Processing and Application of Ceramics 10 [1] (2016) 41–46
DOI: 10.2298/PAC1601041M

Study of lead free ferroelectrics using overlay technique on thick film microstrip ring resonator

Shridhar N. Mathad[1,*], Maruti K. Rendale[2], Roopali N. Jadhav[3], Vijaya R. Puri[3]

[1]*Department of Physics, K.L.E. Institute of Technology, Hubli, Karnataka, India*
[2]*Department of Physics, KLS Gogte Institute of Technology, Belagavi, India*
[3]*Thick and Thin Film Device Lab, Dept. of Physics, Shivaji University, Kolhapur, India*

Received 12 February 2016; Received in revised form 27 February 2016; Accepted 27 March 2016

Abstract

The lead free ferroelectrics, strontium barium niobates, were synthesized via the low cost solid state reaction method and their fritless thick films were fabricated by screen printing technique on alumina substrate. The X band response (complex permittivity at very high frequencies) of Ag thick film microstrip ring resonator perturbed with strontium barium niobates ($Sr_xBa_{1-x}Nb_2O_6$) in form of bulk and thick film was measured. A new approach for determination of complex permittivity (ε' and ε'') in the frequency range 8–12 GHz, using perturbation of Ag thick film microstrip ring resonator (MSRR), was applied for both bulk and thick film of strontium barium niobates ($Sr_xBa_{1-x}Nb_2O_6$). The microwave conductivity of the bulk and thick film lie in the range from 1.779 S/cm to 2.874 S/cm and 1.364 S/cm to 2.296 S/cm, respectively. The penetration depth of microwave in strontium barium niobates is also reported.

Keywords: *thick films, microstrip ring resonator, complex permittivity, strontium barium noibates*

I. Introduction

The interaction of electromagnetic waves with matter is governed by its complex permittivity. All electromagnetic phenomena – transmission, scattering, refraction diffraction, and reflection can be described in terms of the complex permittivity (ε) and permeability (μ). The dielectric constant and loss tangent of materials are important inputs to RF engineering tasks. Knowledge of the dielectric constant (ε_r) and loss tangent ($\tan\delta$) of a dielectric material is required to understand how that material will reacts with electromagnetic fields and behave in RF circuits. The permittivity of dielectrics gauges on characteristics of the medium like density (ρ), chemical composition, frequency (f) and temperature (T) [1]. Dielectric permittivity measurements provide important input to engineering and scientific disciplines due to the effects of permittivity on the interactions between electromagnetic energy and materials. They define the interaction between static or dynamic electric and magnetic fields and the materials [2].

* Corresponding author: tel: +919886347873,
e-mail: *physicssiddu@gmail.com* (S.N. Mathad)
e-mail: *vijayapuri@gmail.com* (V.R. Puri)

Measurements of microwave dielectric properties are broadly classified as resonant method and non-resonant methods [3–5]. In general, dielectric measurement techniques can be categorized as reflection or transmission type, using resonant or non-resonant systems, with open or closed structures. Various techniques can be used for measurement of microwave properties of materials in the time and frequency domain [3–7]. Material characterization is an important field in microwave engineering and is used in the system design of high-speed circuits to satellite and telemetry applications. Every material has a unique set of electrical characteristics that are dependent on its dielectric properties. Accurate measurements of these properties can provide scientists and engineers with valuable information to properly incorporate the material into its intended application for more materials designs or to monitor a manufacturing process for improved quality control. The resonance techniques are more accurate than the transmission techniques, especially in calculating small loss tangent or loss factors, therefore resonance techniques are widely used [3]. The ring resonator is simply a transmission line formed in a closed loop. The basic circuit consists of the feed lines, coupling gaps and the resonator (Figure 1). When the mean circumference of the ring resonator is equal to an integral multiple of a guided wavelength, resonance is established. This may be expressed by following equation [3,8]:

$$2\pi \cdot r = m \cdot \lambda_g \tag{1}$$

for m = 1, 2, 3... integer , where r is the mean radius of the ring that equals the average of the outer and inner radii and λ_g is the guided wavelength.

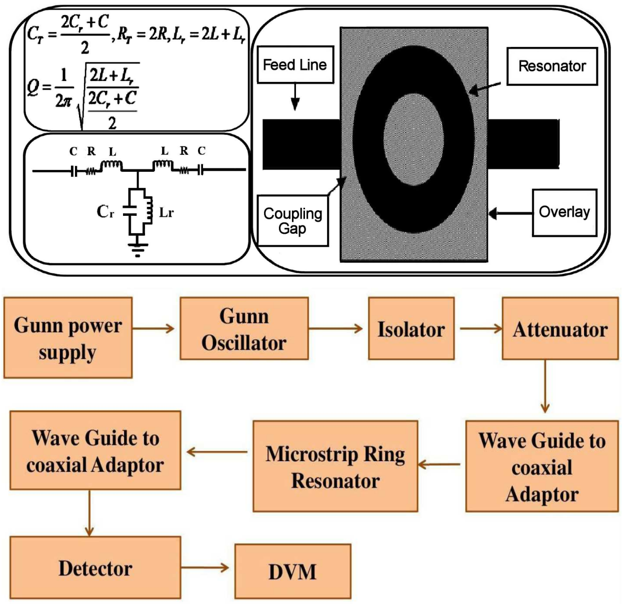


Figure 1. Schematic structure, equivalent lumped circuits of MSRR and block diagram of experimental setup used for Ag thick film MSRR

In this paper, we report the perturbation of Ag thick film microstrip ring resonator (MSRR) with superstrate of both bulk (pellets form) and thick film of strontium barium niobates (SBN - $Sr_xBa_{1-x}Nb_2O_6$) at very high frequencies (microwave frequencies) in absence of external DC magnetic field. The influence of strontium content in $Sr_xBa_{1-x}Nb_2O_6$ (x = 0.4, 0.5, 0.61, and 0.75) on microwave conductivity and penetration depth of bulk and thick films was also investigated. No reports are found in the literature relating to the microwave dielectric properties of strontium barium niobates (both in bulk and thick film form).

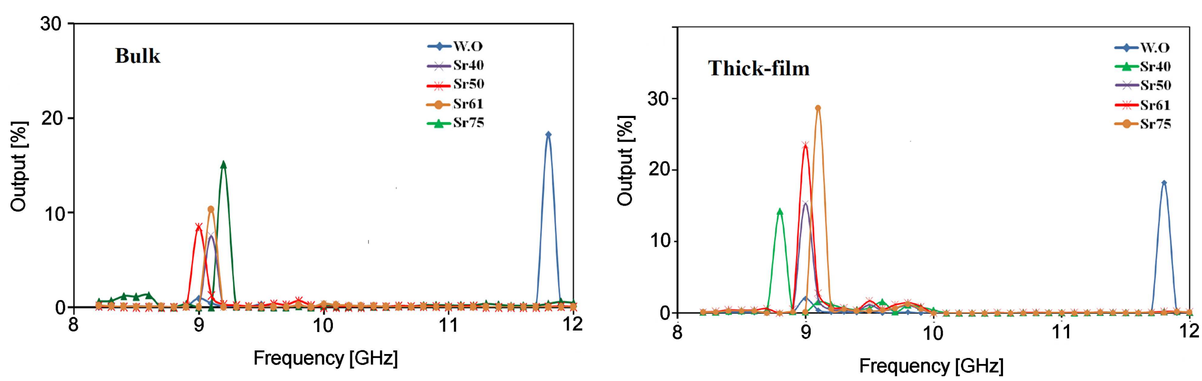
II. Methodology

A series of polycrystalline samples of lead free ferroelectric $Sr_xBa_{1-x}Nb_2O_6$ (where x = 0.40, 0.50, 0.61 and 0.75, and corresponding notation Sr40, Sr50, Sr61 and Sr75, respectively) were prepared by a standard ceramic method, using analytical grade highly pure $SrCO_3$ (99.95%), $BaCO_3$ (99.95%) and Nb_2O_5 (99.999%) in stoichiometric ratio. The tetragonal structured (*P4bm*) powders were characterized in earlier publications [9,10]. Finally, the calcined powder was also used for the preparation of thick paste and pellets. The SBN ceramic paste was prepared using the strontium barium niobate powder, bismuth oxide (Bi_2O_3) binder, and organic vehicle in suitable proportion. They were screen printed on alumina substrates and fired in muffle furnace for 4 h at 900 °C. This screen printing technique is highly conducive for the planarization of bulk materials and is cost effective compared to other techniques. The thick films were fabricated on alumina substrate with dimensions 1 cm × 2.5 cm and thickness of ~32 µm. The Ag thick film ring resonator was also delineated using screen printing on alumina (Kyocera Japan). The used X band setup consisted of signal generator, isolator, attenuators, SMA connector, RF detector and launchers. The output was measured in terms of voltage. The SBN samples (bulk and thick films) were kept in touch with the microstrip ring resonator (MSRR) at its centre. In the case of the SBN films the film side was in contact with the MSRR. The change in the resonance frequency and amplitude was measured.

III. Results and discussion

3.1. Microwave studies

For transmittance measurements, the bulk and thick film of SBN ceramics were kept in touch with the MSRR covering the coupling gaps. The variations of output with frequency for the microstrip ring res-



(a) (b)

Figure 2. Output of MSSR as a function of frequency for: a) SBN ceramics and b) thick films

onator were shown in Fig. 2 for both SBN bulk and thick film samples. For the sample WS, without superstrate (microstrip ring resonator which is not perturbed with SBN), the resonant frequency was observed at 11.7 GHz, with peak output of 18.26%. Due to superstrate, the observed resonance peaks due to both bulk and thick films of all SBN ceramics shifts towards lower frequency. The output (transmission coefficient) resonance increases substantially for the thick film samples, whereas the output resonance peaks for the bulk samples become lower as compared to the sample without superstrate (WS). The resonant frequency increases with the increase of strontium content for both bulk and thick film. In the case of superstrate with the bulk SBN samples the peak output decreases from 18% (WS) down to 15% (bulk) whereas in the thick films peak output increases up to 28.6%.

The resonator shows broadband characteristics due to the bulk and thick film superstrate. The variations of peak output, quality factor and band width with Sr content are shown in Fig. 3. There is an enhancement in the bandwidth (67.5 MHz) of the Sr50 thick film sample as compared to other samples, which has the lowest Q quality factor of 133.33. The Sr61 bulk sample has the highest quality factor in all bulk compositions, together with very low peak output of ~10.3%. The Sr40 thick film has the highest quality factor 153.04. These variations in bandwidth and quality factor are due to the perturbation and change of the even mode characteristics. The peak shifting towards lower frequencies

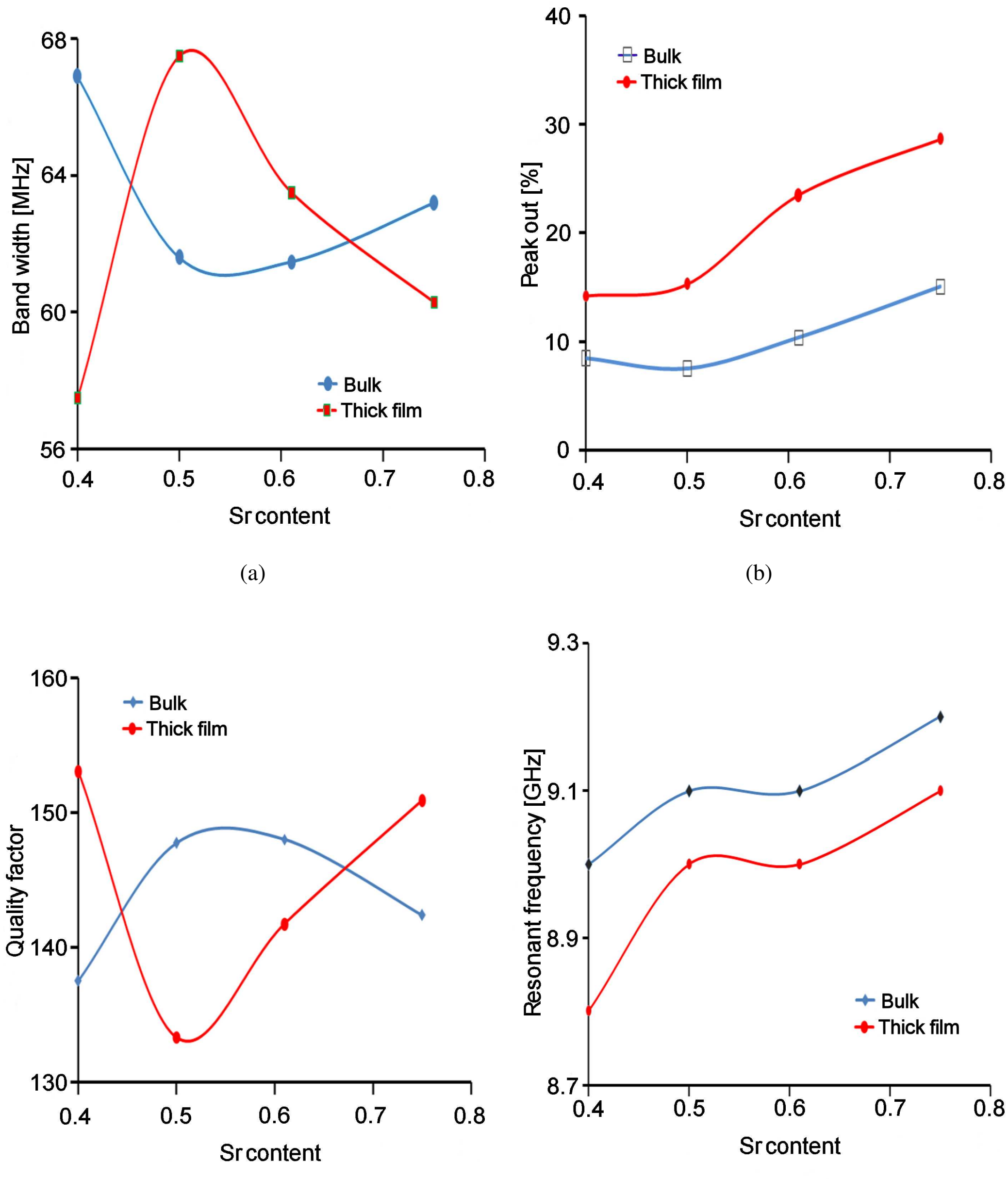


Figure 3. Output of MSSR as a function of frequency for SBN: a) ceramics and b) thick films

Table 1. Resonant frequency, cut off frequency, band width, quality factor and peak output of microstrip ring resonator due to superstrate with SBN bulk ceramics

Sample	Resonant frequency [GHz]	Cut off frequency f_1 [GHz]	f_2 [GHz]	Band width [MHz]	Quality factor	Peak output
Sr40	9.0	8.87	9.04	66.9	137.52	8.50
Sr50	9.1	9.07	9.13	61.6	147.73	7.54
Sr61	9.1	9.07	9.13	61.5	148.02	10.38
Sr75	9.2	9.17	9.23	63.2	142.41	15.09

Table 2. Resonant frequency, cut off frequency, band width, quality factor and peak output of microstrip ring resonator due to superstrate with SBN thick films

Sample	Resonant frequency [GHz]	Cut off frequency f_1 [GHz]	f_2 [GHz]	Band width [MHz]	Quality factor	Peak output
Sr40	8.8	8.77	8.83	57.5	153.04	14.23
Sr50	9.0	8.97	9.04	67.5	133.33	15.30
Sr61	9.0	8.97	9.04	63.5	141.73	23.47
Sr75	9.1	9.07	9.13	6303	150.91	28.65

is due to even mode coupling, in which contribution of transmission is also prominent [11]. This cause changes in transmission (S_{21}) and reflection (S_{11}) shift in resonance frequencies, variations in Q and bandwidth. The detailed resonant frequency, cut-off frequencies, band width, quality factor, and peak output of MSRR due to SBN bulk and thick films superstrate are tabulated in Table 1 and Table 2, respectively.

3.2. Complex permittivity studies

The dielectric superstrate perturbs the fringing field of the microstrip component. The dominant mode for the microstrip ring resonator is TM_{11}. The complex permittivity of SBN ceramics was calculated from the frequency shift of the microstrip ring resonator due to the superstrate with SBN ceramics [12]:

$$\varepsilon' = 1 + \frac{C_0}{K}\left[\left(\frac{f_a}{f_r}\right)^2 - 1\right] \tag{2}$$

$$\varepsilon'' = \frac{N'}{f_r \cdot r_0} \tag{3}$$

where, the ratio C_0/K is a constant term related to the standard alumina sample (substrate) given as:

$$\frac{C_0}{K} = \frac{\varepsilon'_s - 1}{\left(\frac{f_a}{f_s}\right)^2 - 1} \tag{4}$$

where, ε'_s is the permittivity of the standard sample, f_a, f_r and f_s are the resonant frequencies without superstrate, with superstrate of SBN ceramic samples and standard alumina samples, respectively. Also:

$$N' = \varepsilon'_s \cdot f_s \cdot r_{0s} \tag{5}$$

where r_0 and r_{0s} are the reflection coefficient of the SBN samples and standard sample, respectively.

The dielectric constant (ε') and dielectric loss (ε'') of all SBN samples in function of strontium content were shown in Figs. 4 and 5. In case of the thick films, the dielectric constant and loss values range from 16.10 to 18.69 and 2.69 to 4.9, respectively. For the bulk samples the dielectric constant and loss values vary in the range from 15.30 to 16.99 and 3.51 to 5.74, respectively. The dielectric constant of the bulk samples increases with the increase of Sr content. On the other hand, the dielectric constant of the thick films decreases as Sr content increases and the sample Sr40 has the highest dielectric constant with magnitude of 18.69. It is also noticed that for the bulk and thick films loss tangent does not follow any relation with strontium content. The observed increase of the resonant frequency with in-

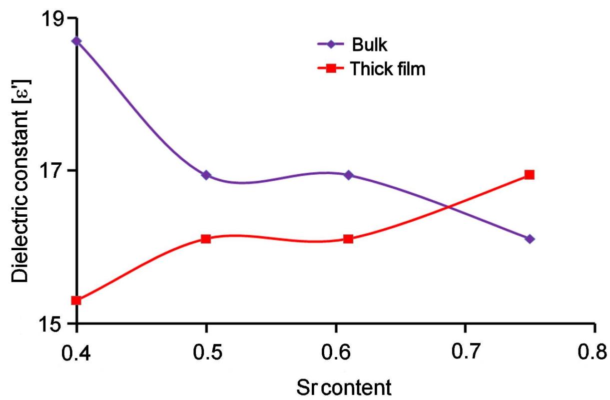


Figure 4. Variation of dielectric constant with Sr content

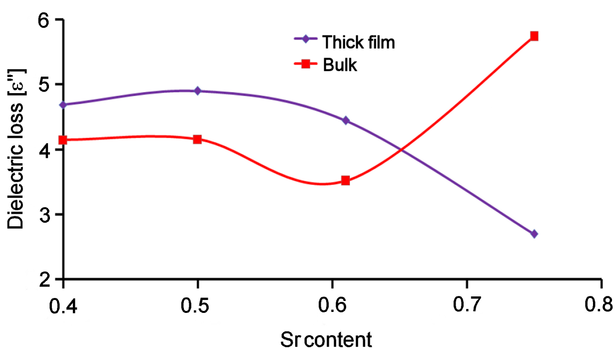


Figure 5. Variation of dielectric loss with Sr content

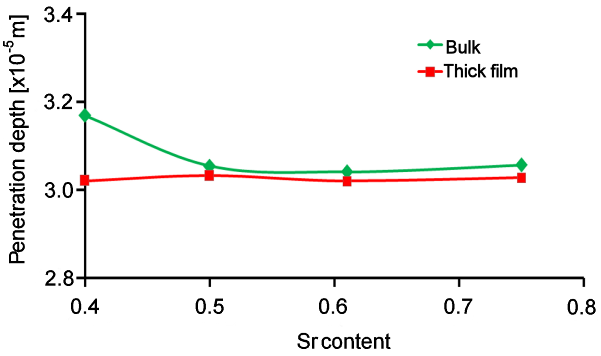


Figure 6. Variation of penetration depth with Sr content

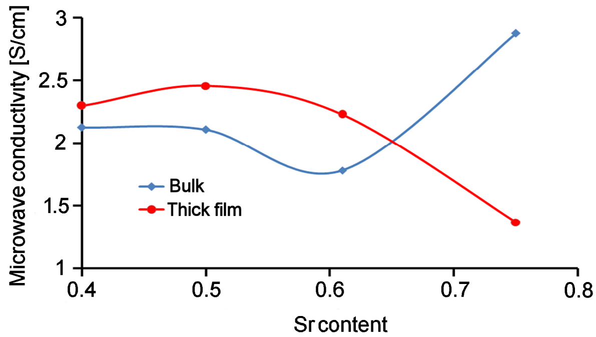


Figure 7. Variation of microwave conductivity with Sr content

crease of Sr content is attributed to the rise in the effective dielectric constant. So, the fringing field lines get concentrated due to the dielectric constant of the superstrate material, thus increasing the fringing field capacitance which results in decrease in resonance frequency. Owing to the larger thickness of the bulk material and larger dielectric constant, the attenuation of the fringing fields might be more prominent leading to the suppression of the radiation loss which ultimately results in decreased power output [11,12]. The superstrate on Ag thick film microstrip ring resonator is an efficient tool capable of detecting the composition dependent changes in microwave properties of ceramic bulk and thick films. The bulk and thick film superstrate decreases the resonance frequency of MSRR at lower frequencies. In this technique even minor change in the superstrate material properties changes the MSRR response.

The penetration depth was determined to be the depth at which the power density has dropped to 37% of its initial value at the surface. The penetration depth of the bulk and thick film SBN samples were presented in Fig. 6. It shows that the penetration depth is greater in the bulk as compared to the thick films. The penetration depth of microwave in the ceramics can be calculated by using the following relation [13]:

$$D = \frac{c}{\omega}\left[2\varepsilon'\left(1+\left(\frac{\varepsilon''}{\varepsilon'}\right)^2\right)^{\frac{1}{2}} - 1\right]^{\frac{1}{2}} \tag{6}$$

where c is speed of light, ω is angular frequency, ε' and ε'' permittivity and dielectric loss of material. The reciprocal of the effective attenuation coefficient (penetration depth) ranges from 3.053×10^{-5} to 3.169×10^{-5} m for the bulk, and from 3.02×10^{-5} to 3.032×10^{-5} m for the thick film samples as function of strontium content (Fig. 6).

The microwave conductivity was calculated using the equation [14]:

$$\sigma = \omega \cdot \varepsilon'' \cdot \varepsilon_0 \tag{7}$$

where ω is angular frequency, ε_0 and ε'' are permittivity of free space and dielectric loss of material, respectively.

The variation of microwave conductivity with strontium content is indicated in Fig. 7. We observe that the microwave conductivity decreases with the increase of strontium content in the thick film, and increases in the bulk SBN samples. The microwave conductivity of the bulk and thick film lie in the range of 1.779 S/cm to 2.874 S/cm and 1.364 S/cm to 2.296 S/cm, respectively. The microwave conductivity of the bulk samples Sr61 and Sr75 were 1.779 S/cm and 2.874 S/cm, respectively. The microwave conductivity of the thick film samples Sr40 and Sr75 were 2.296 S/cm and 1.364 S/cm, respectively.

IV. Conclusions

In this paper we have reported the microwave properties of microstrip ring resonator perturbed with strontium barium niobate bulk and thick films. The influence of strontium content on the microwave properties was investigated. This overlay tool can be conveniently utilized to determine the complex permittivity of ceramics at very high frequencies (X band). Variation of microwave conductivity and penetration depth was also reported.

References

1. A. Kraszewski, "Microwave aquametry – A review", *J. Microwave Power*, **15** (1980) 209–220.
2. I. Waldron, *Ring Resonator Method for Dielectric Permittivity Measurement of Foams*, Degree of Master of Science, Worcester Polytechnic Institute, Electrical and Computer Engineering, Worcester England, 2006.
3. L.F. Chen, C.K. Ong, C.P. Neo, V.V. Varadan, V.K. Varadan, *Microwave Electronics Measurement and Materials Characterization*, John Wiley & Sons Ltd., England, 2004.
4. S.N. Mathad, R.N. Jadhav, P. Jadhav, V. Puri, "Modification of Ag thick film microstripline due to superstrate strontium barium niobate thick films", *Int. J. Comput. Technol.*, **1** [1] (2014) 100–103.
5. R.S. Pawar, S.N. Mathad, N.D. Patil, V.R. Puri, "Modification of Ag thick film microstripline due to superstrate Ni-Cu-Fe-Mn-O ceramics", *Int. J. Eng. Innovative Technol.*, **3** [4] (2013) 220–223.

6. U.C. Hasar, O.E. Inan, "A position - invariant calibration - independent method for permittivity measurement", *Microwave Opt. Technol. Lett.*, **51** [6] (2009) 1406–1408.
7. U.C. Hasar, "Unique permittivity determination of low-loss dielectric materials from transmission measurements at microwave frequencies", *Progr. Electromagn. Res.*, **107** (2010) 31–46.
8. K. Chang, L.-H. Hsieh, *Microwave Ring Circuits and Related Structures*, 2nd edition, John Wiley & Sons Ltd., New Jersy, 2004.
9. S.N. Mathad, R.N. Jadhav, R.P. Pawar, V. Puri, "Electromagnetic behavior of lead free ferroelectrics at microwave frequencies", *Adv. Sci. Eng. Medicine*, **5** (2013) 1–7.
10. S.N. Mathad, V. Puri, "Structural and dielectric properties of $Sr_xBa_{1-x}Nb_2O_6$ ferroelectric ceramics", *Archives Phys. Res.*, **3** [2] (2012) 106–115.
11. R.N. Jadhav, V. Puri, "Response of Ag thick film microstrip straight resonator to perturbation of bulk and thick film $Ni_{(1-x)}Cu_xMn_2O_4$ ($0 \leq x \leq 1$) ceramics", *Microelectro. Int.*, **28** (2011) 53–57.
12. S.N. Mathad, R.N. Jadhav, V. Puri, "Microwave studies by perturbation of Ag thick film microstrip ring resonator using superstrate of bismuth strontium manganites", *Microelectro. Int.*, **30** [2] (2013) 85–91.
13. S.N. Mathad, R.N. Jadhav, R.P. Pawar, V. Puri, "Dielectric spectroscopy and microwave conductivity of bismuth strontium manganites at high frequencies", *Electro. Mater. Lett.*, **9** [1] (2013) 87–93.
14. S.O. Nelson, "Fundamentals of dielectric properties measurements and agricultural applications", *J. Microw. Power Electromagn. Energy*, **44** [2] (2010) 98–113.