe CP atomic force microscope (AFM) and a Hitachi H-8010 transmission electron microscope (TEM) were the equipments employed to characterize the microstructure, components, crystallinity, and surface morphological properties of the ceramic thin films. The cross-sectional morphologies of the thin films were examined by SEM (S-4800) to evaluate the thickness of the thin film.

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Fig. 1. XRD spectrum of the thin film.

3. Results and discussion

Fig. 1 is the X-ray diffraction patterns of the thin films, exhibiting the presence of the well-crystallized thin films. No preferential orientation for the thin film is observed. The thin films contain a mixture of Ba_{0.5}Sr_{0.5}Nb₂O₆ (as compared with the JCPDS card of No. 39-0265, International Center for Diffraction Data, 2002), Ba_{0.67}Sr_{0.33}Nb₂O₆ (No. 73-0126), Sr_{0.744}Ba_{0.247}Nb₂O₆ (No. 70-3747), and $SrNb_2O_6$ (No. 72-2088), which can be written as $Ba_xSr_{1-x}Nb_2O_6$. And ZnO is also observed, as compared with the characteristic peak of ZnO [9]. As we all know, the various sputtering yields of Ba, Sr, Nb, Zn, and O elements are a knotty problem in the deposition of $(Ba_{0.3}Sr_{0.7})(Zn_{1/3}Nb_{2/3})O_3$ -based thin films. The discrepancy between thin film and target probably results from the volatilization of ZnO during the process of sputtering and annealing, because Zn has a significant larger vapor pressure than the other metal elements (Ba, Sr, Nb) [2]. Therefore, ZnO may either not stick to or be re-evaporated from the growing surface [2], worsening the composition deviation with the target.

According to the Scherrer's formula [10], the grain size in the (013) plane can be estimated by the following expression:

$$L_{(0\,1\,3)} = \frac{\kappa\lambda}{\beta_0\cos\theta}$$

where κ is a constant with a value of about 0.89 for the Cu target, λ is the X-ray wavelength with a value of about 1.54718 Å, β_0 is the FWHM of the (013) peak, and θ is the diffraction angle. In the Scherrer's formula, $\beta_0 = 0.003$ rad and $\theta = 29.065^\circ$; therefore, the grain size in the (013) plane of the thin film is about 52.76 nm.

Fig. 2 shows the XPS spectrum of the thin film in the binding energy range from 0 eV to 1200 eV, and the binding energies at various peaks were calibrated using the C1s (284.6 eV) as a standard sample. The inset images in Fig. 2 show the XPS spectrum of O1s peak. All the XPS spectra of Ba3d, Nb3d, Sr3d, Sr3p, and Zn2p of the thin films consist of two peaks corresponding to their angular momentum of electron. Only one spin-orbit doublet is observed for the individual element, i.e., Ba3d_{5/2} and Ba3d_{3/2} peaks at 780.67 and 795.95 eV, Nb3d_{5/2} and Nb3d_{3/2} peaks at 206.57 and 209.47 eV, Sr3d_{5/2} and Sr3d_{3/2} peaks at 133.07 and 135.02 eV, Sr3p_{3/2} and Sr3p_{1/2} peaks at 267.84 and 278.37 eV, Zn2p_{3/2} and Zn2p_{1/2} peaks at 1023.27 and 1046.51 eV, indicating that only one chemical state exists in the thin films for each element of Ba, Nb, Sr, Zn, i.e., chemical state of Ba^{2+}, Sr^{2+}, Nb^{5+}, and Zn^{2+}. A doublet structure is observed in the XPS spectrum of O1s peak. Its component peak in the spectrum is fitted to a Gaussian-type distribution with the lower binding



Fig. 2. XPS spectrum of the thin film.

energy of 530.45 eV and the higher binding energy of 532.65 eV, as shown in the inset spectrum, corresponding to the lattice oxygen and the adsorbed oxygen [10], respectively. In general, the peak of adsorptional oxygen is much weaker than that of lattice oxygen, which would be favorable for the dielectric property of the thin films, because the oxygen vacancies result in the dielectric loss [11]. The quantitative analysis using Ba3d, Sr3d, Zn2p, Nb3d, and O1s peaks reveals that the Ba:Sr:Zn:Nb:O atomic ratio is 3.01:5.33:0.96:27.90:62.80, respectively.

Fig. 3 shows the SEM image of the thin film, which shows that fine spherical particles are distributed on the sample's surface. The surface morphology of the thin film is of dense structure with less small holes. It is evident that the grains are of uniform grain size, and are distributed homogenously on the surface of the thin film.

Fig. 4 shows the thickness of the thin film is about $1.38 \,\mu$ m with dense structure and homogeneous grains and well-defined grain boundaries, which are consistent with the results shown in Fig. 3.

Fig. 5 shows the AFM images of the sample, with a scan area of $10 \,\mu m \times 10 \,\mu m$. As seen in Fig. 5(a), the sample is regular in



Fig. 3. SEM image of the thin film.

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