Effect of temperature on the structural properties of barium titanate nanopowders synthesis via sol-gel process

MOHAMMED TIHTIH ■ Institute of Ceramics and Polymer Engineering, University of Miskolc, Hungary ■ medtihtih@gmail.com

ALEKSEI V. PONARYADOV ■ Institute of Geology, Komi Science Center, Ural Branch of the Russian Academy of Sciences, Russian Federation ■ avponaryadov@geo.komisc.ru

JAMAL ELDIN F. M. IBRAHIM ■ Institute of Ceramics and Polymer Engineering, University of Miskolc, Hungary ■ jamalfadoul@gmail.com

EMESE KUROVICS ■ Institute of Ceramics and Polymer Engineering, University of Miskolc, Hungary ■ fememese@uni-miskolc.hu

ELENA L. KOTOVA ■ Saint-Petersburg Mining University, Russian Federation ■ kotova_el@pers.spmi.ru

LÁSZLÓ A. GÖMZE ■ Institute of Ceramics and Polymer Engineering, University of Miskolc, Hungary, IGREX Engineering Service Ltd ■ femgomze@uni-miskolc.hu

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Abstract

Sol-gel processed barium titanate (BaTiO_3) ceramic has been characterized by X-ray diffraction (XRD) and Fourier Transformation Infrared (FT-IR) techniques. The results confirm that BaTiO_3 crystallizes in the perovskite structure with Ba_2TiO_4 pyrochlore phase when calcined at relatively low temperature (800 °C) for 3h. Furthermore, XRD reveals the formation of a single-phase perovskite BaTiO_3 without presence of secondary phases when calcined at the temperature of 900 and 1000 °C with the following lattice parameters: $a = 3.9978\text{Å}$, $c = 4.0197\text{Å}$ and $a = 3.9967\text{Å}$, $c = 4.0220\text{Å}$ respectively. the calcination temperature is confirmed to has a large influence in BaTiO_3 structure, several parameters including, the tetragonality, volume (V) and crystallite size were found to be increased with increasing calcination temperature.

Keywords: Sol-gel, Barium titanate nanoceramics, Calcination temperature, X-ray Diffraction.

Kulcsszavak: Szol-gél, bárium-titanát nanokeramika, kalcinációs hőmérséklet, röntgendiffrakció.

1. Introduction

The popularity of the research in the area of high-tech ceramics has attracted a great interest lately [1-22]. Barium titanate (BaTiO_3) is the most frequently utilized ferroelectric material, and even after seventy-five years of its disclosure, it remained as the most essential material which has excellent dielectric, optical, piezoelectric and ferroelectric properties [23]. Since the discovery of the phenomenon of ferroelectricity, BaTiO_3 has been a standard material in the family of ferroelectric perovskites. This compound has been used for a long time in many industrial sectors such as Multilayer Ceramic Capacitors (MLCCs) [24]. An interesting and very current application of BaTiO_3 concerns the creation of computer memories FRAMs (Ferroelectric Random Access Memories) [25]. The manufacture of thermistors and the detection of polluting gases such as CO also constitute applications of BaTiO_3 . The properties, both optical and electrical, have proved to be very promising technologically, for example, the linear resonator, the sensors, the actuators, the phasing of laser sources, spatial light modulators, the guides of waves, holographic storage and high filters frequencies etc...) [26]. However, multilayer capacitors have long occupied the first place in BaTiO_3 applications.

Also, it is one of the most extensively studied lead-free ferroelectric materials due to its broad range of device applications including, capacitors, transducers, non-volatile memories, positive

Mohammed TIHTIH

Is graduated from Sidi Mohamed Ben abdellah University /Faculty of sciences Dhar El Mahraz, Fez, Morocco, Department of Physics, for the time being, he is a PhD student in the University of Miskolc, Institute of Ceramics and Polymer Engineering, under supervision of Prof. L. A. Gömze

Aleksei V. PONARYADOV

Is a Researcher of Laboratory of Technology of Mineral Raw, Institute of Geology, Komi Science Center, Ural Branch of the Russian Academy of Sciences. Author and co-author of more than 30 scientific articles. Russian Mineralogical Society

Jamal Eldin F. M. IBRAHIM

Is a lecturer in the University of Bahri, Khartoum, Sudan, he graduated from University of Marmara, Istanbul, Turkey, Institute of Pure and Applied Sciences, Department of Metallurgical and Materials Engineering, for the time being, he is a PhD student in the University of Miskolc, Institute of Ceramics and Polymer Engineering, under supervision of Prof. L. A. Gömze

Emese KUROVICS

Is graduated from the University of Miskolc, Department of Ceramics and Silicate Engineering as a material engineer, where she continues her study as PhD student under supervision of Prof. L. A. Gömze.

Elena L. KOTOVA

Is a Scientific Director of the Mining Museum of the Saint-Petersburg Mining University. Author and co-author of chapter in book, patent and more than 30 articles. The member of the Russian Mineralogical Society, Oil and Gas Historical Society of Russia

László A. GÖMZE

Is establisher and professor of the Department of Ceramics and Silicate Engineering in the University of Miskolc, Hungary. He is author or co-author of 2 patents, 6 books and more than 300 scientific papers.

temperature coefficient (PTC) thermistors and many more. Due to the importance of BaTiO_3 which meet the requirements of miniaturization of integrated circuits in the field of electronics, new preparation techniques have been developed and successfully used to produce BaTiO_3 at lower temperatures compared to the solid path, which results in an appreciable gain in energy and enable the production of cost-effective metal or alloy electrodes. Among these techniques: it's worth mentioning the sol-gel method [27-29], the hydrothermal method [30] and the co-precipitation [31]. Through these methods a high purity BaTiO_3 with very fine and controllable particles size can be obtained. Furthermore, it is possible to easily modify the physical properties of BT since it is very sensitive to the doping on Ba site, Ti site and or a substitution (co-doping) coupled to the two sites, which enable their adaptations to very specific applications. In this work, BT materials were synthesized using sol-gel method and after it was calcined at temperatures of 800 °C, 900 °C, and 1000 °C. The aims of this research work are to determine the effects of calcining temperatures on Physical parameters such as lattice parameters, crystallite size and bands absorption of BT using X-ray diffraction (XRD) and Fourier Transformation Infrared (FT-IR).

2. Experimental procedure

In the present work, BaTiO_3 was prepared using sol-gel method. Barium acetate ($\text{Ba}(\text{C}_2\text{H}_3\text{O}_2)_2 \cdot \text{H}_2\text{O}$, Aldrich, 99.9%) and titanium alkoxide ($\text{Ti}[\text{OCH}(\text{CH}_3)_2]_4$, Aldrich, 97.9%) have been used as precursors, lactic acid ($\text{CH}_3\text{CH}(\text{OH})\text{CO}_2\text{H}$, Aldrich, 90%) was used as peptizing agent, acetic acid was added to dissolve the barium acetate, and distilled water as a solvent. First, the titanium alkoxide was added to lactic acid aqueous solution and H_2O with continuous stirring at 70°C . After 24 hours of reaction, the obtained white precipitate is transformed into clear homogeneous solution. In the second step the barium acetate was added to this colloidal solution in stoichiometric quantities. The obtained nanopowders were calcined in air for 3hrs at 800, 900 and 1000°C with a heating rate of $5^\circ\text{C}/\text{min}$. The different processing steps for the synthesis of BaTiO_3 nanopowders is shown in Fig. 1.

1. ábra A BaTiO_3 nanoporok szintézisének folyamatábrája
Fig. 1 Flow chart of synthesis of BaTiO_3 nanopowders

3. Results and discussion

3.1 X-ray Diffraction

The XRD analysis of the prepared samples was carried out at room temperature using (XPRT-PRO) diffractometer with $\text{CuK}\alpha$ ($\lambda = 1.5406 \text{ \AA}$) radiation. The XRD patterns of the calcined pure BT powders recorded at 800°C , 900°C and 1000°C are presented in Fig. 2. It is clearly observed that the XRD diffractogram of the powder prepared at 800°C is characterized by the appearance of the Ba_2TiO_4 pyrochlore phase [32]. Moreover, at temperatures of 900°C and 1000°C , the disappearance of the pyrochlore phase and the appearance of the pure perovskite phase are noticed. However, the analysis of the XRD spectra shows the juxtaposition of the two peaks (002) and (200) at the position $2\theta = 44.93^\circ$ for the temperature of 800°C (Fig. 3). When the calcination temperature increases ($T = 1000^\circ\text{C}$) the two peaks separate. The presence of these two peaks shows a transition to the quadratic phase based on the temperature [33].

2. ábra A BaTiO_3 nanorészecskék röntgendiffrakciós spektrumai különböző hőmérsékleteken: 800°C , 900°C és 1000°C

Fig. 2 X-ray diffraction spectra of BaTiO_3 nanoparticles at different temperatures: 800°C , 900°C and 1000°C

3. ábra A (002) és (200) csúcsok XRD mintája

Fig. 3 XRD patterns shifting of the peak (002) and (200)

To better highlight the effect of the calcination temperature on the structural properties of BT, the lattice parameters, as well as the crystallite size for each calcination temperature were calculated. The prominent peaks were used to estimate the size of the nanoparticles through the Scherrer equation, expressed as follows [34]:

$$D = \frac{0.9\lambda}{\beta \cos \theta}$$

Where β is the full width at half maximum (FWHM) of the (101) peak, θ is the diffraction angle and λ is the X-ray wavelength ($\lambda = 1.542 \text{ \AA}$).

The evolution of the lattice parameters a and c , tetragonality, the volume (V) of the unit cell, and the crystallite size as a function of the calcination temperatures are listed in Table 1 and represented in Fig. 4. This evolution is in agreement with those obtained by Teraoka et al [35] and Lombardo et al [36];

a higher calcination temperature results in the formation of larger crystallite size. In particular, the increase in particle size is relatively large for the sample calcined at 1000 °C. In Table 1, the estimated average size is 26.1455 nm and 32.4634 nm for the samples calcined at 800 and 1000 °C respectively. The characteristics of the crystallite size and the volume (V) of the unit cell have shown great similarity which indicates that the defects stimulated by the temperature can highly influence all the physical behaviours of BaTiO₃.

Annealed temperature (°C)	a=b (Å)	c (Å)	c/a	Volume (Å) ³	Crystallite size (nm)	Phase / Space group
800	4.0021	4.0107	1.0021	64.2385	26.1455	-
900	3.9978	4.0197	1.0054	64.2444	29.6457	Tetragonal/ P4mm
1000	3.9967	4.0220	1.0063	64.2458	32.4634	Tetragonal/ P4mm

1. táblázat Az BaTiO₃ egységcellák paraméterei, kristály mérete és szerkezete a hőmérséklettől függően

Table 1 Unit cell parameters, Crystallite size and structure phase of BaTiO₃ versus temperature

4. ábra A BaTiO₃ nanorészecskék rács paraméterei és kristály mérete a hőmérséklet függvényében

Fig. 4 Variation of the lattice parameters and the crystallites size of BaTiO₃ nanoparticles as a function of temperature

3.2 FT-IR analysis

5. ábra A BaTiO₃ nanorészecskék FT-IR spektrumai különböző hőmérsékleteken: 800 °C, 900 °C és 1000 °C

Fig. 5 FT-IR spectrums of BaTiO₃ nanoparticles at different temperatures: 800 °C, 900 °C and 1000 °C.

Fig. 5 shows the infrared curves of barium titanate calcined at different temperatures. it can be noticed that these spectra present a set of absorption bands: a wide band of high-intensity located between 3700 and 2500 cm⁻¹ (3369 cm⁻¹) at 800 °C, as well as a fine peak located at 1772 cm⁻¹ of low-intensity indicating the presence of molecular water in the samples. The two more intense peaks located around 441 and 543 cm⁻¹ and the low-intensity peak correspond to the symmetrical stretching of the Ti-O bond [37], the characteristic of these peaks confirm the formation of pure tetragonal phase. These results are in agreement with the literature [38]. The intensity peak at 859 cm⁻¹ corresponds to a Ba-O elongation [39]. The two peaks located around 1440 and 1433 cm⁻¹ correspond to the symmetrical stretch of the C-O bond and the Ti-O-C bond respectively [37]. Note that the intensity of the peaks located at around 1772, 1644 and 859 cm⁻¹ decreases with increasing calcination temperature, this indicates that the barium titanate preparation was completed at 1000 °C.

4. Conclusions

Nanopowders of BaTiO₃ perovskite materials have been successfully prepared by using the sol-gel method at a temperature of 800 °C, 900 °C and 1000 °C. The prepared samples were characterized using XRD and FT-IR spectroscopy. The prepared samples were calcined at 900 °C, XRD analysis of the samples shows the presence of a single tetragonal phase without any secondary phase. At 1000 °C a slight distortion occurred, as showed on the determined unit cell parameters calculated from the corresponding XRD spectrum, which gave rise to the single tetragonal perovskite phase (space group P4mm). The observed change in the structural properties of BaTiO₃ was also examined via FT-IR spectrum and then confirmed by XRD analysis that show a gradual change in the recorded spectra in the temperature range of 800 °C to 1000 °C.

The disappearance of the absorption bands indicated the formation of BaTiO₃ structure and shows the effect of annealed temperature on the structural properties of BaTiO₃.

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