Ceramics International **22** (1996) 305–307

0272-8842(95)00107-7
0272-8842/96/$15.00

Microwave Processing of $Ba_2Ti_9O_{20}$ Ceramic

Ion Teoreanu, Ecaterina Andronescu & Antoaneta Folea

University "Politehnica", Bucharest, Romania

(Received 6 April 1995; accepted 21 July 1995)

Abstract: Microwave synthesis of $Ba_2Ti_9O_{20}$ was realized. The dependence between microwave characteristics, the duration of treatment and the degree of transformation of initial reacting powder was examined. At the same time, features of samples sintered by microwaves were compared to those sintered by conventional methods (in electric furnaces).

1 INTRODUCTION

The development of knowledge related to thermal effects of microwave (MW) radiation leads to the extension of their applications in the fields of:

— dewatering, drying;
— production of high temperatures and the possibility of heat treatment to related temperatures.[1]

When conventional processing is used, the heat is transmitted to the surface of the sample and the energy is then transferred within the bulk by thermal conductivity. The heating requires long periods of time because the coefficients of thermal conductivity are small and in order to reduce thermal stresses. These methods of indirect heating lead to an important growth of crystal within the materials obtained in this way.

Microwave radiation is characterized by volume energy. Its conversion in thermal energy leads to a rapid heating of the whole mass of the solid body (in particular, ceramics). In this way it is possible to reduce thermal stresses and also the product's grain size.

2 CONSIDERATIONS ON MICROWAVE ENERGY USED FOR DIELECTRIC CERAMIC SYNTHESIS

Dielectric materials are able to absorb MW due to the polarisation that takes place within the material when an electromagnetic field is applied.

For MW frequencies (10^7–10^{13} Hz), two mechanisms of polarisation are important for MW heating, namely atomic or ionic polarisation and dipole (orientation) polarisation.

The polarization is delayed when an electric field is applied. This loss of kinetic energy is dissipated as heat.

The equation that governs the heating of a dielectric in an electric field is:[2]

$$P = 2\pi f \epsilon'' E^2$$

where P is the dissipated power, f is the frequency of the electric field, ϵ'' is the dielectric loss factor and E is the electric field within the dielectric.

It could be observed that the heating of a dielectric material using MW energy (the amount of power that can be dissipated within the body), for a given electric field intensity, depends directly on the dielectric loss of that material.

Previous research works[3–7] have mentioned the possibility for a wide range of oxide ceramic materials (with a high capacity to absorb MW energy and to convert it into thermal energy) to be heated and even melted using MW energy. Also, the sintering of $BaTiO_3$ by MW processing has been reported.[8]

The purpose of the present work is to initiate the study of MW synthesis of TiO_2-rich barium titanates, and also to compare them with compounds that were synthesized by conventional methods.[9]

3 CONDITIONS AND EXPERIMENTAL PROCEDURE

Until now, after the knowledge of authors, there have not been reported works on MW synthesis of $Ba_2Ti_9O_{20}$ ceramics. Due to this reason it was interesting to carry out a comparative study of $Ba_2Ti_9O_{20}$ material manufactured by conventional and MW processing.

By conventional processing, powders of $BaCO_3$ and TiO_2 were mixed according to $Ba_2Ti_9O_{20}$ stoichiometry. The samples were obtained by uniaxial pressing and sintered in an electric firing kiln at temperatures between 900 and 1400°C. The duration of heating up to the maximum firing temperature was between 6 and 10 h, depending on the value of sintering temperature. The samples were maintained at maximum temperatures for 3 h.

The $Ba_2Ti_9O_{20}$ specimen obtained by MW technique also started from $BaCO_3$ and TiO_2 powders, according to $Ba_2Ti_9O_{20}$ stoichiometry. The equipment used for this heat treatment consists of a microwave source (2450 MHz magnetron), a waveguide and a cavity resonator. The batch for the reaction, as powder, was placed in the centre of the cavity. The samples were exposed to different powers, for different periods of time (1–5 min).

The $Ba_2Ti_9O_{20}$ ceramic materials produced by conventional and MW techniques were characterized by X-ray diffraction (in order to get information regarding phase evolution and crystal symmetry) and optical microscopy (for microstructure determination).

4 EXPERIMENTAL RESULTS

The diffraction patterns (considered by the most important lines between $d = 3.18$ Å and $d = 2.97$ Å) for the samples prepared by the conventional method have shown that $Ba_2Ti_9O_{20}$ lines are present at 1000°C, together with those belonging to TiO_2. $Ba_2Ti_9O_{20}$ is stable up to 1400°C (Fig. 1).

$Ba_2Ti_9O_{20}$ synthesized using MW energy was identified by X-ray diffraction in samples obtained for incident powers between 800 and 1000 W. The temperature within the material was difficult to establish, although it could be shown to lie in the approximate interval between 1000 and 1300°C.

Figure 2 illustrates the formation of $Ba_2Ti_9O_{20}$ using MW energy, corresponding to 1000 W incident power. The amount of $Ba_2Ti_9O_{20}$ formed by using MW energy increases when the incident power increases from 800 to 1000 W (Fig. 3), and also when the reaction time, for the same incident power, is longer (Fig. 4).

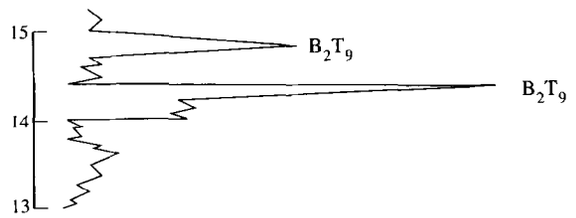

Fig. 1. $Ba_2Ti_9O_{20}$ synthesized by the conventional method. (Duration of heat treatment: 14 h).

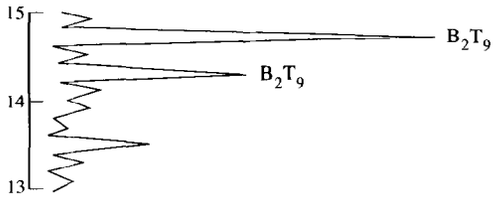

Fig. 2. $Ba_2Ti_9O_{20}$ synthesized by MW processing. (Duration of heat treatment: 5 min).

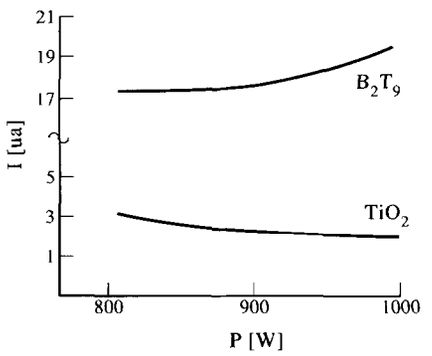

Fig. 3. Phase evolution ($Ba_2Ti_9O_{20}$ and TiO_2) versus power. (Time was constant: 5 min).

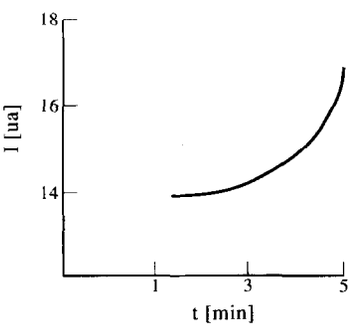

Fig. 4. Time influence on the amount of $Ba_2Ti_9O_{20}$ at 900 W.

It was observed that the amount of conventionally synthesized $Ba_2Ti_9O_{20}$ increased with the time of maintaining maximum temperature (from 1 to 3 h), and also with the increasing of temperature up to 1400°C.

For the ceramics synthesized by the MW technique, the microstructure changes due to the processing conditions (corresponding to the values of reaction time and incident powers) are illustrated in Fig. 5(a)–(e).

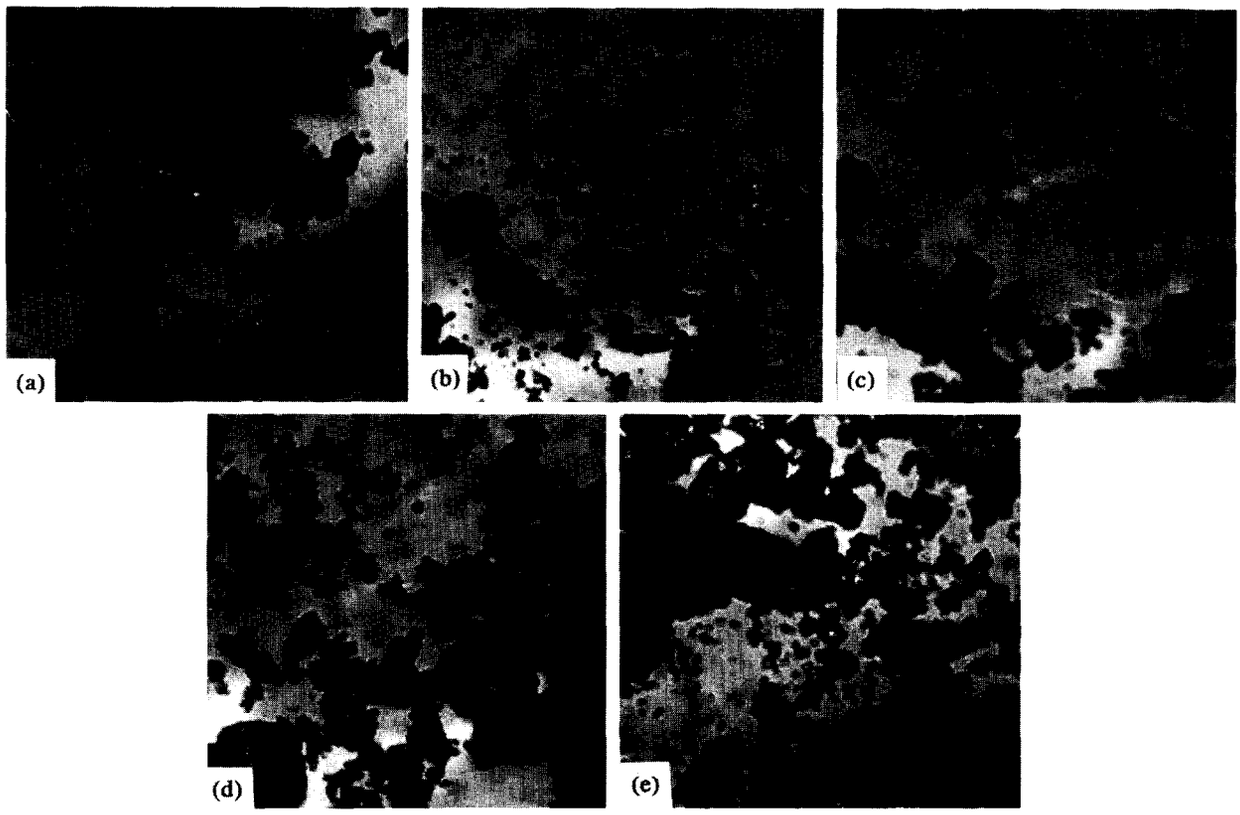

Fig. 5. Evolution of microstructure for different values of MW power and time. (a) 800 W-5 min; (b) 900 W-1 min; (c) 900 W-3 min; (d) 900 W-5 min; (e) 1000 W-5 min.

As was expected, for the same incident power, the increasing of reaction time leads to large grains. The same influence, less evident, was also observed when increasing incident MW powers, for the same reaction time (5 min). Probably this influence is more important when the reaction time is shorter.

5 CONCLUSIONS

The present work showed the possibility of $Ba_2Ti_9O_{20}$ synthesis using MW energy, when certain parameters are fixed. MW processing would allow an important decrease of the time required for the heat treatment and also produce a dense microstructure, with small grain size within the ceramic specimen.

It is also possible for other TiO_2-rich barium titanates to be obtained in the same way.

REFERENCES

1. BADOT, J. C., *Rev. Int. Hautes Temp. Refract.*, **19** (1982) 65–76.
2. HAMLYN, M., *Brit. Ceram. Rev.*, (83) (1990) 34–9.
3. FREUDENBERG, B. & MOCELLIN, A., *J. Am. Ceram. Soc.*, **70**(1) (1987) 33–8.
4. PILUSO, P., LEQUEUX, N. & BOCH, P., Microwave sintering of low-loss dielectric ceramics. In *Proc. 2nd Conf. European Ceramic Society*, Augsburg, Germany, 1991.
5. BOCH, P., LEQUEUX, N. & PILUSO, P., Reaction sintering of ceramic materials by microwave heating. In *Microwave Processing of Materials III*, ed. R. L. Beatty, W. H. Sutton & M. F. Iskander. Mat. Res. Proc. (Vol. **269**), Pittsburgh, PA, 1992, pp. 211–16.
6. QUEMENEUR, L., DESGARDIN, G. & RAVEAU, B., *Silicates Industriels*, **1–2** (1985) 7–15.
7. BOUIRDENE, A., LEFEUVRE, S. & AUDHUY, M., *L'Industrie Ceram.*, **862**(2) (1992) 98–100.
8. DESGARDIN, G., ALIOUAT, M., MAZO, L. & RAVEAU, B., *R.G.E.*, **7**(5) (1988) 434–6..
9. TEOREANU, I., ANDRONESCU, E., FOLEA, A. & CATOI, G., *Mat. de Constr.*, **20**(2/3) (1990) 139–41.