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Materials of Nanocrystalline $Sr_{1-x}Ba_xBi_4Ti_4O_{15}$ for Piezoelectric Sensor

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Abstract. The nanocrystalline Strontium Barium Bismuth Titanate (SBBT) thin films with structure of $Al/TiO_2/SBBT/TiO_2/RuO_2/SiO_2/Si$ were fabricated using sol-gel technique. The source materials are bismuth acetate $Bi(CH_3COO)_2$, barium acetate $Bi(CH_3COO)_2$, strontium acetate $Sr(CH_3COO)_2$ and titanium butoxide ($Ti(OC_4H_9)_4$). Different nanostructures of the films were prepared with un-annealed condition as well as after annealing at three different temperatures of 400, 500 and 600^0C, in air for 2 minutes. The structure of SBBT thin films have been systematically studied by XRD, AFM, SEM and dielectric constant measurement. For the sensor device measurement, the SBBT thin film pressure sensors were tested by pneumatic loading method at pressure range between 0 to 450 kPa. It was found that the sensing properties of the films were affected by the crystalline nature of the films. It is shown that there is a linear relationship between the crystallization, grains size and dielectric properties with the sensing response of the film towards pressure.

Introduction

Pressure sensors are one of the most common micro-sensors devices used in various industries such as automotive, medical, military, aeronautical, hydraulics, instrumentation, process and industrial control [1]. Pressure or stress sensing materials, that is piezoelectric material is the most frequently used material in pressure gauge because of it's special capabilities such as fast response, ruggedness, high stiffness, ability to measure the quasi-static pressure and good thermal stability.

At present, in commercial sector, lead zirconate titanate (PZT) based ceramic are most widely used as a sensing material. However the evaporation of toxic lead during the fabrication of their ceramic causes an environmental problem. Therefore, there is an increasing interest of investigating lead free of piezoelectric materials to replace PZT based piezoelectric ceramics. In the previous research we choose bismuth titanate (BTO) to replace PZT since it has a large piezoelectric coefficient. BTO is one of the family called Aurivillius phase, generally formulated as $A_{m-1}B_mO_{3m-1}$ [2]. However, the most problem found in the performance of BTO pressure sensor are slow recovery time, low repeatability and raising current leakage at high annealing temperature. Several approaches have been made to solve these well known problems. One critical approach is altering the previous BTO solution by doping strontium and barium in proper molar composition. Strontium and barium have been identified as potential stabilizing materials in BTO ceramics. Strontium barium bismuth titanate (SBBT) $Sr_{(1-x)}Ba_{(x)}(Bi_4Ti_4O_{15})$ based ceramics is chosen because of its high Curie temperature (T_c= 620^0C), good piezoelectricity and thermal stability [3]. In this paper, we report the fabrication and characterization of $Sr_{(1-x)}Ba_{(x)}(Bi_4Ti_4O_{15})$ thin films pressure sensors with various annealing temperature.

Experimental

The solution of $Sr_{(1-x)}Ba_{(x)}(Bi_4Ti_4O_{15})$ was prepared by sol gel technique. The raw starting materials were bismuth acetate $Bi(CH_3COO)_2$, barium acetate $Bi(CH_3COO)_2$, strontium acetate $Sr(CH_3COO)_2$ and titanium butoxide $(Ti(OC_4H_9)_4)$. Initially, the powder of bismuth, barium and strontium were dissolved with a proper molar in 4 ml acetic acid containing 20% volume of deionize water at room temperature.

The SBBT films were spin coated on silicon wafer at 400 rpm for 30 second followed by heating process at 300 ^{0}C in air for 15 minutes. The films were then annealed at temperature of 400, 500, 600 and 700 ^{0}C. The crystal structure of the SBBT thin films were examined using X-ray diffractometer (XRD). Dielectric measurements were carried out using impedance spectroscopy Solarton-Schlumberger, while the homogeneities and grains size of the samples were characterized by Scanning Electron Microscope (SEM).

The SBBT pressure sensors were fabricated with the structure of $Al/TiO_2/SBT/TiO_2/SiO_2/Si$. The raw material is silicon wafer and the silicon oxides were deposited by electron beam evaporation with thickness of 1500 Å. Ruthenium Oxide as bottom electrode was deposited using RF magnetron sputtering for 2 hours. In order to reduce the crystallization temperature for the formation of SBBT ferroelectric phase, the buffer layer, TiO_2 was then deposited on these thin films.

Results and Discussion

Thin film characterization

Typical XRD patterns SBBT thin films annealed at various temperatures are shown in figure 1. It is shown that the films were in amorphous phase and it began to converse to crystalline phase after annealing at 600 ^{0}C. Figure. 2 shows SEM micrographs of SBBT thin films. The images indicated that annealed films were in orderly forms, where the nano sized grains were formed with a clear grain boundary. This feature is very important, since the electromechanical responds can be arose from the orderly oriented grains. The electrical behavior of SBBT thin films measured in terms of the dielectric constant. The value of the dielectric constant was calculated from the capacitance of the film using Debye distribution formula, measured at 1 kHz at 20 mV. Dielectric constant of un-anneal SBBT film was 72.81 and increased to 187.4 at annealing temperature 700 ^{0}C. The higher dielectric constant is due to the increasing number of domain population and its wall motion [4] hence improved crystallization process of the thin film after annealing.

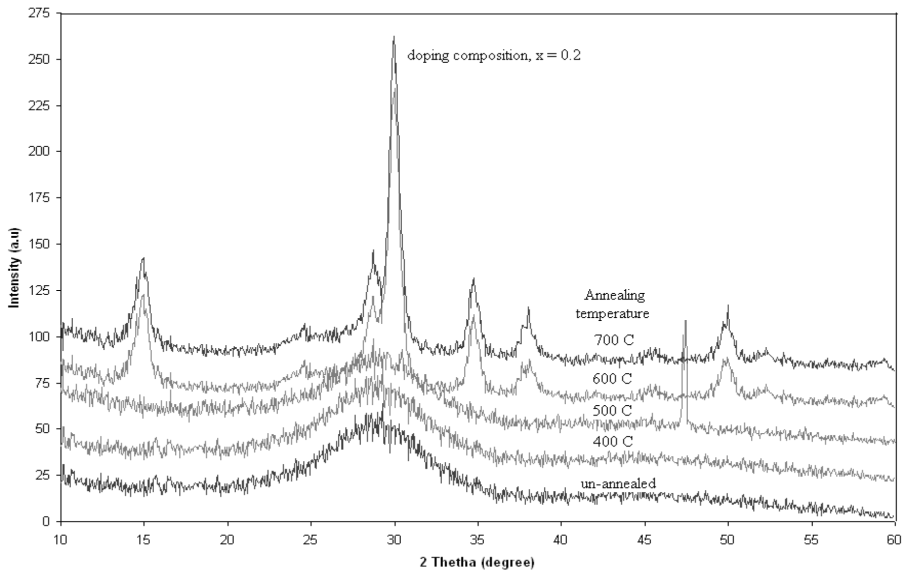

Figure 1. XRD patterns of x = 2 with different annealing temperature.

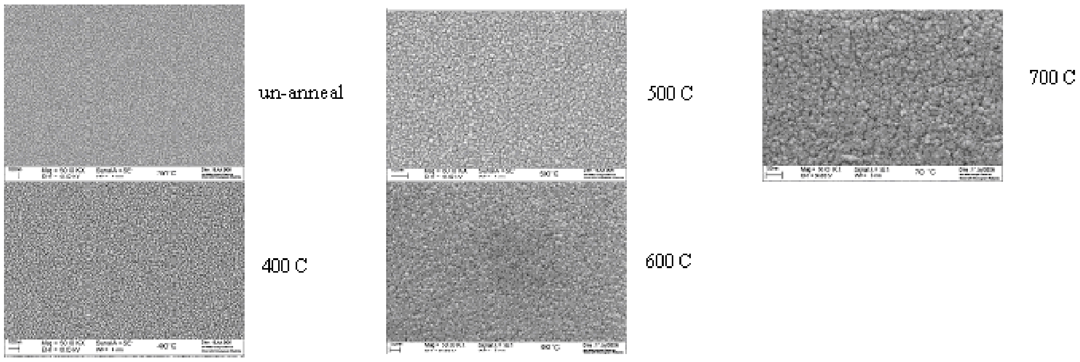

Figure 2. SEM images of surface morphology of $Sr_{(1-x)}Ba_{(x)}(Bi_4Ti_4O_{15})$ thin film for un-anneal and anneal at 400^0 C, 500^0 C, 600^0 C, 700^0 C.

Piezoelectric pressure sensor measurement

The piezoelectric measurement system was designed using the concept of direct piezoelectric effect. When pressure is applied to piezoelectric materials, it create a strain or deformation in the materials, due to the deflection of the lattice in a naturally piezoelectric quartz crystal [5]. The response repeatability of SBBT sensor is shown in figure 3. The measurements were made by seven cycles at 450 kPa. The graph indicated that the 600 ^{0}C anneal sensor showed the best repeatability compared to others since the similar respond was maintained until the seventh cycles. The SBBT pressure sensor achieved a linear characteristic response between 50 kPa to 450 kPa pressure load. It was found that the performance of sensors could be influence by internal structure of SBBT films. This structure is applicable for better electromechanical responds in the ferroelectric's materials. The small dielectric constant for low annealed sensor gave low capacitance that would present poor repeatability during continuous high pressure measurement. The 700 ^{0}C annealed sensor presents poor repeatability. Based on the SEM result, it is suggests that nanostructure of the films would play an important role to generate the high voltage and stable electromechanical response. When the grain size increase, the domain that carrying the dipoles will be increased as well [6].

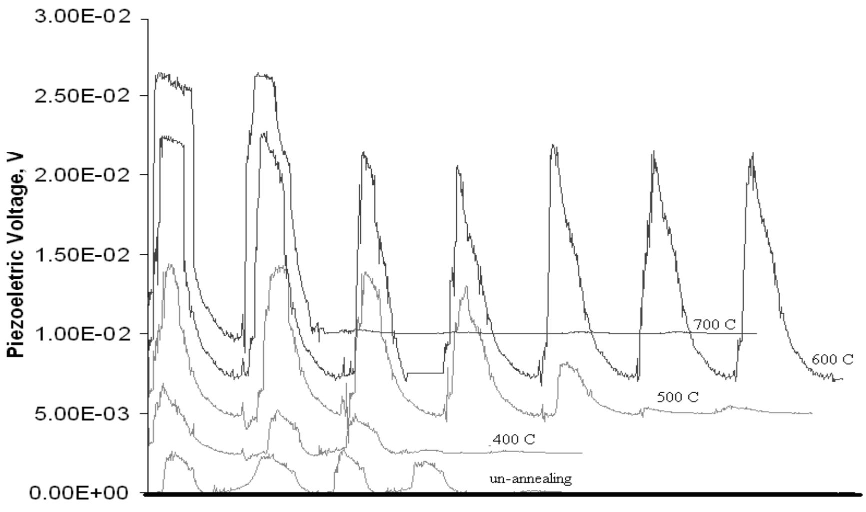

Figure 3. The piezoelectric response SBBT pressure sensors measured at 450 kPa pressure load for un-annealed and annealed at 400, 500, 600 and 700 ^{0}C.

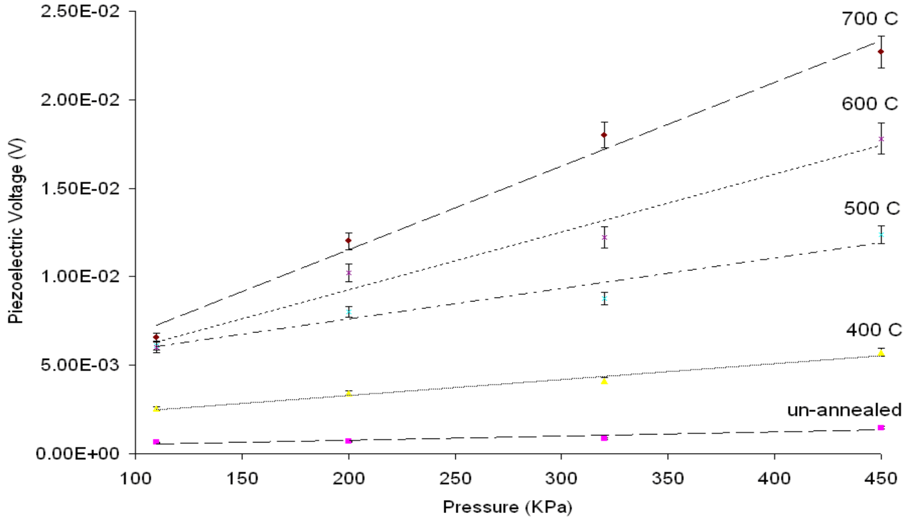

Figure 4. Graph of linearity for pressure sensor performance for un-anneal and anneal at 400^0 C, 500^0 C, 600^0 C, 700^0 C

The figure of merit of this sensor that is the sensitivity of 20-40×10^{-3} mV/Pa is slightly higher compared to that of $PbTiO_3$–PVDF(TeFE) piezoelectric composites which is greater than 1 mV/Pa [7]

Summary

The microstructure and piezoelectric responds of $SrBi_4Ti_4O_{15}$ thin films pressure sensors have been systematically studied in as-prepared condition as well as after annealing at 400, 500, 600 and 700 ^{0}C. The XRD results indicated that the SBBT film starts to converse from amorphous phase to crystalline after annealing at 600 ^{0}C. The dielectric constant and grain size were found to increase with annealing temperature.

Acknowledgement

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