

Synthesis, structural and dielectric properties of SrBi₂Ta₂O₉ prepared via co-precipitation method

Mohamed Afqir^{1,2}, A. Tachafine², D. Fasquelle², M. Elaattmani¹, J. C. Carru², A. Zegzouti¹, and M. Daoud¹

¹Équipe Sciences des Matériaux Inorganiques et leurs Applications, Faculté des Sciences Semlalia, Université Cadi Ayyad, Marrakech, Maroc

²Unité de Dynamique et Structure des Matériaux Moléculaires, Université du Littoral Côte d'Opale, Calais, France

Copyright © 2016 ISSR Journals. This is an open access article distributed under the **Creative Commons Attribution License**, which permits unrestricted use, distribution, and reproduction in any medium, provided the original work is properly cited.

ABSTRACT: Strontium Bismuth Tantalate (SrBi₂Ta₂O₉) was synthesized by a co-precipitation technique and characterized by X-ray diffraction, infrared spectroscopy and dielectric measurements. The powder was synthesized from aqueous solution of Bi(NO₃)₃, Sr(NO₃)₂ and TaO₅ by a precipitation in a strong alkaline solution. By X-ray diffraction, the single-phased perovskite SrBi₂Ta₂O₉ (SBT) was obtained at 800 °C. The trace element analysis was contributed by infrared spectroscopy. The dielectric properties were carried out in the frequencies 100 Hz, 1 kHz and 10 kHz.

KEYWORDS: Aurivillius, Co-precipitation, X-ray diffraction, Infrared, Dielectric proprieties.

1 INTRODUCTION

The crystal structure of strontium bismuth tantalate SBT belongs to the layered perovskite ferroelectrics where the crystal consists of stacking of alternating layers of Bi₂O₂ and pseudo-perovskite SrTa₂O₇ units with double TaO₆ octahedral layers along c-axis [1]. Layered perovskite-like ferroelectric of SBT has an attraction for the development of non-volatile random access memories (NVRAMs) because of its excellent fatigue characteristic.

There are a lot of synthesis methods available for SBT, From the Baorang Li et al. [2], bulk SrBi₂Ta₂O₉ ceramic can be prepared by solid-state reaction at 1200°C. In the studies of Wen Wang et al. [3], SrBi₂Ta₂O₉ nanowires can be prepared by a sol-gel process, using ethoxy tantalum, strontium acetate, and bismuth subnitrate as starting materials. Single-phased perovskite phase SBT was obtained at 800 °C after following chart of the preparation. From the studies of Asit. B. Panda et al. [4], where the nano-sized SrBi₂Ta₂O₉ has been synthesized by four different chemicals based solution methods using metal salts, EDTA, triethanolamine, poly-vinyl alcohol, sugar and ammonium tartarate. It has been observed that SBT powders, prepared at around 950°C. S.R. Dhage et al. [5], the ammonium hydroxide was used to precipitate Sr²⁺, Bi³⁺ and Ta⁵⁺ cations simultaneously. Pure SrBi₂Ta₂O₉ phase was found to be formed at 800°C.

There are several reasons advocate doing this paper. First, SrBi₂Ta₂O₉ (SBT) ceramic prepared by solid-state reaction results in decomposition of SrBi₂Ta₂O₉. Second, few studies talk on dielectric properties of SrBi₂Ta₂O₉ prepared by chemical routes and finally, the chemical routes are not easy to recover.

The objectives of this paper are: (a) to synthesis bismuth strontium tantalate powder, (b) to characterize single phase materials using X-Ray Diffraction and Fourier transform infrared (FT-IR), and (c) To study the dielectric properties.

2 EXPERIMENTAL

A stoichiometric amount of SrCl₂.6H₂O was dissolved in distilled water, Bi(NO₃).5H₂O was dissolved in a minimum amount of dilute HNO₃ to avoid precipitation of Bi ions and Ta₂O₅ was dissolved in a minimum amount of HF. The three limpid

solutions were mixed together, followed by addition of KOH solution (10 M) until pH= 12 to ensure complete precipitation. The precipitate was filtrated with washing several times with distillate water. After drying in the oven, the precursor was heated in a furnace at 400 °C during 12 h, and then at rate 5 °C/min to rise up to 800 °C and kept to this temperature for 12 h.

Structural and dielectric properties were performed by the powder X-ray patterns (XRD), Fourier transform infrared reflection spectroscopy (FT-IR) and LCR meter.

3 RESULTS AND DISCUSSION

The Figure 1 shows the XRD pattern of SBT powder. The crystal structure of SBT is orthorhombic with space group $A2_1am$ (JCPDS no 01-070-4062) and there are no traces of secondary phase. The refined lattice parameters are $a = 5.523$, $b = 5.527$ and $c = 25.077$. The density of the sintered material is 8.83 and 7.064, for theoretical density calculated from XRD data and experimental density measured by pycnometer (Equation 1) respectively. The experimental density achieved above 80% to that of theoretical density, from the latter value, the lower porosity occurred which lead may be to improved dielectric proprieties.

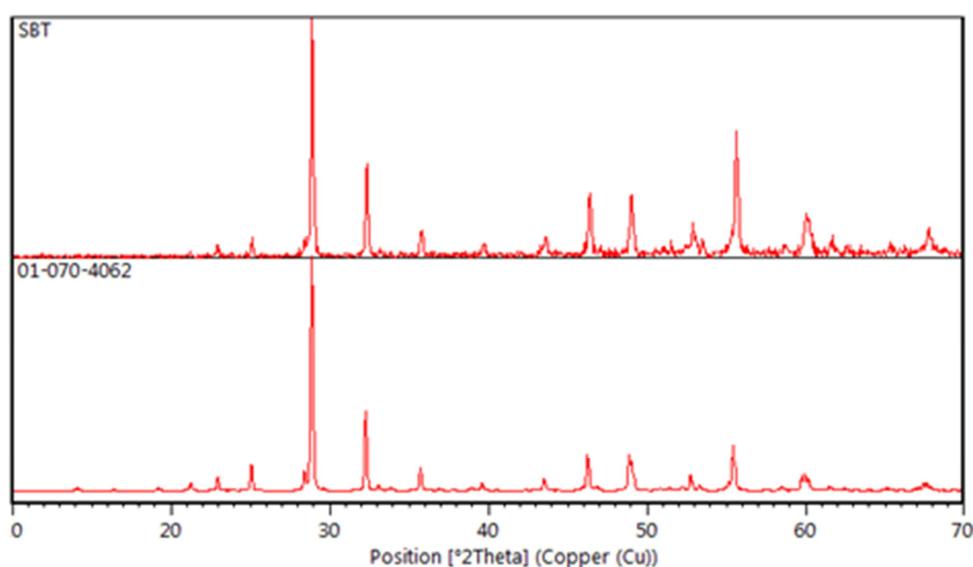


Fig 1. XRD of SBT

From the literature [6], the crystal structure has orthorhombic symmetry with $a = 0.5306$ nm, $b = 0.55344$ nm and $c = 2.49839$ nm; the theoretical density is 8.789 g/cm. The crystallite size calculated from Scherrer's formula is found to be 55 nm.

$$D = \frac{K\lambda}{B \cos \theta} \quad (1)$$

Where $K = 0.9$, λ is the wavelength of X-ray radiation, B is the full width at half maximum of the diffracted peak and θ is the angle of diffraction.

Figure 2, shows the IR spectra. The TO_6 structure could be related to the 600 - 640 cm^{-1} bands which are expected for octahedral stretching [7]. Two infrared absorption bands at 550 and 623 cm^{-1} , indicating the formation of $SrBi_2Ta_2O_9$ oxide [8]. Strange and weak intensity of absorption band at 1416 cm^{-1} and 1576 cm^{-1} respectively, indicate that NO_3^- anions still exist [9], the existence of nitrate ion was not observed in XRD, suggesting the limitation of the analysis. At 3430 and 1630 cm^{-1} assigned to the bending vibration and stretching vibration of the O-H bond in absorbed and coordinated water, showing that there are residual structural O-H [10]. However, several functional and mineral groups are found to be absent.

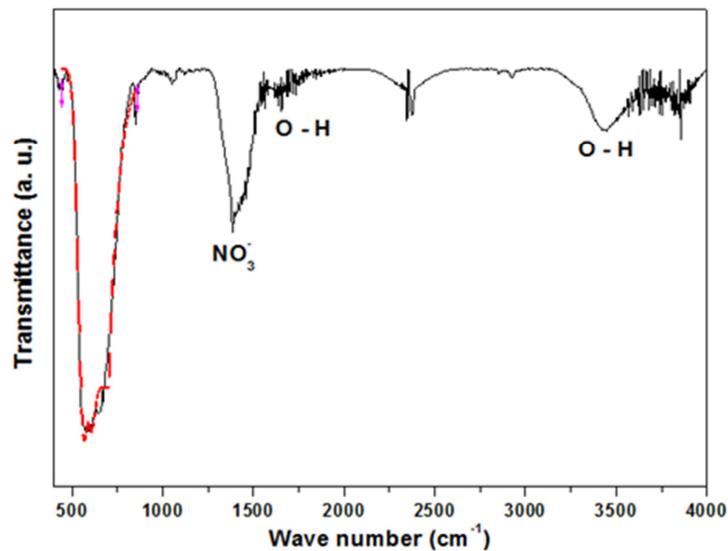


Fig 2. FT-IR of SBT

Figure 3, shows the variation of dielectric constant and tangent loss as a function of temperature at 100 Hz, 1 kHz and 10 kHz. The dielectric constant shows a sharp peak at the phase transition temperature corresponding to the Curie temperature ($T_C = 300^\circ\text{C}$) for all frequencies, which indicated the normal behaviour of the material. The dielectric constant peak (ϵ'_m) was found to be 225, 23 and 12 for 100 Hz, 1 kHz and 10 kHz respectively. The tangent loss increases with increasing temperature and decreasing with increasing frequency. The loss tangent peaks were observed around 290°C for all frequencies with peak values 8.2, 4 and 2.8 for 100 Hz, 1 kHz and 10 kHz respectively. Indicating the normal behavior ferroelectric. A few studies on layered perovskite have reported a sharp peak in the tangent loss at lower frequencies [11, 12].

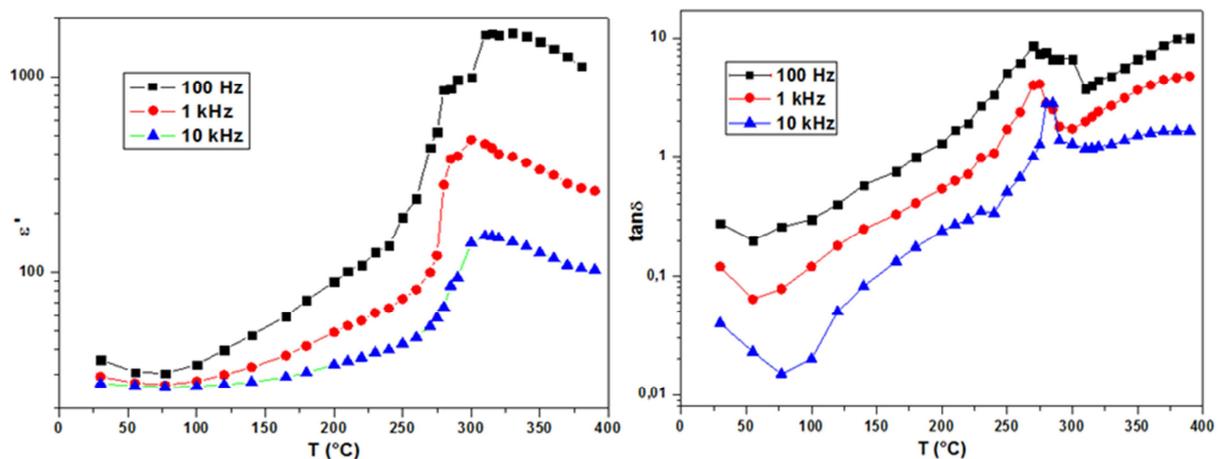


Fig 3. Frequency dependence of dielectric constant and tangent loss as a function of temperature

Figure 4 shows the variations of ϵ' and $1/\epsilon'$ with the temperature at 10 kHz. The dielectric constant data fitted to the Curie–Weiss law:

$$\epsilon' = \frac{C}{T - T_{CW}} \quad (2)$$

Where C is Curie constant and T_{CW} is Curie-Weiss temperature. Note that the Curie temperature ($T_C = 310^\circ\text{C}$) is higher than the Curie-Weiss temperature ($T_{CW} = 177^\circ\text{C}$), and of dielectric constant exhibits a finite maximum at T_C . From the equations depicted in Figure 4, the ratio of positive slope (below T_C) to slope (above T_C) is over 4, thus, predicts a first-order

ferroelectric transition. According to the fitting results, Curie constant is at a magnitude of around 10^5 , indicating the displacement-type nature of the ferroelectric transition.

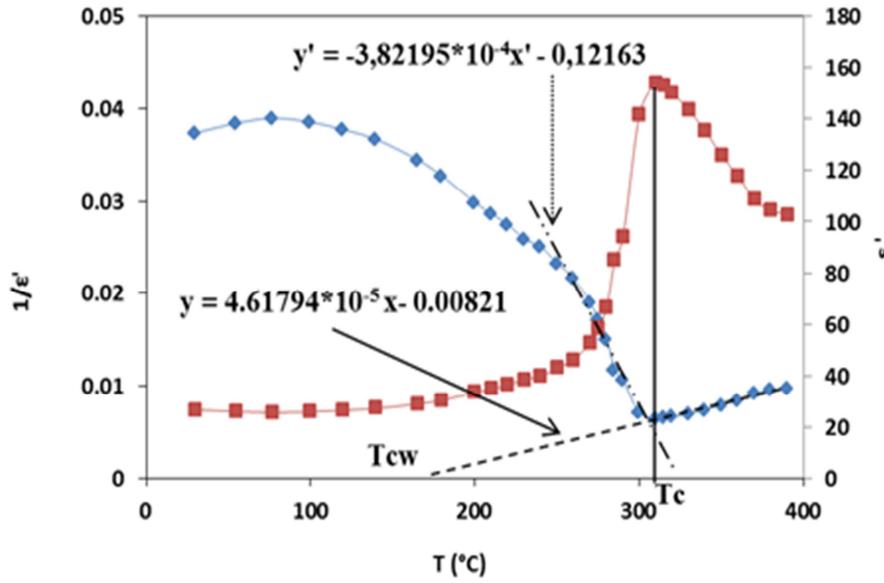


Fig 4. Curie–Weiss fitting of dielectric constant above and below T_c at 10 kHz

The plot of conductivity as a function of temperature are depicted in Figure 5, at low temperatures, the conductivity is seen to be independent of temperature, but shows different values at various frequencies indicating that main contribution to the conductivity result from the presence of space charges. As the temperature increases, the conductivity increase and a mere slope change are observed around T_c . The activation energy value calculated using the Arrhenius equation is found to be 0.56 eV at 10 kHz.

$$\sigma = \sigma_0 \exp\left(\frac{-E_a}{k_B T}\right) \tag{3}$$

Where σ is the conductivity, E_a the activation energy and k_B is Boltzmann constant.

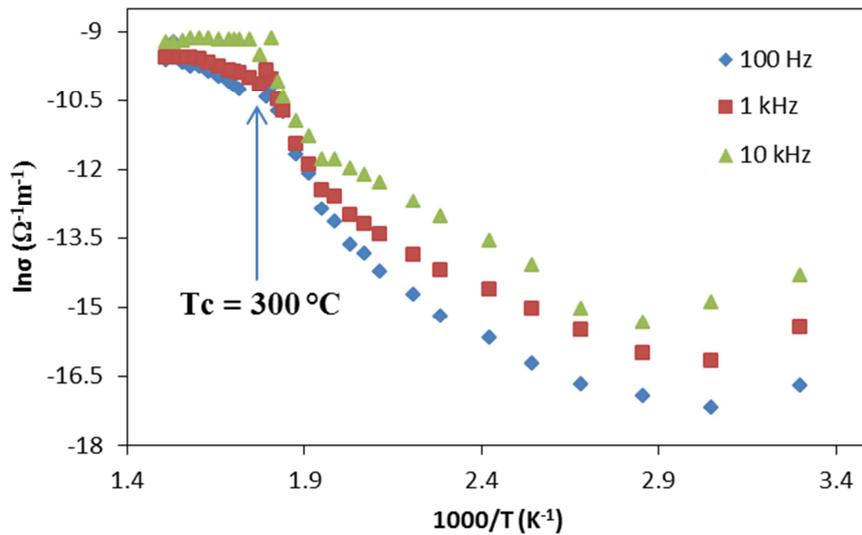


Fig 5. Arrhenius plots for the conductivity of SBT

4 CONCLUSION

A simple co-precipitation technique is described for the preparation powders of SrBi₂Ta₂O₉. XRD confirms that a single-perovskite phase SrBi₂Ta₂O₉ powder. The FT-IR in spectra dictates the presence of the tracers of nitrate ions. The parameters of the Curie–Weiss fitting, suggest the characteristic of the first order ferroelectric transition.

REFERENCES

- [1] Panda, A. Tarafdar, A. Pathak, and P. Pramanik, "Chemical synthesis and characterization of nanocrystalline ABi₂Ta₂O₉ (A = Sr, Ba and Ca) powders," *Ceram. Int.*, vol. 30, pp. 715-720, Mar. 2004.
- [2] Li, L. Li, and X. Wang, "Sintering behavior of bulk SrBi₂Ta₂O₉ prepared by solid-state reaction," *Ceram. Int.*, vol. 29, pp. 351-353, Mar. 2002.
- [3] W. Wang, H. Ke, J. C. Rao, M. Feng, and .Y. Zhou, "Sol–Gel synthesis of SrBi₂Ta₂O₉ nanowires," *J. Alloys Compd.*, vol. 504, pp. 367-370, Aug. 2010.
- [4] B. Panda, A. Tarafdar, and P. Pramanik, "Synthesis, characterization and properties of nano-sized SrBi₂Ta₂O₉ ceramics prepared by chemical routes," *J. Eur. Ceram. Soc.*, vol. 24, pp.3043–3048, Sept. 2004.
- [5] H. Lu, and Y. C. Chen, "Sintering and decomposition of ferroelectric layered perovskites: Strontium bismuth tantalate ceramics," *J. Eur. Ceram. Soc.*, vol. 19, pp. 2909–2915, Dec. 1999.
- [6] S.R. Dhage, Y.B. Kholam, S.B. Deshpande, and V. Ravi, "Co-precipitation technique for the preparation of nanocrystalline ferroelectric SrBi₂Ta₂O₉," *Mater. Res. Bull.*, vol. 38, pp. 1601–1605, Oct. 2003.
- [7] M. Jussara, S.d. Rocha, M. Costa, C. Filho, K. Rhaissa B. Theophilo, J. C. Denardin, I. Frota de Vasconcelos, E. Borges de Araujo, and A. Sergio B. Sombra, "Ferrimagnetism and Ferroelectricity of the Composite Matrix: SrBi₂Nb₂O₉(SBN)_x-BaFe₁₂O₁₉(BFO)_{100-x}," *Mater. Sci. Appl.*, vol. 3, pp. 6-17, Dec. 2012.
- [9] F.F. Oliveira, S. Da Dalt, V.C. Sousa, and C.P. Bergmann, "Synthesis of SrBi₂Ta₂O₉ by solution combustion and its characterization," *Powder Technol.*, vol. 225, pp. 239–243, July. 2012.
- [10] E.N. Muhamad, R. Irmawati, A.H. Abdullah, Y.H. Taufiq-Yap, and S.B. Abdul Hamid, "Effect of number of washing on the characteristics of copper oxide nanopowders," *Malaysian J. Anal. Sci. Spectros.*, Vol. 11, pp. 294–301, 2007.
- [11] J. Ortiz-Landeros, M. E. Contreras-García, and H. Pfeiffer, "Tailored Microporous ZrO₂-Al₂O₃ Mixed Oxides by Template-Assisted Method: Novel Materials for Catalytic Applications," *A Zojomo.*, vol. 4, July. 2008.
- [12] Coondoo, A.K. Jha, and S.K. Agarwal, "Enhancement of dielectric characteristics in donor doped AurivilliusSrBi₂Ta₂O₉ ferroelectric ceramics," *J. Eur. Ceram. Soc.*, Vol. 27, pp. 253–260, July. 2006.
- [13] S. Swain, P. Kumar, and R. B. Choudhary, "Electrical and ferroelectric studies of the 2-layered SrBi₂Ta₂O₉ based ceramics," vol. 477, pp. 56–63, Nov. 201.