Effect of Increasing Calcination Temperature and Partial Doping of Cobalt on the: Phase, Microwave Dielectric Properties, and Microstructure of the $MgTiO_3$ Solid Ceramics.

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Abstract:

Cobalt-doped magnesium titanate ceramics were synthesized using the conventional solid-state route with high-purity oxides from Aladdin Company. Cobalt was added in small amounts to enhance the unloaded quality factor of the material. The mixed powders were preheated at 1150°C for phase formation but failed to eliminate the MgTi2O5 secondary phase. Specimens were sintered between 1325°C and 1425°C for pellet solidification. XRD and SEM analyses confirmed the development of two phases in the ceramics. The highest bulk density was achieved at 1400°C, with a relative density of 91.90%. Theoretical density was also calculated for the cobalt-doped samples. Cobalt doping significantly improved the dielectric properties of the ceramics, with a Qufo value reaching 197,000 GHz. Additionally, slight increases in the dielectric constant (εr) and temperature coefficient of resonant frequency (tf) were observed, demonstrating the beneficial effects of cobalt doping on magnesium titanate ceramics.

Keywords: MgTiO3, Cobalt doping, Calcination temperature, Microwave dielectric properties, Microstructure, Quality factor (Qufo), Temperature coefficient of resonant frequency (tf).

1. INTRODUCTION

The fast development of emerging wireless communication systems caused to a growing demand of small high-frequency resonators, In the GHz range filters and antennas can be operated [1,2]. Uniqueness of the electrical characteristics of the ceramic dielectric resonators have transformed the microwave-based wireless communications industry with smaller and cheaper filter and oscillator elements of circuit systems [3, 4].

In the present century, the use of solid ceramics because of exhibiting the three main properties (i.e. high **εr** for the size reduction of devices, tf which assure the stability of the devices against the temperature alteration, and the last one is the high unloaded Q_uf_o value needed for the selectivity) which are essential for the use in the wireless devices increases its value in the modern world [5].The solid ceramics were the best materials to become the alternative of the air filled resonators due their high dielectric constants. The main property which is desirable to reduce the practical applications dimension, is the **εr** value of the solid ceramics [6].

$MgTiO_3$ is one of the reported solid ceramics materials which is much cheaper and exhibits these 3 main properties (Q_uf_o ~ 160,000 GHz at 7 GHz, **εr** ~ 17 & tf ~ −50 ppm/°C) needed for the practical applications [7]. One of the ways is to enhance the properties of the existing materials with the partial replacement on either A-site or B-site, since this could cause an enormous effect on the microwave properties exhibited by the main composition [8]. J.H. Sohn and his team observed that the partial replacement at A-site with Mn^{2+} and calcined at 1100°C for 4h effectively enhanced the Q_uf_o value of the $MgTiO_3$ solid ceramics beside with the decreased in the tf value [9]. Similarly, the Q_uf_o value of the $MgTiO_3$ solid ceramics was reported to be increased from 16×10^4 to 18×10^4 with fractional replacement of Mg^{2+} with Ni^{2+} at A-site [10]. C.F.

Tseng and C.H. Hsu were also among those researchers who did research on the B-position replacement of the $MgTiO_3$ material & they exchanged Ti^{4+} by Sn^{4+} because of having a small difference in the ionic difference i.e., 0.605 Å and 0.69 Å respectively [11]. This method of doping has been adopted by many researchers and they effectively enhanced the microwave properties of other solid ceramics composition as well. One of them used cobalt as doping agent and doped on the A-site of low dielectric constant $MgSiO_3$ solid ceramics and the reported values were much attractive than the base material [12].

Nickel was selected by another research team for the replacement of magnesium at A-site of the Mg_2TiO_4 solid ceramics. The Q_uf_o value which was reported by the research group for 0.02 doping of nickel was 172,800 GHz which was about 22,000 greater than the pure Mg_2TiO_4 solid ceramics. [13]. In 2003 another research also used cobalt for A-site doping in order to improve the Q_uf_o value of Zinc titanate, which was reported initially by M. Sugiura and K. Ikeda [14].

The research team doped different ratio of cobalt in the solid ceramics but the properties which noted to be good for the practical combination with this cobalt doping were observed at ratio of 0.5 cobalt doping and the resultant solid ceramics was sintered at 1150°C. The research team also stressed that sintering this solid ceramics at much higher temperature than 1150°C with doping ratio of 0.5 caused the downfall in the different values of the dielectric properties at microwave frequencies rather than enhancement. Finally, the property for which the research team conducted this research was successfully enhanced and was reported to have 70,000 GHz for the solid ceramics with the cobalt ratio of 0.5 [15].

C.L. Huang and his research fellow also selected cobalt for doping in the Mg_2TiO_4 solid ceramics and they reported that cobalt doping in this solid ceramics can effectively increase the quality factor up to 286,000 GHz with the partial replacement of 5% of cobalt with Mg^{2+} {i.e., $Mg_{0.95}Co_{0.05})_2TiO_4$} [16]. In another study, C.L. Huang et al. used cobalt again for the enhancement of Q_uf_o value of the $(Mg_{1.8}Ti_{1.1})O_4$ solid ceramics and reported that this doping of Co^{+2} over A-position in the aforementioned material could brought far enough increase of the Q_uf_o value in the chief composition. The considered microwave properties for the composition $(Mg_{1-x}Co_x)_{1.8}Ti_{11}O_4$ after doping 5% cobalt were: Q_uf_o ~ 207,500 GHz at 10.72 GHz, **εr** ~ 16.11, & tf ~ −52.6 ppm/°C [17]. In the same year 2009 the same research group doped manganese on the A-site of the aforesaid solid ceramics and reported that this doping of manganese efficiently enhanced the Q_uf_o value from 150,000 GHz to 276,200 GHz [18].

Since, it is clear from the above discussion that the partial Co^{+2} replacement had very remarkable outcome over the dielectric properties at microwave frequencies; especially on the Q_uf_o value of the various reported solid ceramics. So in our research study, we worked to find the effect of partial cobalt replacement over the microwave properties of the $MgTiO_3$ material which were calcined at somewhat upper calcination temperature of 1150°C/5h.

2. EXPERIMENTAL METHOD

There are several ways in which researchers come up with new ceramic materials. The most popular of these is the solid-state route, as it is easy, reliable, and cheap. We have also used the solid-state method to prepare ceramic samples in this work. Our sample preparation process is presented in the following steps.

For the preparation of cobalt-doped magnesium titanate ceramics, high-purity oxides were chosen to avoid impurities and defective phases. Magnesium oxide (MgO), cobalt oxide (CoO), and titanium dioxide (TiO_2) with 99.99% purity (Aladdin) served as the starting materials. The oxides were weighed according to the stoichiometric composition of $(Mg_{0.95}Co_{0.05})TiO_3$, mixed with propanol, and milled with zirconia balls for 24 hours. The slurry was dried at 100 °C for 12 hours, sieved to break agglomerates, and re-milled to obtain a fine, homogeneous powder.

To suppress secondary phases and stabilize the magnesium titanate structure, the mixed powders were calcined at 1150 °C for 5 hours. This step removed impurities and produced phase-pure ceramics suitable for further processing. The calcined powders were re-milled, dried again at 100 °C, and lightly ground using a granite mortar. To improve compactness and prevent cracking, 5 wt% polyvinyl alcohol (PVA) was added as a binder. The powders were then pressed at 150 MPa into cylindrical pellets of 5–6 mm diameter, as shown in (Fig. 1.1).

Sintering was carried out in two steps. The pellets were first heated at 600 °C for 30 minutes to burn out the binder, followed by final sintering in the temperature range of 1300–1425 °C for 4 hours. Sintering just below the melting point ensured high densification by closing pores while avoiding abnormal grain growth. This step was essential to achieve dense microstructures, as density directly influences dielectric performance (Fig. 1.2).

Phase formation and crystallographic structure were analyzed by X-ray diffraction (XRD). The technique is based on Bragg's law (2dsinθ = nλ), which describes the diffraction of X-rays from atomic planes [19]. Polished and powdered samples were examined to determine crystalline phases, lattice parameters, unit cell volume, and symmetry.

The microstructural features were studied using scanning electron microscopy (SEM/FESEM). This technique provided high-resolution images of grain morphology, porosity, and secondary phases as a function of sintering temperature. Prior to observation, samples were polished, slightly reheated below sintering temperature to reveal grain boundaries, fractured, and coated with a thin conductive layer to avoid charging under the electron beam.

Bulk density was determined using Archimedes' method according to equation (1.1):

$$\rho_{exp} = \frac{x1}{x3 - x2} \qquad (1.1)$$

Where x_1, x_2, and x_3 represent the sample weights in air, water, and air after immersion, respectively. The theoretical density was calculated using equation (1.2):

$$\rho_{theo} = \frac{ZA}{V_{Cell} \times N_A} \qquad (1.2)$$

And relative density was obtained from equation (1.3):

$$\rho_{rel} = \frac{\rho_{exp}}{\rho_{theo}} \times 100 \qquad (1.3)$$

Dielectric performance was evaluated using the Hakki–Coleman cylindrical cavity resonance method. Polished cylindrical pellets were placed between metallic plates inside a cavity, and their dimensions were precisely measured using a Vernier caliper before input into the network analyzer software. The dielectric constant (εr) and unloaded quality factor (Quf_0) were determined by recording the resonant frequency in the range of 300 MHz to 40 GHz with a network analyzer.

The temperature coefficient of resonant frequency (τf) was calculated from the resonant frequencies measured at 25 °C and 85 °C using the relation (1.4).

$$\tau_f = f_2 - f_1 / f_1 \, (T_2 - T_1) \qquad (1.4)$$

Where f_1 and f_2 are the resonant frequencies at T_1 (25 °C) and T_2 (85 °C), respectively.

Fig. 1.1: Picture of the cylindrical pellets produced from pressing the milled and calcined powders.

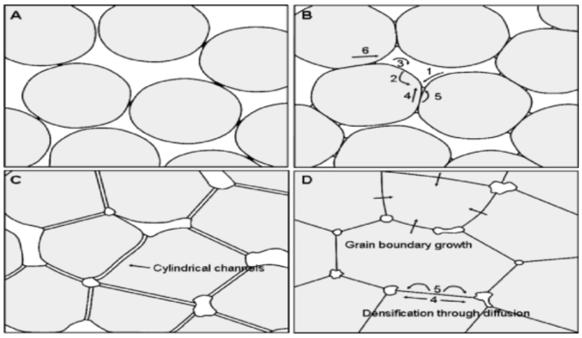

Fig. 1.2: Grain sizes enlargement and removal of the pores from the solid ceramics samples with the enhancing sintering temperature.

3. RESULTS AND DISCUSSION

3.1: XRD Results

XRD analysis of $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics calcined at 1150 °C and sintered at 1325–1425 °C confirmed the primary perovskite phase (peaks marked "*"), with a minor secondary phase $MgTi_2O_5$ detected at 2θ = 25.48° (marked "+") (Fig. 1.3). Peak positions showed no significant variation with sintering temperature. Literature reports indicate that Mg:Ti = 1:1 compositions consistently exhibit $MgTi_2O_5$, which is difficult to eliminate [20–22]. In this study, higher calcination (1150 °C, 5 h) reduced but did not fully suppress the phase, as reflected by its lower intensity compared to earlier reports. Since secondary phases degrade microwave dielectric properties, minimizing $MgTi_2O_5$ remains essential for enhancing performance.

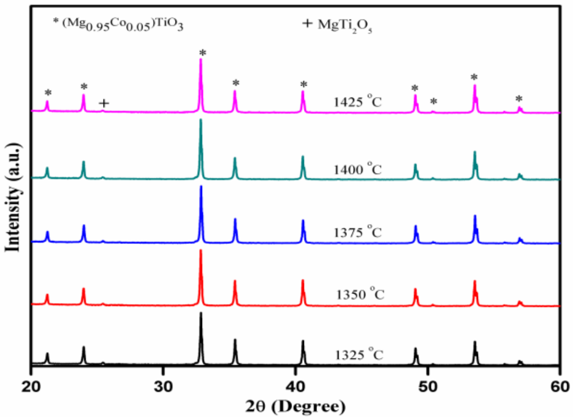

Fig. 1.3: All the solid $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics sintered in the range of 1325-1425 °C powder XRD patterns.

3.2: Micrograph of the solid material

FESEM micrographs of $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics sintered at 1400 °C are shown in Fig. 1.4. The microstructure revealed two distinct grain types: large grains (labeled 1), attributed to the primary perovskite phase, and smaller needle/rod-like grains (labeled 2), likely corresponding to the $MgTi_2O_5$ secondary phase, consistent with previous reports [11, 23]. This dual grain morphology supports the XRD findings [24]. Furthermore, the micrograph showed well-developed grains without visible porosity, confirming densification at 1400 °C/4 h and correlating with the high experimental density measured. These results validate both the phase analysis and microstructural integrity of the prepared ceramics.

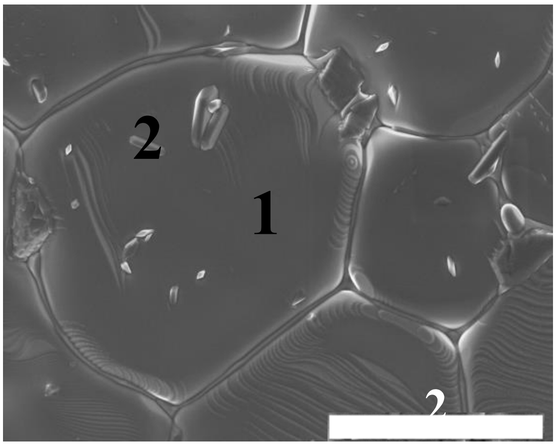

Fig. 1.4: Solid $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics microstructure; well-polished and thermally etched after the sintering temperature.

3.3: Bulk Density of the prepared solid ceramic

Experimental densities of $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics, measured by Archimedes' method, are plotted in Fig. 1.5. Density increased with sintering temperature, from 3.36 g/cm³ at 1325 °C to a maximum of 3.63 g/cm³ at 1400 °C, due to pore elimination and grain growth. A decline was observed at 1425 °C, likely from re-formation of pores. The theoretical density was calculated as 3.95 g/cm³, giving a maximum relative density of ~91.9% at 1400 °C. These results indicate optimal densification at 1400 °C/4 h, while higher temperatures negatively affect microstructural integrity.

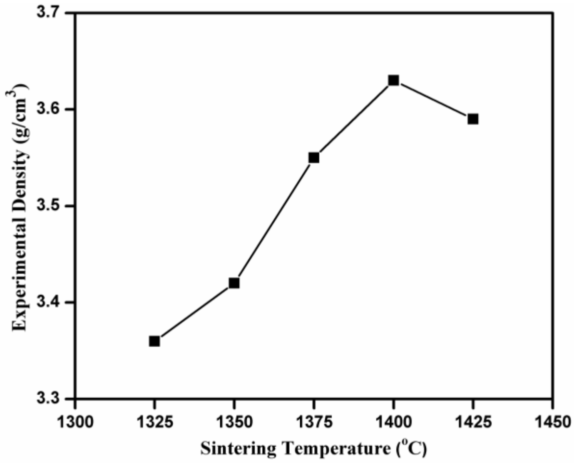

Fig. 1.5: Results of the calculated Experimental densities of the solid $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics at various sintering Temperatures.

3.4: Relative permittivity of the prepared cobalt doped magnesium titanate solid ceramics

Figure 1.6 shows the variation of relative permittivity (εr) of $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics with sintering temperature. εr increased from 15.9 at 1325 °C to a maximum of 17.1 at 1400 °C, then slightly declined to 17.0 at 1425 °C, following the same trend as density. This behavior is attributed to pore elimination at intermediate temperatures and pore re-formation at higher temperatures. εr is strongly influenced by density, composition, secondary phases, and porosity [25–31]. The observed values were higher than undoped $MgTiO_3$, likely due to the presence of $MgTi_2O_5$ (εr ≈ 17.4), which contributed to the enhanced permittivity in cobalt-doped samples.

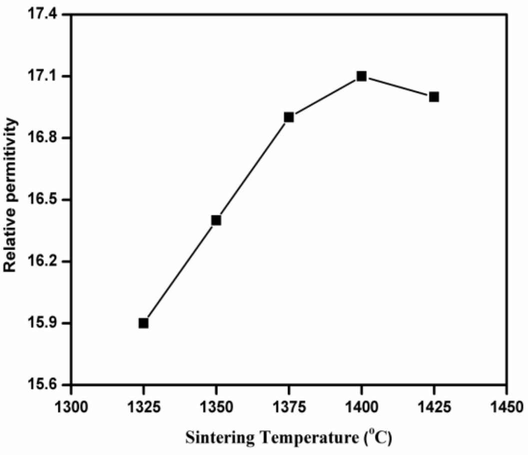

Fig. 1.6: Variation in the □r of the cobalt doped magnesium titanate solid ceramics as function of change in the solidification temperature.

3.5: Quality Factor of the Material Sintered in the Range of 1325-1425°C.

Figure 1.7 shows the unloaded quality factor (Qufo) of $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics measured by the Hakki–Coleman method. Q_uf_0 increased sharply from 148,000 GHz at 1325 °C to a maximum of 197,000 GHz at 1400 °C, then decreased at 1425 °C due to pore formation and reduced density. Variations in Q_uf_0 are strongly influenced by density, porosity, secondary phases, and sintering temperature [32–36]. The presence of $MgTi_2O_5$, which has a lower Q_uf_0 than $MgTiO_3$, may also contribute to degradation. Nevertheless, the high Q_uf_0 achieved indicates that partial cobalt substitution effectively enhances microwave dielectric performance.

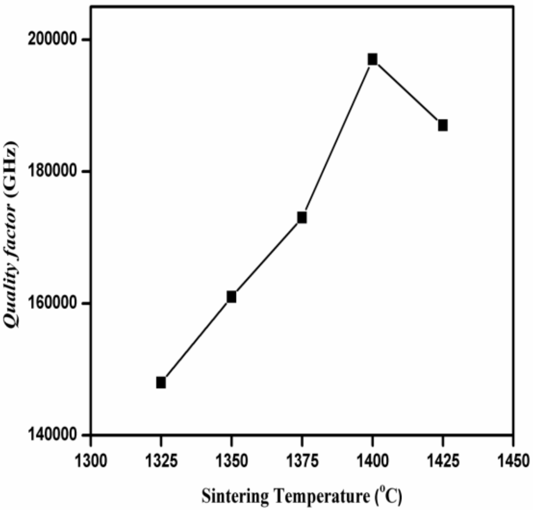

Fig. 1.7: Variation noted in the Q_uf_o value

3.6: Ceramics Material □f Value.

Figure 1.8 shows the temperature coefficient of resonant frequency (τf) of $(Mg_{0.95}Co_{0.05})TiO_3$ ceramics as a function of sintering temperature. τf increased with temperature, reaching a maximum of −49 ppm/°C at 1400 °C, then decreased to −53 ppm/°C at 1425 °C. Across the 1325–1425 °C range, τf fluctuated between −61 and −49 ppm/°C. The improved τf values compared to pure $MgTiO_3$ are attributed mainly to intrinsic effects of cobalt substitution, as the secondary $MgTi_2O_5$ phase, with even lower τf, did not significantly influence results [37].

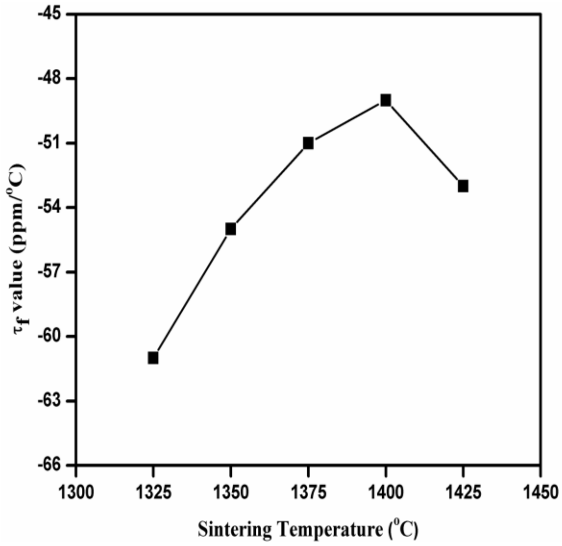

Fig. 1.8: Increment and reduction in the □f value of prepared solid ceramics by means of variation in the heating temperature.

CONCLUSION

Cobalt-doped $MgTiO_3$ ceramics were successfully synthesized using high-purity oxides and sintered between 1325–1425 °C. XRD and microstructural analysis confirmed the coexistence of a primary $(Mg_{0.95}Co_{0.05})TiO_3$ phase with a minor $MgTi_2O_5$ phase. Optimal densification was achieved at 1400 °C, yielding the highest bulk density (3.63 g/cm³), relative permittivity ($\varepsilon r \approx 17.1$), and quality factor (Qufo ≈ 197,000 GHz), despite the presence of a secondary phase. The temperature coefficient of resonant frequency (τf) reached −49 ppm/°C at this temperature. These results demonstrate that partial cobalt substitution effectively enhances the dielectric performance of $MgTiO_3$ ceramics, making them promising candidates for microwave applications.

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