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Short communication

Microwave-hydrothermal synthesis of barium titanate under stirring condition

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Abstract

The role of in situ stirring under microwave-hydrothermal (M-H) conditions on the synthesis of barium titanate was investigated for the first time by powder X-ray diffraction and scanning and transmission electron microscopy. Stirring under M-H conditions in the temperature range of 150–200 °C led to enhanced crystallization of Ba titanate as revealed by yields compared to the static condition. In addition, stirring led to smaller and more uniform crystals under M-H conditions compared to those crystallized without stirring. Powder X-ray diffraction revealed the formation of only cubic polymorph of Ba titanate at or below 200 °C in 4 h with or without in situ stirring under M-H conditions. These results show that stirring is an important parameter during M-H synthesis of nanophase Ba titanate.

Keywords: Barium titanate; Cubic phase; Ferroelectric; Microwave-hydrothermal synthesis; Stirring

1. Introduction

Barium titanate is by far the largest amount of dielectric material used in the ceramic capacitor industry. It is also a ferroelectric material with piezoelectric properties although lead zirconate titanate (PZT) replaced Ba titanate because PZT has better piezoelectric properties. Ba titanate material is widely used in multi-layer ceramic capacitors (MLCCs), piezoelectric and ferroelectric devices, positive temperature coefficient (PTC) thermistors, field-effect transistors, etc. Ba titanate has been prepared by the traditional solid-state process, sol–gel process and many other techniques including the hydrothermal process [1–3]. An excellent review of the various BT synthesis techniques was previously published [1]. Among these, the hydrothermal process is ideally suited for preparing nanopowders of Ba titanate (BT) and in fact, hydrothermally produced BT has been commercially sold [4] for several years. Nanophase Ba titanate is a necessity for making compact MLCCs. The main advantages of the hydrothermal process are (a) it is a low temperature process, (b) it is an environmentally benign process as the processing is done under closed system conditions and (c) it is easy to control the size and shape of the particles. The conventional-hydrothermal process has been extensively used in single crystal as well as powder preparation for over a century because of the above advantages [5,6]. Over the years, there are a few modifications to the conventional-hydrothermal (C-H)

Table 1
Microwave-hydrothermal (M-H) and conventional-hydrothermal (C-H) syntheses of $BaTiO_3$.[a].

Reaction	Yield[b] (%) of $BaTiO_3$	
	No stirring	Stirring at 157 rpm
M-H 150 °C—10 min	90.7	96.4
M-H 150 °C—60 min	91.3	97.7
M-H 180 °C—10 min	86.7	91.9
M-H 180 °C—30 min	89.5	94.5
M-H 180 °C—120 min	95.9	96.4
M-H 200 °C—15 min	93.1	97.2
M-H 200 °C—2 h	–	97.4
M-H 200 °C—4 h	–	97.9
C-H 200 °C—24 h	96.6	–
C-H 200 °C—48 h	97.2	–

[a] Only cubic phase formed in all cases.
[b] Yield (%) is calculated as the weight ratio of product obtained to that expected upon complete crystallization.

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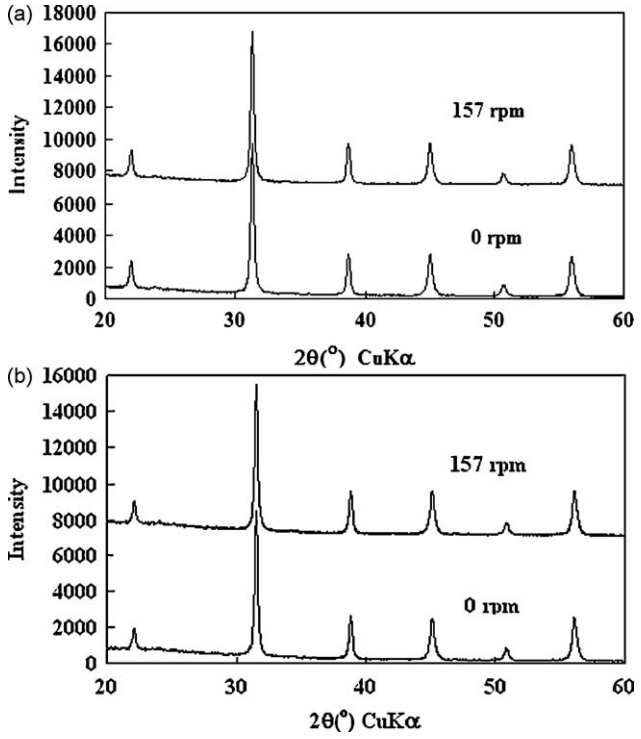

Fig. 1. XRD patterns of $BaTiO_3$ powder prepared at 150 °C (a) and 200 °C (b) for 4 h with and without stirring using microwave-hydrothermal process.

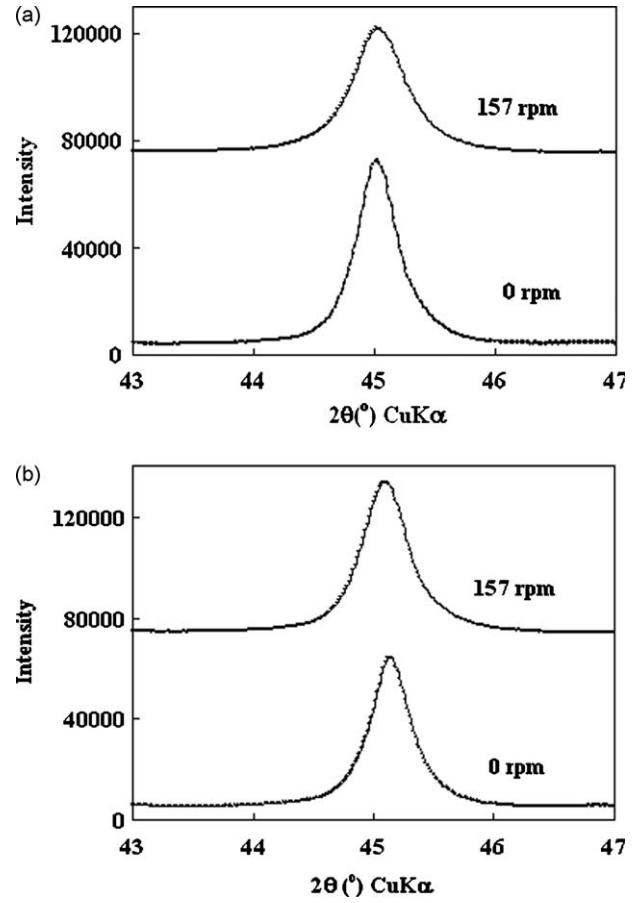

Fig. 2. (0 0 2) XRD pattern of cubic $BaTiO_3$ prepared at 150 °C (a) and 200 °C (b) for 4 h with and without stirring treatment under microwave-hydrothermal conditions.

process involving the addition of microwave [7], electric [8], and ultrasonic [9] fields to the hydrothermal vessels to enhance the reaction rates. Among these, the addition of microwaves to the hydrothermal system became popular starting in the eighties and the term ''microwave-hydrothermal'' (M-H) process was coined by us and documented in a publication in 1992 [7]. Although C-H process was used for the synthesis of Ba titanate as early as in 1970 [10–13], the M-H process was used by us for the first time in 1992 [7], which led to the synthesis of 100–200 nm Ba titanate particles. Subsequent studies by us and others [14–18] optimized the M-H process for Ba titanate synthesis. One of the disadvantages of the M-H process is the aggregation of particles because of extremely rapid crystallization [7,19,20] if no stirring or mixing was applied during synthesis. All the Ba titanate syntheses to date by the M-H process were done without stirring during the reaction [13–18] because stirred M-H reactors were not available until recently. With the advent of stirred M-H reactors, it would be useful to see the role of stirring on the M-H synthesis of Ba titanate because one recent study [21] showed that nanophase $BaTiO_3$ powders with narrow size distribution and high tetragonality could be synthesized by the rotary-hydrothermal process where the contents were mixed during C-H reaction. Thus the objective of this study was to investigate the

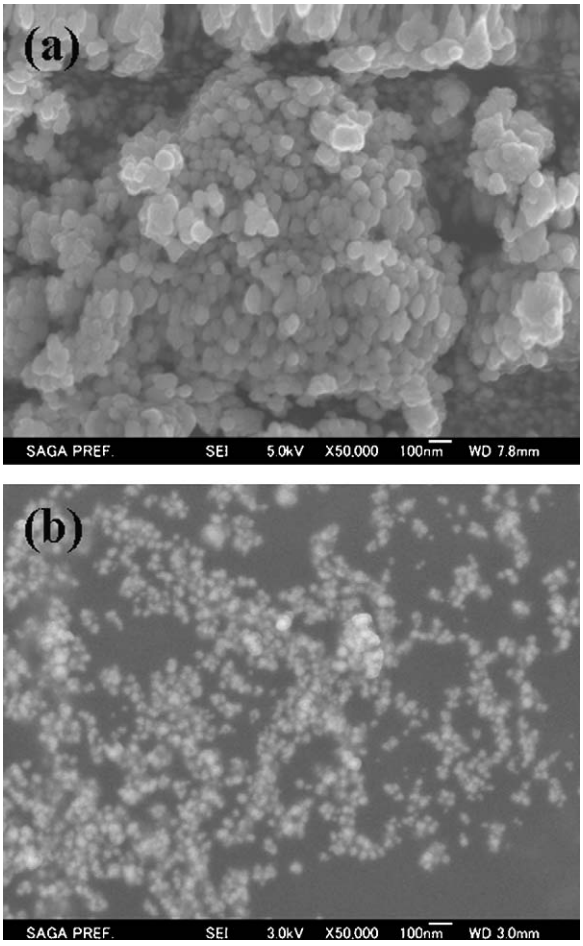

Fig. 3. Scanning electron micrographs showing the effect of stirring on the morphology of $BaTiO_3$ particles prepared at 180 °C for 4 h: (a) without stirring and (b) with stirring of 157 rpm.