BARIUM TITANATE BASED ACOUSTIC SENSING ELEMENTS

N. Motsi[1,a], N. Siafakas[1], A.Mamalis[2,b], E.Hristoforou[1,c]

[1]*School of Electrical and Computer Engineering, National Technical University of Athens, Iroon Polytechniou 9, 15780 Zografou, Greece,* [a]*nefelimotsi@mail.ntua.gr*, [c]*hristoforou@ece.ntua.gr*
[2]*Project Center for Nanotechnology and Advanced Engineering (PC-NAE), NCSR Demokritos, Athens, 15310, Greece,* [b]*agmamalis@yahoo.com*

Abstract. Materials with the ability to generate voltage when subjected to mechanical stress, such as PZT and $BaTiO_3$ are used in various applications such as supercapacitors, dielectrics ceramics and catalysts. Barium titanate ($BaTiO_3$) possesses various advantageous properties in terms of its structure, piezoelectricity and ferroelectricity. Yet, to further enhance these properties, barium was substituted with strontium using both sintering and sol-gel methods. Barium strontium titanate (BST) ferroelectric materials have attracted significant interest due to their chemical stability, high permittivity, excellent tunability and minimal dielectric losses. This study demonstrates the synthesis of perovskite $Ba_xSr_{(1-x)}TiO_3$ materials, with varying strontium content ranging from 0% to 50%, using the sintering and the sol-gel processes. In addition, the samples were examined using scanning electron microscopy (SEM), X-ray diffraction (XRD), differential thermal analysis (DTA) and thermogravimetry analysis (TGA).

1. Introduction

Barium titanate ($BaTiO_3$), a commonly used piezoelectric ceramic material, possesses a high dielectric constant and low dielectric loss, which render it an exceptional choice for electronic applications and piezoelectric sensors [1-2]. By replacing barium with strontium in $BaTiO_3$ through sintering and sol-gel techniques, the development of Barium Strontium Titanate (BST) material aimed to improve its sensitivity and response. The properties of BST ceramics, such as ferroelectricity and dielectricity, are affected by factors including sintering conditions, grain size, porosity, doping amount, and structural defects [3]. The enhancement of dielectric properties can be achieved by optimizing the doping and sintering conditions. BST is a solid solution that combines barium titanate ($BaTiO_3$) and strontium titanate ($SrTiO_3$), and its Curie temperature varies depended on the Ba/Sr ratio. To determine the most effective approach, this study synthesized $Ba_xSr_{(1-x)}TiO_3$ with different Sr contents and analyzed their structure and properties.

2. Experimental procedure and sample preparation

To synthesize $Ba_xSr_{(1-x)}TiO_3$ materials via sintering, a powder mixture consisting of barium carbonate ($BaCO_3$), strontium carbonate ($SrCO_3$), and titanium dioxide (TiO_2) was used. The mixture was ball milled in distilled water, and then the samples underwent Differential Thermal Analysis (DTA) to identify the temperatures at which the reactions resulting in the formation of $BaCO_3$ and $SrCO_3$ occur. Subsequently, the calcined powders were subjected to cold isostatic pressing at a pressure of 200 MPa, forming discs with a weight of 3g, a diameter of 3cm, and a width of 1.5mm. These discs were then sintered in an air atmosphere at 1500°C for 4 hours.

For the production of $Ba_xSr_{(1-x)}TiO_3$ using the sol-gel method, the raw materials consisted of barium acetate ($Ba(CH_3COO)_2$), strontium acetate ($Sr(CH_3COO)_2$), and titanium isopropoxide ($Ti(OC_3H_7)_4$). Solvents such as acetic acid (CH_3COOH), acetylacetone ($CH_3COCH_2COCH_3$), and propylene glycol ($C_3H_8O_2$) were used, along with distilled water as an additional agent. Initially, the solutions were subjected to calcination at 120°C to form xerogels. Subsequently, the xerogels were analyzed using Thermogravimetric Analysis (TGA) and Differential Thermal Analysis (DTA). The xerogels were then sintered in an air atmosphere at 750°C for 90 minutes, with a heating rate of 5°C/min. Following this, the calcined xerogels were cold isostatically pressed at a pressure of 200 MPa, resulting in the formation of discs weighing 3g, with a diameter of 3cm and a width of 1.5mm. Finally, the disks were sintered at 1500°C for 2 hours.

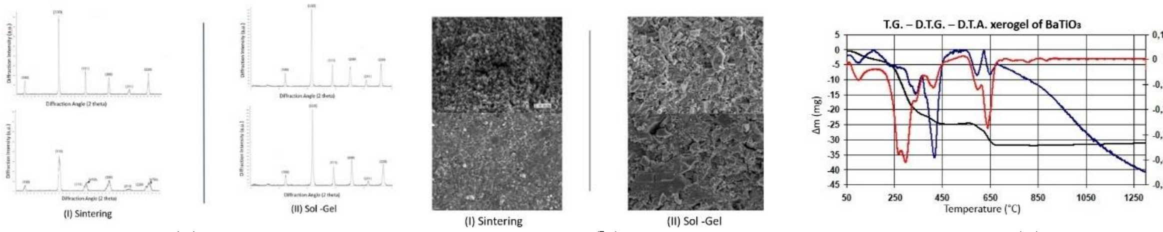
All the samples produced via sintering and sol-gel method, were examined by X-ray diffraction (XRD) and scanning electron microscopy (SEM).

3. Results of characterization techniques

Based on the XRD graphs presented in Figure 1(a), it can be observed that all the samples produced through the sintering and the sol-gel methods exhibit a perovskite structure. The predominant crystalline orientation is directed towards the [111] crystallographic axis across various Sr concentrations. It is evident that the formation of barium-strontium titanate has been successfully achieved. Yet, when examining the samples with 30% Sr and 50% Sr in $Ba_xSr_{(1-x)}TiO_3$ prepared via the sintering process, the presence of a $SrTiO_3$ peak indicates an unsuccessful blending of $BaCO_3$ and $SrCO_3$ powders prior to the calcination step.

Figure 1(b) illustrates SEM micrographs of piezoelectric materials produced by both the sintering and the sol-gel methods. One the one hand, the sintering process results in materials that possess uniform microstructures and the formation of necks between particles. On the other hand, samples produced via the sol-gel method exhibit nonuniform microstructures with the presence of aggregations. However, as the amount of Sr increases in the barium-strontium titanate (BST) material, the microstructure appears to improve and become more uniform.

Figure 1(c) displays that at temperatures above approximately 160°C, water and other liquids undergo vaporization. At temperatures below 300°C, the deposits of organic solutions onto xerogel powders vaporize. The peaks observed in the DTA curve correspond to the deconstruction of carboxyls and alkoxides. Within the temperature range of 300°C to 400°C, the formation of carbides and oxides (such as $BaCO_3$ or TiO_2) occurs, followed by the eventual production of the desired perovskite structure of BST through the sol-gel method. Between 450°C and 550°C, the plateau in the plot signifies a loss of mass, which correlates with the peaks observed in the DTA curve.



(a) (b) (c)

Fig. 1 Characterization techniques: (a) XRD diagrams, (b) SEM micrographs and (c) T.G. - D.T.G. - D.T.A of $Ba_xSr_{(1-x)}TiO_3$ piezoelectric materials.

Conclusions:

- The formation of barium-strontium titanate has been successfully accomplished.
- The Differential Thermal Analysis (DTA) indicates that the process for creating $BaTiO_3$ occurs at a temperature of 950°C. Additionally, the formation of $SrTiO_3$ takes place at a higher temperature of 1050°C.
- Sintered BST materials exhibit a more consistent microstructure and the formation of necks between particles, whereas samples produced by the sol-gel method display a nonuniform microstructure with the presence of aggregations.
- Based on the XRD analysis, samples synthesized using the sol-gel method demonstrate superior crystallographic properties compared to those obtained through sintering.

References:

[1] Duong N.X., Bae J.S., Jeon J., Lim S.Y., Oh S. H., Ullah A., ... & Kim T.H. Polymorphic phase transition in $BaTiO_3$ by Ni doping, Ceramics International, (2019), **45(13)**, 16305-16310.

[2] Mi L., Zhang Q., Wang H., Wu Z., Guo Y., Li Y., ... & Qi X. Synthesis of $BaTiO_3$ nanoparticles by sol-gel assisted solid phase method and its formation mechanism and photocatalytic activity, Ceramics International, (2020), **46(8)**, 10619-10633.

[3] Reddy S. B., Rao K. P., & Rao M. R. Effect of La substitution on the structural and dielectric properties of $BaZr_{0.1}Ti_{0.9}O_3$ ceramics. Journal of alloys and compounds, (2009), **481(1-2)**, 692-696.