ELSEVIER

Ceramics International 33 (2007) 297–299

CERAMICS
INTERNATIONAL

www.elsevier.com/locate/ceramint

Short communication

A co-precipitation technique to prepare $BaTa_2O_6$

S.C. Navale [a], Violet Samuel [b], A.B. Gaikwad [c], V. Ravi [a,*]

[a] Physical and Materials Chemistry division, National Chemical Laboratory, Pune 411008, India
[b] Catalysis division, National Chemical Laboratory, Pune 411008, India
[c] Center for Materials Characterization, National Chemical Laboratory, Pune 411008, India

Received 11 July 2005; received in revised form 20 July 2005; accepted 29 August 2005
Available online 1 December 2005

Abstract

A mixture of ammonium oxalate and ammonium hydroxide was used to co-precipitate barium and tantalum ions as barium oxalate and tantalum hydroxide under basic conditions. On calcination at 750 °C, this precursor yielded $BaTa_2O_6$ (BT) ceramics. This is much lower temperature as compared to that prepared by traditional solid-state method (1000 °C) as reported for the formation of $BaTa_2O_6$. Transmission electron microscope (TEM) investigations revealed that the average particle size is 70 nm for the calcined powders. The room temperature dielectric constant at 1 kHz is found to be 100.

Keywords: B. Electron microscopy; Ceramics; Oxides; X-ray diffraction; Barium tantalate

1. Introduction

$Sr_xBa_{1-x}Nb_2O_6$ ($0.25 < x < 0.75$) (SBN) ceramics are ferroelectric materials with tungsten bronze type structure, which have excellent electro-optic, pyroelectric and photorefractive applications [1–5]. However, there are not much studies [6–8] reported on barium or strontium tantalum oxides in the literature. There was a report [6] for $BaTa_2O_6$ (BT) as a possible photocatalyst for water purification. The crystal structure of BT is orthorhombic and the reported lattice parameters are $a = 12.33$ Å, $b = 10.26$ Å and $c = 7.67$ Å. Recently, we have been working on these materials [1–5] and here we communicate a simple coprecipitation procedure to prepare $BaTa_2O_6$ ceramics at low temperatures. The advantages of wet chemical techniques over conventional high temperature ceramic processing such as small particle size of the resultant powders, large surface area and compositional homogeneity are well known in the literature. Among chemical methods, co-precipitation is one of the more successful techniques for synthesizing ultrafine ceramic powders having narrow particle size distribution. This process can avoid complex steps such as refluxing of alkoxides, resulting in less time consumption compared to other techniques. The only drawback of co-precipitation process is that required cations should have similar solubility product.

2. Experimental

For preparing BT, tantalum (V) oxide and barium chloride were used as the starting materials, which were of AR grade (LOBA cheme). Ta_2O_5 was dissolved in minimum amount of HF after heating at hot water bath for 10 h. To this TaF_5 solution, required quantity of $BaCl_2{\cdot}2H_2O$ is added and mixed well. A mixture of ammonium oxalate and ammonium hydroxide is added dropwise to precipitate niobium and barium as hydroxide and oxalate respectively. The pH was maintained around 9 to ensure completion of the reaction. Then, the precursors powder precipitated was filtered and oven dried. The dried powders are calcined at different temperatures ranging from 400 to 1000 °C for 6 h. Various techniques such as XRD (Philips PW 1710 diffractometer) and TEM were employed to characterize these powders. For lattice parameter and interplanar distance (*d*) calculation, the samples were scanned in the 2θ range of 10–80° for a period of 5 s in the step scan mode. Silicon was used as an internal standard. Least squares method was employed to determine the lattice parameters.

The TEM picture was recorded with JEOL model 1200 EX instrument at the accelerating voltage of 100 kV. The fine

---

* Corresponding author. Tel.: +91 20 5893300; fax: +91 20 5893044.
E-mail address: ravi@ems.ncl.res.in (V. Ravi).

0272-8842/$32.00 © 2005 Elsevier Ltd and Techna Group S.r.l. All rights reserved.
doi:10.1016/j.ceramint.2005.08.012

powders were dispersed in amyl acetate on a carbon coated TEM copper grid. For comparison, BT samples are also prepared by ceramic method. The barium carbonate and tantalum oxide are taken in stoichiometric ratio and mixed, ground several times and heated at 1000 °C for 24 h. The calcined powders (from both precursor derived and ceramic method) were mixed with few drops of 1 wt.% solution of poly vinyl alcohol and pelletized at 1–2 tons. The green pellets were sintered at 1300 °C for 2 h. The densities of the sintered samples are measured by Archimedes technique. The surfaces of the sintered pellet were polished and electroded with low-temperature curing silver paint. A LCR meter was used to measure the dielectric constant at 1 kHz.

3. Result and discussion

Fig. 1 shows the XRD pattern of $BaTa_2O_6$ powder formed after calcining the precursor at 750 °C. The crystal structure of BT is orthorhombic and all the d-lines pattern match with reported values (JCPDS: 20–146). The calculated lattice parameters by least square fit (two decimal accurate) are $a = 12.353$ Å, $b = 10.275$ Å and $c = 7.672$ Å. This is the lowest temperature reported so far for the formation of $BaTa_2O_6$ by coprecipitation technique. The particle size and morphology of the calcined powders were examined by transmission electron microscopy. Particle morphology of calcined powder (750 °C for 6 h) prepared by this technique was irregular in shape and agglomerated, with an average primary particle size around 70 nm (Fig. 2).

The crystallite size calculated from Scherrer's formula ($t = K\lambda/B\cos\theta_B$), where t is the average size of the particles, assuming particles are spherical, $K = 0.9$, λ is the wavelength of X-ray radiation, B is the full width at half maximum of the diffracted peak and θ_B is the angle of diffraction) is 80 nm. Conventional solid-state method also forms $BaTa_2O_6$ phase at 1000 °C after prolonged heating (24 h) with comparatively larger particle size of ~1 μm (Fig. 3). The densities of all the sintered samples are above 92% of the single crystal values. It is well known that dielectric properties obtained depends on sinter-density, grain size and defects present in the sample. The room temperature dielectric constant measured at 1 kHz is 100 for the specimens prepared by the wet chemical method as compared to 80 for specimens prepared by traditional solid-state method.

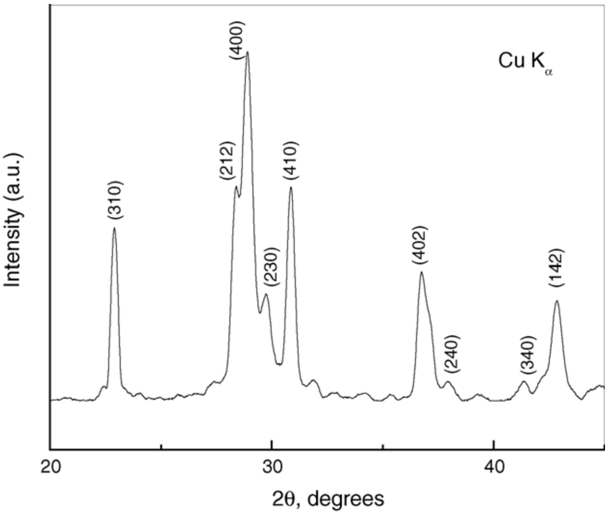

Fig. 1. XRD of barium tantalate after annealing the precursor at 750 °C.

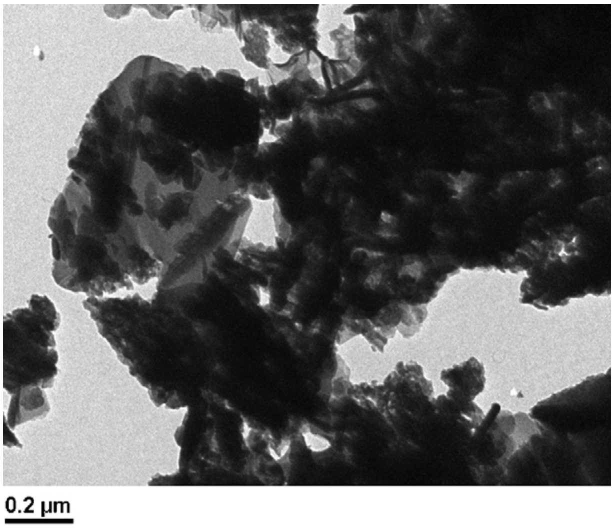

Fig. 2. TEM micrograph of barium tantalate powders prepared by coprecipitation and calcined at 750 °C.

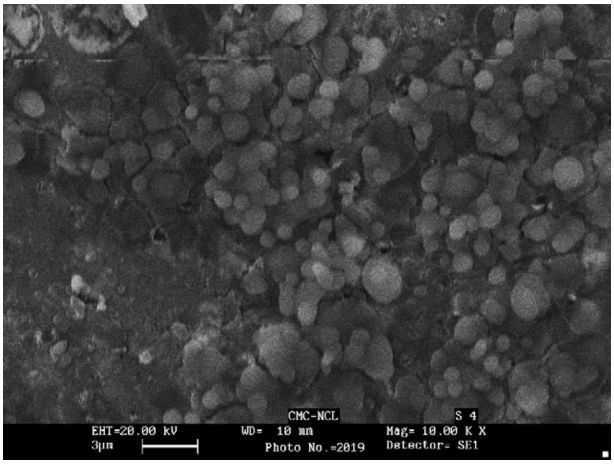

Fig. 3. TEM micrograph of barium tantalate powders prepared by the high temperature ceramic method.

4. Conclusions

A simple coprecipitation process is adopted for the preparation of ultrafine powders of $BaTa_2O_6$. The average particle size and morphology of thesr powders were investigated by TEM.

Acknowledgement

One of the authors (V.R.) acknowledges the Department of Science and Technology, (DST) India, grant no. SP/S1/H-19/2000 for financial assistance.

References

[1] S.P. Gaikwad, V. Samuel, R. Pasricha, V. Ravi, Mater. Lett. 58 (2004) 3699.

[2] N. Natarajan, V. Samuel, R. Pasricha, V. Ravi, Mater. Sci. Eng. B 117 (2005) 169.

[3] V. Ravi, Mater. Character. 55 (2005) 92.

[4] V. Ravi, S.C. Navale, Ceram. Inter., in press.

[5] R. Pasricha, V. Ravi, Mater. Chem. Phys. 94 (2005) 34.

[6] T.A. Vanderah, R.S. Roth, T. Siegrist, W. Fabo, J.M. Loezos, W.W. Ng, Solid State Sci. 5 (2003) 149.

[7] H. Kato, A. Kudo, Chem. Phys. Lett. 295 (1998) 487.

[8] Y.S. Kim, Y.H. Lee, M.Y. Sung, Solid State Electron. 43 (1999) 1189.