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# Synthesis and Dielectric Properties of $MgTiO_3$ Ceramic Material

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***Abstract*** **- Magnesium Titanate ceramic materials are synthesised by conventional Solid-state diffusion method. Synthesized powder is characterized by X-ray diffraction for phase purity. Then the powder was pressed into pellets. These pellets were sintered at $1300^0$C respectively for 4h. The microstructure was studied by SEM. The dielectric constant from room temperature to $350^0$C was calculated using HIOKI 3532-50 LCR HiTESTER in the frequency range of 1KHz-5MHz. The dielectric constant at room temperature at a frequency of 1KHz for pure Magnesium Titanate was found to be 15. The AC conductivity, Dielectric loss of the sample was measured and thermoelectric properties were studied. XRD studies revealed that the Magnesium Titanate compound exhibits single phase with Hexagonal structure.**

***Keywords: Solid-state diffusion method, Dielectric Constant, X-ray diffraction, AC conductivity***

## I. INTRODUCTION

Magnesium Titanate ($MgTiO_3$) ceramics is a popular dielectric material which is used at microwave frequencies. $MgTiO_3$ has ilmenite type structure belonging to the Hexagonal space group R3 [1]. Magnesium Titanate ceramics play an important role in microwave technologies such as global positioning system operating at microwave frequencies, resonators, filters, antennas for communication system and multilayer capacitors [2-5]. It is a versatile material of low dielectric loss with high quality factor (Q above 20000 at 8GHz) and intermediate dielectric constant ($\varepsilon_r$=17) [6]. Various techniques such as thermal decomposition of peroxide precursors, hydrothermal mechano-chemical complexation routes, Sol-gel method, MOSD and auto- igniting combustion technique [7-11], Chemical coprecipitation [12], metalorganic chemical vapour deposition [13] are used for the preparation of $MgTiO_3$.

## II. EXPERIMENTAL PROCEDURE

Magnesium Titanate was synthesized by conventional Solid State diffusion route method using Magnesium Oxide, Titanium Dioxide ($TiO_2$). High Purity Oxide powders were taken according to their compositions and were ball milled (PM 200) for 12h. Then the powder was taken in alumina crucibles and placed in a programmable furnace. Magnesium Titanate was calcined at a temperature of $1200^0$C for 36h with intermediate grindings. The calcined powder was again milled for 10h. The powders were pressed into pellets using PVA binder. The Magnesium Titanate pellets were sintered at $1300^0$C for 4h with a heating rate of $10^0$C/min and then cooled to room temperature. The sintered samples were used for structural analysis and dielectric characterization. The crystalline phases and lattice parameters of the sintered samples were identified by X-Ray diffractometer (Rigaku) using CuK α radiation. Surface Morphology and microstructural studies were done on the sintered samples by SEM and Energy Dispersive X-Ray Spectroscopy (EDS). The SEM images were analysed using ImageJ software. The dielectric constant, dielectric loss and AC conductivity were carried using by LCR meter HIOKI 3252-50 in the frequency range 1KHz to 5MHz. The dielectric constant is calculated by using the equation

$$\varepsilon_r = \frac{C * d}{\varepsilon_0 * A} \qquad (1)$$

where C is capacitance of the pellet, d is thickness of the pellet; A is the area of the cross section of the pellet and $\varepsilon_0$ is the permittivity of free space. The AC conductivity of the samples was estimated from dielectric parameters. The AC conductivity ($\sigma_{AC}$) was measured using the relation

International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
Vol. 5 Issue 05, May-2016

$$\sigma_{AC} = \omega\varepsilon\varepsilon_0\tan\delta \qquad (2)$$

where $\varepsilon_0$ is the permittivity of the free space, $\omega$ is the angular frequency and tanδ is the loss tangent.

## III. RESULTS AND DISCUSSIONS

### *3.1 XRD analysis*

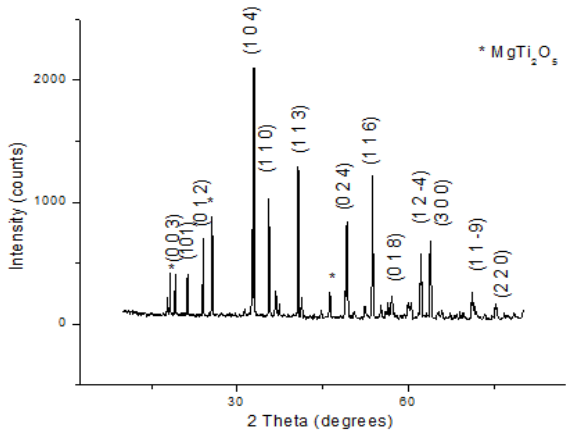


Fig.1 XRD pattern of $MgTiO_3$

Fig.1 represent the XRD analysis of Magnesium Titanate ceramic material respectively. The XRD pattern is indexed accordingly. The crystalline structure of $MgTiO_3$ ceramic sample was evaluated by XRD analysis at room temperature using Rigaku X-Ray Powder Diffract Meter (Japan) is shown in fig.1. and Table 1. Hexagonal crystalline structure having single reflection peaks was observed which confirms the formation of $MgTiO_3$ with the exception of few additional phases corresponding to the presence of $MgTiO_3$ (MT) and $MgTi_2O_5$ exhibited by the sample was observed. The $MgTi_2O_5$ requires a high sintering temperature of $1450^0$C. So second phase $MgTi_2O_5$ was formed as intermediate phase and difficult to eliminate from sample prepared by mixed oxide route [14]. The intensity of the diffraction pattern depends on structure factor shows the XRD data of present sample and the average crystallite size ($D_p$) of 70.44nm was obtained using Scherer formula

$$D_p = k\lambda/\beta\text{Cos}\theta \qquad (3)$$

where k is a constant and is equal to 0.9, θ is diffraction angle, λ=0.154056 nm (CuKα) and β is full width half maxima. The dislocation density is calculated by the formula $\rho=D^{-2}$ where its value is calculated to be $2.015\text{x}10^{14}$ $m^{-2}$. The lattice constants are found to be a=5.053628 $A^0$, b=5.053628 $A^0$ and c=13.903004 $A^0$ and Unit cell volume is found to be 307.500529$(A^0)^3$ are calculated for $MgTiO_3$ are in good agreement with the literature.

Table 1. XRD profile data of $MgTiO_3$ sample.

| 2θ | d-space $A^0$ | FWHM | (h k l) | $D_p$ (nm) |
|---|---|---|---|---|
| 19.196046 | 4.619892 | 0.069783 | (0 0 3) | 120.5 |
| 21.312117 | 4.165725 | 0.075317 | (1 0 1) | 112 |
| 24.053901 | 3.696741 | 0.095657 | (0 1 2) | 88.6 |
| 32.893371 | 2.720713 | 0.090102 | (1 0 4) | 96 |
| 35.530027 | 2.52462 | 0.100249 | (1 1 0) | 86.9 |
| 40.666769 | 2.216795 | 0.124214 | (1 1 3) | 71.2 |
| 49.196152 | 1.850561 | 0.135907 | (0 2 4) | 67.1 |
| 53.655546 | 1.706801 | 0.13204 | (1 1 6) | 70.4 |
| 56.98786 | 1.614655 | 0.134999 | (0 1 8) | 69.9 |
| 59.862653 | 1.543799 | 0.372503 | (0 0 9) | 25.7 |
| 62.115251 | 1.493112 | 0.113963 | (1 2 -4) | 84.9 |
| 63.759867 | 1.458505 | 0.137385 | (3 0 0) | 71.1 |
| 65.347153 | 1.426866 | 0.791972 | (2 1 -5) | 12.4 |
| 71.114376 | 1.324637 | 0.149167 | (1 0 10) | 68.3 |
| 71.536254 | 1.317858 | 0.159517 | (1 1 -9) | 64.1 |
| 75.14304 | 1.263296 | 0.228144 | (2 2 0) | 45.8 |

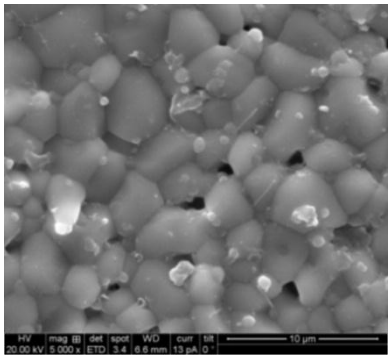

Fig.2. SEM image of $MgTiO_3$ of magnification 10μm

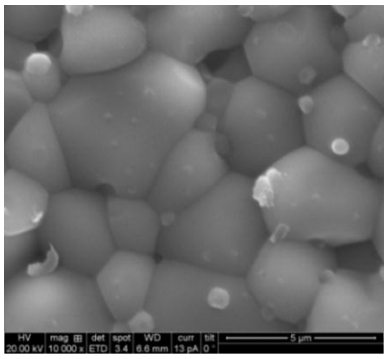

Fig. 3.SEM images $MgTiO_3$ ceramic sample of magnification 5μm.

$$\text{Average grain size} = \frac{1.5L}{MN} \qquad (4)$$

Where L=the total test line length, M=the magnification, N=the total number of intercepts which the grain boundary makes with the line.

International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
Vol. 5 Issue 05, May-2016

Table 2. EDS profile data of $MgTiO_3$ sample.

| *Element* | *Wt%* | *At%* |
|---|---|---|
| OK | 31.61 | 46.55 |
| MgK | 41.51 | 40.23 |
| TiK | 26.88 | 13.22 |
| Total | 100 | 100 |

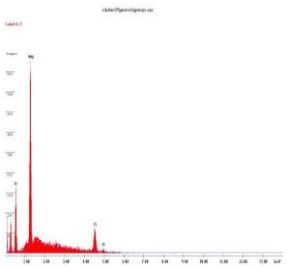

Fig. 4. EDS spectra of the $MgTiO_3$ sample.

*3.3. Dielectric properties*

The dielectric properties were performed on disks for pure $MgTiO_3$ in the temperature range from room temperature to $350^0$C. The dielectric constant found was 15 for $MgTiO_3$ at 1kHz at room temperature which is in good agreement with the literature. The dielectric constant at different frequencies for the sample $MgTiO_3$ was plotted in the fig. 5. Due to the accumulation of charge at the grain boundary and at the interface of electrode sample and electrode which is called space –charge polarization, the dielectric constant is high at low frequency. Fig.6 represents the variation of tangent loss with temperature for $MgTiO_3$ ceramic sample at different frequencies ranging from 1kHz, 10kHz, 1MHz, and 5MHz. At low frequency there is increase in dielectric loss with temperature nearly at $200^0$C and then decreases. Higher the frequencies, the dielectric loss is very less and almost stable which may be an extrinsic loss due to oxygen vacancies, grain sizes, secondary phases, and densification. The variation ac conductivity as a function of frequency at different temperatures is plotted in the fig. 7. As the temperature goes on increasing, the dielectric relaxation gets thermally activated and hence this can be governed by the Arrhenius equation.

$$\sigma = \sigma_0 \, e^{-\frac{E_a}{K_B T}} \qquad (5)$$

where K=8.6X10$^{-5}$eV, $\sigma_o$= pre exponential factor, and T= absolute temperature.

Fig. 8 depicts the variation of ac conductivity with reciprocal of temperature and reveals almost a linear relation between ln $\sigma_{ac}$ and 1000/T. The activation energies are determined by the slopes of the curves and were achieved as 0.3051eV, 0.2499 eV, 0.1988 eV at 1 kHz, 10 kHz, 100 kHz and 1MHz frequencies respectively. At 5 MHz $E_a$ was obtained as 0.1988eV. This clearly shows a fact that conductivity increases with increasing frequency and is almost independent of temperature at high frequencies. At lower frequencies the conductivity increases with increase of temperature due to thermal activation process and this may be due to hopping of charge carriers.

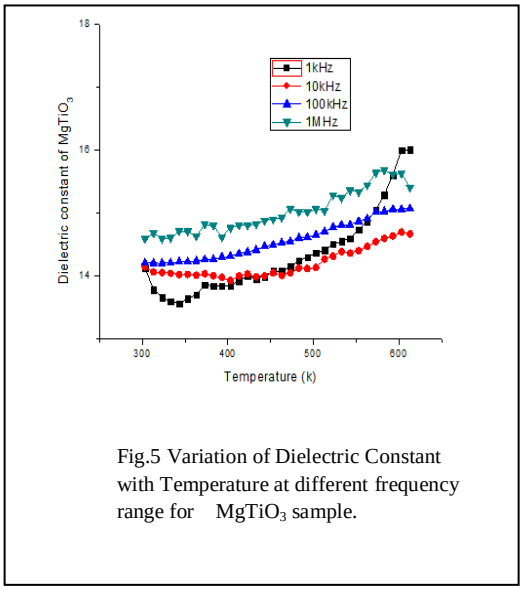


Fig.5 Variation of Dielectric Constant with Temperature at different frequency range for $MgTiO_3$ sample.

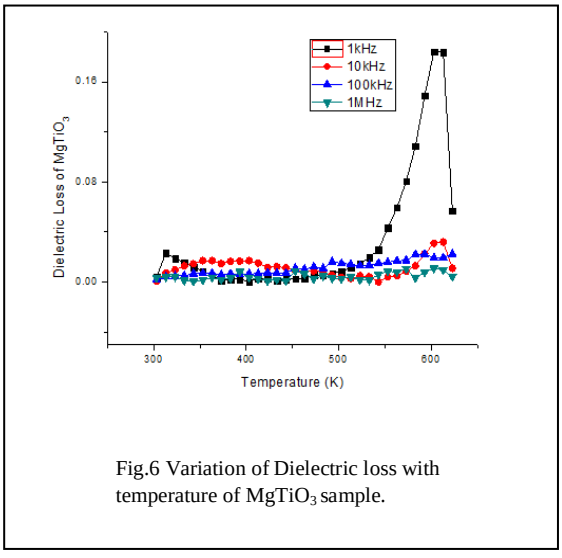


Fig.6 Variation of Dielectric loss with temperature of $MgTiO_3$ sample.

International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
Vol. 5 Issue 05, May-2016

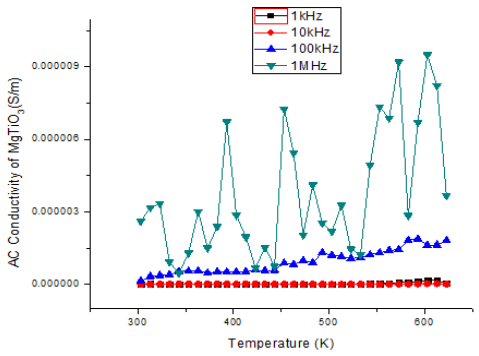


Fig.7. Variation of AC conductivity with temperature of $MgTiO_3$ sample.

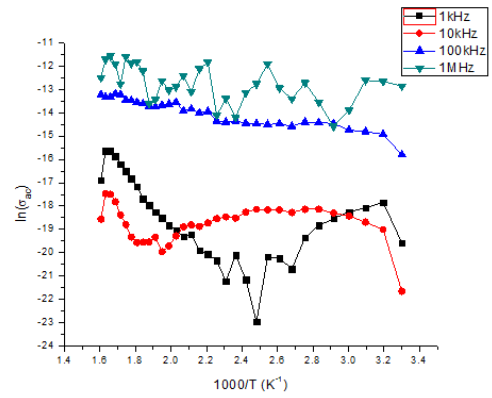


Fig.8 Variation of $\ln(\sigma_{ac})$with 1000/T of $MgTiO_3$ sample.

*3.4 Electrical Studies*

Fig. 9 shows the variation of Thermoemf (E) with temperature. Fig. 10 gives the variation of Seebeck coefficient with Temperature.The Seebeck coefficient increases with increase of temperature and is positive which reveals that holes are introduced. At high temperatures the Seebeck coefficient remains constant and later slightly increased with an overall slope of 214.75μV/K.. The carrier concentration (n) was calculated for the $MgTiO_3$ pellet of thickness 0.281cm and radius 0.598cm as $0.2639X10^{22}$ cm-3 using the following equation.

$$n = \frac{N}{V}\exp(\frac{-Se}{K}) \qquad (6)$$

Where $N=10^{22}$ $cm^{-3}$ (Density of states),
$V=0.315cm^3$(Vvolume of the sample),

$$\frac{k}{e} = 86.4\ \mu V/K$$

S=Seebeck Coefficient

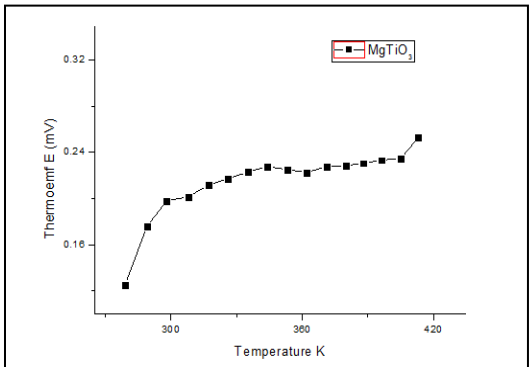


Fig.9. Variation of Thermoemf with temperature of $MgTiO_3$ sample.

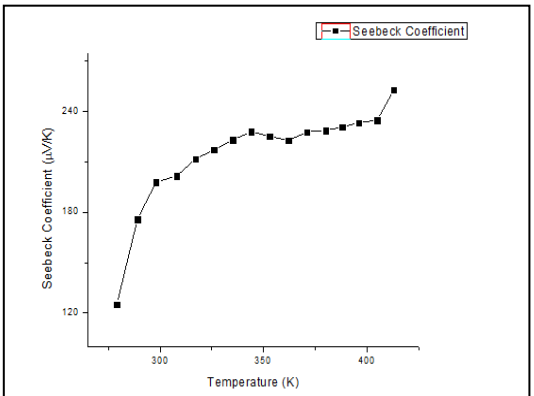


Fig.10 Variation of Seebeck coefficient with temperature of $MgTiO_3$ sample.

Fig. 11. shows the variation of impedence(Z) with temperature. At higher temperature, lower is the impedance for low frequencies, which indicate the larger polarization. On further increase in frequency Impedance shows independent behaviour. The moderate Z-values were achieved at higher frequencies.

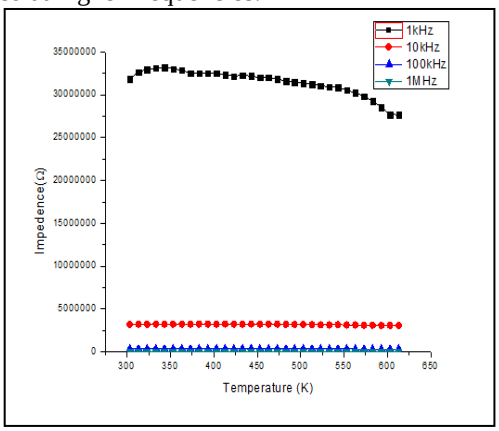


Fig.11 Variation of Impedance with temperature of $MgTiO_3$ sample.

International Journal of Engineering Research & Technology (IJERT)
ISSN: 2278-0181
Vol. 5 Issue 05, May-2016

## IV. CONCLUSION

Magnesium Titanate was prepared by solid state diffusion method. The crystalline size and phase of hexagonal structure was determined by X-ray diffraction. The particle size obtained for sample by ImageJ software is about 175nm. The average gain size is found to be 5µm. The Magnesium Titanate shows good dielectric properties of dielectric constant $\varepsilon_r \approx 15$ where sample is sintered at 1300°C. The dielectric loss is very low at room temperature. The activation energy at 1MHz frequency was found to be 0.1278eV. Thus magnesium titanate is good for microwave dielectric properties.

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