PHYSICAL CHEMISTRY 2006

Proceedings

of the 8th International Conference on Fundamental and Applied Aspects of Physical Chemistry

September 26-29,
Belgrade, Serbia

ISBN	86-82139-26-X
Title:	Physical Chemistry 2006. (Proceedings)
Editors	Prof. dr A. Antić-Jovanović
Published by:	The Society of Physical Chemists of Serbia, Studentski trg 12-16, P.O.Box 137, 11001 Belgrade, Serbia
Publisher:	Society of Physical Chemists of Serbia
For publisher:	Prof. dr S. Anić, president of the Society of Physical Chemists of Serbia
Printed by:	"Jovan" Printing and Published Comp; 250 Copies; Number of Pages: x + 442; Format B5; Printing finished in September 2006.
Text and Layout:	Aleksandar Nikolić

250 – copy printing

ULTRASOUND MODIFICATIONS OF PHYSICAL PROPERTIES OF $BaTiO_3$ POWDERS

S. Marković[1], Lj. Kandić[1], M. Mitrić[2], G. Starčević[3] and D. Uskoković[1]

[1]*Institute of Technical Science of the Serbian Academy of Sciences and Arts, Knez Mihailova 35/IV, Belgrade, Serbia (smilja@ffh.bg.ac.yu)*
[2]*Institute of Nuclear Sciences "Vinča", Belgrade, Serbia*
[3] *NIS – Oil Refinery, Pančevo, Serbia*

Abstract

The starting $BaTiO_3$ powder was prepared by solid state reaction, and then modified by ultrasound into powders with different physical characteristics. The prepared barium titanate powders were characterized by X-ray powder diffraction (XRD), laser particle size analyzer, also, specific surface area and powders density were determined. The studies indicate that physical properties of $BaTiO_3$ powders were changed by sonochemical treatment. However, tetragonal crystal structure was maintained. It is shown that sonochemical method can be used for easy modification of barium titanate powder characteristics.

Introduction

Due to its ferroelectric and dielectric properties $BaTiO_3$ based materials have many applications in electronic devices such as multilayer ceramic capacitors, pyroelectric detectors, ferroelectric memories, sensors and positive temperature coefficient (PTC) thermistors [1].

The importance of the starting powders characteristics on the properties of the final ceramics is very well known from the literature [2]. The classical route to prepare barium titanate powder is the solid state reaction between $BaCO_3$ and TiO_2 at temperature interval 1000-1200 °C. This is a widely used method for large bath processing for barium titanate based powders. Powders prepared by solid state reaction are agglomerated and aren't highly reactive toward sintering. A narrow particle size distribution and a small average size may be achieved by powders milling before pressing and sintering. During milling impurities can be introduced into system. Instead milling, sonochemical treatment can be used for powders activation.

In this study, we prepare barium titanate powder by solid state reaction, and then modified them by sonochemical treatment into powders with different physical characteristics. We investigated powders crystal structure, density, particle size, crystallite size, particle size distribution and specific surface area.

Experimental

The $BaTiO_3$ (BT) starting powder was prepared by conventional solid state reaction. Mixture of $BaCO_3$ and TiO_2 was homogenized for 24 hours in ethanol, dried and calcined at 1100 °C for 2 h. After calcinations, starting barium titanate powder was modified by ultrasound treatment. The powder was dispersed in isopropanol and treated for

10, 60 and 180 minutes, with high-intensity ultrasound radiation using a direct-immersion titanium horn (Sonics VCX – 750, 20 kHz, 750 W). After the ultrasound process was stopped, the powders were filtered and dried.

The crystal structure of the barium titanate powders were investigated by XRD measurements, obtained on Philips PW-1050 automatic diffractometer using Cu K_α radiation. The diffraction measurements were done over scattering angle from 20 to 120 °2θ with a step of 0.02° and a counting time of 15s. The FullProf program was used for structural refinement. The average particle size and particle size distribution were determined in isopropanol using a laser particle size analyzer (Mastersizer 2000, Malvern Instruments Ltd, UK). The specific surface area (BET) and pore size of powders were measured by N_2 adsorption-desorption isotherms at -195.8 °C on a Micromeritics® analyzer, while powders density were measured by pycnometry in hexane.

Results and Discussion

After XRD measurements, we noticed that all powders were high crystalline, pure barium titanate with tetragonal symmetry. The crystallite size in the BT powders were calculated from the half-width of the XRD peaks of (002) and (200) planes, using by the Scherer's equation, and are shown in Table 1, as well as the tetragonality (*c*/*a* ratio). It can be noticed that these values are very similar, i.e. negligibly dependent on duration of ultrasound treatment. Fig. 1 shows Rietveld refinement of starting $BaTiO_3$ powder structure.

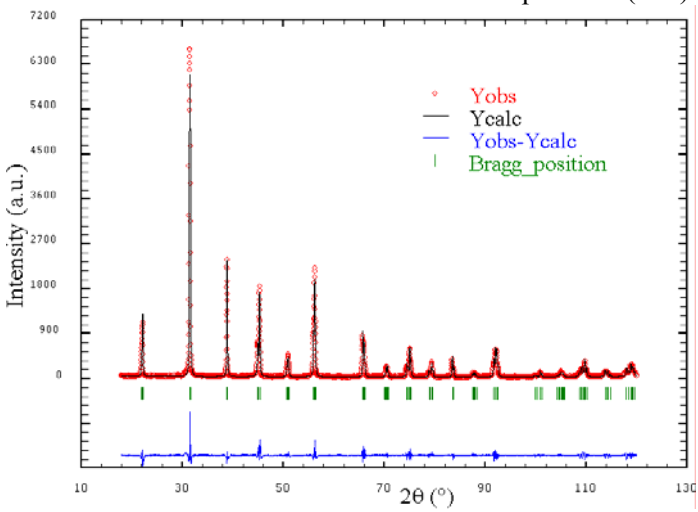

Fig. 1. Observed (◦ Y_{obs}), calculated (- Y_{calc}) and residual (lower) XRD of starting $BaTiO_3$ powder.

Fig. 2 shows the particle size distribution of $BaTiO_3$ powders (distribution based on numbers). The particle size distribution of the starting barium titanate powder, prepared by solid state reaction, was very narrow, and average particle size was 1.399 μm. It is noticeable that during ultrasound treatment particles are de-agglomerated, and average particle sizes decreases, while, the width of the particle size distribution is broadened. Values of average particle size, specific surface area and powders density are shown in Table 1.

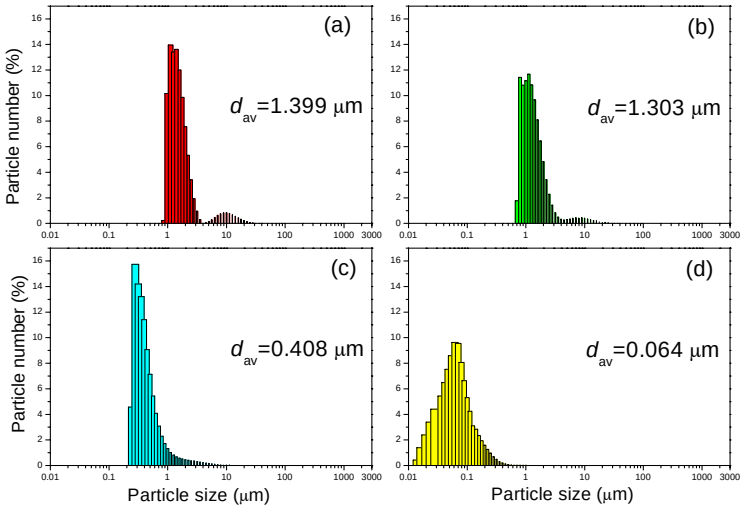

Fig. 2. Particle size distribution of $BaTiO_3$ powders (distribution based on number). Sonication time: (a) 0; (b) 10; (c) 60 and (d) 180 min.

Table 1. Characteristics of $BaTiO_3$ powders

Sonication time (min)	*c*/*a*	Crystalite size (Å)	Density (g/cm^3)	Average particle size (μm)	Specific surface area (m^2/g)
0	1.0077	**(002)** 318.6 **(200)** 695.7	5.58	1.399	1.01
10	1.0077	**(002)** 307.4 **(200)** 635.1	5.54	1.303	4.61
60	1.0077	**(002)** 327.4 **(200)** 645.3	5.63	0.408	4.79
180	1.0079	**(002)** 293.3 **(200)** 562.1	5.06	0.064	1.31

Results of our investigations indicated that using by ultrasound irradiation we could improve physical characteristic of $BaTiO_3$ powder previously prepared by solid state reaction, without changing of powder's crystal structure. After three hours of ultrasound treatment of tetragonal $BaTiO_3$ we obtained nanometer-sized, uniformly distributed particles, suitable for preparation of high density ceramics, and consequently, for ceramics with good dielectric properties.

Acknowledgments

The Ministry of Science and Environmental Protection of the Republic of Serbia provided financial support under grant no. 142006.

References

[1] S, Gablenz, C. Damm, F.W. Müller, G. Israel, M. Rössel, A. Röder and H.-P. Abicht, Solid State Science, 2001, **3,** 291-299.

[2] T. Tsurumi, T. Sekine, H. Kakemoto, T. Hoshina, S.-M. Nam, H. Yasuno, S. Wada, J. Amer. Ceram. Soc., 2006, **89** [4], 1337-1341.