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Effects of CeO_2 nanoparticles and annealing temperature on the microwave dielectric properties of $MgTiO_3$ ceramics

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Abstract

$MgTiO_3$ (MTO) ceramics have been prepared by the conventional solid-state reaction method. The effects of CeO_2 nanoparticles up to 1.5 wt% as a sintering aid and annealing temperature on the crystal structure, microstructure and microwave dielectric properties of MTO ceramics were investigated. It is found that the addition of CeO_2 nanoparticles to the MTO ceramics leads to improvement in the relative density, reduction in sintering temperature. However, $Q \times f_o$ values are found to decrease marginally. The driving forces for the improved sintering process are the large surface energy of CeO_2 nanoparticles and their defect energy, which are activated during the sintering process. The addition of CeO_2 nanoparticles does not change the dielectric constant (ε_r), and the unloaded quality factor (Q_u) but the τ_f values are altered significantly. The annealing temperatures of the pure MTO ceramics and MTO+$x$$CeO_2$ (x=0.5, 1.0 and 1.5 wt%) were optimized. The samples annealed at 1100 °C for 48 h exhibit the best microwave dielectric properties. The maximum value of $Q \times f_o$ for the pure MTO was achieved by sintering at1400 °C and significantly such maximum value was achieved in x=0.5 wt% sample by sintering at 1300 °C itself. The values of $Q \times f_o$ are in the range of 52250–140800 GHz. Pure MTO ceramics sintered at 1400 °C and annealed at 1100 °C for 48 h exhibit the best microwave dielectric properties (ε_r=17.21, $Q \times f_o$=165500 GHz). The observed results are correlated to the reduction in oxygen vacancies, relaxation of strain during annealing, increase in grain size and relative densities.

Keywords: A. Powders: solid-state reaction; B. X-ray methods; C. Dielectric properties; E. Substrates; E. Capacitors

1. Introduction

Recently, dielectric resonators are being widely investigated due to the fast growth of microwave telecommunication and satellite broadcasting. Dielectric resonators play a significant role in the miniaturization of microwave components, such as oscillators, amplifiers, tuners and filters [1,2]. The miniaturization of microwave circuits intensely requires materials with high dielectric constant, low dielectric loss and with good temperature stability [3]. $MgTiO_3$ (MTO) is an attractive resonator material for high frequency applications due to its promising microwave dielectric properties, with moderately high values of quality factors and due to the low cost of raw materials. MTO ceramics exhibit an ilmenite structure with rhombohedral crystal system and belong to $R\bar{3}$ space group [4]. $MgTiO_3$ ceramics exhibit a dielectric constant of (ε_r~17), high $Q \times f_o$ factor of about ~150,000 GHz at 8 GHz and a negative τ_f value of ~−50 ppm/°C [5]. However, the sintering temperature is as high as 1450 °C and it is difficult to densify the material at lower sintering temperatures without any sintering aids. To reduce the sintering temperature of the ceramics three methods are generally adopted: (a) addition of fluxes with low melting point, (b) chemical processing, and (c) starting with smaller initial particle sizes. Nevertheless, there are only a few reports on the synthesis of MTO ceramics by reducing the sintering temperature by adding the impurities with low melting point [6–8] and by chemical processing methods [9–10].

Recent past, oxide nanoparticles have attracted much attention, as they are efficiently acting as a link between bulk materials and atomic structures. A sole aspect of nanoscale materials is the tremendously increased ratio of surface area to volume, which opens new possibilities on the surface dependent phenomena. At these reduced dimensions, oxide nanoparticles provide a tremendous driving force during the sintering, particularly at

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elevated temperatures and hence the surface energy of the nanoparticles substantially affects the interior bulk properties of the host materials. With these smaller particles, the sintering can take place at decreased temperatures over shorter time scales than for the larger particles. Since the microwave dielectric properties of the ceramics are very much sensitive to their density and the microstructures, the understanding on the control of the above properties of the ceramics with respect to the ceramic processing parameters, such as initial powder natures, types of additives, and the sintering temperature is necessary. However, there is no systematic study with the addition of CeO_2 nanoparticles to MTO ceramics. Hence, in this study, we report the effect of CeO_2 nanoparticles, as additives, on the sintering ability, densification, crystal structure, microstructure, and the microwave dielectric properties of MTO ceramics.

Minimization of the dielectric loss is one of the important tasks for improving the performance of materials and hence the main interest of many researchers is in understanding the fundamental mechanism of the dielectric loss at microwave frequencies. It is generally known that both intrinsic and extrinsic factors contribute to dielectric loss. The intrinsic loss is mostly due to anharmonic forces that mediate the interaction between crystal lattice and phonons, which leads to damping of optical phonons. The extrinsic loss comes in to picture due to the presence of defects, porosity, impurities, secondary phases and lattice strain. It is well known that the minimization of extrinsic loss is much easier as compared to inherent losses. The extrinsic loss can be minimized by achieving maximum density, larger grain size, ordered structure, slow cooling rate and by reducing the oxygen vacancies [11–15]. A careful literature [1,5,16–18] survey on the titanate based ceramics reveal that the extrinsic losses of titanate based ceramics can be reduced by systematic annealing of the samples. However, the effect of annealing on the improvement of microwave dielectric properties of MTO has not been reported in detail. Hence, in the present study we have carried out the study of the effects of (i) addition of CeO_2 nanoparticles as a sintering aid (smaller concentrations) and (ii) the annealing conditions on the material properties such as crystal structure, densification, microstructure and microwave dielectric properties of MTO ceramics.

2. Experimental techniques

$MgTiO_3$ samples were prepared by the conventional solid-state reaction method using the high-purity oxide powders (> 99.99%): MgO and TiO_2 (Sigma-Aldrich, USA). The starting materials were mixed according to the stoichiometry of $MgTiO_3$ and the mixture was grinded under distilled water for 5 h by using a planetary ball mill (Fritsch GmbH, Germany). The prepared mixtures were dried and calcined at 1100 °C for 2 h in air. The calcined powders were re-milled with an inclusion of x=0.5, 1.0 and 1.5 wt% of CeO_2 nanoparticles. The CeO_2 nanoparticles with an average particle size of 80 nm were procured from M/S Alfa Aesar, USA. Further, the powders were added with an organic binder (PVA) and pressed into pellets with 10 mm in diameter and 4–5 mm in thickness under a pressure of 2000 Kg/cm^2 isostatically. The pellets were subsequently sintered at 1250–1400 °C for 3 h in air. To optimize the annealing temperature, MTO ceramics were annealed separately at 1000 °C, 1100 °C and 1200 °C for 48 h followed by a cooling rate of 1 °C per minute.

The phase purities of sintered and annealed MTO ceramics were examined by recording the XRD patterns using Rigaku high power X-ray Diffractometer (RINT 2500 system TTRAX) with CuK_α radiation (λ=1.5406 Å). The surface morphology of sintered and annealed samples was observed by scanning electron microscope (Leo 1430 vp) and Energy dispersive spectroscopy (EDS). The relative densities of the sintered specimens were measured by the Archimedes's method. The dielectric constant (ε_r) and Quality factors (Q_0) of the samples at microwave frequencies were measured using the Hakki and Coleman dielectric resonator method, as modified and improved by Courteny [19,20]. The temperature coefficient of resonance frequency (τ_f) was measured in the temperature range of 25–80 °C using an invar cavity.

3. Results and discussions

To study the effect of addition of CeO_2 nanoparticles on crystal structure of MTO ceramics, XRD patterns were recorded. The XRD patterns of $MgTiO_3$ ceramics with x=0.5–1.5 wt%, sintered at 1300 °C for 3 h are shown in Fig. 1a and for x=0–1.5 wt% annealed at 1100 °C for 48 h are shown in Fig. 1b. It is observed that the samples with x=0 wt% reveal $MgTiO_3$ as a main crystalline phase along with a minor secondary phase of $MgTi_2O_5$. On the other hand the secondary phases for the MTO ceramics with x=0.5–1.5 wt% are found to be Ce_2TiO_5 and $MgTi_2O_5$. The crystal structure of $MgTiO_3$ is found to be trigonal (ICDD # 06–0464). The intensities of the peaks corresponding to Ce_2TiO_5 phase is found to enhance with increase in x wt%. $MgTi_2O_5$ usually forms as an intermediate phase and it is difficult to eliminate this phase in the samples prepared by the mixed oxide route. The $MgTi_2O_5$ phase exhibits a dielectric constant of 17, $Q \times f_o$ value of 41,000 GHz (at 9 GHz) and a τ_f value of −66 ppm/°C [21]. The lattice constants of pure and MTO+x wt % ceramics were calculated. No appreciable variations in lattice constants with increase in x wt% are observed and it suggests that CeO_2 nanoparticles are not forming a solid solution with MTO matrix and the obtained lattice constants of the as prepared and annealed samples are tabulated in Table 1. This is expected due to the mismatching of ionic radii of Ce^{4+} ions (1.02 Å) with Mg^{2+} (0.72 Å) and Ti^{4+} (0.605 Å) ions. Hence the Ce^{4+} ion cannot be substituted in the Mg^{2+} and Ti^{4+} sites. The annealing gives rise to reduction in the intensity of XRD peaks of $MgTi_2O_5$ impurity phase in x=0 sample and the intensities of both Ce_2TiO_5 and $MgTi_2O_5$ impurity phases in x=0.5 wt% samples. However, the intensities of Ce_2TiO_5 and $MgTi_2O_5$ phases are found to increase up on annealing for x=1.0 and 1.5 wt%.

The relative densities of MTO ceramics sintered at different sintering temperatures with x=0.0–1.5 wt% were measured using the Archimedes method and are given in Table 2. The improvement in the relative densities of MTO ceramics with an increase in x wt % is observed with the increase in the sintering temperature up to 1300 °C, and beyond that it is found to decrease. However, pure MTO ceramics exhibited the maximum density of 97.5% of theoretical density only at 1400 °C. Further, the values of relative

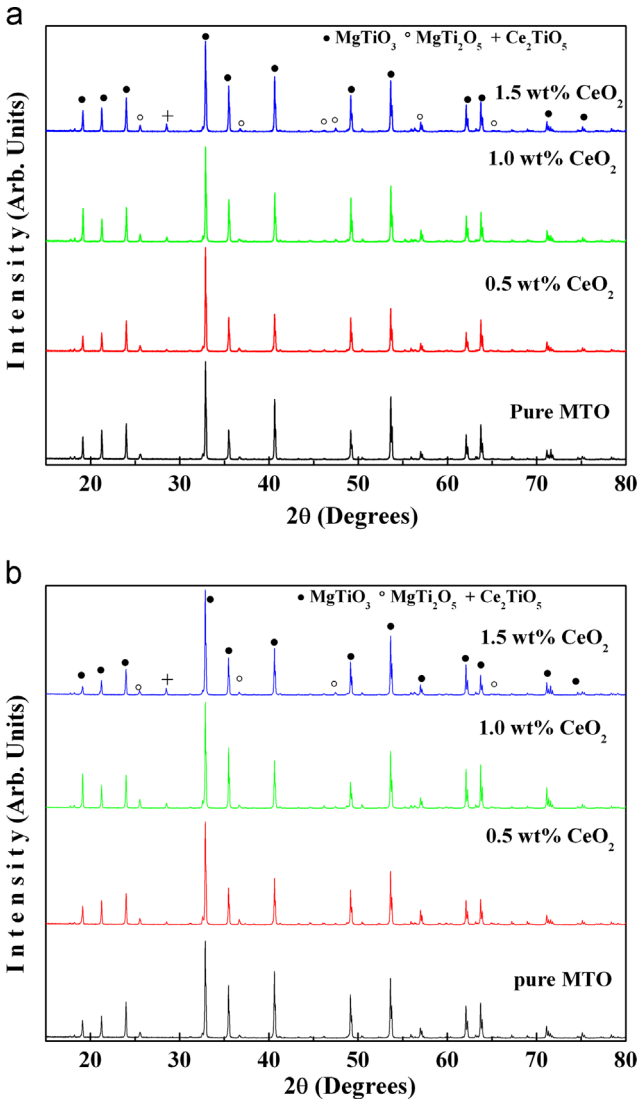

Fig. 1. XRD patterns of MTO ceramics with $x=0.0$–1.5 wt% (a) before annealing (b) after annealing.

Table 1
The calculated lattice constants of the as prepared and annealed samples.

Composition	Obtained lattice parameters (Å)
As prepared	
Pure MTO	$a=b=5.0547$, $c=13.9003$
MTO+0.5 wt% of CeO_2	$a=b=5.0554$, $c=13.9001$
MTO+1.0 wt% of CeO_2	$a=b=5.0562$, $c=13.9010$
MTO+1.5 wt% of CeO_2	$a=b=5.0559$, $c=13.9008$
After annealing	
Pure MTO	$a=b=5.0554$, $c=13.9000$
MTO+0.5 wt% of CeO_2	$a=b=5.0561$, $c=13.9012$
MTO+1.0 wt% of CeO_2	$a=b=5.0555$, $c=13.9014$
MTO+1.5 wt% of CeO_2	$a=b=5.0543$, $c=13.9009$

density of MTO ceramics are found to decrease with an increase in x wt% beyond $x=0.5$ wt%. The maximum relative density of 97.4% was obtained for the sample with $x=0.5$ wt %, and sintered at 1300 °C. The relative density values are found to be in the range of 97.5–94.2% for all the samples sintered in the temperature range

Table 2
Relative density, measured microwave dielectric constant (ε') and dielectric constant corrected for porosity (ε_m) values of pure and MTO ceramics added with CeO_2 nanoparticles.

Material composition	Sintering temperature (°C)	Percentage of relative density	ε' (measured)	ε_m (corrected)
$MgTiO_3$	1400	97.50	17.21	17.82
	1350	95.90	17.04	17.70
	1300	95.10	16.95	17.80
	1250	94.22	16.80	17.95
$MgTiO_3$+0.5 wt% of CeO_2 nanoparticles	1400	96.50	16.70	17.54
	1350	96.66	16.96	17.80
	1300	97.40	17.12	17.76
	1250	95.05	16.69	17.91
$MgTiO_3$+1.0 wt% of CeO_2 nanoparticles	1400	95.56	16.60	17.67
	1350	96.05	16.67	17.63
	1300	96.47	16.77	17.63
	1250	95.58	16.52	17.59
$MgTiO_3$+1.5 wt% of CeO_2 nanoparticles	1400	95.07	16.52	17.72
	1350	95.59	16.58	17.65
	1300	95.97	16.59	17.56
	1250	95.25	16.35	17.49

of 1250–1400 °C. The optimum sintering temperature for the maximum relative densities for MTO+x wt% ceramics is 1300 °C and the reduced density for the samples sintered above 1300 °C could be due to the possible evaporation of CeO_2 nanoparticles, secondary phases and non uniform grain growth (SEM image Fig. 2(d)). The reduction in sintering temperature can be explained as follows: it is known that nanoparticles have more surface to the volume ratio than the bulk materials and the higher surface energy lowers the melting point [22]. Moreover, sintering velocity is inversely proportional to particle size, i.e., sintering is faster for the finer particles. In addition, the sintering velocity gets enhanced, when the diffusion across the grain boundary increases. Therefore, during the sintering process, surface defect atoms of powders tend to become normal atoms with lower energy within the crystalline grains. When the CeO_2 nanoparticles (80 nm) were added to MTO powders (1.2 μm), the high temperature provides external activation energy to release the internal energy contained in powders thus enabling the mass transfer and reduction in surface energy [23]. Similar studies of reduction of sintering temperature by the addition of nanoparticles were reported by Pamu et al. [24], and Shanker et al. [25]. In order to prove the effect of CeO_2 nanoparticles, we have also prepared by adding bulk CeO_2 powder (average particle size is ~1.5 μm) of different wt% and by sintering at 1300 °C and 1350 °C. It is found that the densities of these samples are found to be in the range of 82–88%. Thus CeO_2 nanoparticles play a crucial role in enhancing the density of MTO ceramics.

In order to study the effect of CeO_2 nanoparticles on the microstructure of MTO ceramics, the SEM images were recorded in pure and CeO_2 nanoparticles added. Those SEM images of pure

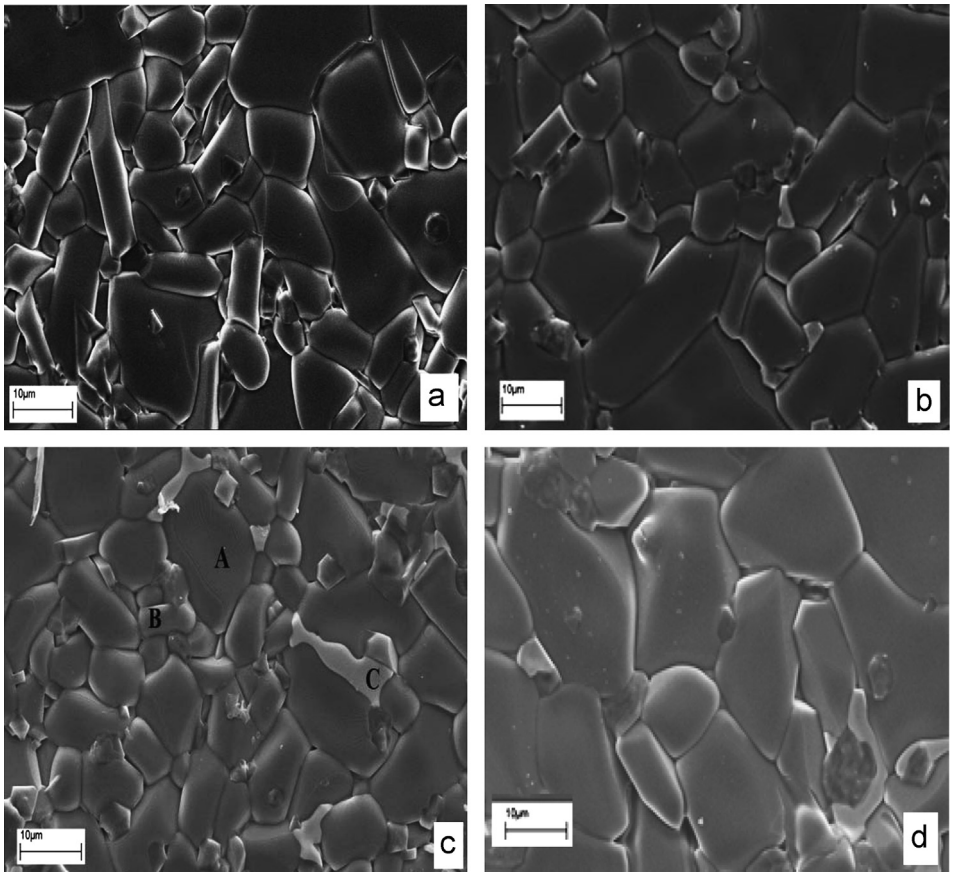

Fig. 2. SEM images of (a) pure MTO ceramics sintered at 1400 °C and MTO–CeO_2 (x wt%) ceramics with (b) $x=0.5$, (c) $x=1$ and (d) $x=1.5$, sintered at 1300 °C.

MTO ceramics sintered at 1400 °C is shown in Fig. 2a and MTO ceramics with $x=0.5$–1.5 wt%, sintered at 1300 °C for 3 h are shown in Fig. 2(b–d). The microstructures revealed that all the samples are crack free and highly densified and the samples with $x=0.5$ wt % exhibited a microstructure with mixed grain sizes. Further, as the x wt% increases, grain morphology of dense MTO ceramics exhibited two different types of grains: at the grain boundary of the large grains, a segregation of brown and white colored phases was observed. In order to understand the chemical compositions of these contrast phases, EDS analysis was performed. The chemical composition for MTO+$x=1.0$ wt% (Fig.2c) contrast phases obtained from EDS is shown in Table 3. The larger grains (spot A) exhibit the chemical composition of $MgTiO_3$, whereas the chemical compositions of spot B and spot C correspond to $MgTi_2O_5$ and Ce_2TiO_5, respectively. Further, as the amount of x wt% increases, magnitude of the secondary phase is intensified, and the grain growth is non uniform. The average grain sizes of the sintered specimens for $x=0.5$, 1.0, 1.5 wt% are 8.7, 7.1 and 9.7 μm, respectively.

To see the effect of annealing on the microstructure of MTO +$x=0.0$–1.5 wt%, SEM micrographs of the samples were obtained. The microstructures of MTO ceramics annealed at 1100 °C are shown in Fig. 3 (a–d) for different values of x. Where (a) corresponds to pure MTO ceramics, (b) with $x=0.5$, (c) with $x=1.0$ and (d) with $x=1.5$ wt%, respectively. Pure MTO ceramics did not show any noticeable change in the microstructure. However, in the case of MTO ceramics with $x=0.5$–1.5 wt%, segregation of secondary phases at the grain boundary was observed. The chemical compositions of contrast phases obtained from EDS for MTO+$x=1.0$ wt% (Fig.3c) annealed at 1100 °C for 48 h is shown in Table 3. During the annealing process, the secondary phases were segregated on to the surface of the samples through the grain boundary which is confirmed from SEM images (Figs. 3b–d). This is in good agreement with the earlier reports on similar systems [26]. Further, in the case of as prepared samples, all the secondary phases may be dispersed/suppressed in MTO matrix. After annealing, the secondary phases precipitated on to the surface of MTO ceramics.

To comprehend the influence of CeO_2 nanoparticles on MTO ceramics, microwave dielectric properties were measured. The measured dielectric constants and the porosity corrected dielectric constants are tabulated in Table 2. As the sintering temperature increases up to 1300 °C, the dielectric constant increases and beyond that it decreases. Further, pure MTO ceramics exhibit the maximum value of dielectric constant only after sintering at 1400 °C. On the other hand, MTO with $x=0.5$ wt% showed the highest value of dielectric constant even sintering at 1300 °C. The relationship between the ε_r and sintering temperature follows the same trend as that of relative density vs sintering temperature, and it shows the dependence of dielectric constants with relative densities. The measured values of dielectric constants are in the range of 16.35–17.21. To see the effect of porosity on the dielectric constant, the measured values of the

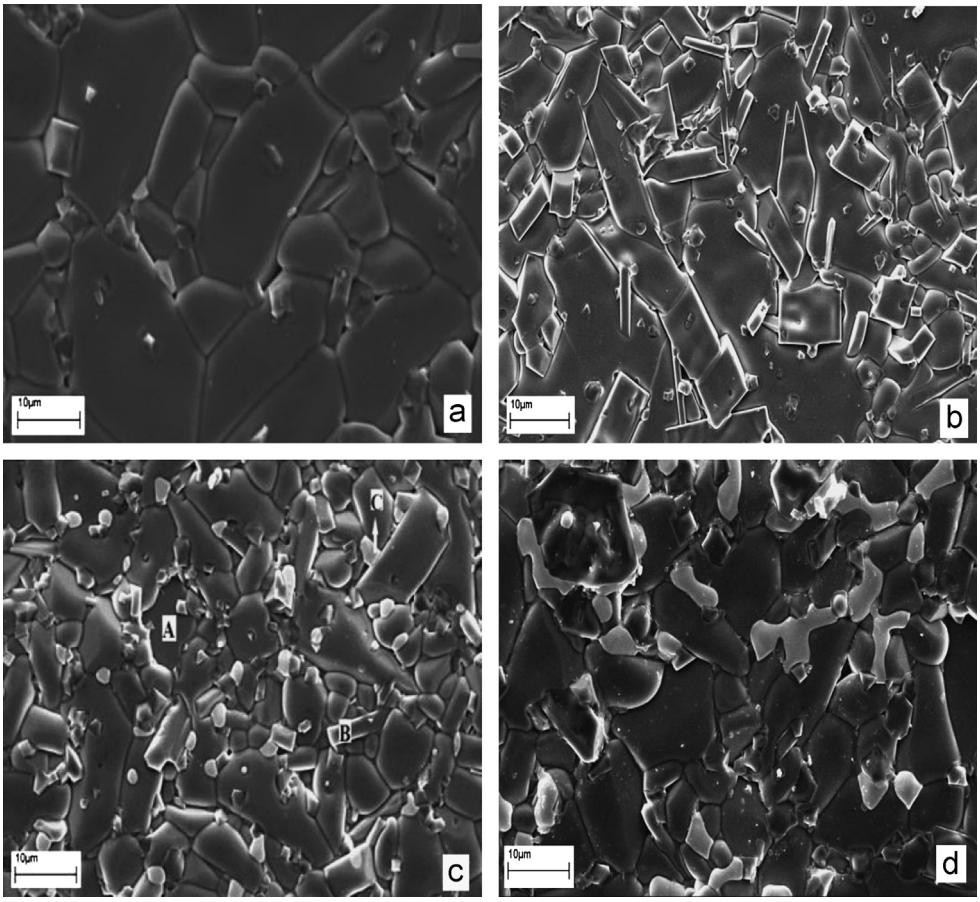

Fig. 3. SEM images of MTO ceramics with $x=0.0$–1.5 wt% annealed at 1100 °C for 48 h (a) pure MTO ceramics and MTO–CeO_2 (x wt%) ceramics with (b) $x=0.5$, (c) $x=1$ and (d) $x=1.5$.

Table 3
EDS spectra results of the as prepared MTO sample with $x=1.0$ wt%, sintered at 1300 °C.

Spot	Atom (%)			
	Mg K	Ti K	Ce L	O K
A	19.66	19.79	–	60.55
B	12.21	23.93	–	63.86
C	–	11.86	24.95	63.19

EDS spectra results of the MTO sample with $x=1.0$ wt%, sintered at 1300 °C and annealed at 1100 °C for 48 h.

Spot	Atom (%)			
	Mg K	Ti K	Ce L	O K
A	19.45	19.23	–	61.32
B	13.23	24.91	–	61.86
C	–	11.98	25.32	62.90

microwave dielectric constants corrected for porosity using the following equation:

$$\varepsilon_r = \varepsilon_m\left(1-\frac{3P(\varepsilon_m-1)}{2\varepsilon_m+1}\right) \quad (1)$$

Where ε_m is the dielectric constant corrected for porosity, ε_r is the experimental dielectric constant and P is the fractional porosity [27]. The obtained values are tabulated in Table 2.

The dependence of $Q\times f_o$ values of MTO+x wt% ceramics as a function of sintering temperatures are shown in Fig. 4a. The $Q\times f_o$ values of pure MTO ceramics are found to increase with increase in sintering temperature and the maximum value is achieved at 1400 °C. However, for MTO with $x=0.5$–1.5 wt% the best $Q\times f_o$ value is obtained for samples sintered at 1300 °C. The trend of relative density as a function of sintering temperature is found to be comparable to that of $Q\times f_o$. The maximum $Q\times f_o$ values obtained for the pure

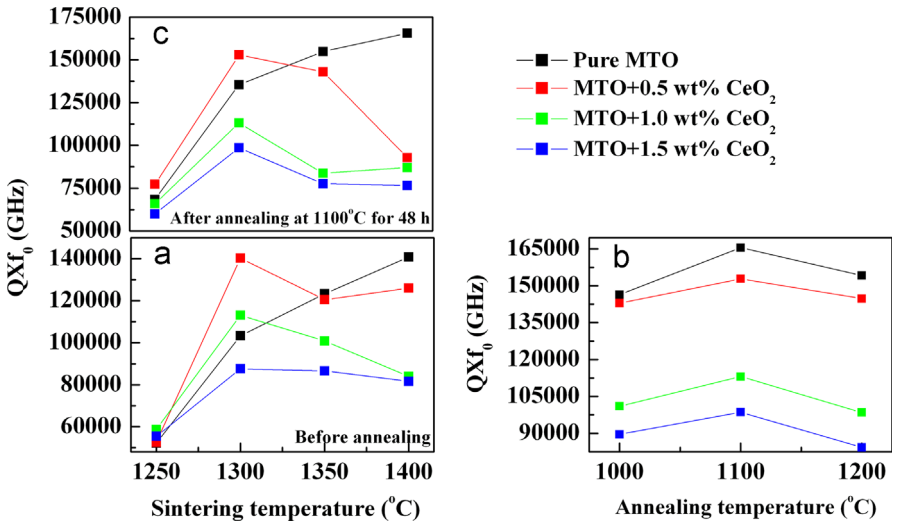

Fig. 4. Plots of variation in (a) $Q \times f_o$ values and (b) $Q \times f_o$ values of the samples used to optimize the annealing temperature. (c) $Q \times f_o$ values after annealing as a function of sintering temperature for MTO ceramics added with x=0.0–1.5 wt%.

MTO and MTO+x=0.5 wt % were found to be 1,40,800 and 1,40,350, respectively. Besides, it is also observed that for a particular sintering temperature the value of $Q \times f_o$ is found to decrease with increase in x wt% and it is due to the presence of secondary phase of Ce_2TiO_5. Further, it is also noticed that $Q \times f_o$ values did not show considerable dependence on average grain sizes.

To optimize the annealing conditions, four sets of samples with maximum relative densities and the best microwave dielectric properties (pure MTO sintered at 1400 °C and MTO+x=0.5–1.5 wt% sintered at 1300 °C) were chosen. Each set was annealed at 1000, 1100 and 1200 °C for 48 h separately. After annealing, the microwave dielectric properties were measured again. It was found that the best microwave dielectric properties were obtained for the samples annealed at 1100 °C. It was observed that the samples annealed at 1100 °C exhibited the maximum $Q \times f_o$ values compared with other conditions and the measured $Q \times f_o$ values are depicted in Fig. 4b. The variations of $Q \times f_o$ as a function of sintering temperature after annealing at 1100 °C for MTO+x wt% ceramics are shown in Fig. 4c. No significant change in relative density and dielectric constant values were observed up on annealing whereas $Q \times f_o$ values strongly affected. On annealing, pure MTO ceramics exhibited large $Q \times f_o$ values compared to MTO ceramics with x wt% and they are found to be in the range of 59,950–1,65,500 GHz. It is interesting to note that the as prepared pure MTO ceramics and MTO with x=0.5 wt% exhibit almost the same dielectric properties. Nevertheless, on annealing, pure MTO ceramics exhibited the best microwave dielectric properties as compared to MTO with x wt%.

In order to understand the effect of the strain on the $Q \times f_o$ values, the strain in the samples was estimated using the Williamson–Hall plot method. The strain values of the samples correspond to the as prepared and the annealed samples are found to be in the order of 10^{-3} and 10^{-4}, respectively. Further, it is well known that the slow rate of cooling process improves the $Q \times f_o$ values [28]. The strain values in the annealed samples are found to be lower compared to those of the as prepared samples and it could influence the dielectric loss.The improvement in $Q \times f_o$ values of pure MTO ceramics after annealing can be described as follows: when MTO ceramics are sintered at elevated temperatures, oxygen loss occurs due to the defect equilibrium. Further, these oxygen vacancies may trap the electrons which partially reduce Ti^{4+} to Ti^{3+} as expressed as follows:

$$O_o^{\times} \leftrightarrow V_o\bullet\bullet + 2e' + \frac{1}{2}O_2 \quad (2)$$

$$Ti_{Ti}^{\times} + e' \leftrightarrow Ti_{Ti}^{'} \quad (3)$$

The above equations imply that the frozen defects of oxygen vacancies, elelctrons and Ti^{3+} produced during the sintering process are a result of oxygen loss. On annealing, these oxygen vacancies would be reduced and it leads to increase in $Q \times f_o$ values [16–18,29]. In addition, the increase in $Q \times f_o$ values can also be attributed to the relaxation of strain and slow cooling rate [28]. On annealing, the microwave dielectric loss in pure MTO ceramics considerably is decreased as compared to MTO ceramics with CeO_2 nanoparticles. The increase in microwave dielectric loss is due to the precipitation and diffusion of secondary phases during the annealing process. The temperature coefficient of resonant frequency (τ_f) is known to be related to the composition and the presence of secondary phase in the material. The pure MTO exhibited a negative value of τ_f −49 ppm/°C and the τ_f values for the samples with the addition of CeO_2 nanoparticles are in the range of −30 to −44 ppm/°C. The variations in the τ_f values can be attributed to the presence of secondary phases.

4. Conclusions

The microwave dielectric properties of $MgTiO_3$ ceramics with the addition of CeO_2 nanoparticles as a sintering aid were investigated. The addition of CeO_2 nanoparticles give rise to mixed phases with $MgTiO_3$ as the main phase and $MgTi_2O_5$ and Ce_2TiO_5 as minor impurity phases after sintering at 1300 °C. The values of relative density, dielectric constant

and $Q \times f_o$ are found to detoriate with increase in the amount of CeO_2 nanoparticles beyond $x=0.5$ wt% due to the formation of secondary phases. The as prepared MTO ceramics with $x=0.5$ wt%, sintered at 1300 °C exhibit excellent microwave dielectric properties i.e $\varepsilon_r = 16.55$ and $Q \times f_o = 1{,}39{,}350$ GHz. The annealing process significantly improved the microwave dielectric properties of pure MTO ceramics as compared to the MTO with CeO_2 nanoparticles. The improvement in the dielectric properties was attributed to the reduction in oxygen vacancies, relaxation of strain and the suppression of secondary phases.

Acknowledgments

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