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Barium silicate modified strontium bismuth tantalate ferroelectric thin films

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ABSTRACT

Novel strontium bismuth tantalate ($Sr_{0.8}Bi_{2.2}Ta_2O_9$ (SBT)) modified with 3 and 5 mol% ratio barium silicate (Ba_2SiO_4) thin films were grown on Pt(100 nm)/Ti(50 nm)/SiO_2/Si(100) substrates by spin coating technique. The influence of barium silicate doping in SBT was studied from the view point of changing dielectric and ferroelectric properties like dielectric constant (ε_r) and remnant polarization (P_r). Well crystallized thin films showed convenient ferroelectric properties with comparatively lower P_r in the range between 1.52 and 0.44 μC/cm^2 and smaller ε_r value of 163. Thus, with such reduced values of P_r and ε_r barium silicate modified SBT offers a useful potential to be used in Ferroelectric Field Effect Transistor (FeFET) type (1T-type) Ferroelectric Random Access Memories (FeRAMs) upon improving insulation properties.

1. Introduction

Strontium bismuth tantalate ($SrBi_2Ta_2O_9$ (SBT)) is one of the most popular ferroelectric materials utilized in Ferroelectric Random Access Memories (FeRAMs). The robust remnant polarization versus fatigue [1], the high read/write speed, and the capability of low voltage operation have made SBT superior compared to other popular ferroelectrics like lead zirconate titanate (PZT) in FeRAM applications. One transistor (Ferroelectric Field Effect Transistor (FeFET)) type (1T-type) FeRAM is different than from the capacitor type one with respect to the utilization and functionality of the ferroelectric film. Actually, concept of ferroelectric gate in FeFET yields enabling a nondestructive readout operation. On the other hand, directly grown ferroelectric films on Si have some challenges in order to achieve the required ferroelectric properties and where the interface can results in mobile charges which consequently lead to a decrease in the retention time. Such alternative operation concept necessitates for different requirements for ferroelectric films such as relatively small values of remnant polarization and dielectric constant. As a popular member of bismuth layered structures SBT consists of $(SrTaO_7)^{2-}$ perovskite like layers [2] which are responsible for the ferroelectric properties. These layers are sandwiched between two $(Bi_2O_2)^{2+}$ layers which are charged for insulation character. Considering the structure of SBT remnant polarization can be changed by reasonable elemental doping/substitution or through structural twisting mechanisms by the addition of different materials. Orthorhombic crystal structure allows two fold symmetry in a-axis which is the polarization direction. Polarization is emerged from the movement of Ta^{5+} ion from the off center location with respect to the surrounding oxygen ions in the octahedron. Thus, to alter the ferroelectric properties some silicates [3–5] and $BaZrO_3$ [6–8] were added into SBT to utilize 1T-type FeRAM applications.

In the current study, strontium bismuth tantalate ($Sr_{0.8}Bi_{2.2}Ta_2O_9$ (SBT)) doped with 3 and 5 mol% ratio barium silicate (Ba_2SiO_4)

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thin films were successfully fabricated on Pt(100 nm)/Ti(50 nm)/SiO_2/Si(100) substrates by sol–gel spin coating method. Well grown thin films were evaluated to investigate the impact on the remnant polarization (P_r) and dielectric constant (ε_r). It was found that both P_r and ε_r were remarkably reduced in modified SBT which will be offered to use in 1T-type FeRAMs applications with further improvement of insulation properties and investigating realization properties.

2. Experimental

Precursor sol–gel solutions of Ba_2SiO_4 and $Sr_{0.8}Bi_{2.2}Ta_2O_9$, (Toshima MFG Co. Ltd.) were spin coated on Pt(100 nm)/Ti(50 nm)/SiO_2/Si(100) substrates. The solutions were spread on the surface of the substrates at a rotation speed of 2500 rpm and dried on hot plates to remove chemicals according to the following heating sequence: 155 °C for 2 min, 240 °C for 3 min and 400 °C for 3 min. Next, the dried films were successively annealed in a Rapid Thermal Annealing (RTA) furnace at 750 °C for 1 min in oxygen atmosphere. This spin coating, drying, and annealing process was repeated five times until the film thickness reached to 255 and 240 nm for pure and modified SBT, respectively. Later, the films were crystallized in the RTA furnace at 750 °C in oxygen atmosphere for 30 min. After that, platinum round-shape top electrodes with areas of 3.14×10^{-4} cm^2 were deposited by the Electron Beam Evaporation (E-gun) technique through a shadow mask as electrical connections. Pure and 3 and 5 mol% Ba_2SiO_4 doped SBT thin film samples were labeled as SBT, SBT + Ba_2SiO_4-3, and SBT + Ba_2SiO_4-5, respectively. Crystal structures were investigated with a multipurpose X-ray diffractometer (X`Pert-Pro MPD, Philips). Ferrroelectric properties were characterized by the measuring polarization versus electric field (P-E) hysteresis loops at a frequency of 10 kHz by a ferroelectric test system (Toyo Corp., FCE-1A/Fop-100 V). Capacitance behavior was investigated by measuring capacitance versus voltage (C–V) at a frequency of 1 MHz by a LCR Meter (Toyo Corp.). Conduction behavior was characterized by measuring leakage current density versus electric field (J–E) by HP4156C Precision Semiconductor Parameter Analyzer (Agilent).

3. Results and discussion

Fig. 1 shows the X-ray diffraction (XRD) patterns of SBT, SBT + Ba_2SiO_4-3, and SBT + Ba_2SiO_4-5 films grown on Pt/Ti/SiO_2/Si (100) substrates.

XRD diffraction patters clearly demonstrate polycrystalline nature of the films. All the peaks of the layered perovskite phase are observed for the films which indicate that the layered perovskite structure was preserved by the incorporation of Ba_2SiO_4 up to 5 mol %. It is worth noting that the doping improves the crystallinity as the doping ratio increases. On the other hand, it is noticeable that the addition of Ba_2SiO_4 leads to arise some negligible peaks which are probably barium silicates as a secondary phase. This can be attributed to the mixing of precursor solutions where Ba_2SiO_4 and $Sr_{0.8}Bi_{2.2}Ta_2O_9$ may lead to the formation of a secondary phase besides the main one via crystallization process when the temperature is increased to 750 °C according to the fabrication route. Therefore, XRD technique clearly confirms the enhanced SBT crystals with a cost of secondary phase. Hence, XRD spectra is enough for identifying the crystal structure without the need for micrograph which might be difficult to distinguish such a small quantity of a secondary phase. Furthermore, arising (0 0 8) peak causes to degrade polarization as will be discussed later. As a matter of fact, the (115) orientation is significantly important since there is an inherent relationship with the polarization.

Since the main aim of the current research is to decrease the polarization, the impact of the Ba_2SiO_4 addition on the ferroelectric properties is investigated. Hence, the ferroelectric properties of the samples were evaluated by analyzing the polarization vs. electric field (P–E) hysteresis behavior. Fig. 2 presents the comparisons of the P–E loops and the saturation properties of SBT, SBT + Ba_2SiO_4-

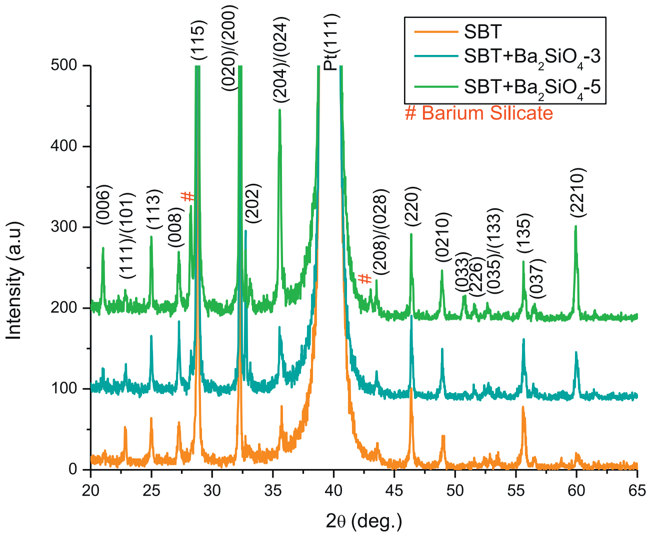

Fig. 1. XRD patterns of SBT, SBT + Ba_2SiO_4-3, and SBT + Ba_2SiO_4-5.