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Research Paper Science

Synthesis Of Nonatitnate Based Dielectric Ceramic Using Various Grain Size of Starting Materials

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ABSTRACT

The technological advancements in the field of microwave communication owes to the availability of suitable and affordable electroceramic materials, called Dielectric Resonators (DRs).
Ceramic dielectrics especially the complex titanates are the promising candidates as good DR material. There exists two important choices, namely; $BaTi_4O_9$ and $Ba_2Ti_9O_{20}$.
The present work investigates the effect of particle size distribution among the reacting crystallites on fabrication of very dense bodies of $Ba_2Ti_9O_{20}$ ceramic with phase purity. The $Ba_2Ti_9O_{20}$ phase development and its dielectric characteristics are found to be highly sensitive to the particle size of starting materials, impurity phases evolved and porosity in the sintered products.
It was found that most promising characteristics of the dielectric ceramics based on $Ba_2Ti_9O_{20}$ were obtained when reagent powders were ground for 9 hrs.

Keywords : Nonatitnate,ceramic,grain size

1. Introduction:
The technological advancements in the field of microwave communication, especially Mobile communication technology, owes to the availability of suitable and affordable electroceramic materials, called Dielectric Resonators (DRs), that function as the important circuit elements, like filters, oscillators with selectable frequencies, amplifiers and tuners (Cava & Chem, 2001). These Electroceramics, in general, can be specially formulated to have desired electromagnetic properties. Their characteristics can be easily tailored to meet specific requirements through exploiting the complex interplay between processing and chemistry, structure at many levels and device physics.

A good DR material must have low loss (i.e large Q value), high temperature stability with a reasonably high relative permittivity and a very small change in resonant frequency as a function of temperature. Ceramic dielectrics and ferroelectrics (i.e. dielectrics with spontaneous polarization) are the obvious choices for this application. The dielectric microwave materials like simple titanate ceramics i.e. $BaTiO_3$ and its derivatives, have been investigated extensively and beside a number of research reports, a few patented technologies are also available in literature (Petrov, Peter, Alford & McNeill, 2006). However the complex titanates have not been given much importance though they have enormous research and application potential. Two important candidates for DRs are $BaTi_4O_9$ and $Ba_2Ti_9O_{20}$ as both have reasonably high dielectric constants and quality factors (~39 and >10,000, respectively) (NEGAS, YEAGER, BELL, COATS & MINIS, 1993), (MOULSON & HERBERT, 1990). However, $Ba_2Ti_9O_{20}$ has been reported to have a much smaller value of temperature coefficient (~4 ppm/K) as compared to $BaTi_4O_9$ (~14 ppm/K). This makes $Ba_2Ti_9O_{20}$ a more favorable choice as DR for use in mobile communication technology. While $BaTi_4O_9$ has an orthorhombic Pnmn crystal structure (LUKASZEWICZ, 1957), Ba2Ti9O20 resembles a hexagonal close-packed cell (DAVIES & ROTH, 1987) of Ba^{2+} and O^{2-} with Ti^{4+} in octahedral sites. This pseudohexagonal arrangement has a nine layer stacking sequence with a primitive triclinic cell. There are two coordinations of barium ions within the layers: half are 12-coordinated with oxygen and the rest are 11-coordinated with vacant Ba^{2+} sites adjacent to them. This crystal structure plays an important role in the dielectric properties of this electroceramic (TAMURA, 1994).

The difficulty in realization of $Ba_2Ti_9O_{20}$ phase has daunted its development for years (LIN, GERHARDT, SPEYER & HSU, 1999), (Purohita & Tyagi, 2002). The advent of monophasic $Ba_2Ti_9O_{20}$ had been a challenge. The synthesis of phase pure $Ba_2Ti_9O_{20}$ materials with improved characteristics (i.e. narrow particle size distribution among the crystallites, higher surface area and better sinterability) the processes employed at synthesis step are of vital significance. The fabrication of very dense bodies of $Ba_2Ti_9O_{20}$ ceramic with phase purity is of great practical importance for improvement in dielectric characteristics as these are highly sensitive to the particle size, impurity phases and porosity in the sintered products. In the present work an investigation on influence of grain size on synthesis and phase development of $Ba_2Ti_9O_{20}$ dielectric ceramic has been done using mechanical grinding of starting materials and conventional mixed oxide synthesis technique.

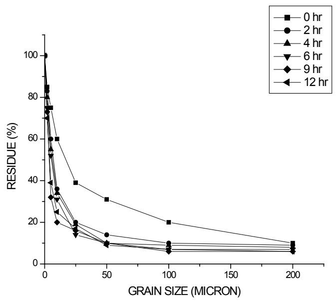
2. Experimental:
The dielectric ceramics based on $Ba_2Ti_9O_{20}$ were synthesized by using AR grade titanium dioxide and barium carbonate (purity 99.2%) as starting materials. The starting powder batch was prepared according to the compound stoichiometry (Ba:Ti = 2:9 in molar ratio). The grinding of the starting powder was then carried out for 2,4, 6, 9 and 12 hr in a ball mill having Al2O3 as grinding media in acetone. The grain-size measurements were done on resulted powder slurry by means of a Zeta Potential Particle Size Analyzer; Nicomp 380 ZLS. The oxide powders were homogenized, dried and pressed into cylindrical shapes of 12 mm diameter and 10 mm height, using a hydraulic press under 6-8 ton/cm^2 pressure. Pre-sintering of samples was carried out in a muffle furnace at 1150°C for 4 hr. Then the samples were ground to powders for 6 and 12 hr first using agate pastel mortar followed by ball milling. The powders were then prepared for sintering after pressing into a cylindrical form of 12 mm diameter and 10 mm height. The final sintering was carried out in an electric furnace at 1330°C for 3 hr. The cooling of the samples was done slowly at a rate of 10°C/min.

The samples so obtained were then characterized for shrinkage, porosity, absolute and apparent density using standard methods. Structural and phase development studies were done by X-Ray Diffractometer; BRUKER-D-8-ADVANCE. The functional properties of the dielectric ceramic samples, Polerizability and Dielectric Constant were determined in a high-frequency field varying from 2 to 8 GHz by means of a Hioki measuring apparatus with a parallel plate arrangement using the samples in cylindrical shape of 6 mm in diameter and 4 mm in height.

3. Result and Dicussion:

The grain size distribution, expressed as residual percentage versus grinding times (0,2,4,6,9 and 12 hrs) for the starting materials $BaCO_3$ and TiO_2 are shown in Figure 1 and 2 respectively.

Figure 1: Grain Size Distribution Expressed As Residual Percentage As Function Of Grinding Time For BaCO3

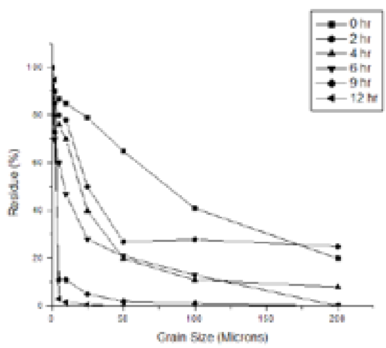


The investigation into grain-size distributions versus grinding time for $BaCO_3$ (see Figure 1) shows that the optimum $BaCO_3$ grain-size distributions requires 9 hr of grinding to result in dimensional uniformity of size less than 20 µm. With increasing the grinding duration, the greater than 50 µm fractions get converted into the less than 20 or even 10 µm fractions.

For TiO_2, on the other hand, the optimum grinding times suitable for achieving a continuous grain-size distribution as homogeneous as possible were found out to be with 9 and 12 hrs (see Figure 2). The resulted powders after grinding were found to have particles prominently less than 20 µm in size. With increasing the grinding duration, the greater than 20 µm fraction is found to change into the less than 20 µm fraction.

It has been found at the presintering stage of powder mix that weight and volume losses show slight increase with increase in grinding time, which obviously indicates that the grain size intervenes in increasing the physico-chemical interactions between reactant particles. This is an advantageous effect for proper phase development. The densities of sintered ceramic samples show an increase, except for the ceramics obtained from 9 hr ground starting powders, but the sintering degree has shown an increasing tendency for sintered materials with 12 hr grinding time.

Figure 2: Grain Size Distribution Expressed As Residual Percentage As Function Of Grinding Time For TiO_2



The X-ray Diffraction patterns of the sintered samples are shown in Figure 3. These studies were done to investigate the effect of grain size of starting powder mix on phase evolution. The sample synthesized using unground powder mix shows the characteristic peaks of $BaTi_4O_9$ and TiO_2 (most intense peak) with $Ba_2Ti_9O_{20}$ in vary small amount. The characteristics peaks of $Ba_2Ti_9O_{20}$ start to appear with grinding time ≥3 hr, becoming major peaks in the sample with 6 hr grinding. This sample shows a mixture of $BaTi_4O_9$, $Ba_2Ti_9O_{20}$ and TiO_2 phases. The intensity of $Ba_2Ti_9O_{20}$ peaks is found to increase rapidly with the increasing grinding time the cost of the peak height of $BaTi_4O_9$ and TiO_2 phases. The XRD patterns of the sintered samples of 9 and 12 hrs are found to be identical with no detectable impurities. The monophasic $Ba_2Ti_9O_{20}$ could be thus

Figure 3: The X-ray Diffraction patterns of the sintered samples with varying grinding times [0 hr (a), 6 hr (b) and 9 hr (c)].

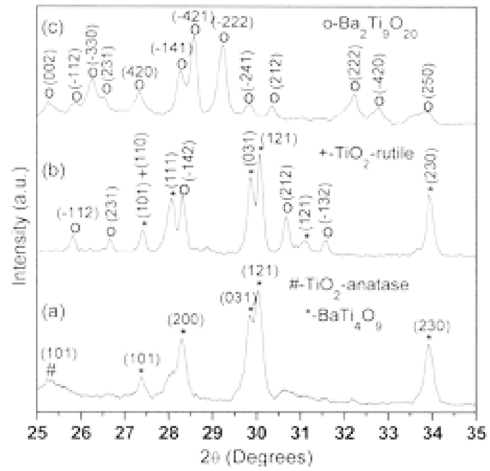
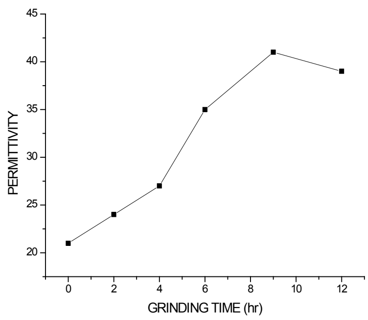


Figure 4: Dependance of Permittivity (εr) of Sintered Ceramic Ba2Ti9O20 on Grinding Time (hrs).



obtained with at least 9 hr grinding of the starting powder mix. Therefore, the most promising characteristics of the dielectric ceramics based on $Ba_2Ti_9O_{20}$ were obtained when reagent powders were ground for 9 hr, to have particle size in the range of less than 20 µm.

The polerizability and dielectric measurements on these materials showed that the optimized grinding of starting materials resulted in increase of permittivity (see Figure 4), maximum value being exhibited by the sample with 9 hr grinding, having particle size<20 µm. The resonant frequency is found to increase by 1.5 GHz, with notable narrowing in band width by 2.5 MHz. The quality factor is found to improve by 1250 and 1400 for that under load and for that specific to the resonator, respectively, whereas, the loss angle tangent exhibits a decrease by 0.4x10-3. The variation coefficient versus temperature is found out to be of +20 ppm/^{o}C at the frequency of 8 GHz.

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