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Fişa suspiciunii de plagiat / Sheet of plagiarism's suspicion	
Opera suspicionată (OS)	**Opera autentică (OA)**
Suspicious work	**Authentic work**
OS — Ioachim Andrei, Toacşăn Mariana Irina, Banciu Marin Gabriel, Nedelcu Liviu, Dutu Constantin Augustin, Alexandru V. Horia, Antohe Stefan, Andronescu E, Jinga Sorin, Niţă Petru. Synthesis and properties of Ba (Zn 1/3 Ta 2/3) O3 for microwave and millimeter wave applications. *Thin Solid Films*. 2008 Feb 15;516(7):1558-1562. **Note**: a) the source of suspicion: www.integru.org. b) This work was supported by Ministry of Education and Research under contract CEEX No. 4 / 2005. c) The papers was RETRACTED by the editor.	
OA — Ioachim A, Toacşăn M.I, Banciu M.G, Nedelcu L, Duţu C.A, Feder M, Plăpcianu C, Lifei F, and Niţă P. Effect of the sintering temperature on the Ba (Zn 1/3 Ta 2/3) O3 dielectric properties. *Journal of the European Ceramic Society*. 2007 Dec 31;27(2):1117-1122. (Available on-line in 12 June 2006). Note: This work was supported by Ministry of Education and Research under contract nr: 09N/505.2003.	
Incidenţa minimă a suspiciunii / Minimum incidence of suspicion	
p.1559:02s – p.1559: 35s	p.1117:10d – p.1118:23s
p.1559:37s – p.1559:08d	p.1118:26s – p.1118:35s
p.1559:Fig.2	p.1118:Fig.1
p.1559:09d – p.1560:14s	p.1118:37s – p.1118:25d
p.1560:19d – p.1560:31s	p.1119:21s – p.1119:32s
p.1560:04d – p.1560:14s	p.1119:33s – p.1119:45s
p.1560:21d – p.1561:03s	p.1119:34s – p.1119:03d
p.1561:10s – p.1561:21s	p.1120:08d – p.1120:19d
p.1561:22s – p.1561:23d	p.1120:21d – p.1120:44d
p.1561:Fig.7	p.1121:Fig.9
p.1561:Fig.6	p.1121:Fig.7
p.1562:34d – p.1561:38d; p.1562:10s – 1562:03d	p.1121:06d – p.1122:05s
Fişa întocmită pentru includerea suspiciunii în Indexul Operelor Plagiate în România de la Sheet drawn up for including the suspicion in the Index of Plagiarized Works in Romania at www.plagiate.ro	

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Note: By „p.72:00" one understands the text ending with the end of the page 72. By „p.00:00" one understands the taking over from the initial point till the last page of the current chapter, entirely.

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Fişa de argumentare a calificării

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b) Plagiatul este definit prin textul legii[1].

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Tehnic, plagiatul are la bază conceptul de **piesă de creaţie** care[2]:

> „…este un element de comunicare prezentat în formă scrisă, ca text, imagine sau combinat, care posedă un subiect, o organizare sau o construcţie logică şi de argumentare care presupune nişte premise, un raţionament şi o concluzie. Piesa de creaţie presupune în mod necesar o formă de exprimare specifică unei persoane. Piesa de creaţie se poate asocia cu întreaga operă autentică sau cu o parte a acesteia…"

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> iii) Cele două opere conţin piese de creaţie identificabile comune care posedă, fiecare în parte, un subiect şi o formă de prezentare bine definită.
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> vii) In opera suspicionată se identifică un fir sau mai multe fire logice de argumentare şi tratare care leagă aceleaşi premise cu aceleaşi concluzii ca în opera autentică…"

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[1] Legea nr. 206/2004 privind buna conduită în cercetarea ştiinţifică, dezvoltarea tehnologică şi inovare, publicată în Monitorul Oficial al României, Partea I, nr. 505 din 4 iunie 2004

[2] ISOC, D. *Ghid de acţiune împotriva plagiatului: bună-conduită, prevenire, combatere*. Cluj-Napoca: Ecou Transilvan, 2012.

[3] ISOC, D. *Prevenitor de plagiat*. Cluj-Napoca: Ecou Transilvan, 2014.

Effect of the sintering temperature on the Ba(Zn$_{1/3}$Ta$_{2/3}$)O$_3$ dielectric properties

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Abstract

Preparation of dielectric materials with optimal properties for wireless applications requires special thermal treatments due to the difficult control of the cationic ordering. The BZT samples prepared by solid-state reaction were sintered at temperatures in the range 1400–1600 °C for 4 h. For compositional, structural and morphological characterization, XRD, SEM and EDX were employed. The order–disorder transition of the BZT material was analyzed. With the increase of the sintering temperature, a long-range order with a 2:1 ratio of Ta and Zn cations on the octahedral positions of the perovskite structure was observed. The dielectric parameters were measured in the microwave range and were correlated with morphological and structural properties. In order to improve the microwave properties, annealing treatment at 1400 °C for 10 h was performed. Sintering temperatures greater than 1550 °C are required for undoped BZT compositions, in order to obtain low microwave loss (Qf > 100,000). © 2006 Elsevier Ltd. All rights reserved.

Keywords: Tantalates; Sintering; Microstructure; Dielectric properties; Super-high—Qf

1. Introduction

Ba(Zn$_{1/3}$Ta$_{2/3}$)O$_3$ (BZT) is a perovskitic material of the A(B′B″)O$_3$ type, having ultra high values of the quality factor Q. It has potential for applications in satellite broadcasting at frequencies higher than 10 GHz and as a super-high-Q dielectric resonators (DR) in mobile phone base stations or combiner filter for PCS applications.[1] The BZT ceramics exhibit a dielectric constant of 29, a Qf product of 100,000–150,000 and a low temperature coefficient of resonant frequency (τ_f).

The factors influencing Q values of perovskites Ba(M$_{1/3}^{2+}$Ta$_{2/3}^{5+}$)O$_3$ (M = Mg, Zn) have been considered to be long-range ordering (LRO) of cations, zinc oxide evaporation, point defects and stabilization of microdomain boundaries.[2,3] This explained the high-Q values from the point of view of the lattice vibrations of its hexagonal superstructure. Sagala and Nambu[4] calculated the dielectric loss tangent at microwave frequencies from the equation of ion motions which was a function of B-site ordering. Gallasso and coworkers[5] concluded that the B-site ordering increased as the difference in charge and size between B″ and B″ atoms increased. The ordering of complex perovskite BZT is important because the 1/3 <1 1 1> (1:2) ordering is believed to be closely related to the high-Q property of BZT. There is a strong correlation between the cation ordering, domain growth, zinc loss and sintering parameters. In this paper we report the synthesis of a BZT material, its structural and morphologic characteristics as well as its microwave dielectric properties.

2. Experimental

Ceramics with molar formula Ba(Zn$_{1/3}$Ta$_{2/3}$)O$_3$ were prepared by solid-state reaction. The starting materials were BaCO$_3$, ZnO and Ta$_2$O$_5$. Stoichiometric quantities were weighted, ground, homogenized and milled in an agate mill in water for 2 h. The powders were calcined at T = 925 °C for 2 h. Then the powders were milled for 2 h and calcined at 1000 °C for 2 h. The calcined powders were mixed with 1% polyvinylalcohol (PVA) and dried at T = 80 °C were pressed into cylindrical samples of 12 mm diameter and 10 mm height. The pellets were slowly dried at 80 °C in order to eliminate the PVA. The third calcination was carried out at T = 1150 °C in air for 2.5 h.

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The density of green ceramics was $\rho = 4.4\ g/cm^3$. The sintering treatment was performed in air for 4 h at six temperature values: 1400, 1450, 1500, 1525, 1550 and 1600 °C. In order to increase the dielectric properties, especially the quality factor Q, the samples were treated supplementary for 10 h at 1400 °C. The pellets were polished to remove the superficial zone, which is porous and poor in Zn, in order to obtained correct values of the microwave dielectric parameters.

The relative bulk density of sintered cylinders was measured by the Archimedes method. Morphological, structural and compositional analyses were performed on six sets of samples by X-ray diffraction (XRD) analysis, and electron microscopy (SEM, EDX). The patterns were recorded in a 2θ range from 20° to 90° on a Seifert Debye Flex 2002 diffractometer into the $2\theta - \theta$ mode. Measurements were performed at room temperature using Cu Kα radiation, Ni filter and a detector scan speed of 7°/min. In order to obtain information on cation ordering state and unit cell distortion a detector scan speed of 5°/min was used.

The dielectric parameters, i.e. the dielectric constant ε, the quality factor Q and the loss tangent $\tan\delta$ were measured in the microwave range by Hakki-Coleman method. A computer aided measurement system containing a HP 8757 C scalar network analyzer and a HP 8350 B sweep oscillator was employed.

3. Results and discussion

3.1. Bulk density

The bulk density of the fired BZT ceramics was measured after grinding and polishing. The temperature dependence of densification after 4 h sintering in air is shown in Table 1. BZT ceramics sintered between 1400 and 1600 °C were well sintered.

The X-ray density of $Ba(Zn_{1/3}Ta_{2/3})O_3$ compound with stoichiometric composition was considered as $\rho_t = 7.92\ g/c^3$[6] The most dense ceramic presents a porosity value of $P = 10\%$. An abnormal grain growth together with an accentuated ZnO evaporation occurred above 1525 °C, so the bulk density slightly decreased.

3.2. X-ray diffraction analysis

The crystal structure of $Ba(Zn_{1/3}Ta_{2/3})O_3$ is of a classic ordered perovskite with two transition metals (B$'$ and B$''$) on the octahedral site. The B$'$ and B$''$ ions occur on alternating (1 1 1) planes in a 1:2 ratio. Each (1 1 1) plane contains only one type of small oxygen octahedron, and the planes are ordered in the 1:2 stoichiometric ratio, in the sequence Zn–Ta–Ta–Zn–Ta–Ta–.... The resulting symmetry, due to the (1 1 1) type plane stacking and accompanying small oxygen displacement strongly depends on processing parameters. So, the ordering of Zn and Ta depends on firing temperature and of the sintering time. The BZT sintered at lower temperatures ($T_s < 1300$ °C) exhibits a pseudo-cubic perovskite type structure where Zn and Ta are in disorder. At higher sintering temperatures ($T_s > 1300$ °C) BZT has a hexagonal perovskite type structure, with Zn and Ta showing 1:2 order in the B site. The order of Zn and Ta expands the original perovskite unit cell along the <1 1 1> direction and contracts the unit cell in the plane normal to <1 1 1>. Consequently, c/a has a value greater than $\sqrt{3/2} \approx 1.2247$ and the unit cell is distorted.

The increase of cation ordering on B site in BZT compounds creates a superlattice with a hexagonal structure. The degree of long-range-structural order (LRO) can be measured by X-ray methods and it can be expressed as the ratio between the intensity of the super structure reflection and that of a basic unit cell reflection. The X-ray diffraction patterns for BZT compound, sintered at six sintering temperatures, using Cu Kα radiation are given in Fig. 1. The X-ray patterns confirm the formation of the hexagonal structure, which is the majority phase. For the BZT compounds sintered at $T_s = 1500$ and 1525 °C the patterns reveal the presence of secondary phases $Ba_7Ta_6O_{22}$, Ba_8ZnTa_{24} and BaO, which disappear at higher sintering temperatures. The order structure was examined in the XRD patterns by observing

Table 1
Density vs. sintering temperature for BZT samples sintered in air for 4 h

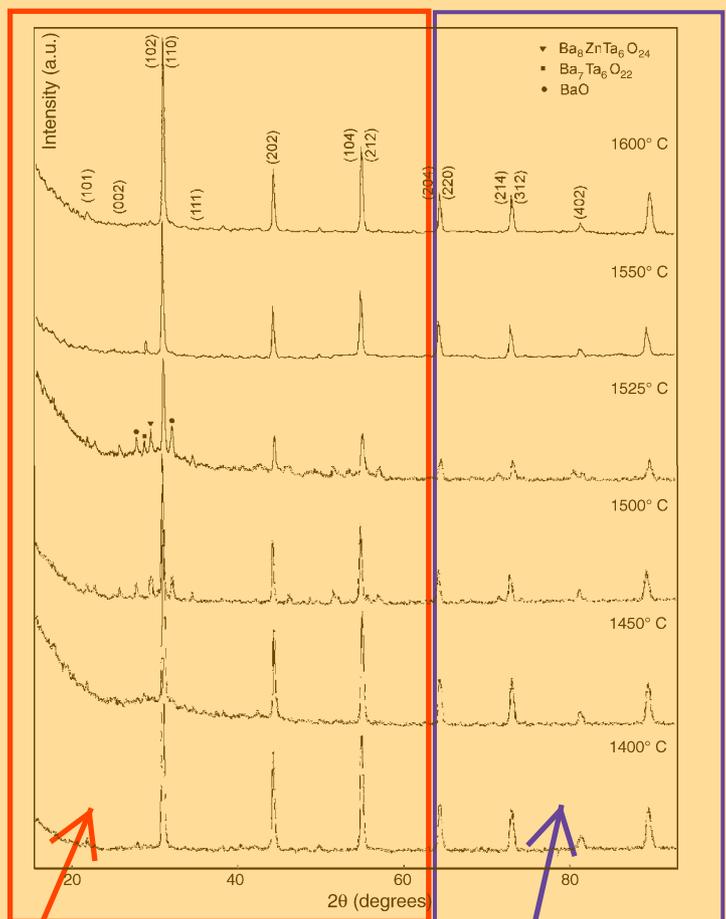| Sintering temperature (°C)	Bulk density ρ_0 (10^3 kg/m³)	Absorption capacity (%)	Porosity P (%)
1400	5.37	5.3	32.1
1450	6.22	2	21.4
1500	7.10	0.9	10.3
1525	6.90	0.6	12.8
1550	6.93	0.7	12.5
1600	7.04	0.6	11.1

Fig. 1. XRD patterns of $Ba(Zn_{1/3}Ta_{2/3})O_3$ system vs. sintering temperature.

In the TSF paper, these values are the same but are expressed in nm rather than Angstroms.

the reflections due to Zn and Ta order (superstructure reflection) and the line split, i.e. the c/a differs of $\sqrt{3/2}$. The former shows the amount of Zn and Ta ordering and the latter shows the lattice distortion by Zn and Ta ordering. In the superstructure lines, the reflections whose index $(2h + k + l)/3$ does not equal the integer, are caused by the Zn and Ta. Therefore, a change in intensity of the superstructure reflection corresponds to the amount of Zn and Ta order. The $(1\,0\,0)$ reflection has the strongest intensity among the superstructures lines. Fig. 1 shows the increase in intensity of $(1\,0\,0)$ reflection versus sintering temperature for 4 h treatments time. LRO gradually increase up to 1600 °C.

Lattice distortion caused by Zn and Ta ordering can be evaluated by the split of the $(4\,2\,2)$ and $(2\,2\,6)$ reflections. If no distortion occurs, then the $(4\,2\,2)$ and the $(2\,2\,6)$ reflections are not separated. Fig. 2 shows the profile change of $(4\,2\,2)$ and $(2\,2\,6)$ reflections versus sintering temperature. Diffraction patterns show that the ceramics fired up to $T_s = 1500$ °C consist of half distorted and half non-distorted structure. In ceramics fired from 1525 to 1600 °C, a split of $(4\,2\,2)$ and $(2\,2\,6)$ reflections can be observed.

The perfectly ordered structure is produced by the following process:

1. A perovskite-type structure is formed, but Zn and Ta are in disorder.
2. Zn and Ta are partially in order.
3. Zn and Ta are completely in order and the lattice is distorted.

A supplementary distortion can be produced by the ZnO loss. At high sintering temperatures, ZnO evaporates. The Zn^{2+} ions on B′-site are partially replaced by Ba^{2+}, which have a much larger ionic size 1.61 Å comparatively to 0.74 Å for Zn. This substitution creates a supplementary unit cell distortion,[7] even when the LRO is saturated and leads to an increase of Q.

3.3. Analysis of microstructure and composition

The microstructure of BZT ceramics sintered in air at 1550 °C/4 h was investigated by using SEM. The images are presented in Figs. 3–6. The BZT samples sintered at $T_s > 1500$ °C present grains with concentric ordered shells. BZT sample has a porous structure due to ZnO evaporation during the sintering process. In Fig. 3 is presented a micrograph of the sample periphery. The SEM image reveals an extended porosity, with pore size in the range (10–30) μm, inter-aggregates. Smaller pores, between 3 and 5 μm, are located between grains. Submicron pores appear only on the grain surface. The large grains of an octahedron shape, well faceted and with sizes in the range (10–20) μm are present in all the samples. Micron grains are located on the grain boundaries or on the grain surfaces.

The SEM image of the central zone of BZT sample sintered at 1550 °C for 4 h is shown in Fig. 4. In this case the porous structure is reduced comparatively with the spherical zone. The large pores disappear. There are inter-aggregates pores with sizes between 5 and 10 μm. Moreover, there are smaller pores ones located between three and four grains with sizes (1–3) μm. The micrograph presents a bimodal distribution of
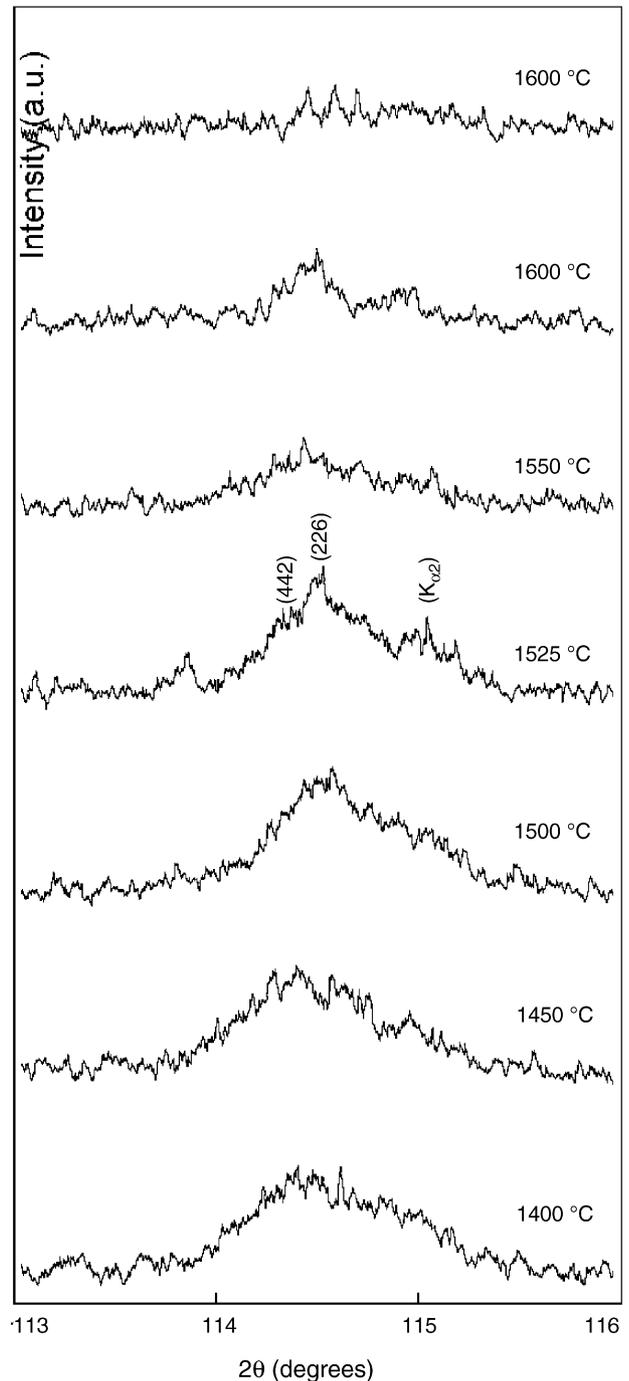

Fig. 2. The $(2\,2\,6)$ and $(4\,2\,2)$ reflection profiles of BZT ceramics vs. sintering temperature.

grains size. Well faceted, polyhedral grains with dimensions in the range (20–40) μm are present with smooth surfaces and edges. It appears that the dislocations climbed and dissociated (region a in Fig. 5) when the material was partially in ordered state and that the dislocation configurations became frozen when disorder set in during granular growth. In most cases, the less-ordered area was associated with at least one dislocation, weak 1:1 order, surrounded by areas with 1:2 order. Few micron grains (1–3) μm, are located on the smooth layer grains surfaces with a cubic-deformed shape (region b in Fig. 5). Other micron grains
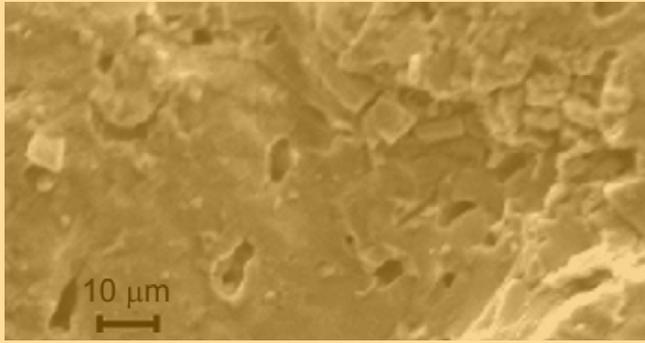
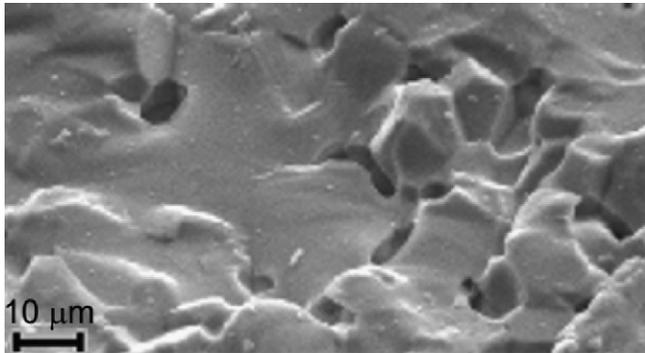Fig. 3. Micrograph of the periphery of the BZT sample sintered 1550 °C/4 h.

Fig. 4. SEM micrograph of BZT sintered in air for 4 h at 1550 °C—care zone.
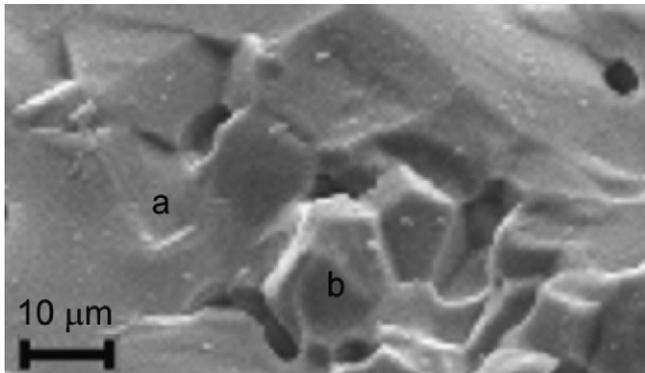

Fig. 5. Detail of SEM image from Fig. 4: (a) dislocations climbed and dissociated in a BZT grain; (b) small cubic-micron grains located on the surface of large octahedral well faceted ones.
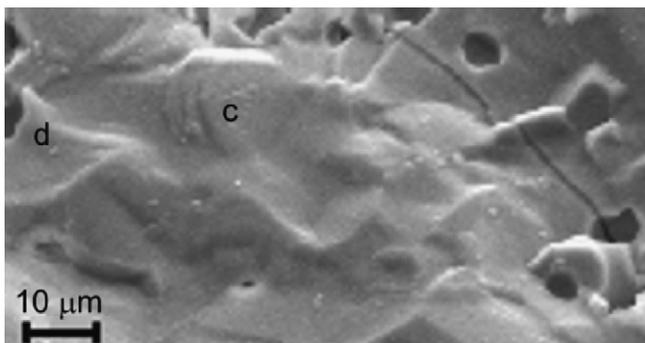

Fig. 6. Micrograph of 1:2 ordered compound BZT: (c) region containing very large domains of order; (d) very large grains with curved edges.

Table 2
EDX data: atomic concentration percents of basic elements of BZT specimen sintered for 4 h at 1550 °C in air

Ba (%)	Ta (%)	Zn (%)	Position on the disk
57.18	41.97	0.86	Outer zone
53.53	45.43	1.04	Inner zone

with spherical shape are agglomerate in aggregates of (6–8) μm near the boundaries of larger grains.

The micrographs reveal the presence of few very large grains ($d > 50$ μm), with curved edges, corresponding to the abnormal granular growth (region d in Fig. 6). The abnormal granular growth together with the ZnO evaporation can explain the bulk density decrease of BZT specimens sintered at $T_s > 1500$ °C. We used microanalysis (EDX) in an attempt to examine whether a composition trend such as a "paucity" of the B′ element could explain the correlation between microstructure, long-range ordering and dielectric properties. The inner and outer zone compositions are presented in Table 2. The Zn atomic concentration indicates a sub-stoichiometric composition, accentuated at the sample periphery where the ZnO evaporation is more possible. On the other hand, the experimental data indicate a variation of Ba^{2+} content with higher concentration at the sample boundary. In this case, Ba^{2+} can enter on the free Zn-octahedron sites, producing a supplementary deformation of the unit cell.

3.4. Microwave measurements

Microwave measurements on dielectric constant and loss tangent were performed on the BZT 1–5 samples. The data obtained revealed a determinant influence of the sintering temperature as well as of the annealing on the complex dielectric constant. The increase of the dielectric constant with the increase of the sintering temperature can be observed in Table 3. This can be considered as an effect of the reduced porosity resulting in a better densification at high sintering temperatures, as can be seen in Table 1.

The loss tangent exhibits values around 2×10^{-4} for samples sintered at 1400 and 1450 °C. The dielectric loss tangent increases to 7×10^{-4} with the sintering temperature increases to 1500–1550 °C, then it decreases 2×10^{-4} for samples sintered at 1600 °C. This variation could be related with the Zn and Ta cation ordering process and with the secondary phases, which appear for sintering temperatures between 1450 and 1525 °C as shown in Fig. 1. For temperatures higher than 1550 °C these phases disappear and, consequently, the dielectric loss became lower.

The additional thermal treatment at 1400 °C for 10 h for all samples resulted in the reduction of the BZT dielectric loss as can be seen in Table 4 and Fig. 7. The quality factor Q of annealed samples is higher than for samples without thermal treatment, especially for samples sintered beyond 1550 °C. The quality factors Q of the annealed BZT4 and BZT5 samples take values up to 18,000 and the dielectric permittivity is around 31 as shown in Fig. 8 and Table 4.

Table 3
Dielectric parameters of BZT ceramics vs. sintering temperature

Sample	Sintering temperature (°C)	Resonance frequency f (GHz)	Dielectric permittivity ε_r	Dielectric losses $tg\delta$ ($\times 10^{-4}$)	Quality factor Q	Product Qf (GHz)
BZT 1	1400	7.18	20.8	1.9	5130	36830
BZT 2	1450	6.66	26.6	1.8	5550	36960
BZT 3	1500	6.33	31.1	7	1230	7790
BZT 4	1525	6.29	32	6.6	1230	7740
BZT 5	1600	6.23	34.4	2.1	4600	28660

Table 4
Dielectric parameters of BZT ceramics annealed at 1400 °C/10 h vs. sintering temperature

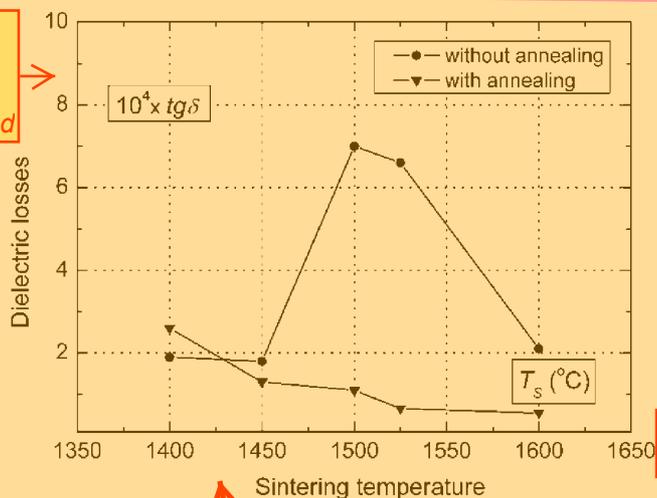| Sample	Sintering temperature (°C)	Resonance frequency f (GHz)	Dielectric permittivity ε_r	Dielectric losses $tg\delta$ ($\times 10^{-4}$)	Quality factor Q	Product Qf (GHz)
BZT 1	1400	7.73	20.7	2.6	3910	30220
BZT 2	1450	7.12	26.4	1.3	7710	54800
BZT 3	1500	6.75	30.7	1.1	9090	61360
BZT 4	1525	6.73	31.6	0.66	15210	102360
BZT 5	1600	7.51	31.8	0.55	18220	136830

Fig. 7. Modification of dielectric losses curve vs. sintering temperature for BZT ceramics without and with annealing treatment at 1400 °C/10 h.
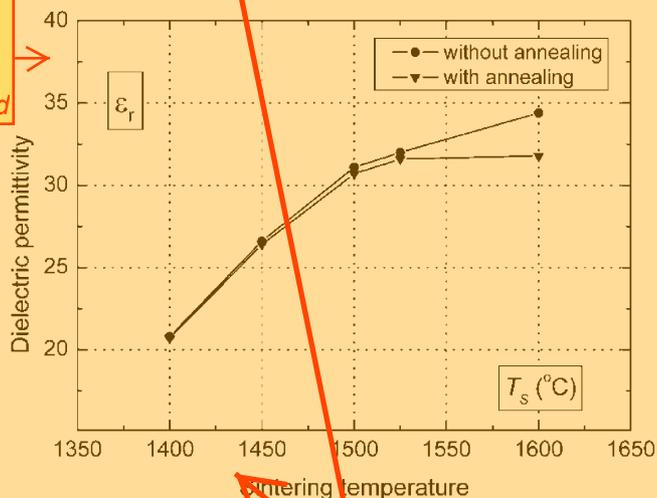

Fig. 8. Modification of dielectric permittivity curve vs. sintering temperature for BZT ceramics without and with annealing treatment at 1400 °C/10 h.
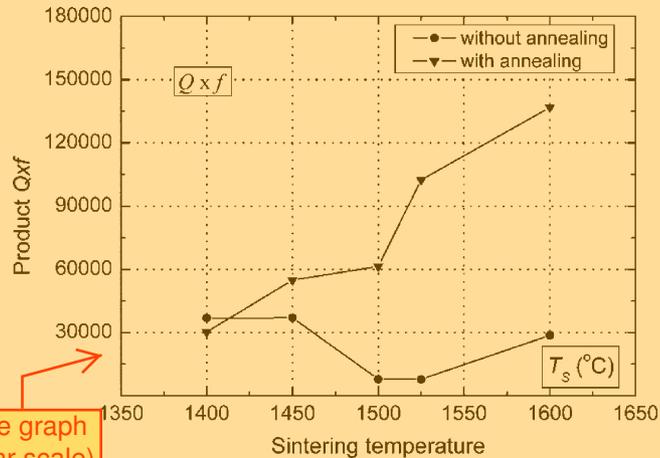

Fig. 9. Modification of the product Qf curve vs. sintering temperature for BZT ceramics without and with annealing treatment at 1400 °C/10 h.

The product Qf between the quality factor Q and the measurement frequency f also increases with the sintering temperature for annealed samples as shown in Fig. 9. This could indicate the necessity of an increased sintering time.

4. Conclusions

Ba(Zn$_{1/3}$Ta$_{2/3}$)O$_3$ ceramics with low loss in microwave domain were obtained by solid-state reaction. The investigations on dielectric properties revealed that the increase of sintering temperature decrease the dielectric loss especially when an annealing of 10 h at 1400 °C is performed.

Qf product strongly depends on the BZT crystalline structure: the unit cell distortion and cationic order. Dielectric loss decreases with the increase of ordering degree in the structure and with the disappearance of secondary phases. Lowest loss is obtained for a Zn and Ta completely ordered BZT ceramic with a strongly distorted unit cell. Porosity has small effect on dielectric loss of BZT material.

Well-sintered and annealed BZT samples exhibit a dielectric constant around 31 and a *Qf* product up to 130,000. Super-high quality factor of BZT materials recommend them for microwave and millimeter wave such applications as resonators and filters for wireless communication systems.

Acknowledgement

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