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Solution combustion synthesis: Effect of calcination and sintering temperature on structural, dielectric and ferroelectric properties of five layer Aurivillius oxides

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ABSTRACT

The effect of calcination temperature on phase formation and sintering temperature on structural, dielectric, electrical and ferroelectric properties of $Ba_2Bi_4Ti_5O_{18}$ (BBT), $Pb_2Bi_4Ti_5O_{18}$ (PBT) and $Sr_2Bi_4Ti_5O_{18}$ (SBT) ceramics prepared by solution combustion route using glycine as a fuel are investigated in this paper. Calcination temperature was optimized at 650 °C for BBT and 750 °C for SBT and PBT, at which these compounds showed pure phase formation. It was observed that density and grain size of the sintered pellets increases with increasing sintering temperature. The dielectric constant was found to be dependent on grain size and density. Transition temperature, activation energy and remnant polarization were found to increase with an increase in sintering temperature. Porosity and conductivity decreases with an increase in the sintering temperature. Thus improving the dielectric, electrical and ferroelectric properties of five layered Aurivillius oxides.

1. Introduction

Bismuth layered structure ferroelectric (BLSF) compounds were first synthesised by Aurivillius in 1949, hence are also known as Aurivillius oxides [1]. The general formula of BLSF compounds is $(Bi_2O_2)^{2+}$ ($A_{m-1}B_mO_{3m+1}$)²⁻, where the second term in the formula is interleaved between bismuth oxide layer along the c-axis. The A-site is occupied by mono, di, trivalent ions or a combination of them, the Bsite is occupied by tetra-, penta- or hexa- valent cation and *m* indicates the number of perovskite slabs. It was observed that the ferroelectric property and structural stability is closely related to the value of *m*. This may be due to the fact that with an increase in the value of *m* lattice parameters along the a-b plane increases, which increases the strain as a result of size mismatch between the $(Bi_2O_2)^{2+}$ and perovskite layers, thus making the structure unstable [2].

The kind of ion occupied at A-site or B-site influences the structural stability, dielectric, and ferroelectric properties. A-site substitution also affects the phase transition of bismuth layered ferroelectric compounds, with an increase in the ionic radii of A-site substitution, the transition temperature increases and the compounds show diffused phase transition [3,4].

Bismuth layer-structured ferroelectric ceramics possess attractive electrical properties such as excellent fatigue endurance, fast switching

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speed, good polarization retention, relatively high Curie temperature, low aging rate and low operating voltage. High Curie temperature makes BLSFs attractive for high temperature piezoelectric applications under high temperature and high frequency range. BLSF compounds have fatigue free nature. They are chemically and mechanically stable [2]. These materials exhibit good ferroelectric properties above room temperature and having comparatively high transition temperature and low crystallization temperature. They find a wide range of applications in ferroelectric random access memory (FeRAM), piezoelectric devices, sensors and as photo catalysts [3].

 $Ba_2Bi_4Ti_5O_{18}$, $Pb_2Bi_4Ti_5O_{18}$ and $Sr_2Bi_4Ti_5O_{18}$ are members of the fifth group of the BLSF family. The structure of five layered bismuth titanate compounds consist of five layers of $[(A_2Bi_2)Ti_5O_{16}]^{2^-}$ situated between the $(Bi_2O_2)^{2^+}$ layer, where A = Ba, Pb, Sr. The transition temperatures as reported earlier for $Ba_2Bi_4Ti_5O_{18}$, $Pb_2Bi_4Ti_5O_{18}$ and $Sr_2Bi_4Ti_5O_{18}$ are 267 °C, 285 °C, 329 °C respectively [3,4].

Ferroelectricity in five layered (m = 5) BLSF compounds was first reported by Subbarao [5]. Fang et al. described the formula of Ba₂Bi₄Ti₅O₁₈ in the form of Bi₄Ti₃O₁₂.2BaTO₃ and suggested that this compound has a layered structure and shows ferroelectric properties [6]. Aurivillius phases A₂Bi₄Ti₅O₁₈ (A = Ba, Pb, Sr, Ca) possess orthorhombic (Pseudo tetragonal) structures belonging to B2eb space group [7].





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 $\label{eq:Fig. 1. X-ray diffraction patterns of (a) Ba_2 Bi_4 Ti_5 O_{18}, (b) Pb_2 Bi_4 Ti_5 O_{18} and (c) Sr_2 Bi_4 Ti_5 O_{18} ceramics calcined at different temperatures.$

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