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Dielectric Properties of $Ba_5Nb_4O_{15}$ Ceramic

By

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Introduction Ceramic dielectrics with high dielectric constant (relative permittivity ϵ_r) in the microwave frequency range are used as filters, oscillators [1], etc. in microwave integrated circuits (MICs) particularly in modern communication systems like cellular telephones and satellite communications. Such ceramics, known as 'dielectric resonators (DRs)', do not only offer miniaturisation and reduce the weight of the microwave components, but also improve the efficiency of MICs.

DRs chosen for practical applications possess high dielectric constant ($\epsilon_r > 30$), low loss ($\tan \delta < 2 \times 10^{-3}$), and small temperature coefficient of resonant frequency ($\tau_f < \pm 20$ ppm/K). In pursuit of high ϵ_r materials, a number of ceramics such as $Ba_2Ti_9O_{20}$ [2], (Zr, Sn)TiO₃ [3], Ba(Zn, Ta)O₃ [4], and $A(B_{1/2}^{3+}B_{1/2}^{5+})O_3$ perovskites [5, 6] have been reported. In the present note, we report preparation, characterisation, and dielectric properties of $Ba_5Nb_4O_{15}$ (B_5N_4).

Experimental The material B_5N_4 is prepared by the conventional solid state ceramic preparation route. High purity (>99.9%) $BaCO_3$ and Nb_2O_5 are mixed in the molar ratio 5:4 in an agate mortar for about 1 h in the presence of acetone. The dried mixture is then calcined in an alumina crucible at 1200 °C for 6 h. The calcined powder is thoroughly ground for 90 min. PVA is added to it as binder and again ground for 30 min. The slurry is then pressed into the form of thick cylinders by applying 1.5 to 2.0 t in a Carver press. The cylindrical pellets are slowly heated to about 200 °C to expel the binder and then sintered at 1420 °C for 4 h. The sintered pellets are polished.

The bulk density of the pellets is measured by the dimensional method. From a thick cylinder one thin disc is cut and powdered. The powder sample is studied using a Rigaku powder X-ray diffractometer. Another thin disc cut from the thick pellet is used as a capacitor by applying silver paste to the faces. The ϵ_r and $\tan \delta$ are obtained using an HP 4192 impedance analyser in the range 100 Hz to 10 MHz.

The dielectric constant in the microwave frequency region is measured using the 'dielectric post resonator' method proposed by Hakki and Coleman [7] and later modified by Courtney [8]. The test sample which is in the form of a cylinder, $D/L \approx 2.0$ is placed between two large conducting plates. The TE_{011} resonance is identified using the network analyser (HP 8510 B) as f_0 . From the sample dimensions D and L , and the mode chart given in [7],

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ϵ_r is calculated using the formula

$$\epsilon_r = 1 + \left(\frac{\lambda_0}{\pi D} \right)^2 (\alpha_1^2 + \beta_1^2),$$

where $\lambda_0 = c/f_0$, c being the velocity of electromagnetic waves in free space.

The sample is then placed inside an aluminium cylindrical cavity whose D and L , are approximately 10 times larger than the sample. The whole set-up is heated slowly using a hot plate. The resonant frequency shift is noted every 10 K, from 25 to 75 °C. A graph is drawn with temperature as abscissa and frequency as ordinate. The temperature coefficient is then obtained from

$$\tau = \frac{1}{f_0} \frac{\Delta f_0}{\Delta T} \text{ [ppm/K]}.$$

The unloaded quality factor (Q_u) of the resonator is determined using the stripline method of Khanna and Garault [9]. The resonator is placed symmetrically near a 50 Ω stripline inside a brass cavity of size $6 \times 5 \times 5 \text{ cm}^3$ which serves as shield. From the transmission curve, the width of the TE_{010} resonance, corresponding to the transmission coefficient

$$S_{21u} = S_{21} \sqrt{\frac{2}{1 + S_{21}^2}}$$

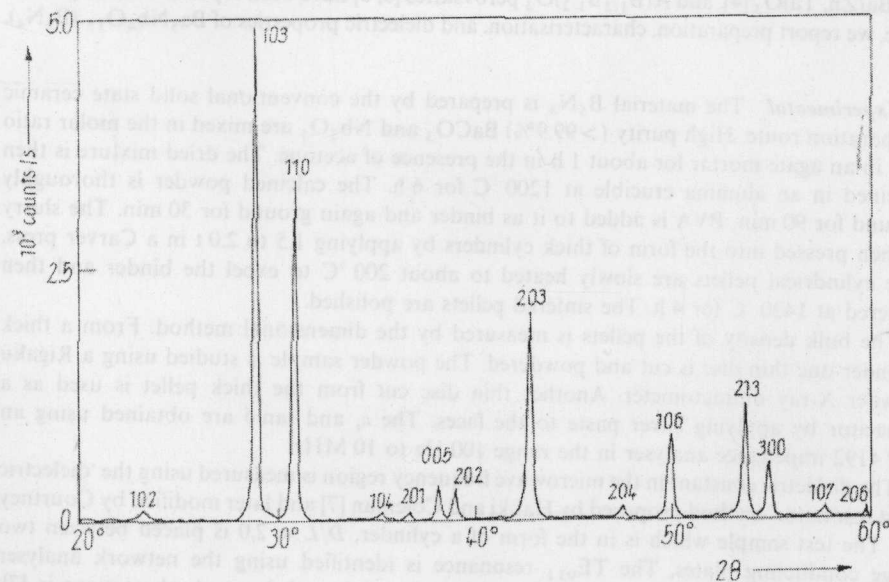


Fig. 1. X-ray powder diffraction pattern recorded from $\text{Ba}_3\text{Nb}_4\text{O}_{15}$ using $\text{CuK}\alpha$ radiation

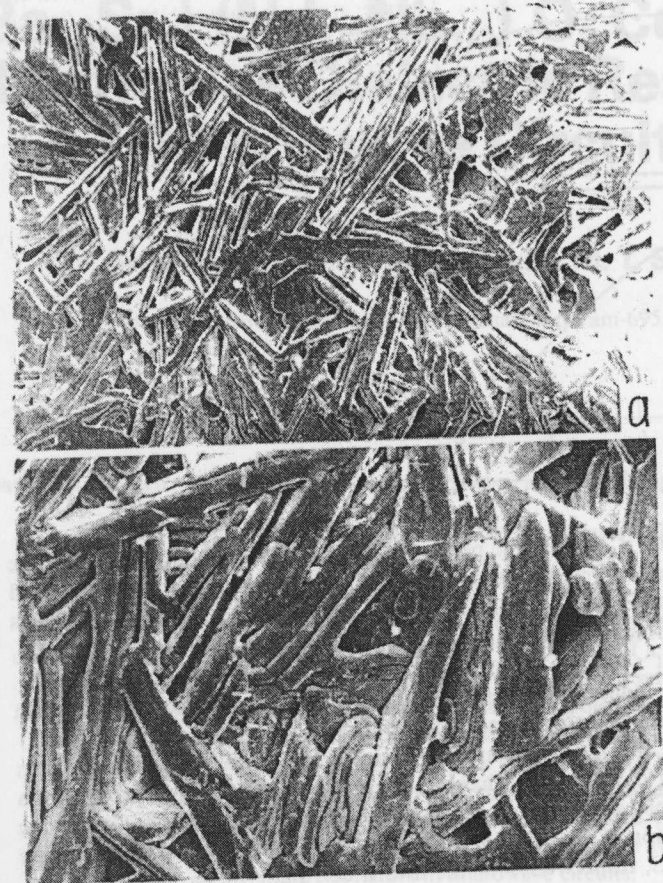


Fig. 2. Scanning electron micrographs recorded from the polished surface of a thermally etched sample

is calculated. If this width is Δf and the resonant frequency is f_0 , Q_u can be calculated using the relation

$$Q_u = \frac{f_0}{\Delta f}$$

Results and discussion Fig. 1 shows a powder X-ray diffraction pattern recorded from $Ba_5Nb_4O_{15}$. The observed diffraction pattern is in good agreement with JCPDS File No. 14-28. The structure of B_5N_4 is hexagonal. The lattice parameters as calculated from Fig. 1 are $a = 0.576$ and $c = 1.177$ nm. The theoretical density is 6.42 g/cm³. The experimental bulk density is 5.9 g/cm³ (92% TD). Fig. 2 shows the microstructure of a sintered pellet recorded after polishing and thermal etching. The variations of ϵ_r and $\tan \delta$ in the region 100 Hz to 10 MHz are shown in Fig. 3a and b, respectively.

At 7.2 GHz, the ϵ_r obtained is 41.0 and $Q_u > 1.000$. The temperature coefficient of resonant frequency τ_f is +120 ppm/K.

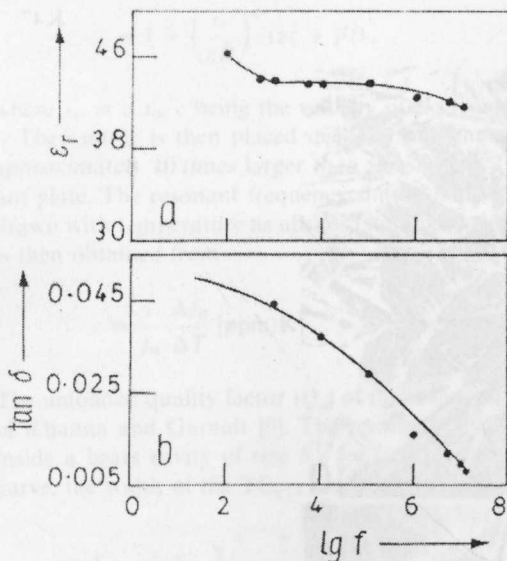


Fig. 3. The dielectric property variation of $\text{Ba}_x\text{Nb}_3\text{O}_{7-x}$ ceramic in the 100 Hz to 10 MHz range: a) ϵ_r vs. frequency, b) $\tan \delta$ vs. frequency

Conclusion To conclude, B_5N_4 ceramic possesses high ϵ_r suitable for use as a DR. The relatively large τ_r precludes its present practical use in high precision devices. Takata and Kageyama [5] reported that $\text{Ba}(\text{Nd}_{1/2}\text{Nb}_{1/2})\text{O}_3$ has positive τ_r and $\text{Sr}(\text{Nd}_{1/2}\text{Nb}_{1/2})\text{O}_3$ has negative τ_r . More recently, Sreemoolanadhan et al. [10] prepared $(\text{Ba}_{1-x}\text{Sr}_x)(\text{Nd}_{1/2}\text{Nb}_{1/2})\text{O}_3$ ceramics and reported that τ_r can be adjusted to a minimum by varying the value of x . Thus, it may be possible to reduce τ_r by suitable substitution and solid solution formation with similar materials having negative τ_r and such work is in progress.

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