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Microstructure of microwave dielectricthin films by RF magnetron sputtering

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ABSTRACT

The article describes the microstructure and morphological properties of microwave dielectric ceramic thin films. These thin films were successfully prepared on SiO_2 (1 1 0) single-crystal substrates by radio frequency magnetron-sputtering system. The microstructure and morphology of the thin films were characterized by X-ray diffraction, scanning electron microscopy, atomic force microscopy, and transmission electron microscopy. The results show that the main phase is $Ba_{0.5}Sr_{0.5}Nb_2O_6$,which has a tetragonal perovskite structure, a long strip pattern, and uniform crystal-grain size of about 2–3 μm in length when annealed under 1150 °C for 30 min in an O_2 atmosphere. These thin films are of excellent crystallization quality, with a polycrystalline and dense structure.

1. Introduction

In recent years, microwave dielectric ceramic materials of the $A(B'_{1/3}B''_{2/3})O_3$ type with their complex perovskite structures have become prospective candidates for microwave applications due to their excellent dielectric properties [1–5]. $(Ba_{0.3}Sr_{0.7})(Zn_{1/3}Nb_{2/3})O_3$ has many advantages, such as extremely low dielectric loss and near-zero temperature coefficient of resonance frequency, under very high microwave frequency [1]; thus, they have a potential application value in the fields of satellite communication and radar and mobile communication system. However, the minimum size is λ/4 for the microwave dielectric ceramic bulk material, which cannot meet the need for integration, thus block the application of the microwave dielectric apparatus. Because it is the general tendency to use frequency apparatuses made of dielectric films, there are significant implications for the study on microwave dielectric ceramic thin films as promising materials for use in microelectronic and microwave integration circuit, which will attract great attention in the near future [6].

The radio frequency (RF) magnetron-sputtering method is a dominant growth technique for the thin films because a large quantity of the thin films can be prepared with relatively high purity and low cost and this is likely to be of interest in a commercial-scale production. The fabrication of microwave dielectric ceramic thin films using $(Ba_{0.3}Sr_{0.7})(Zn_{1/3}Nb_{2/3})O_3$ as target material has not been reported before, and there is vey little information that can be direct conference. In this article, ceramic thin films have been prepared on SiO_2 (1 1 0) single-crystal substrates by the RF magnetron-sputtering method, using $(Ba_{0.3}Sr_{0.7})(Zn_{1/3}Nb_{2/3})O_3$ microwave dielectric ceramic as target materials, which have been prepared by conventional solid-state sintering technique. The microstructure and morphology of the thin films have been analyzed by X-ray diffraction (XRD), scanning electron microscopy (SEM), atomic force microscopy (AFM), and transmission electron microscopy (TEM).

2. Experimental procedure

In this experiment, the thin films were successfully deposited by RF magnetron sputtering, with a JGP450 magnetron-sputtering system, using $(Ba_{0.3}Sr_{0.7})(Zn_{1/3}Nb_{2/3})O_3$ (BSZN, in short) microwave dielectric ceramic as target (62 mm × 3 mm in size) and SiO_2 (1 1 0) as substrates (19 mm × 7 mm × 0.5 mm in size). The substrates were ultrasonically cleaned in acetone and propanol and the chamber pressure was maintained at 1.0×10^{-3} Pa to remove impurities from the chamber. Presputtering was carried out for 10–20 min in an Ar (99.99%) atmosphere to remove the defects on the target and to obtain stable plasma at the same time. During the experimental process, the orthogonal analysis was applied to optimize the parameters, such as sputtering power, sputtering pressure, substrate temperature, and annealing process, to prepare the films with good quality. The thin films were deposited according to the optimum conditions listed in Table 1.

A post-deposition annealing is needed so that the deposited films have a well-crystallized structure. After the furnace was

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Table 1
Optimum deposition conditions of thin films.

Sputtering conditions	The value of parameters
Target–substrate distance (cm)	11
Sputtering powder (W)	200
Sputtering gas	Ar (≥99.99%)
Working pressure (Pa)	0.25
Deposition time (min)	180
Substrate temperature (°C)	610

heated up to 1150 °C quartz boat containing the thin-film samples was placed into the constant-temperature region. Subsequently, flowing O_2 (99.999%) was introduced into the tube, and the samples were annealed in O_2 atmosphere at a flow rate of 500 ml/min at 1150 °C for 30 min. After annealing, the samples were taken out for characterization.

A Rigaku D/max-rB X-ray diffractometer with Cu Kα radiation, a Hitachi S-450 scanning electron microscope, a PARK AUTOPROBE CP atomic force microscope, and a Hitachi H-8010 transmission electron microscope were applied for the characterization of the microstructure and surface-morphology properties, in addition to study the crystallinity of the thin films. The cross-sectional morphology of the thin films was examined by SEM (JEOL JSM-6390).

3. Results and discussion

3.1. XRD analysis

Fig. 1 shows the XRD pattern of the thin films annealed at 1150 °C for 30 min. In Fig. 1, the major peaks are identified that the main phase is $Ba_{0.5}Sr_{0.5}Nb_2O_6$, as compared with the JCPDS card No. 39-265 (International Center for Diffraction Data, 1990), with a tetragonal-perovskite type structure of $a = b = 12.456$ nm; and $c = 3.952$ nm. The main phase does not conform to the target composition, which maybe result from the volatilization of ZnO during the process of sputtering and annealing. No preferential orientation for the thin film is observed. The diffraction peaks in every crystal plane of the thin films are apparent, which indicates that the grains have developed completely. Moreover, in the XRD pattern, there are no diffraction peaks of any other phases, establishing the excellent purity of the thin films.

To investigate the thin film growth and the formation of grains, the full-width at half-maximum (FWHM) of the peak (4 1 0) of the $Ba_{0.5}Sr_{0.5}Nb_2O_6$ phase has been analyzed. According to the

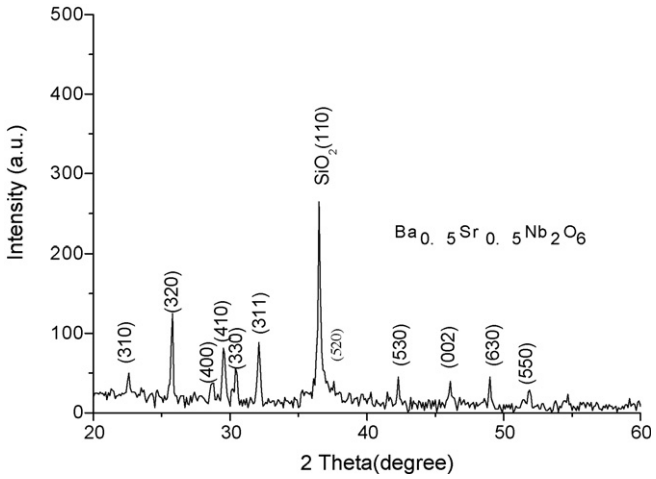


Fig. 1. XRD spectrum of thin-film samples annealed at 1150 °C for 30 min.

Scherrer's formula [7], the grain size in (4 1 0) orientation can be estimated by the following expression:

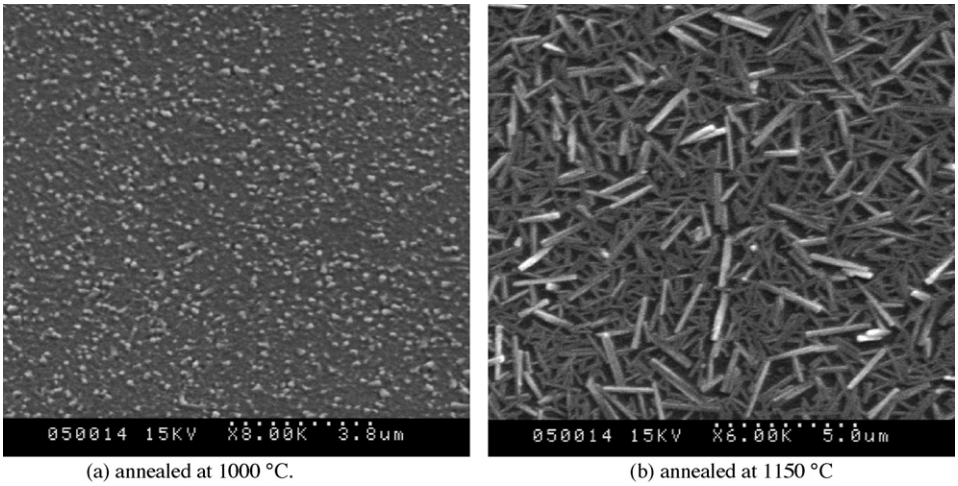
$$L_{(4\,1\,0)} = \frac{\kappa\lambda}{\beta_0 \cos\theta},$$

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 (4 1 0) peak, and θ is the diffraction angle. In the Scherrer's formula, $\beta_0 = 0.0043$ rad and $\theta = 29.48°$; therefore, the grain size in the (4 1 0) orientation of the thin films is about 36.3 nm.

3.2. SEM and AFM analyses

Figs. 2 and 3 are the surface morphologies of the samples fabricated at 200 and 250 W at different annealing temperatures of 1150 °C and 1000 °C, respectively.

As seen in Figs. 2 and 3, it is evident that the crystalline quality is improved when the sputtering power decrease from 250 to 200 W and the annealing-temperature increase from 1000 to 1150 °C. In Fig. 2a, when the annealing temperature is 1000 °C, there are some uniformly distributed fine particles on the sample surface. This reason maybe come from the fact that during the coalescence stage of the annealing process, atoms do not have sufficiently high kinetic energies to grow continuously. Fig. 2b



(a) annealed at 1000 °C. (b) annealed at 1150 °C

Fig. 2. SEM morphology of the thin films fabricated at 200 W at different annealing temperatures for 30 min.