Pergamon

Materials Research Bulletin, Vol. 30, No. 6, pp. 653-658, 1995

0025-5408/95 $9.50 + .00

0025-5408(95)00070-4

# HIGH PERMITTIVITY AND LOW LOSS CERAMICS IN THE $BaO-SrO-Nb_2O_5$ SYSTEM

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(Received March 24, 1995; Refereed)

**ABSTRACT**

A new group of compounds with composition $(Ba_{5-x}Sr_x)Nb_4O_{15}$, having high permittivity and low loss have been prepared and characterised in the microwave frequency region. X-ray diffraction studies showed that monophase compound existed for all values of x from 0 to 5. Microwave dielectric properties such as $\varepsilon_r$ and $\tau_f$ showed smooth variation with x, while the unloaded quality factor ($Q_u$) showed remarkable improvement with x. A range of ceramic dielectric resonators(DR) with $40< \varepsilon_r <50$, $-10<\tau_f<+10$ and $Q \times f >10{,}000$ can be obtained in this system.

**MATERIAL INDEX**: dielectric resonators, microwave ceramics, niobates, oxide ceramics.

## Introduction

The importance of ceramic dielectrics in the microwave technology is increasing. With the progress in microwave integrated circuits, the need for miniaturisation of components like filters and oscillators has stimulated the development of high permittivity ceramics known as `dielectric resonators (DR)'(1,2). The DRs possess excellent dielectric properties which make them suitable for a wide range of applications in microwave communication, down from 500 MHz to 20GHz (3,4) (which include Cellular mobile radio, Satellite communication etc.). No single ceramic compound is, so far, equally good in all aspects for this whole band of operation. Since the desired frequency of operation (usually $TE_{01\delta}$) is dependent on the dimension of the DR and the $\varepsilon_r$ of the material, ceramics with different $\varepsilon_r$ are used in different bands. For example, ceramics with $\varepsilon_r$ as high as 80 ( 5,6) are more suited below 1 GHz band while those with $\varepsilon_r$ <25(7,8) are useful above 13 GHz band. Also, in practical applications, DRs with different shapes and configurations are optimised for better performance efficiency. Hence, the requirement now for any DR ceramic can be briefly put as follows:

1. $\varepsilon_r$ >20- For miniaturisation since the size of the resonator is proportional to $\varepsilon_r^{-1/2}$.
2. $Q_u$>2,000- Quality factor is a measure of efficiency and determines the frequency selectivity.
3. $\tau_f$ <20 ppm/$^{O}$C- $\tau_f$ is the coefficient of thermal variation of resonant frequency and must be close to 0. $\tau_f$<20 ppm/$^{O}$C make DRs competent with Copper cavities.

Accordingly, a few materials like $Ba_2Ti_9O_{20}$ (9,10), $(Zr,Sn)TiO_4$(5,11,12), $Ba(Zn,Zr,Ta)O_3$ (13) etc. are found to be useful. Still the search for new and new ceramics are progressing. The possibility of obtaining new ceramic dielectric resonator materials in the system, $(Ba_{5-x}Sr_x)Nb_4O_{15}$ was identified when we studied the microwave dielectric properties of $Ba_5Nb_4O_{15}$(14). In this paper we present the preparation and microwave characterisation of the (5-x)BaO-xSrO-2$Nb_2O_5$ solid solutions.

## Ceramic preparation

The materials are prepared by the solid state ceramic route. High purity (99.9 %) $BaCO_3$, $SrCO_3$ and $Nb_2O_5$ are mixed in the appropriate stoichiometry. 6 compositions, having 0, 8.3, 17.3, 27.1, 37.7 and 49.4 mol % of SrO (x=0, 1...5) are wet mixed using distilled water for about 40 minutes, dried and calcined in alumina crucibles. The compound $Ba_5Nb_4O_{15}$ ($B_5N_4$) calcined at 1250 $^{O}$C in 4 hours while the strontium counterpart $S_5N_4$ calcined only at 1350 $^{O}$C. Calcined powders are thoroughly ground into fine powders and pressed into cylindrical discs with 10 mm diameter and 6-7 mm thickness for microwave measurement. Thin discs are also prepared for XRD and SEM analyses. The compacts are sintered between 1380 $^{O}$C and 1440 $^{O}$C for 4 hours. The $S_5N_4$ is found to densify at 1450 $^{O}$C after adding1 wt.% of $La_2O_3$ to the calcined powder. However, the sinterability of this ceramic is poor. The sintered pellets had a light cream colour with a slight greenish tint for the barium-rich composition.

## Characterisation

I. Powder X-ray diffraction:- Thin sintered pellets are crushed and used for powder XRD using CuK$\alpha$ radiation ($\lambda$=1.5406 A) (Model: Rigaku, Japan). The d-values of prominent reflecti ons are calculated using the interfaced computer, and intensities compared.

II. Scanning Electron Microscopy:- Selected compacts are polished and thermally etched at 1200 $^{o}$C and the surface is analysed using SEM (Model: Jeol, Japan). Also, for selected compositions fracture surface has been used.

The bulk densities of all the sintered compacts are measured using thick cylinders. Density is directly calculated from the volume and mass.

III. Microwave characterisation:- The $\varepsilon_r$ and $Q_u$ at microwave frequencies are measured by using HP 8510 B Network Analyser, HP 8341 B Sweep Oscillator and HP 8514 B Reflection-Transmission Unit. The whole set up is controlled using a HP 9000,300 series computer. $\varepsilon_r$ is calculated from the $TE_{011}$ resonance of the end-shorted dielectric cylinder like that done by Courtney(15). Since the DR applications involve coupling using striplines, the resonant mode of ($TE_{01\delta}$) a band rejection filter, constructed using 50$\Omega$ microstrip transmission line coupled with the cylindrical DR, is taken. A brass enclosure of size 5X5X3 cm$^3$ provides adequate shielding. By the method of Khanna and Garault(16), the unloaded quality factor ($Q_u$) of the DR is obtained. Then the DR is heated slowly in an aluminium cavity of size $\phi$ 4.5 cm x 7 cm in the temperature range 25 to 80 $^{O}$C. Using HP 8410 C Network Analyser and HP 8350 B Sweep Oscillator the shift in TE $_{01\delta}$ mode of the resonator with respect to temperature is noted and the $\tau_f$ is calculated. To reduce the effect of coupling probe, cavity wall expansion and other tuning effects(1), the DR is placed at the centre of the bottom plate, the cavity top is open(covered with thermocoul) and a coupling of 10 dB is given.

## Results and Discussion

The X-ray powder diffractograms obtained for the six samples are shown in Figure1. The patterns are identical and match with that reported for $B_5N_4$ by Galasso *et al.*(17) (JCPDS File No.14-28). The unit cell structure of the compound is hexagonal with an $AO_3$ layer packing. The $Nb^{5+}$ ions are positioned at the octahedral sites of the five $AO_3$ layers which altogether constitute the unit cell. From Figure1 it is clear that strontium substitutes for all values of barium without any secondary phase formation. The $B_5N_4$ is reported to be polytypic(18); but no such polytype forms are seen from XRD. Figure 2 shows the typical microstructures obtained for $(Ba_{5-x}Sr_x)Nb_4O_{15}$ ceramics with x=0 and 4. They have elongated grains. The SEM picture of x=0 is taken from the surface and that of x=4 is from fracture. The microwave dielectric

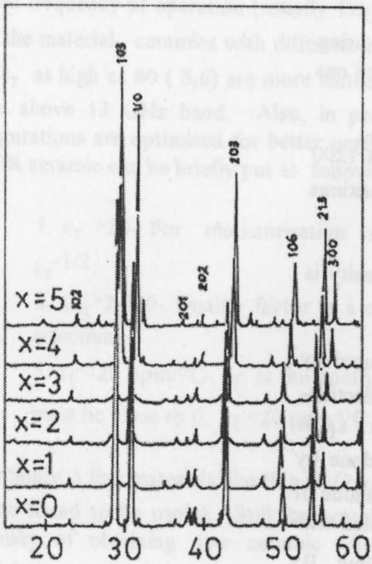


**FIG.1**

Powder XRD patterns of $Ba_{5-x}Sr_xNb_4O_{15}$ ceramics.

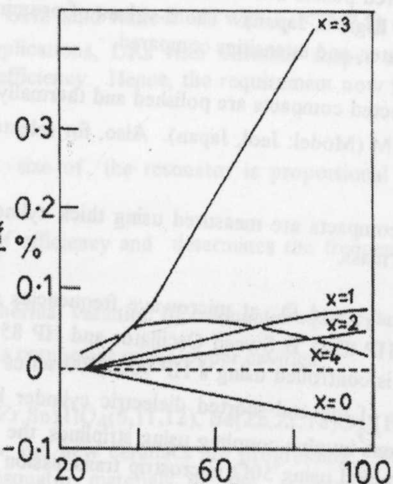


**FIG.3**

Variation of resonant frequency with temperature.

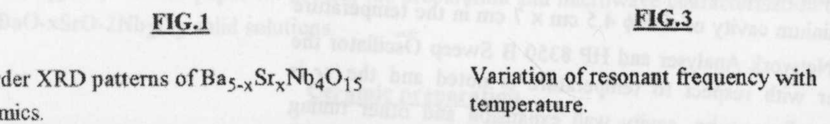


**FIG.2**

SEM photographs of $Ba_{5-x}Sr_xNb_4O_{15}$ ceramics for x=0 (left) and x=4 (right).

properties such as $\varepsilon_r$ , $Q_u$ and $\tau_f$ are shown in Table I. It can be seen that a small percentage substitution of strontium for barium (~20 %) increases

**TABLE 1**

Microwave dielectric properties of $(Ba_{5-x}Sr_x)Nb_4O_{15}$ ceramics.

| Material | $\varepsilon_r$ | $Q_u$ (at GHz) | $\tau_f$(ppm/$^O$C ) |
|---|---|---|---|
| $Ba_5Nb_4O_{15}$ | 38.4 | 2,500 (5.07) | -9 |
| $Ba_4SrNb_4O_{15}$ | 48.4 | 2,100 (4.70) | +11 |
| $Ba_3Sr_2Nb_4O_{15}$ | 49.5 | 3,500 (4.71) | +7 |
| $Ba_2Sr_3Nb_4O_{15}$ | 50.6 | 4,600 (4.61) | +85 |
| $BaSr_4Nb_4O_{15}$ | 44.9 | 8,800 (5.00) | +7 |

both the $\varepsilon_r$ and $Q_u$ and reduces the $\tau_f$. Further substitution increases the $\varepsilon_r$ only slightly and increases the $\tau_f$. The composition, $(Ba_2Sr_3)Nb_4O_{15}$ shows maximum $\varepsilon_r$ and $\tau_f$ values. The variation of the resonant frequency with temperature is shown in Figure 3. The high $Q_u$ and $\varepsilon_r$ are sufficient to make these ceramics useful for practical applications.

## Conclusion

The $(Ba_{5-x}Sr_x)Nb_4O_{15}$ ceramics have been prepared and characterised in the microwave frequency region. Dielectric Resonators with $\varepsilon_r$ as high as 48 and $Q_u$ > 15,000 (1 GHz) with $\tau_f$ in the range -10 to +10 ppm/$^O$C can be obtained with a small percentage of Sr substitution.

## Acknowledgement

The authors are grateful to Dr.A.D.Damodaran and Prof.K.G.Nair for the encouragement and help rendered in the work. H.Sreemoolanathan is thankful to CSIR, Govt. of India for the Resesrch Fellowship.

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