MICROSTRUCTURE AND MICROWAVE CHARACTERIZATION OF SrO_2-$Ba_2Ti_9O_{20}$ DIELETRIC RESONATORS FOR TELECOMMUNICATIONS APPLICATIONS

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Abstract This work deals with the influence of strontium oxide (SrO_2) in the dielectric properties of the $Ba_2Ti_9O_{20}$ ceramics as microwave oscillators for telecommunications applications. The main requirements of these dielectric resonators are the high selectivity and stability in frequency, the high value of both the dielectric constant (ε) and of the quality factor (Q), as well as the low temperature coefficient of resonance frequency (τ_f). The ceramics were prepared using suitable powder mixtures, without and with SrO_2 addition, varying the content from 0.2 to 1.0 mol %. The powders were mixed, compacted by uniaxial and isostatic pressing, producing cylindrical samples with specified dimensions in order to obtain dielectric resonators operating at frequency range around 7.0 GHz. Finally, they were sintered at 1250ºC for 6 hours and 1360ºC for 6 hours. The ceramics characterization was carried out using X-ray diffraction technique (for the formation study of the $Ba_2Ti_9O_{20}$ chemical compound), and scanning electron microscopy – SEM (for microstructure densification degree and grain size analyses). The dielectric parameters in microwaves (ε, Q and τ_f) were measured using a suitable microwave system specially mounted to accomplish such measurements. SEM analyses showed a high densification degree and a grain size increase with the amount of SrO_2 added to the ceramics. This fact has increased the quality factor due to dielectric losses, although the dielectric constant was kept practically unchanged.

Introduction

Several kinds of dielectric ceramic have been developed for applications in microwave telecommunications, satellite broadcasting systems and in many kinds of personal communication systems. These devices, which are similar to the metallic resonant cavities, present advantages due of the high dielectric constant, the quality factor high values, frequency high stability, small dimension, low cost, ease of assembling and compatibility with microwave integrated circuits [1].

Several ceramic compositions based on barium and titanium compounds [1-5] have presented good dielectric properties, such as $BaTi_4O_9$, $BaTi_5O_{11}$, and $Ba_2Ti_9O_{20}$.

Microstructures of barium nanotitanate ($Ba_2Ti_9O_{20}$) dielectric resonators (DRs) doped with strontium (Sr) for microwave application are here studied. These DRs should fulfill the requirements of the dielectric constant as well as the quality factor high values and frequency high stability, for application as local oscillator of an INPE Communication Satellite. It has been demonstrated that the ceramic porosity affects the dielectric constant (ε_r), the quality factor (Q) and the thermal coefficient (τ_f) of the $Ba_2Ti_9O_{20}$ DR, as shown in Fig. 1 [5].

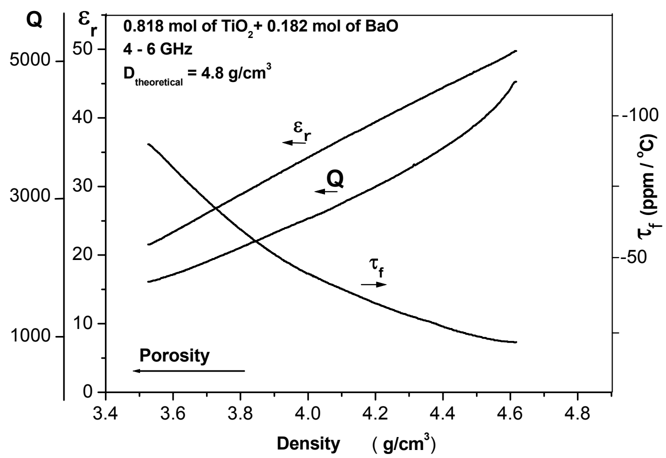


Figure.1 Effects of the ceramic density on the dielectric properties of the $Ba_2Ti_9O_{20}$ DR

Experimental Procedure

Five different samples were prepared: a $Ba_2Ti_9O_{20}$ specimen and others with 0.2, 0.4, 0.6, 0.8 and 1.0 mol % strontium contents. For their preparation, the suitable 18.2% BaO and 81.8 TiO_2% mol composition fraction were used (Fig. 2). The raw materials used for the preparation of these $Ba_2Ti_9O_{20}$ ceramics with addition of strontium were: $BaCO_3$, TiO_2 and $SrCO_3$, with purity degree more than 98%. Since the stability region of the $Ba_2Ti_9O_{20}$ phase occurs in a narrow range of composition BaO - TiO_2 it is possible to obtain it reacting 81.8 mol % TiO_2 and 18.8 mol % BaO (Fig. 2) for temperature lower than 1300°C.

The manufacturing process of these DRs requires the suitable temperature both for the formation and the stabilization of the $Ba_2Ti_9O_{20}$ phase and for the sintering and densification of the ceramic. The thermochemistry reaction for the formation of the desired phase is given by:

$$2\ BaCO_3\ (s) + 9\ TiO_2\ (s) \rightarrow Ba_2Ti_9O_{20}\ (s) + 2\ CO_2\ (g)$$

Cylindrical samples with a preestablished relationship of H/D = (0.40 ± 0.02) (where H is height and D the diameter) are produced in order to operate at the desired frequency range around 7 GHz. Defined the composition, the powders of $BaCO_3$ and TiO_2 with and without addition of $SrCO_3$ plus the compacting addictive PVAL (2% of the mass in weight) were wet mixed in a ball centrifugal mill for 4 hours at 150rpm. The powders were mixed and dried using a rotative evaporator at vacuum. This process aided the total homogeneity and the break of the formed agglomerates, thus, minimizing the formation of kind of particles. The powder was compacted by a uniaxial (40 MPa) and isostatic (300 MPa) pressings, producing cylindrical test bodies. Finally, they were synthesized and sintered at 1255ºC for 6 hours and 1360ºC for 3 hours.

Afterwards, the ceramic crystallographic phases were characterized by X-rays diffraction. The fracture surfaces were observed by scanning electronic microscopy (SEM).

The dielectric parameters were measured at microwave frequencies: resonance frequency (*f*), dielectric constant (ε) and, the unloaded quality factor Q_o, which in its turn corresponds to fator Q due to dielectric losses. These characteristics were measured using a test box made from copper. The testing device consists of a cylindrical dielectric radius *a* and height *H*, placed between two parallel conducting plates, in the case of resonant frequency [3, 4]. This configuration allows that DR can operate at the electromagnetic mode TE_{011}. From the experimental value of the resonant frequency we can determine the electrical constant by a field equation relating the resonant frequency, the dielectric constant and the resonator dimensions. The calculation was made running the software "Mathematica" in a microcomputer [3,5].

As for the quality factor, providing the test box dimensions are at least three times the size of the dielectric resonator and inserting the resonator between low-loss teflon spacers, then the metallic losses can be ignored.

The microwave measurements were performed according to the test setup shown in Figure 3 [5]. The DR was excited by means of an electric probe with optimum coupling. Another electric probe is used as a receiving device to detect the sign radiated by the resonator. The measured factor quality is the unloaded Q (Q_o), based on bandwidth at the half-power frequencies on the detected frequency spectrum.

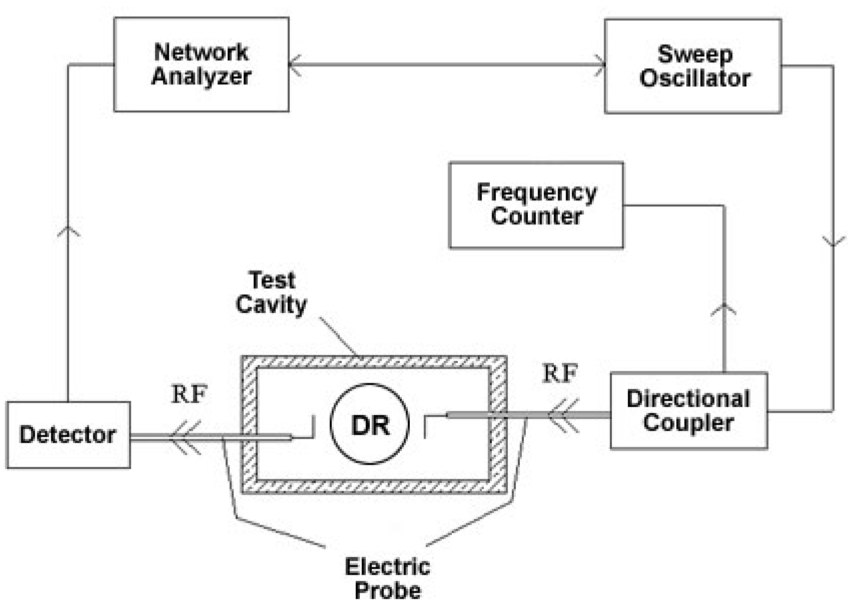


Figure. 3 Experimental setup for measuring the microwave parameters

Results

The X-rays diffraction analyses showed that all investigated ceramics have the $Ba_2Ti_9O_{20}$ as the major phase content (Fig. 4). However, the strontium addition did not prove to affect this phase formation.

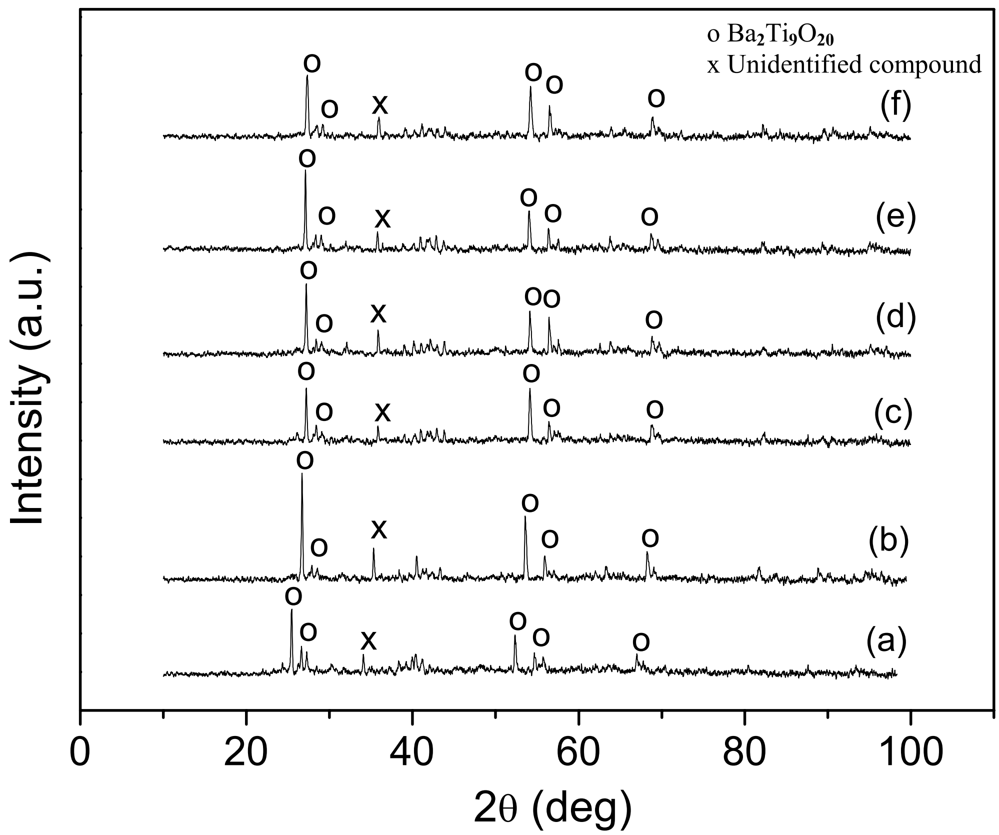
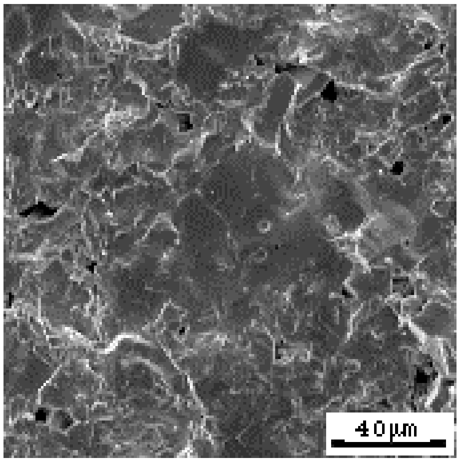


Figure. 4 X-rays diffraction patterns for the $Ba_2Ti_9O_{20}$ ceramics: (a) without Sr addition, (b) with 0.2 mol % Sr, (c) with 0.4 mol % Sr, (d) with 0.6 mol % Sr, (e) with 0.8 mol % Sr and (f) with 1.0 mol % Sr

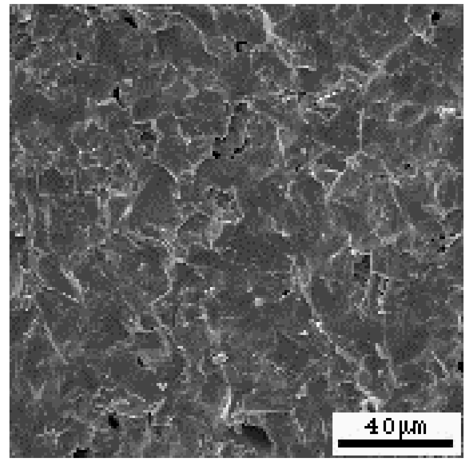
The scanning electronic microscopy (SEM) analyses of the ceramic fracture surfaces showed a high densification degree of the microstructure and the presence of few pores (Fig. 5). These defects are due to the packing powder flaws occurred during the compacting step. The defects can be minimized by optimizing the grain size distribution of these powder mixtures which can be obtained by a suitable procedure in the grinding step. The grain size increases with the amount of added strontium in the $Ba_2Ti_9O_{20}$ ceramic.

The measuring results at microwave frequencies for investigated DRs are presented in the Table 1. SEM analyses showed a grain size increase with the amount of

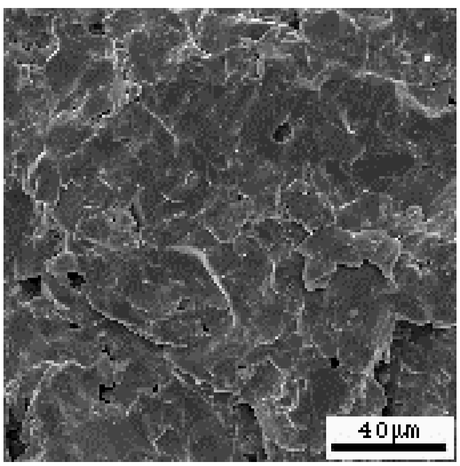
SrO_2 added to the ceramics. This fact has increased the quality factor due to dielectric losses, although the dielectric constant was kept practically unchanged.



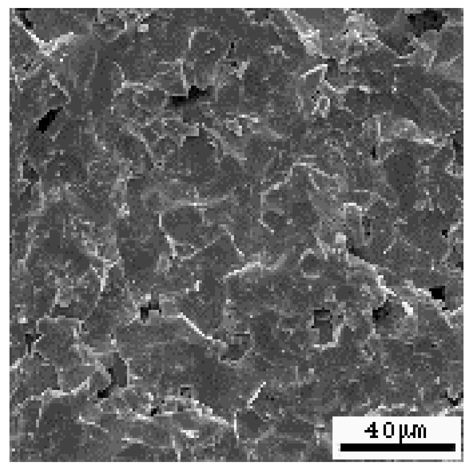
(a)



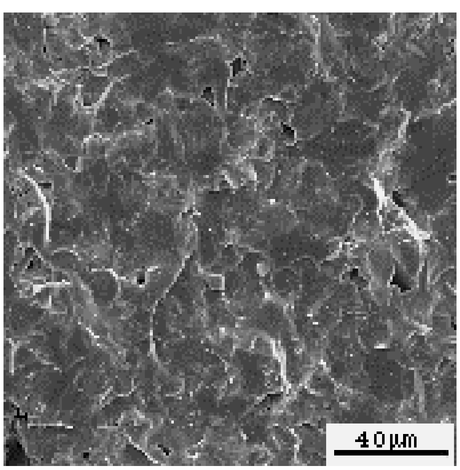
(b)



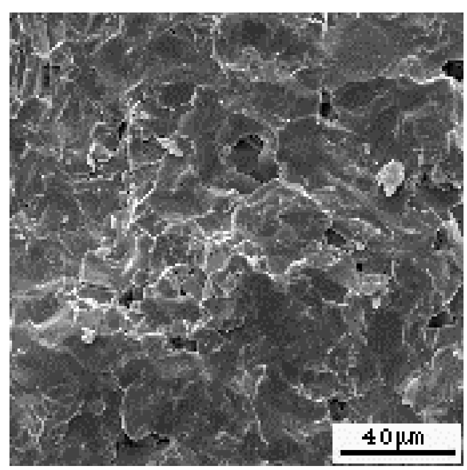
(c)



(d)



(e)



(f)

Figure. 5 SEM micrographs of $Ba_2Ti_9O_{20}$ ceramic fracture surfaces: (a) without Sr addition, (b) with 0.2 mol % Sr, (c) with 0.4 mol % Sr, (d) with 0.6 mol % Sr, (e) with 0.8 mol % Sr and (f) with 1.0 mol % Sr.

Table 1 – The measuring results at microwave frequencies

DR code	***H*** ± 0,04 [mm]	**1.** *a* ± **0,04** [mm]	***f*** ± $5{,}0\times10^{-3}$ [GHz]	ε ± 0,5	***Q***
00Sr	4. 10	5.45	7.64540	35.5	3770
02Sr	4. 20	5.45	7.68061	35.2	3824
04Sr	4. 15	5.45	7.67426	35.2	3929
06Sr	4. 10	5.45	7.65263	35.3	4495
08Sr	4. 25	5.40	7.67644	35.2	4610
10Sr	4. 20	5.40	7.69248	35.0	5002

Conclusions

The Brazilian raw materials ($BaCO_3$, TiO_2 and SrO_2) used without any previous processing resulted in sintered ceramics with a densification degree suitable for the DR manufacturing. The synthesizing and simultaneous sintering procedures make possible an increase of the suitable densification degree in comparison to the ceramics obtained by other authors. The strontium addition did not affect directly the desired stabilization of the $Ba_2Ti_9O_{20}$ phase as well as the densification degree of the ceramics. The obtained quality factor showed tohave been influenced by the amount of strontium added to the ceramic. Ceramic microstructures and their associated microwave characteristics have found good performance.

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