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Study of Dielectric Constant of (1-x) Zn.xMg.TiO_3 (ZMT) Ceramic Material at Microwave Frequencies as a Function of Composition x and Processing Temperature

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ABSTRACT

In this paper measurement of real part of permittivity and loss tangent of (1-x) *Zn.xMg.*TiO_3 (*ZMT*) *material in powder form in S-band of microwave frequencies is presented and the associated accuracy estimated. This approach is based on the measurement of transmitted power from the cavity at and off resonance. Details of the design and fabrication of the rectangular cavity and the input and output coupling are given. The variation of dielectric properties of* (1-x) *Zn.xMg.*TiO_3 (*ZMT*) *with* x*-value and calcination temperature* (*for* x = 0.1) *is presented. The effect of doping of* V_2O_5 *is also studied. The results of present work may provide useful design guidelines for development of microwave components including dielectric resonator antennas.*

Keywords: *Cavity Perturbation Method, Permittivity, Loss Tangent And Ceramic Material*

1. Introduction

The measurement of electromagnetic properties of materials at microwave frequencies is considered important since it provides the relaxation time, dipole moment in liquids, characteristics of materials for device applications, microwave conductivity, effective mass in semiconductors etc. [1].

The microwave measurement techniques for electromagnetic characterization of materials generally are divided into two groups: Non-resonant methods and resonant methods. Non-resonant methods mainly involve reflection or transmission methods. The reflection methods include open-circuit and short-circuit line methods. The transmission/reflection methods have many varieties, including free-space, coaxial line, waveguide, and planar structures such as microstrip and stripline. These methods have achieved success in permittivity and permeability measurements of high- and medium-loss materials. However, non-resonant methods usually require large size samples and have strict requirements on sample preparation, and their measurement accuracy is not very high, especially for low-loss materials. Resonant methods including resonant perturbation methods can provide higher accuracy as compared with non-resonant ones for these low loss materials [2]. The resonance techniques can be further divided into two categories: The first one is a resonance technique, basically supported by the dielectric sample itself. The sample acts as a dielectric resonator. Metal shields with different geometries are always introduced to prevent radiation loss. This type is called dielectric resonance technique. The second one is perturbation technique, where the resonance is supported by the metal walls of the cavity. The presence of small size sample in the cavity causes only a "perturbation" on the field distributions in the metal cavity [3]. In resonator methods, the sample under measurement is excited as a resonator in the measurement circuit, and its electromagnetic properties are deduced from its resonant properties. In resonant perturbation methods, the sample under measurement is introduced into a resonator (meas-

urement fixture) thus altering the electromagnetic boundaries of the resonator and the electromagnetic properties of the sample are deduced from the change in the resonant properties of the resonator. Due to its high accuracy and flexibility in sample preparation, resonant. perturbation method is widely used for low-loss samples, samples in powder form, small size samples and samples of irregular shapes. Resonant perturbation methods mainly include wall replacement methods and cavity perturbation methods. In wall replacement methods, part of the resonator wall is replaced by sample surfaces. Wall-replacement methods are often used in the study of the electro-dynamic properties of conductors after the discovery of high Tc superconductors. Many methods have been developed based on wall-replacement method to measure the surface resistance of superconducting thin films. The cavity perturbation method is based on the change in the resonant frequency and quality factor of the cavity due to the insertion of a sample inside the cavity at the position of maximum electric field or maximum magnetic field depending on the nature of the parameter to be studied. In this method, the sample under study is introduced into a resonant cavity, and its complex permittivity or complex permeability is determined from the change of resonant frequency and quality factor of the cavity due to the introduction of the sample. Cavity perturbation methods are popular in the study of the electromagnetic properties of dielectrics, semiconductors, magnetic materials and composite materials [4].

Ceramic materials are widely used in the design and development of microwave circuits. Further, low loss ceramic materials find applications in the development of stable microwave sources and dielectric resonator antennas.

In this paper cavity perturbation technique, which is highly accurate and advantageous in the determination of small loss tangents is used to measure the dielectric constant and the loss tangent of ZMT material in powder form in S-band of microwave frequencies. The accuracy of the present technique is determined through measurement on Teflon and comparison of results with that available in the literature [5]. The variations in dielectric properties of the ZMT material with the x-value, the calcination temperature and the V_2O_5 doping for x = 0.1 are also studied experimentally.

2. Theory

The cavity perturbation technique for measurements of dielectric constant of material has been analyzed in the literature [4,6-10]. A brief description of the analysis is given in the following:

Consider two almost identical cavities which are distinguished by subscripts 1 and 2. Cavity 1 is empty and cavity 2 contains very small size dielectric material. Both cavity walls are assumed to be lossless. Maxwell's equations for these cavities can be written as

$$\nabla \mathrm{X} \overline{E}_j = -j\omega_j \mu_j \overline{H}_j \tag{1}$$

$$\nabla \mathrm{X} \overline{H}_j = j\omega_j \varepsilon_j \overline{E}_j \qquad j = 1,2$$

By taking suitable dot products with the four equations given in Equation (1), subtracting and integrating over the cavity volume (remembering that cavity walls are perfectly conducting), one obtains

$$\frac{\omega_2 - \omega_1}{\omega_2} = \frac{\left(\mu_2 - \mu_0\right)\iiint_{V_S} \overline{H}_1 . \overline{H}_2 dV - \left(\varepsilon_2 - \varepsilon_0\right)\iiint_{V_S} \overline{E}_1 \overline{E}_2 dV}{2\varepsilon_0 \iiint_{V_C} \left|\overline{E}_1\right|^2 dV} \tag{2}$$

where ω_1 and ω_2 are respectively the complex resonant angular frequencies before and after the introduction of the sample; ε_0 and ε_2 are respectively the permittivities of free-space and the sample; μ_0 and μ_2 are respectively the magnetic permeabilities of free-space and the sample; V_C is the region enclosed by the cavity, and V_S is the volume of the sample.

Equation (2) is the basic cavity perturbation formula. Derivation of the formula has been done under the assumptions that the cavity walls are perfectly conducting and the perturbation is small [4,10].

In the present work, non-magnetic ceramic material is considered and therefore Equation (2) reduces to

$$\frac{\omega_2 - \omega_1}{\omega_2} = -\frac{\left(\varepsilon_{r2} - 1\right)}{2} \frac{\int_{V_s} \overline{E}_1 \overline{E}_2 dV}{\int_{V_C} \left|\overline{E}_1\right|^2 dV} \tag{3}$$

The relationship of complex angular frequency to real frequency and Q factor is given by

$$\frac{\omega_2 - \omega_1}{\omega_2} = \frac{f_{r2} - f_{r1}}{f_{r2}} + \frac{j}{2}\left(\frac{1}{Q_{L2}} - \frac{1}{Q_{L1}}\right) \tag{4}$$

where f_{r1} and f_{r2} are respectively the resonant frequencies of the cavity before and after the introduction of sample; and Q_{L1} and Q_{L2} are the corresponding loaded quality factors.

From Equations (3) and (4), we get

$$2\left(\frac{f_{r1} - f_{r2}}{f_{r2}}\right) - j\left(\frac{1}{Q_{L2}} - \frac{1}{Q_{L1}}\right) = \left(\varepsilon_{r2} - 1\right) \frac{\iiint_{V_S} \overline{E}_1 . \overline{E}_2 dV}{\iiint_{V_C} \left|\overline{E}_1\right|^2 dV} \tag{5}$$

3. Dielectric Constant and Loss Factor

The length of very small diameter (as compared to cavity size and the wavelength) cylindrical plastic tube which can hold the sample is slightly greater than the narrow dimension '*b*' of the cavity so that it can occupy the entire narrow dimension of the cavity and can be taken out from or inserted into the TE_{101} mode cavity through a small hole cut in the broad wall centre where electric field is maximum.

From Equation (5) the expressions for real and imaginary parts of the dielectric constant can be written as

$$\varepsilon'_{r2} = \varepsilon'_r = \frac{V_c\left(f_{r1} - f_{r2}\right)}{2V_cV_s} + 1 \tag{6}$$

$$\varepsilon''_{r2} = \varepsilon''_r = \frac{V_c}{4V_s}\left(\frac{1}{Q_{L2}} - \frac{1}{Q_{L1}}\right) \tag{7}$$

The Loss factor $\left(\tan\delta\right)$ can be written as

$$\tan\delta = \varepsilon''_r \Big/ \varepsilon'_r \tag{8}$$

4. Design and Fabrication

4.1. Preparation of Sample

In the present study the sample is prepared by the conventional solid state ceramic route. Stoichiometric amount of $MgCO_3$ [99.5%, Thomas Baker, India], ZnO [99.5%, S. D. Fine. Chem. India] and TiO_2 [98%, S. D. Fine. Chem. India] are weighed accurately to make the total amount equal to 15 grams. Weighed powders are mixed thoroughly using acetone in a mortar with pestle for about 2 hour. After that powders were ball milled for 4 hours using acetone as mixing medium. The mixture is then dried and placed for calcination in platinum crucibles at 1200˚C for 12 hours. The calcined powders were ground and mixed with 2% polyvinyl alcohol as binder. It was then pressed using a hydraulic press at optimized load (= 60 kN) to get the pellets. The pellets of all the compositions (x = 0.1, 0.2, 0.3, 0.4 and 0.5) were sintered at 1300˚C for 24 hours at a heating and cooling rate of 4˚C /min. on a platinum foil. The pellets of all the compositions were crushed in a mortar with the help of pestle to obtain powdered sample for measurement purpose.

Other samples of composition (x = 0.1) were also prepared but the calcination temperatures are different for different samples. The calcination temperatures used are 900, 950 and 1000˚C, and total heating time for each sample was chosen to be 9 hours. The doping of V_2O_5 was done in one of the samples for which calcination temperature is 1000˚C.

4.2. Design and Fabrication of Cavity

Rectangular TE_{101} mode transmission cavity for resonance at 2.62 GHz was designed [11] and fabricated using a standard S-band waveguide (WR-284) (**Figure 1).** The length of the cavity is 9.14 cm ($=\frac{\lambda_g}{2}$ at 2.62 GHz). Two copper plates were used as shorting plates to form the cavity from the standard waveguide. For coupling power into the cavity, a hole of diameter D = b/2.2, where b is the height of waveguide [11], was made at the centre of each of the shorting plates. The diameter of each coupling hole comes out to be 1.54 cm for the designed cavity. To insert the powdered sample material into the cavity, a circular slot of diameter 2.5 mm was cut at the centre of one of the broad walls of the cavity. The inner and outer diameters of the polythene capillary tube used for filling the powdered sample are chosen to be 1.3 and 2.3 mm respectively and its length is equal to 3.6 cm.

5. Experiments, Results and Discussion

The arrangement for measuring the dielectric properties of (1-x)Zn.xMg.TiO_3 ceramic material consists of Agilent Technologies make analog microwave source (Model no. E8257D, 250 kHz-20 GHz), SICO make waveguide/coaxial transition, variable attenuator, fabricated cavity and HP power sensor (Model no. 436A) and meter forming a test bench as shown in **Figure 2.** The standard procedure given in the reference [10] was followed to measure power fed to and transmitted from the cavity with and without the sample as a function of frequency deviation from resonance. Resonant frequency corresponds to maximum transmitted power in each case.Measurement of dielectric properties of (1-x) Zn.xMg. TiO_3 ceramic material was carried out for different x-values and different calcination temperatures for x = 0.1. A number of measurements were performed to determine the resonance frequency and 3-dB frequencies 'f_1' and 'f_2' both for cavity with the capillary tube and cavity with tube containing the powdered sample material. The average values of measured and computed parameters of the cavity with the capillary tube but without the sample are shown in **Table 1.** The average values of the parameters of the cavity with the capillary tube containing the sample for different x-values and calcination temperatures are shown respectively in **Tables 2** and **3**. The loaded quality factor for the cavity with and without the sample material is computed using the formula $Q_L = f_r \Big/ \left(f_2 - f_1\right)$, where f_r is the resonant frequency of TE_{101} mode cavity .The values of ε'_r , ε''_r and tan δ of the sample material are evaluated using Equations (6-8)

and the results are shown in **Tables 2** and **3** respectively for different x-values and calcination temperature.

The variations in the values of ε_r' and $\tan\delta$ with x-value are shown in **Figures 3** and **4** respectively.

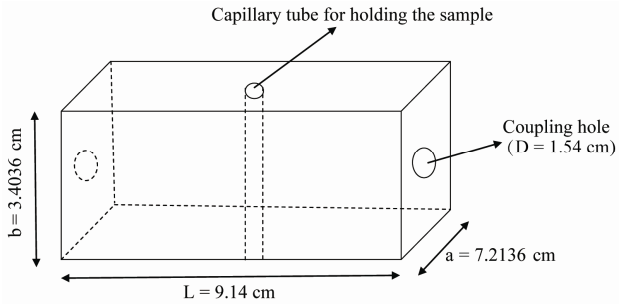
The variations in the values of ε_r' and $\tan\delta$ with calcination temperature for x = 0.1 are shown in **Figures 5** and **6** respectively.

From **Figure 3** and **Table 2** it can be seen that ε_r' value first decreases with increase in x-value and reaches 5.768 at x = 0.3 and then increases for x > 0.3. The loss tangent first increases and reaches 0.004 at x = 0.2, it then decreases and reaches 0.001 at x = 0.3, and again increases for values of x above 0.3, as shown in **Figure 4** and **Table 2**. These variations in the dielectric properties may be attributed to the presence of multiple phases in the material and the variation in the amount of phases determined by the area under peaks with change in x-value as shown in **Figure 7.** The possible 2θ values (Bragg angle) where reflections occur are determined by unit cell dimensions. However, the intensities of the reflections are determined by the distribution of the electrons in the unit cell. The highest electron density is found around atoms. Therefore, the intensities depend on what kind of atoms we have and where in the unit cell they are located. Planes going through areas with high electron density will reflect strongly, planes with low electron density will give weak intensities. From **Figures 5** and **6** and **Table 3**, it is observed that the material ε_r' value increases and tan δ decreases with increase in calcination temperature of the material, though the rates of increase are different. This variation in the dielectric properties with calcinations temperature may be attributed to the presence of single phase in the material. The dielectric constant and loss tangent of V_2O_5 doped and 1000˚C calcined sample were 4.04 and 0.002698. Dielectric constant is higher and loss tangent is lower in comparison with undoped and 1000˚C calcined sample. The change in ε_r' and tan δ of ZMT with the addition of V_2O_5 is due to increase in densification rate of ZMT. The measured dielectric constant values are lower than those reported in reference [5]. This is due to lower value of percentage density of sample material. The sample material used for measurement in [5] was in highly dense pellet form, while in the present study we measured dielectric constant of sample material in powder form. The reported percentage density of solid sample material studied in reference [5] is more than 95%.

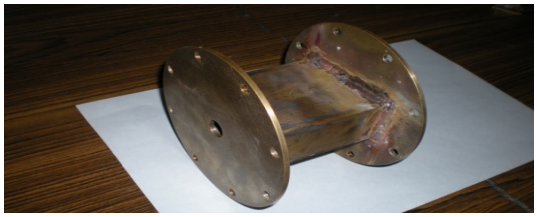
To test the accuracy of the cavity perturbation technique measurement on a sample of Teflon-AF of known dielectric constant ($\varepsilon_r' = 1.93$) [12] was repeated three times. The measured and computed parameters are presented in **Tables 3** and **4**. The average values of ε_r' and $\tan\delta$ are found to be 1.795 and 0.0094.

6. Conclusions

The microwave dielectric properties of Zinc Magnesium Titanate (1-x) Zn.xMg.TiO_3 (x = 0.1, 0.2, 0.3, 0.4, 0.5) in



(a)



(b)

Figure 1. Rectangular S-band cavity (a) Design layout (b) Fabricated cavity.

Figure 2. Experimental setup for dielectric constant measurement.

Table 1. Resonant frequency and quality factor of cavity with only the capillary tube.

Average Centre or Resonance Frequency (f_r) (GHz)	Average Lower 3 dB Frequency (f_1) (GHz)	Average Upper 3 dB Frequency (f_2) (GHz)	Average Bandwidth (f_2-f_1) (GHz)	Average Quality Factor $Q_{L1} = f_r / (f_2 - f_1)$
2.62138	2.62100	2.62174	0.00074	3542.40

Table 2. Dielectric constant of ((1-x) Zn.xMg.TiO_3) for different values of x.

X value	Average Centre or Resonant Frequency (f_r) (GHz)	Average Lower 3dB Frequency (f_1) (GHz)	Average Upper 3 dB Frequency (f_2) (GHz)	Average Quality Factor $Q_{L2} = f_r / (f_2 - f_1)$	ε_r'	ε_r''	$\tan\delta = \varepsilon_r'' / \varepsilon_r'$
0.1	2.61462	2.61422	2.61500	3352.07	6.4648	0.0169	0.0026
0.2	2.61494	2.61454	2.61534	3268.67	6.2055	0.0249	0.0040
0.3	2.61548	2.61546	2.61620	3534.91	5.768	0.00063	0.0001
0.4	2.61504	2.61468	2.61542	3533.83	6.1244	0.00072	0.00011
0.5	2.61456	2.61418	2.61494	3404.21	6.5134	0.0121	0.00185

Table 3. Dielectric constant of ((1-x) Zn.xMg.TiO_3) where x = 0.1, for different values of calcination temperatures.

Sample	Calcination Temperature (^{0}C)	Centre or Resonant Frequency (f_r) (GHz)	Lower 3dB Frequency (f_1) (GHz)	Upper 3 dB Frequency (f_2) (GHz)	Quality Factor $Q_{L2} = f_r / (f_2 - f_1)$	ε_r'	ε_r''	$\tan\delta = \varepsilon_r'' / \varepsilon_r'$
ZMT	900	2.61728	2.61688	2.61770	3191.80	2.94	0.0174	0.005914
ZMT	950	2.61736	2.61696	2.61778	3191.90	3.03	0.0173	0.00507
ZMT	1000	2.61752	2.61712	2.61790	3355.79	3.67	0.0111	0.003026
ZMT +V_2O_5	1000	2.61472	2.61434	2.61514	3268.40	4.04	0.01188	0.002698

Table 4. Resonance frequency and quality factor of cavity without standard sample.

Average Centre or Resonance Frequency (f_r) (GHz)	Average Lower 3 dB Frequency (f_1) (GHz)	Average Upper 3 dB Frequency (f_2) (GHz)	Average Bandwidth (f_2-f_1) (GHz)	Average Quality Factor $Q_{L1} = f_r / (f_2 - f_1)$
2.62794	2.62754	2.62834	0.00080	3284.925

Table 5. Dielectric constant of standard material (Teflon-AF).

Standard Material	Centre or Resonant Frequency (f_r) (GHz)	Lower 3 dB Frequency (f_1) (GHz)	Upper 3 dB Frequency (f_2) (GHz)	Quality Factor $Q_{L2} = f_r / (f_2 - f_1)$	ε_r'	ε_r''	$\tan\delta = \varepsilon_r'' / \varepsilon_r'$	Average Value ε_r' and $\tan\delta$
Teflon-AF (diameter = 2.5 mm and length = 37.25 mm)	2.62386	2.62340	2.62438	2677.408	1.794	0.0176	0.0098	1.795 and 0.0094
	2.62388	2.62340	2.62436	2733.208	1.790	0.0156	0.0087	
	2.62382	2.62344	2.62432	2677.367	1.801	0.0176	0.0097	

powder form have been investigated. The cavity perturbation technique has been used for the evaluation of dielectric properties of the material in S-band of microwave frequencies. In this technique, a cavity has been designed with very small slot at the centre of the broad wall of rectangular waveguide in order to insert the sample material. A cylindrical capillary is designed for holding of the sample. According to Wang [5] dielectric

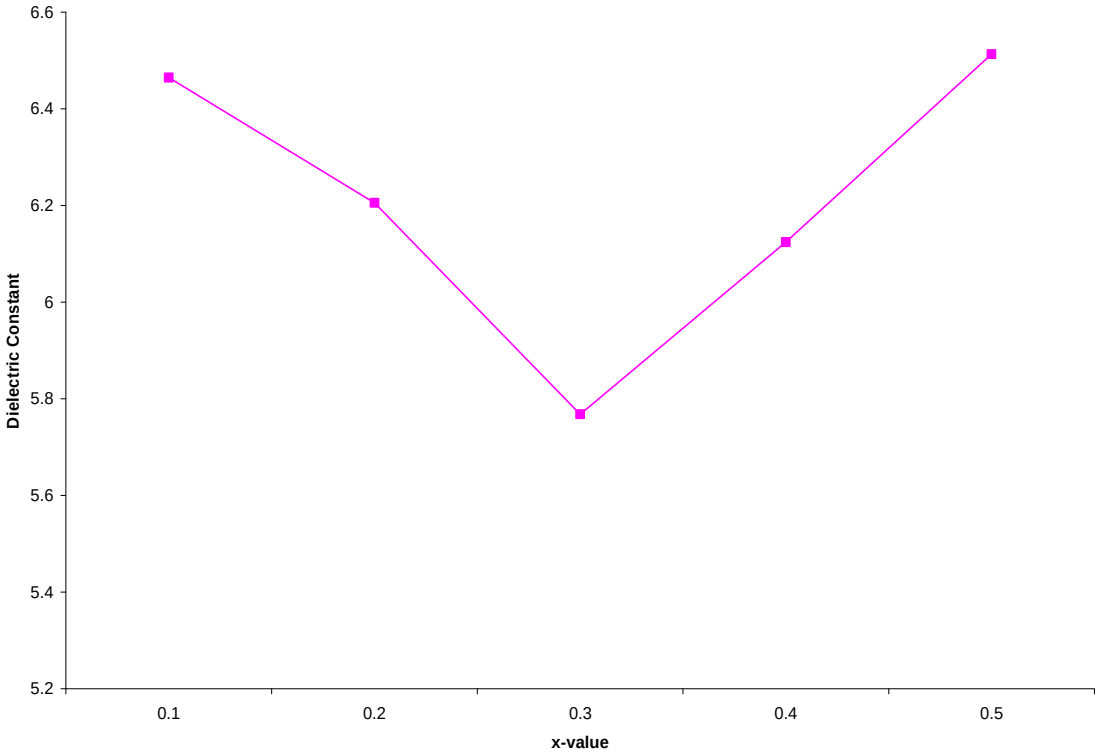


Figure 3. The experimentally observed variation of dielectric constant with x-value.

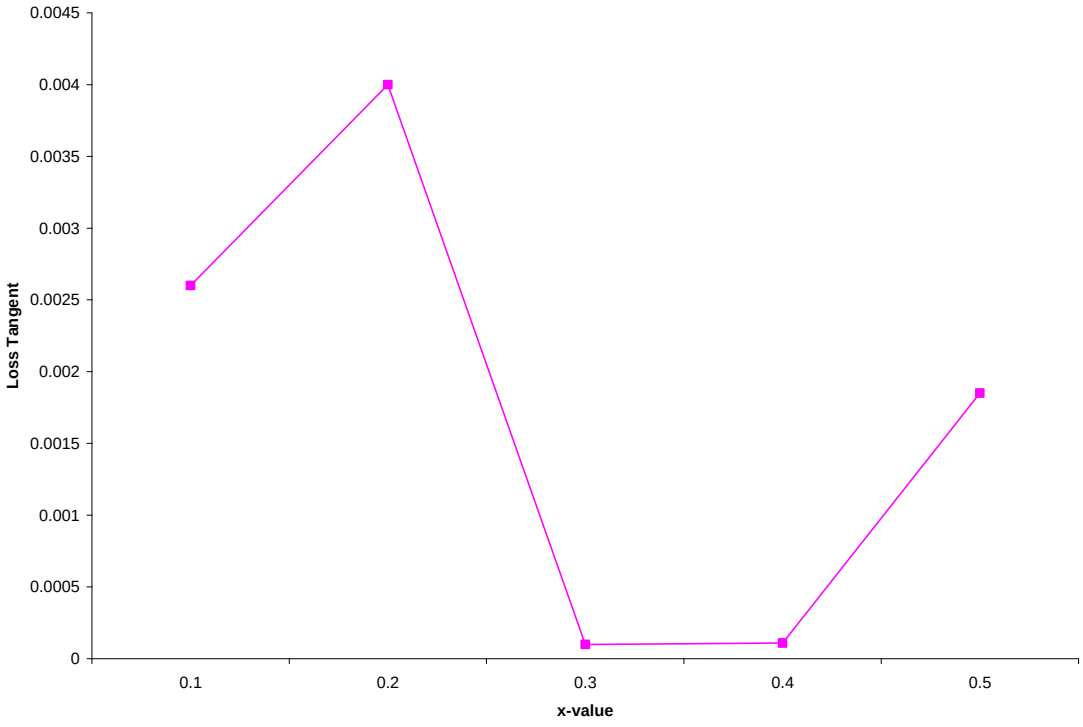


Figure 4. The experimentally observed variation of loss tangent with x-value

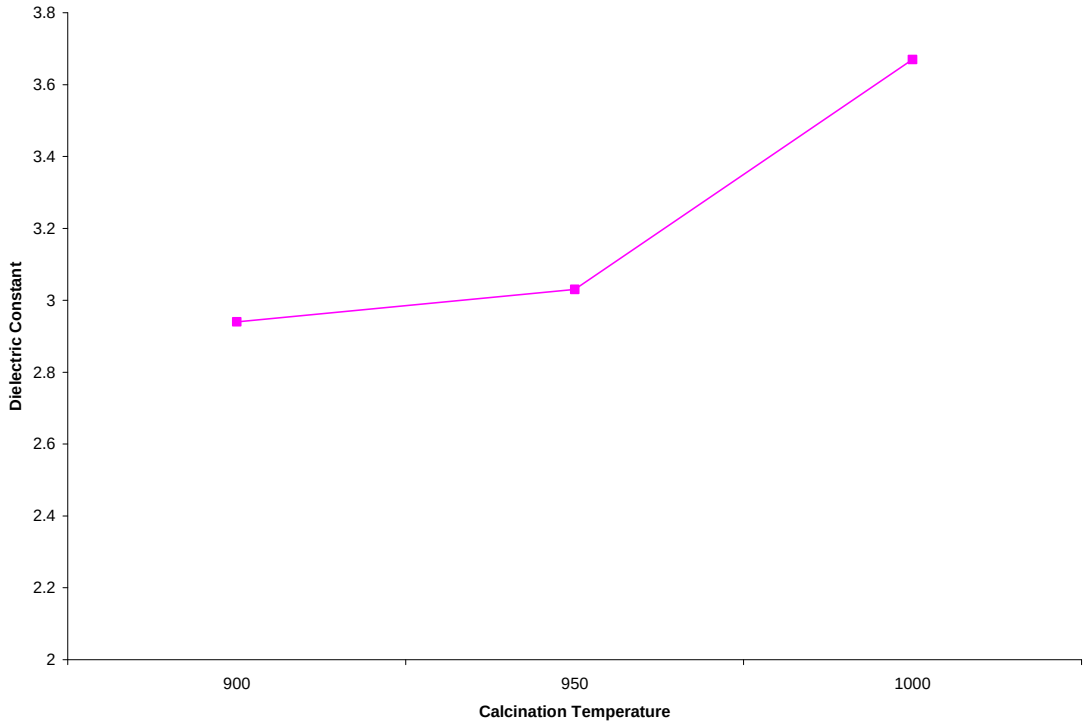


Figure 5. The experimentally observed variation of dielectric constant with calcination temperature for composition x = 0.1.

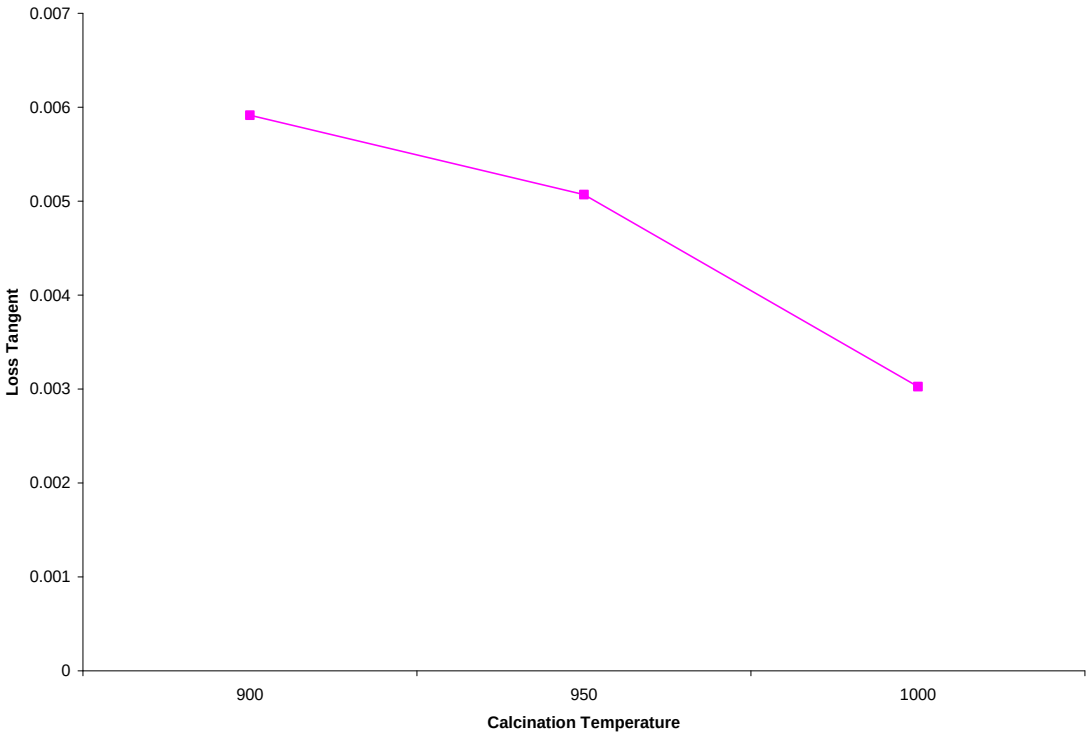


Figure 6. The experimentally observed variation of loss tangent with calcination temperature for composition x = 0.1.

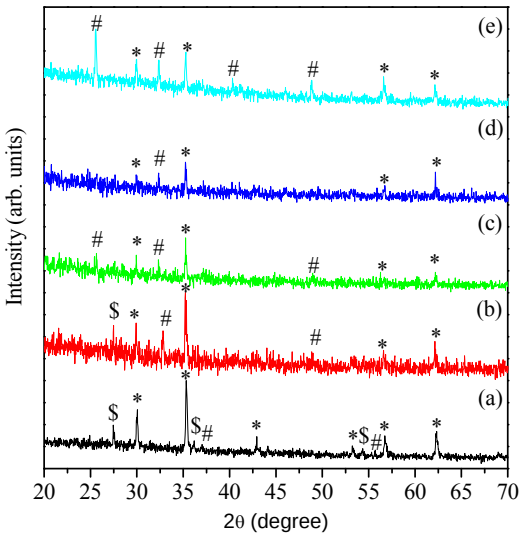


Figure 7. X-ray diffraction patterns of (1-x)Zn.xMg.TiO_3 with (a) x = 0.1, (b) x = 0.2, (c) x = 0.3, (d) x = 0.4 and (e) x = 0.5 (where * (Mg, Zn)TiO_3, # (Mg, Zn)Ti_2O_5 and $ TiO_2).

constant decreases with increase in x-value. In the present study the dielectric constant value decreased for x–values from x = 0.1 to 0.3 and then increased for x > 0.3. This is because of the presence of different phases in the composite material and the variation in the amount of phases with change in x-values as shown in the XRD patterns of the material. The dielectric constant has increased and loss tangent reduced with increase in calcination temperature. This may be attributed to the presence of single phase in the material. The ε_r' value increased and tan δ reduced with the addition of V_2O_5 in ZMT material. This is because of improvement in the rate of densification of ZMT material. The measured dielectric constant values are lower than those reported in reference [5]. This is due to lower value of percentage density of sample material. The reported percentage density of sample materials studied in reference [5] is more than 95%. Sample material used for measurement in reference [5] is in highly dense pellet form while in the present study we measured the permittivity of sample material in powder form.

It is concluded that the results represented here may provide important inputs for the design of devices at microwave frequencies.

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