

Technological regulation for creating a ceramic target from BaTiO₃ gel powders doped with Sn. Temperature dependence of the dielectric permittivity.

Lyuben Lakov¹, Mihaela Aleksandrova¹, Vladimir Blaskov¹, Yordan Marinov², Veselin Petkov¹

¹Bulgarian Academy of Sciences, Institute of Metal Science, Equipment and Technologies with Hydro- and Aerodynamics Centre "Acad. A. Balevski", Shipchenski Prohod Blvd. 67, 1574 Sofia, Bulgaria, e-mail: mihaela.krasimirova@mail.bg

²Bulgarian Academy of Sciences, Institute of Solid State Physics "G. Nadjakov", 72 Tzarigradsko Chaussee, Blvd., 1784 Sofia, BULGARIA

Summary: The research is aimed at creating a target from Sn doped barium-titanate ceramics. A technological regulation for the preparation of test bodies from a material synthesized by a low-temperature sol-gel method has been developed. The physicochemical parameters of the samples were determined and the temperature dependence of the dielectric permittivity was monitored. Physicochemical characterization of the samples was performed by X-Ray diffraction (XRD), and scanning electron microscopy (SEM). The preparation of pure BaTiO₃ cubic phase was established. The dielectric permittivity of Sn doped barium-titanate possesses a maximum value of 45000 at a Curie temperature- T_c 40°C.

Keywords: BaTiO₃, SOL-GEL, CERAMIC TARGET, DOPED, DIELECTRIC PERMITTIVITY

1. Introduction

Modern consumer electronics are being miniaturized quickly. In recent decades, the progress of microelectronics has been remarkable, as a result - all components of the electronic device are miniaturized. For the most of them this creates a problem, but for capacitors whose capacity value depends directly on the size, it is a major problem. [1-3]

In order to overcome this problem, manufacturers have realized two approaches: 1) introduction of dielectric materials with high relative dielectric permittivity to increase the capacity, 2) creation of a new capacitor architecture, which will not only reduce the technological parameters, but will also increase the overall capacity of the device.[2-4]

The typical capacitor consists of two conductive plates and a non-conductive dielectric material. The dielectric material separates the two conductive metal electrode plates. The application of voltage to the electrode plates of the capacitor generates an electric field in the non-conductive dielectric material that which stores energy. The dielectric permittivity, also known as relative dielectric permittivity, is a measure of a material's ability to store electricity and is one of the main properties of dielectric material.[5,6]

1.1 Effects of the dielectric permittivity on the characteristics of a capacitor

The capacitor stores energy when an electrical voltage is applied. The amount of electrical energy stored by the capacitor depends on the dielectric material and is influenced by the polarization generated with applied voltage. Materials with high dielectric permittivity can store more energy than those with low dielectric permittivity. The electrical susceptibility of a material is a measure of the polarizability origin from as a result of applied electric field. Good dielectric materials have a high electrical susceptibility. [7-10]

The dielectric permittivity is one of the main parameters that must be taken into account in the case of choosing a dielectric material for a capacitor. This permittivity is measured in farads per meter and determines the magnitude of the capacitance that the capacitor can achieve. Dielectric materials with high dielectric permittivity are applied when high capacitance values are required, although, as mentioned above, other parameters that determine the capacitance

of the capacitor include the distance between the electrodes and the effective area of the plate.[1-6]

The dielectric permittivity of a dielectric material in relation to relative that of a vacuum is noted the relative dielectric permittivity and is usually denoted by ϵ_r . The following equation connects the absolute dielectric permittivity (ϵ_0), the relative dielectric permittivity (ϵ_r) and the dielectric permittivity of a material (ϵ).

$$\epsilon_r = \epsilon / \epsilon_0 \quad (1)$$

1.2 Dielectric constants of ordinary dielectric materials

All materials are able to store electricity as a result of the effects of the electric field. Storage capacity varies from one material to another. The dielectric permittivity of materials is usually given with respect assigned to the dielectric constant in vacuum, denoted by ϵ_0 .

$$\epsilon_r = \epsilon / \epsilon_0$$

Vacuum dielectric constant is known as absolute dielectric constant and refers to the amount of resistance required to generate an electric field in a vacuum. The absolute dielectric constant in vacuum is approximately $8.85418782 \times 10^{-12} \text{ m}^{-3} \text{ kg}^{-1} \text{ s}^4 \text{ A}^2$.

The dielectric constant of a dielectric material relative to that of a vacuum is called the relative dielectric constant or dielectric constant and is usually denoted by ϵ_r . The following equation connects the absolute dielectric constant (ϵ_0), the relative dielectric constant (ϵ_r) and the dielectric constant of a material (ϵ).

$$\epsilon_r = \epsilon / \epsilon_0$$

The table below shows the dielectric relative permittivity of commonly used dielectric materials.[7-9]

It can be seen that relative dielectric permittivity of Sr doped barium titanate ceramics is significantly higher than the other materials.

Changes in temperature influence significantly on the dielectric permittivity of a given material. For example, an increase in temperature induces sharply decrease in dielectric permittivity at a freezing temperature.

Table 1 The dielectric relative permittivity of commonly used dielectric materials

| Material | Dielectric relative permittivity |
|---------------------------|----------------------------------|
| Air | 1.0006 |
| Aluminum Oxide | 8.5 |
| Barium Strontium Titanate | 500 |
| Ceramic porcelain | 4.5 - 6.7 |
| Glass | 3.7 - 10 |
| Mica | 5.6 - 8 |
| Paper | 3.85 |
| Polyester PET | 3.3 |
| Polypropylene | 2.25 |
| Tantalum oxide | 27.7 |

Nowadays, the miniaturization of the electrical circuit is a subject of intensive studies. Smaller components are required for the production of miniature circuits. Materials with high dielectric permittivity are used in case of smaller capacitors.

2. Experimental part and discussion

The main developments in the present work include the synthesis of barium titanate phases and the preparation of test specimens in order to determine the dielectric permittivity of the material.

The Sn doped BaTiO₃ phase was synthesized by low-temperature sol-gel method. The modes of monophases preparation were presented in previous publications. The test samples were obtained according to a certain technological regulation in the following sequence:

Stabilized sols and gels with desired properties are obtained by sol-gel technology. The mixtures for pressing of the necessary samples are prepared using 10% polyvinyl alcohol. The pressing is carried out in metal molding equipment. The samples are placed in a ceramic vessel with α-Al₂O₃ backfill and thermally treated to 1000°C for 3 hours in order to burn the plasticizer and biscuit the product.

The final sintering is realized at 1250°C in order to achieve the necessary strength. After cooling, additional machining follows until the test samples are obtained. (Fig. 1)



Fig.1. The sample of a ceramic target from doped with Sn BaTiO₃ gel powders.

Some physicochemical parameters of the test samples are presented in the following tables. Table 2 presents data concerning test specimens of Sn doped BaTiO₃ before sintering at 1250°C.

Table 2. Presents data concerning test specimens of Sn doped BaTiO₃ before sintering at 1250°C.

| Diameter [mm] | Thickness [mm] | Mass [mg] |
|----------------|----------------|-----------|
| 17,91 | 2,38 | 2856 |
| 17,63 | 2,35 | 2856 |
| 17,65 | 2,28 | 2856 |
| 17,71 | 2,36 | 2856 |
| Average 17,725 | Average 2,3425 | |

One of the samples shows the best results after annealing and its physical parameters are measured - Table 3.

Table 3. The best results after annealing and its physical parameters

| Diameter [mm] | Thickness [mm] | Mass [mg] |
|---------------|----------------|-----------|
| 18,81 | 2,21 | 3183 |
| 18,98 | 2,25 | 3183 |
| 18,92 | 2,27 | 3183 |
| 18,90 | 2,30 | 3183 |
| Average 18,91 | Average 2,26 | |

The density of the test specimens ($\rho = 5.02 \text{ mg} / \text{mm}^3$) is characteristic for the cubic and tetragonal phase of pure BaTiO₃, obtained by solid phase synthesis. [9-10]

Table 4. Thermal treatment of the test samples of Sn doped BaTiO₃ ceramics test samples

The next step in the study is to determine the dielectric permittivity of the sample

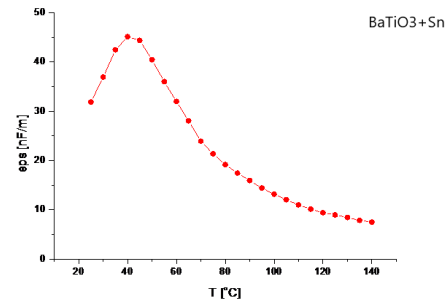


Fig.2 Determination of the relative dielectric permittivity of Sn doped BaTiO₃.

In Fig. 2 shows the dependence of the relative dielectric permittivity of Sn doped BaTiO₃ on the temperature at which the measurement is performed. The resulting curve is typical of ferromagnetic material. The high value of relative permittivity 45000 is impressive. The Curie temperature (Kc) at which the transition from para electric to ferroelectric (40°C) takes place is significantly lower than in comparison to the untreated BaTiO₃ (120°C).

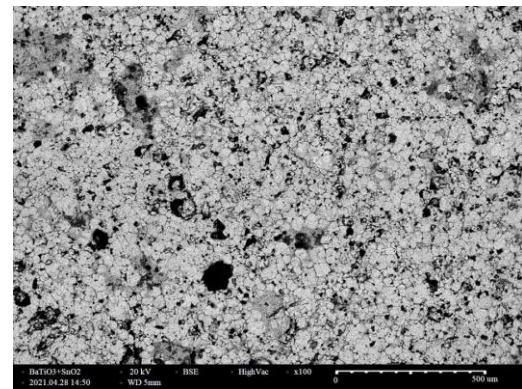
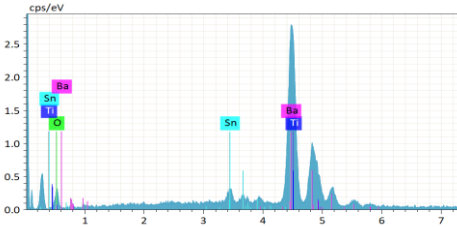


Fig.3 SEM of Sn doped BaTiO₃ phases of ceramic target



| Element | At. No. | Netto | Mass [%] | Mass Norm. [%] | Atom [%] | abs. error [%] (1 sigma) | rel. error [%] (1 sigma) |
|---------|---------|-------|----------|----------------|----------|--------------------------|--------------------------|
| Ba | 56 | 16346 | 53.23 | 59.23 | 25.56 | 1.56 | 2.93 |
| Ti | 22 | 8904 | 21.59 | 24.02 | 29.74 | 0.68 | 3.14 |
| O | 8 | 955 | 10.19 | 11.34 | 42.01 | 2.36 | 23.19 |
| Sn | 50 | 1966 | 4.86 | 5.41 | 2.70 | 0.22 | 4.46 |
| Sum | | 89.88 | 100.00 | 100.00 | | | |

The very good coincidence of the weight (mass percentages of Sn) determined by EDS (5.41) and the calculated ones (5.419) is impressive.

3. Conclusion

Sn doped barium titanate ceramics, with a very high value of dielectric constant and Curie temperature (Kc) close to room temperature was synthesized by sol-gel method. The ceramics thus obtained is promising for use as a supercapacitor in miniature electronic devices, and could also be used in electric vehicles. Sn

| temperature | temperatue value | soaking | temperature value | time for reaching | temperatur e value |
|----------------|------------------|----------------|-------------------|-------------------|--------------------|
| C ₁ | 120 | t ₁ | 60 | u ₁ | 120 |
| C ₂ | 700 | t ₂ | 120 | u ₂ | 300 |
| C ₃ | 1000 | t ₃ | 120 | u ₃ | 120 |
| C ₄ | 50 | t ₄ | 30 | u ₄ | 420 |

doped titanium ceramics possesses many times higher dielectric constant than non-doped BaTiO₃ at a significantly lower Curie temperature.

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