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# Synthesis and role of Nd and Sm on the microwave dielectric properties of $BaNd_{2(1-x)}Sm_{2x}Ti_5O_{14}$ dielectric resonator

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**Abstract**

Microwave dielectric properties of dielectric resonators ($BaNd_{2(1-x)}Sm_{2x}Ti_5O_{14}$) with high dielectric constant ($\varepsilon_r$), low loss (high $Q$) and low temperature coefficient of resonant frequency ($\tau_f$) have been studied by adjusting the Nd/Sm ratio. The composition with $x = 0.7$ gives high $\varepsilon_r$, high $Q$ and low $\tau_f$ which is highly suitable for dielectric resonators (DR). © 1997 Elsevier Science S.A.

*Keywords:* Dielectric resonators; Microwave dielectric properties; Temperature

## 1. Introduction

Microwave dielectric resonators (DRs) are miniature resonant devices used for frequency control in microwave integrated circuits. As the resonant frequency of these devices is inversely proportional to the dielectric constant ($\varepsilon_r$) of the material, to use these devices in the lower end of the microwave range frequency spectrum, materials with higher values of dielectric constant are required. Moreover using low loss materials with higher $\varepsilon_r$ values, other devices like patch antennas which are useful for direct reception of signals from low power transmitters can also be made. But as the $\varepsilon_r$ value increases, the dielectric loss will also increase, and thereby decreasing $Q$. The same is also the case with the temperature coefficient of resonant frequency ($\tau_f$). Only a few materials are identified so far as suitable for meeting all these requirements for microwave frequency applications. They are (i) $BaNd_2Ti_3O_{10}$[1]; (ii) $BdNd_2Ti_5O_{14}$[1]; (iii)BaO,$Bi_2O_3$–$Nd_2O_3$–$TiO_2$[2]; (iv) PbO–BaO–$Nd_2O_3$–$TiO_2$[3]. Among these, single phase composition is only $BaNd_2Ti_5O_{14}$[1].

Hence the synthesis and study of this single phase material assumes good importance. More so, as the properties of the present composition prepared are highly susceptible to Rare-Earth substitutions.

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## 2. Materials and methods

The starting materials were reagent grade Barium Carbonate (99.99% pure), Titanium dioxide (99.99% pure) supplied by CERAC, USA and, Neodymium oxide (99.99% pure), and Samarium oxide (99.99% pure) supplied by Indian Rare Earth Ltd, India. Appropriate proportions of the powdered materials were thoroughly mixed in water, dried and calcined at different temperatures ranging from 900 to 1200°C to find the optimum temperature of calcination. After calcination the powders were again ground thoroughly and subjected to sintering in air at temperatures ranging from 1200 to 1300°C in the form of pellets.

The microwave characteristics of the sintered pellets have been measured using frequency domain methods (reflection method and transmission method). Reflection method was used to find the $Q$ value of the DR and transmission method to find $\varepsilon_r$. The unloaded Q *f*actor of the microwave dielectric resonator is measured by reflection type technique proposed by D. Kajfez [4] using a cylindrical cavity, to within 2% accuracy.

The dielectric port resonator technique [5–7] is convenient for measuring $\varepsilon_r$, since an exact analysis relating the dielectric constant to resonant frequency is available, the sample does not require metallization, and the $TE_{011}$ mode is relatively easy to identify among the various modes. In this method, the dielectric resonators

Table 1
Microwave dielectric properties of $BaNd_{2(1-x)}Sm_{2x}Ti_5O_{14}$

| $x$ | $Q$ | $f_r$ (GHz) | $Qf_r$ | $\varepsilon_r$ | $\tau_f$ (ppm °C$^{-1}$) | Density ($10^{-2}$ g mm$^{-3}$) |
|---|---|---|---|---|---|---|
| 0.0 | 1470 | 6.077 | 8930 | 70.2 | +68 | 5.101 |
| 0.1 | 1410 | 6.120 | 8630 | 68.0 | +53 | 5.144 |
| 0.2 | 1320 | 6.122 | 8080 | 68.0 | +44 | 5.250 |
| 0.3 | 1275 | 6.065 | 7730 | 70.4 | +35 | 5.315 |
| 0.4 | 1315 | 6.061 | 7970 | 69.0 | +30 | 5.300 |
| 0.5 | 1315 | 6.139 | 8070 | 66.2 | +29 | 5.274 |
| 0.55 | 1335 | 6.164 | 8230 | 67.7 | +24 | 5.247 |
| 0.6 | 1500 | 6.119 | 9180 | 68.8 | +22 | 5.288 |
| 0.7 | 1420 | 6.125 | 8700 | 69.5 | +17 | 5.377 |
| 0.8 | 1400 | 6.120 | 8570 | 68.0 | +12 | 5.361 |
| 0.9 | 340 | 6.447 | 2190 | 63.2 | +1 | 5.372 |
| 1.0 | 260 | 6.545 | 1700 | 59.0 | −10 | 5.380 |

in the form of a short cylinder is placed between two parallel metallic plates of which one of them being movable with respect to the sample. The dielectric constant is calculated from both sample dimensions and resonant frequency of the $TE_{011}$ mode.

For temperature coefficient of resonant frequency ($\tau_f$) measurement, a cylindrical cavity made of Invar is used, but the technique is the same as that of $Q$ measurement. The test cavity is placed over a hot plate with an enclosure and the temperature range used is +25°C ($T_1$) to +60°C ($T_2$). The $\tau_f$ (ppm °C$^{-1}$) is calculated by noting the change in resonant frequency ($\Delta f$) by,

$$\tau_f|_{25°C}^{60°C} = \frac{\Delta f}{f_{25°C}\Delta T} \quad \text{or} \quad \tau_f = \frac{f_1 - f_2}{f_1(T_2 - T_1)} \tag{1}$$

where $f_1$ is the resonant frequency at $T_1$ and $f_2$ is the resonant frequency at $T_2$.

## 3. Results and discussion

Table 1 shows the results obtained for different compositions varying in their Nd/Sm ratio. Fig. 1 shows that there is an optimum calcination temperature (1120°C), at which the material density is maximum along with the highest possible values of $\varepsilon_r$.

This is understandable as the densification process depends predominantly on the particle characteristics of the calcined powder which is heavily determined by the calcination treatment. A lower calcination temperature will result in the presence of more unreacted reagents while higher temperatures will result in grain growth during the calcination stage itself resulting in a larger particle size. Both can hamper densification. The pellets calcined at different temperatures were given the same sintering treatment. Fig. 1 shows that the $Q$ value also follows as that of $\varepsilon_r$ for different compositions. Fig. 2 gives the variation of $\varepsilon_r$ with respect to rare earth (RE) substitution parameter '$x$'. The zig-zag variation can be attributed to the differences in maturing the materials. Up to $x = 0.7$, the $\varepsilon_r$ value remains almost constant and then rapidly decreases.

Fig. 2 shows the $Q$ value also follows a similar trend. But $\tau_r$ value steadily decreases from a high positive value and goes to a negative value for $x = 1.0$ (Fig. 2). Sm has a lower ionic radius and lower electronic polarizability parameter [8] compared with Nd. These can influence the structure in such a way that the Nd rich phase will have a higher packing fraction for the structure and hence a high $Q$ value; but, at the same time, higher electronic polarizability will cause higher values of $\varepsilon_r$ [8] as Nd occupies a site which will enhance the dielectric response of the system for a small increase in the electronic polarizability of the ion occupying the site.

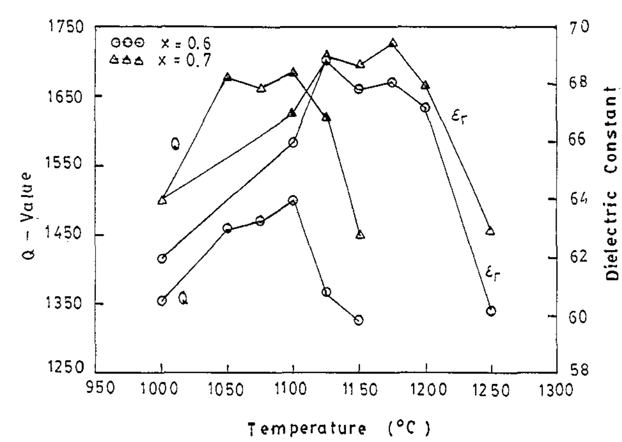


Fig. 1. The effect of calcination and sintering temperature on the $\varepsilon_r$ and '$Q$' value for the sample $x = 0.6$ and 0.7.

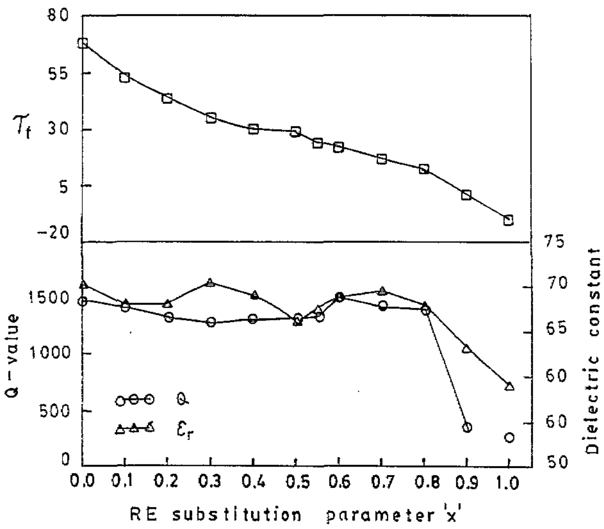


Fig. 2. Variation of $\varepsilon_r$, '$Q$' and $\tau_f$ as a function of rare-earth substitution parameter '$x$'.

## 4. Conclusion

Decrease in $\tau_f$ value means an increase in $\tau_\varepsilon$ value as $\tau_f$ is given by

$$\tau_f = -\left(\frac{\tau_\varepsilon}{2} + \alpha\right) \tag{2}$$

where $\alpha$ is the linear thermal expansion coefficient which is nearly a constant for all the compositions. But a lower value of $\tau_f$ is advantageous for applications. Hence a proper ratio of Nd/Sm will give a high $\varepsilon_r$ value with high $Q$ and at the same time lower $\tau_f$ (conditions essential for a material to be used as a microwave dielectric resonator). The composition with $x = 0.7$ is the best result and can be used for microwave applications.


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