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PREPARATION, CHARACTERISATION AND DIELECTRIC PROPERTIES OF CERAMICS IN THE $BaO-Nd_2O_3-TiO_2$ SYSTEM

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The dielectric ceramics $BaNd_2Ti_3O_{10}$, $BaNd_2Ti_4O_{12}$ and $BaNd_2Ti_5O_{14}$ have been prepared by conventional solid state ceramic route. The sintered ceramic samples have been characterised by X-ray diffraction and Scanning Electron Microscopy (SEM). The dielectric properties in the microwave frequency range have been measured using conventional microwave dielectric resonator methods. The $BaNd_2Ti_3O_{10}$, $BaN_2Ti_4O_{12}$ and $BaNd_2Ti_5O_{14}$ have dielectric constants $(\epsilon_r) \sim 60$, 84 and 77 respectively. They have relatively high quality factors.

Keywords: Dielectric resonator; microwave ceramic; barium-neodymium titanate; dielectric ceramic

INTRODUCTION

The growing importance of ceramic dielectrics for application in microwave integrated circuits has led to great advances in the material research and development of dielectric ceramic systems. [1–9] Dielectric resonators provide significant advantages in terms of compactness, light weight, temperature stability and relatively low cost in the production of a variety

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of microwave devices such as band-pass filters and band-stop filters, and frequency stabilization of solid state oscillators. A high dielectric constant (ε_r), a high quality factor (Q) and small or zero variation of resonant frequency with temperature are the most desirable properties for these ceramics.

In pursuit of high-ε_r ceramics, many researchers have studied the $BaO-Ln_2O_3-TiO_2$ (where Ln is any lanthanide) system. Among the ternary oxide compounds of this system, ceramics containing Nd and Sm with stoichiometry near to $BaLn_2Ti_5O_{14}$ have been widely studied. [2–6, 9] In the $BaO-Nd_2O_3-TiO_2$ compounds, the microwave dielectric properties of $BaNd_2Ti_5O_{14}$ have been widely studied and found to be useful for resonator applications. It has been shown that Sm substitution for Nd in $BaNd_2Ti_5O_{14}$ lowers the temperature coefficient of the dielectric ceramic without significant change of the ε_r value. [10] The $BaNd_2Ti_3O_{10}$ has relatively high dielectric constant, low loss and good thermal stability in the low frequency region. [1] Recent reports show that in the $BaO-Ln_2O_3-TiO_2$ system, 1:1:4 compounds have higher ε_r than the corresponding 1:1:5 compounds. [4, 7] This paper reports the preparation, characterisation and properties of $BaNd_2Ti_3O_{10}$ (N2T3), $BaNd_2Ti_4O_{12}$ (N2T4) and $BaNd_2Ti_5O_{14}$ (N2T5) as dielectric resonators.

METHODS AND MATERIALS

The ceramics were prepared by the solid state ceramic route. Stoichiometric amount of high purity TiO_2, Nd_2O_3 and $BaCO_3$ (purity > 99.9%) were weighed and wet mixed in distilled water for about 1 hour, dried and then calcined in platinum crucibles in the range 1200–1300°C for 4 hours. The calcined powders are ground again for 1 hour and the fine powders were pressed (pressure 120 MPa) into cylindrical discs with 10 mm diameter and a thickness of 6–7 mm for microwave measurements and about 1 mm thickness for low frequency measurements. The compacts were sintered at different temperatures in the range 1350–1450°C for 4 hours. The sintered compacts were polished and the bulk densities were measured. The phase constitution and crystal structure were examined using X-ray diffraction (XRD). The dielectric properties of the DRs in the low frequency range (100 kHz–13 MHz) were measured on metallised samples using a HP 4192 A Impedance Analyser. The dielectric constant and unloaded quality factor at microwave frequency were measured by a HP 8510 B Network Analyser and HP 8514 B Reflection-Transmission unit. The whole set-up was controlled using a HP 9000, 300 series computer. The ε_r was calculated from the TE_{011} resonance mode of the end-shorted samples placed between two conducting plates using the method Hakki and Coleman [11] and modified by Courtney.[12] The Q factors ($TE_{01\delta}$ mode) were measured by microstrip-line method of Khanna and Garault.[13] The coefficient of thermal variation of resonant frequency was measured by noting the temperature variation of resonant frequency of the TE_{011} mode over the temperature range of 20 – 80°C.

RESULTS AND DISCUSSION

The bulk densities of N2T3, N2T4 and N2T5 are given in Table I. The theoretical densities and lattice parameters of these materials reported earlier are also included in the Table I. The microstructures of these ceramics recorded using a Scanning Electron Microscope are shown in Figure 1. The SEM pictures show single phase appearance with grains of 2 to 3 μ size. The SEM photographs do not show any indication of liquid phase formation. The powder XRD patterns of N2T3 and N2T5 compounds (Figs. 1a, c) are in agreement with that reported by Kolar *et al.* [1]. The pattern of N2T4 shows agreement with that reported by Takahashi *et al.* [14, 15]. However, all the three XRD patterns contain small additional peaks due to the presence of secondary phases. Kolar *et al.* [1] had obtained $BaTiO_3$ and TiO_2 as secondary phases along with $BaNd_2Ti_3O_{10}$. In the case of N2T5, TiO_2 and $Ba_2Ti_9O_{20}$ were found as additional phases. [1, 9] The reported crystal structures of these free ceramics is orthorhombic with four molecular formula per unit cell. [1, 9, 15]

Figure 3 shows the variation of ε_r of N2T3 ceramics with frequency in the 100 Hz – 13 MHz region. The N2T4 and N2T5 are well studied [2, 4–6, 8, 9] and so their low frequency values are not shown. The ε_r was obtained from the capacitance of metallised ceramic of about 1 mm thickness and 10 mm diameter using a HP 4192 A Impedance Analyser.

TABLE I Lattice parameters and densities of materials in the $BaO-Nd_2O_3-TiO_2$ system

Material	*Lattice parameters (A°)*			*Density (g/cc)*	
	a	*b*	*c*	*Theoretical*	*Experimental*
$BaNd_2Ti_3O_{10}$	7.62	28.16	3.87	5.85	5.39
$BaNd_2Ti_4O_{12}$	22.35	12.2	3.85	5.15	5
$BaNd_2Ti_5O_{14}$	12.2	22.35	3.84	5.62	5.14

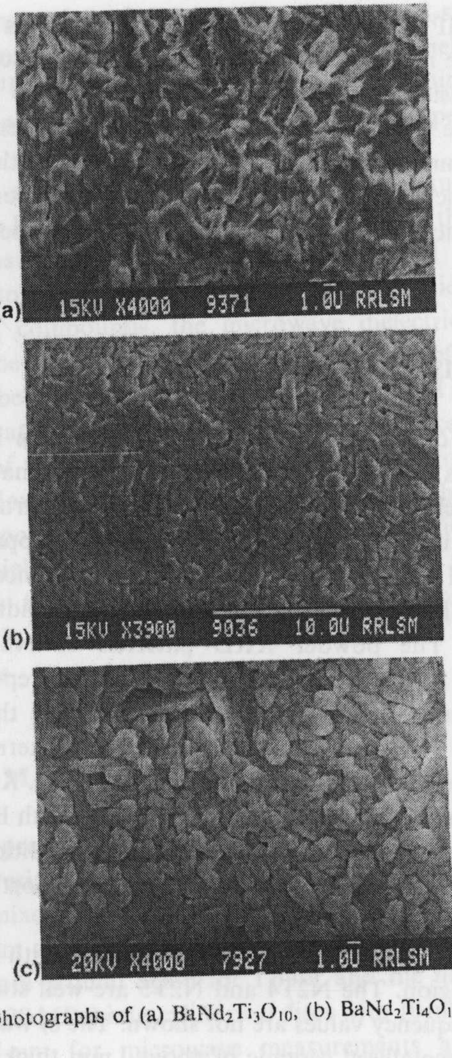


FIGURE 1 SEM photographs of (a) $BaNd_2Ti_3O_{10}$, (b) $BaNd_2Ti_4O_{12}$ and (c) $BaNd_2Ti_5O_{14}$ ceramic.

The results of the microwave measurements are shown in Table II. The ε_r of N2T3 at low frequency region and 4 GHz are nearly the same. When compared with N2T4 and N2T5, N2T3 has the lowest dielectric constant and highest temperature coefficient (+140 ppm/K). The Qu of N2T3 at 4.3 GHz ($TE_{01\delta}$) is 1, 280. Assuming Qu$\sim$1/tan δ, the calculated tan δ is 7.813×10^{-4}. The N2T4 and N2T5 showed Qu of 2, 140 and 1, 190 respectively at 2.89 and 3.88 GHz (Tab. II). The ε_r values of these ceramics are 83.8 and 77.6, higher than N2T3.

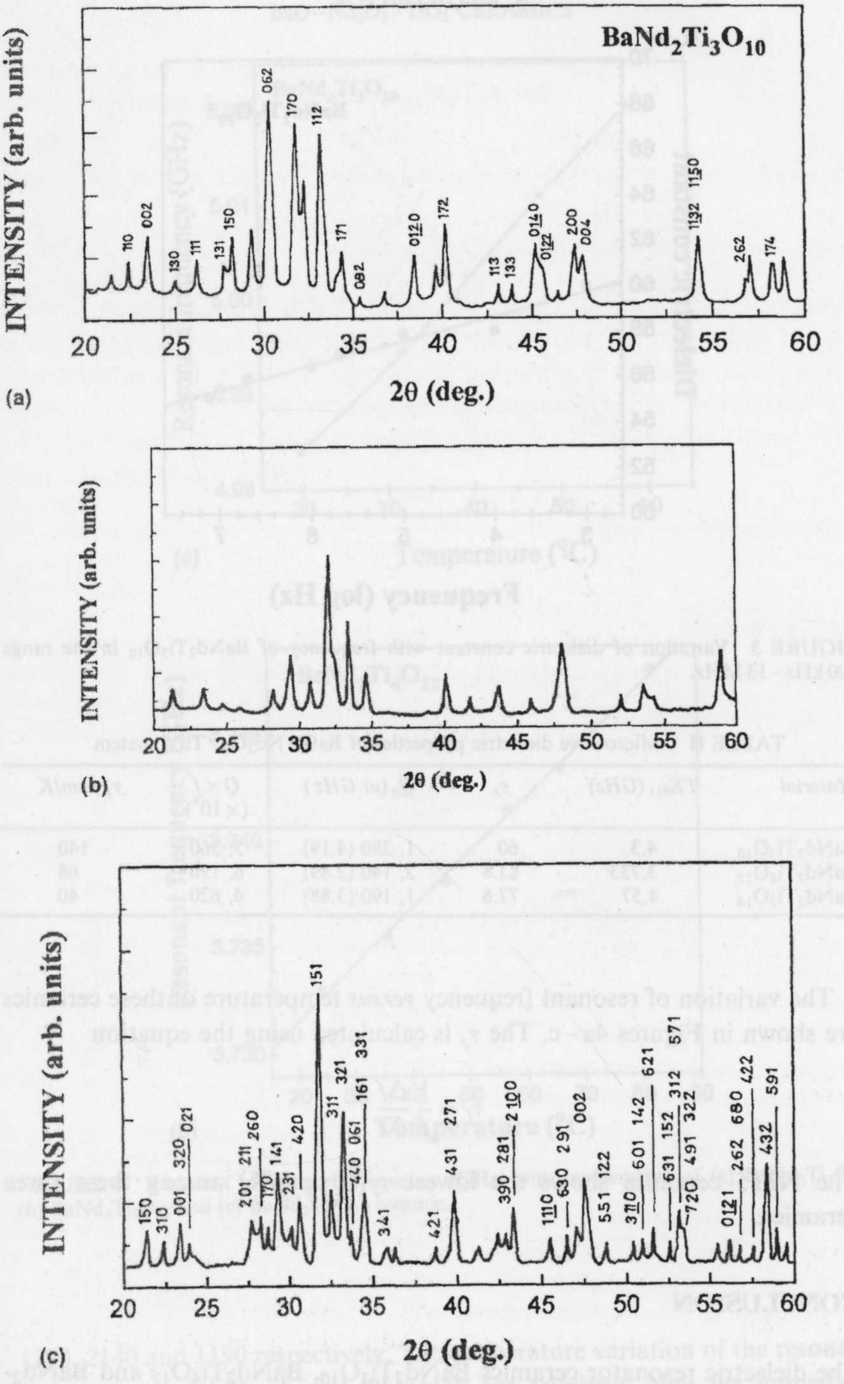


FIGURE 2 Powder X-ray diffraction patterns of (a) $BaNd_2Ti_3O_{10}$ (b) $BaNd_2Ti_4O_{12}$ and (c) $BaNd_2Ti_5O_{14}$ ceramic obtained using CuKα radiation.

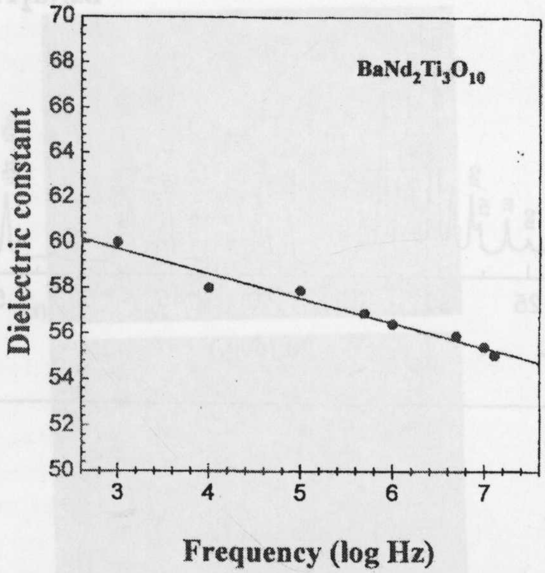


FIGURE 3 Variation of dielectric constant with frequency of $BaNd_2Ti_3O_{10}$ in the range 100 kHz – 13 MHz.

TABLE II Microwave dielectric properties of $BaO-Nd_2O_3-TiO_2$ system

Material	TE_{011} *(GHz)*	ε_r	Q_u *(at GHz)*	$Q \times f$ $(\times 10^9)$	τ_f *ppm/K*
$BaNd_2Ti_3O_{10}$	4.3	60	1, 280 (4.19)	5, 360	140
$BaNd_2Ti_4O_{12}$	3.733	83.8	2, 140 (2.89)	6, 190	68
$BaNd_2Ti_5O_{14}$	4.57	77.6	1, 190 (3.88)	4, 620	40

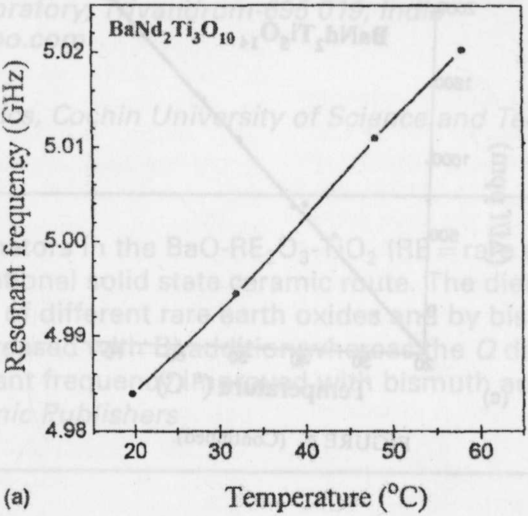
The variation of resonant frequency *versus* temperature of these ceramics are shown in Figures 4a – c. The τ_f is calculated using the equation

$$\tau_f = \frac{1}{f}\frac{\Delta f}{\Delta t}$$

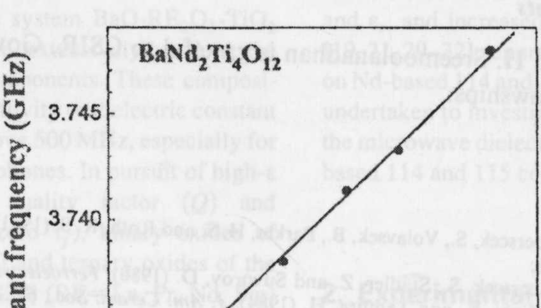
The N2T5 ceramics shows the lowest τ_f(40 ppm/K) among these three ceramics.

CONCLUSION

The dielectric resonator ceramics $BaNd_2Ti_3O_{10}$, $BaNd_2Ti_4O_{12}$ and $BaNd_2$-Ti_5O_{14} have dielectric constant of 60, 84 and 78 and quality factors $(Q \times f)$



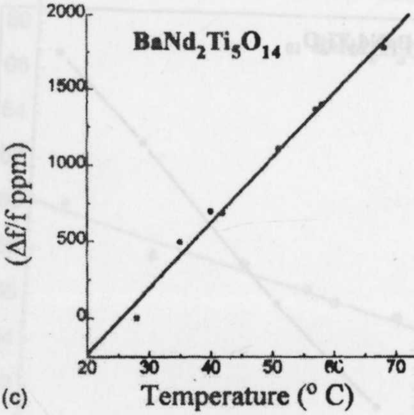
(a)



(b)

FIGURE 4 Variation of resonant frequency (GHz) with temperature of (a) $BaNd_2Ti_3O_{10}$, (b) $BaNd_2Ti_4O_{12}$ and (c) $BaNd_2Ti_5O_{14}$ ceramic.

1280, 2140 and 1190 respectively. The temperature variation of the resonant frequencies are 140, 68 and 40 which are relatively high for immediate practical applications.



(c)

FIGURE 4 (Continued).

Acknowledgements

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