The Response of \((\text{Mg}_{0.6}\text{Zn}_{0.4})\text{TiO}_3\) Ceramic System as A Dielectric Resonator Oscillator at C-Band

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Abstract. Magnesium titanate (\(\text{MgTiO}_3\)) with the ilmenite structure has been recognized as a potential dielectric ceramic material for modern cellular technology, satellite communications in the microwave frequency region, radar systems, filters, resonators and antennas in global positioning systems. This paper demonstrates the response of magnesium zinc titanate \((\text{Mg}_{0.6}\text{Zn}_{0.4})\text{TiO}_3\) ceramic system when it was mounted as a dielectric resonator element in a dielectric resonator oscillator (DRO) circuit when the circuit was tuned in a transverse electric (TE)01δ resonant mode operating at the frequency range of 3 – 12 GHz. The discussion is related to the structure, microstructure and dielectric characteristics.

1. Introduction

In today's information age, the need for modern communication technology for humans, both civilians and military for national defence is very important. Magnesium titanate (\(\text{MgTiO}_3\)) with the ilmenite structure has been recognized as a potential dielectric ceramic material for modern wireless technology, satellite communications in the microwave frequency region, radar systems, filters, resonators and antennas in global positioning systems [1–4]. There are three important aspects that must be fulfilled by a dielectric material for these applications, i.e. high relative permittivity (\(\varepsilon_r\)), high quality factor (\(Q\times f\)) and temperature coefficients of the resonant frequency (\(\tau_f\)) approaches to zero [1].

This high \(\varepsilon_r\) requirement is intended for component miniaturization, i.e. the size of the component can be reduced but still has the same performance as when the component size is not reduced, or even better. The high \(Q\times f\) is aimed to increase the frequency selectivity of the components. The temperature coefficient at the resonant frequency close to zero is intended to increase temperature stability, i.e. the \(\varepsilon_r\) performance of the component remains stable even though it is measured over a wide temperature range, and generally between 20-80 °C. \(\text{MgTiO}_3\)-based dielectric ceramics met these three important aspects. It has been reported that \(\text{MgTiO}_3\) ceramic possesses \(\varepsilon_r \sim 17.5\), \((Q\times f) \sim 21,000-45,000\) GHz at 7 GHz and \(\tau_f \sim (-50)\) - \((-40)\) ppm °C [1,2,5].

The fabrication and development of \(\text{MgTiO}_3\)-based dielectric ceramics to meet the demands of these important aspects has been widely reported [6,7,8,15,16]. The potential use of \(\text{MgTiO}_3\) ceramic as type-I capacitors [17], as microwave dielectric substrates, filters and patch antennas in wireless communication device [18] and as dielectric multilayers for 5G wireless communication technology [6] are available. However, the use of \(\text{MgTiO}_3\)-based ceramic as a dielectric resonator oscillator (DRO) operating at C-band frequency, as far the author concern, is hardly found. This paper is intended to
report the response of the (Mg$_{0.6}$Zn$_{0.4}$)TiO$_3$ ceramic system when the system acting as a dielectric resonator (DR) element in a DRO circuit operating in the C-band frequency. The effect of DR element thickness on the response is also investigated.

2. Method

The (Mg$_{0.6}$Zn$_{0.4}$)TiO$_3$ powder (abbreviated as MZT04) was synthesised from high purity magnesia, zinc and titanium metal powders (Merck) dissolved in 12 M of hydrochloric acid liquid following the route given in [1,3]. The resulting powder was calcined at 550 °C for 2h. The MZT04 ceramic was fabricated by adding 2 wt.% Bi$_2$O$_3$ powder into the MZT04 calcined powder as a liquid agent and mixed these powders using a planetary ball milling and zirconia balls under ethanol medium at 500 rpm for 5 hours. The mixed powder was dried using a rotary evaporator and compacted into pellets using a 5-mm diameter of cylindrical die press and a hydraulic hand press at 25 kg/cm$^2$ for 10 s. Two different thicknesses of the pellets were prepared, i.e. ~1.2 and ~1.5 mm. The pellets were sintered at 1000 °C for 2 h using a Yamato FD 41 furnace.

Crystalline phase identification of the MZT04 powder and the sintered ceramic was analysed by x-ray diffraction (XRD) method using Cu K$_\alpha$ radiation with X’pert ddiffactometer (Phillip) of 2θ from 15° to 65° at the rate of 0.02°/min. Microstructure of a fractured surface of the ceramic was studied using field emission scanning electron microscope (FESEM, FEI Model Inspect F50), the voltage was at 20 kV. Bulk density of the ceramic was examined using the Archimedes technique with a Balance Mettler Toledo ME 403 E and Density kit ME-DNY-43. The relative permittivity ($\varepsilon_r$) and the quality factor (Q$_{sf}$) at 3.3 GHz were measured following the Hakki-Coleman method using 8719ET network analyser (Agilent) and a rectangular resonator. The temperature coefficient of resonance frequency ($\tau_f$) was measured at the resonance frequency of the TE$_{011}$ mode at 20 and 70 °C by open cavity method using a Haeraus chamber.

The frequency response and the output (transmitted) power of the ceramic in a DRO circuit was measured using a N9343C Handheld Spectrum Analyser (Keysight Technologies) operating in the frequency range of 3 – 12 GHz, 9 – 12 DC Volt and 100 - 200 mA and tuned at transverse electric (TE)$_{011}$ resonant mode.

3. Results and Discussion

3.1. Structure and Microstructure

Figure 1 depicts the XRD patterns of MZT04 calcined powder and MZT04+2 wt. % Bi$_2$O$_3$ sintered ceramic. As shown, the spectra of the calcined powder and the sintered ceramic are all MgTiO$_3$ single phase (PDF #06-0494). The inclusion of the zinc fraction in the (Mg$_{0.6}$Zn$_{0.4}$)TiO$_3$ powder system did not cause the appearance of new peaks containing zinc. Instead, zinc contributes to the formation of the single phase MgTiO$_3$. The same is true for the (Mg$_{0.6}$Zn$_{0.4}$)TiO$_3$ ceramic system. This fact suggests that the presence of zinc in the powder system introduces the formation of (Mg$_{0.6}$Zn$_{0.4}$)TiO$_3$ solid solution which can be indexed as the MgTiO$_3$ phase. Meanwhile, in the ceramic system, the zinc fraction maintains the presence of solid solution.

Figure 2 presents the FESEM image of a fractured surface of the MZT04+2 wt. % Bi$_2$O$_3$ ceramic. As seen, microstructure of the ceramic comprises of grains with nearly uniform sizes in polyhedral morphology, pores and grain boundaries connecting the nearby grains. The average grains size measured with Image J software is (1.02 ± 0.03) μm. Upon measuring the Archimedes density, the ceramic possesses the bulk density of (3.8 ± 0.2) g cm$^-3$. The addition of 2 wt. % Bi$_2$O$_3$ in the ceramic helps to compact the ceramic faster, even though there are still pores between the grain boundaries. In our previous results in [1], MZT04 ceramic with similar density had also been attained but with a much higher sintering temperature of 1400 °C for 8 h due to the absence of Bi$_2$O$_3$ addition. Based on this fact, Bi$_2$O$_3$ addition promotes better sinterability of the ceramic. The use of Bi$_2$O$_3$ has also been reported by Hsieh [19] to improve the sinterability of (Zn,Mg)TiO$_3$ ceramic down to 1000 °C and by Belnou [20] as sintering aid for MgTiO$_3$ ceramics and therefore accelerate the densification rate of the ceramics.
Figure 1. The XRD patterns of MZT04 calcined powder and (MZT04+2 wt.% Bi$_2$O$_3$) sintered ceramic. The indexed peaks = MgTiO$_3$ (PDF #06-0494)

Figure 2. Microstructure of MZT04 ceramic taken from a fractured surface and examined using FESEM.
3.2. Dielectric properties

Table 1 lists the relative permittivity ($\varepsilon_r$) and the quality factor ($Q\times f$) of the ceramic measured at 3.3 GHz using the Hakki-Coleman method and the temperature coefficient of resonance frequency ($\tau_f$) at the TE$_{018}$ mode and when measured at 20 and 70 °C.

| $\varepsilon_r$ (at 3.3 GHz) | $Q\times f$ GHz (at 3.3 GHz) | $\tau_f$ (ppm/°C) |
|---------------------------|-------------------------------|--------------------|
| MZT04+2wt% Bi$_2$O$_3$    | 17.9                          | 21,230             |

In Table 1, the $\varepsilon_r$ of the ceramic is 17.9. This value is much higher as compared to the $\varepsilon_r$ of the MZT04 ceramic without Bi$_2$O$_3$ addition reported in [1], i.e. only 13.3. The analysis on this low $\varepsilon_r$ value was due to the zinc content in the MZT04 ceramic hindered the dipoles polarisation of the system (i.e. TiO$_2$ and (Mg/Zn)O$_2$) in the microwave frequencies. This makes sense because zinc has an atomic weight that is almost three times greater than the atomic weight of magnesium so that the presence of zinc in a big fraction as found in the MZT04 ceramic system has the potential to delay the oscillation rate of the (Mg/Zn)O$_6$ dipoles in the system. This in turns reduced the $\varepsilon_r$ value of the system. Further, the low $\varepsilon_r$ value of MZT04 ceramic in [1] was also contributed by the presence of secondary phase (Zn$_6$ Ti$_7$ O$_{23}$) accompanying MgTiO$_3$ as the main phase. In this presence work, the ceramic is single phase MgTiO$_3$ (see Fig. 1). The high $\varepsilon_r$ value of the ceramic suggests that the presence of single phase MgTiO$_3$ and 2 wt.% Bi$_2$O$_3$ has removed the hindering of the dipoles oscillation rate, especially at 3.3 GHz. Belhou in [20] claimed without showing the data that the dielectric properties of investigated MgTiO$_3$ ceramics were similar to the properties of pure MgTiO$_3$ suitable for type-I capacitors.

In Table 1, the ($Q\times f$) factor at 3.3 GHz (i.e. 21,230 GHz) is high indicates that the ceramic is a low loss dielectric material. In addition, the $\tau_f$ value of -40 ppm/°C that close to zero supports this indication. These properties are very advantageous when using the ceramic as a dielectric resonator (DR) element in a dielectric resonator oscillator (DRO) circuit. The response of the ceramic when it was mounted in the DRO circuit is provided in the following sub section.

3.3. The response as DR element in the DRO circuit at C-band

Figure 3 shows the response of MZT04+2 wt.% Bi$_2$O$_3$ ceramics with two different thicknesses (1.2 and 1.5 mm) as a DR element in a DRO circuit operating at C-band (4-8 GHz) and mechanically tuned in a transverse electric (TE)$_{018}$ resonant mode. As seen, the ceramic with the thickness of 1.2 mm displayed the signal with a sharp peak centred at 4.71933 GHz with the output power of -6.41987 dBm which is good since the peak is sharp and the peak height approaches to zero. The performance of the 1.5-mm ceramic is also good because of the same reason, i.e. at 4.69973 GHz with the output power is -3.8595 dBm.

The fact in Fig. 3 suggests three important things, i.e. first, the ceramics with two different thicknesses respond positively when using as a DRO element in the DRO circuit. These frequency responses are in the range of the C-band frequencies, i.e. 4-8 GHz, meaning that the MZT04+2 wt.% Bi$_2$O$_3$ ceramics are promising for using as DR elements in the DRO circuit operating at the C-band. Second, thicker ceramic responds at a slightly lower frequency than the thinner one. Third, with the ceramic dimension is only in the order of millimetres, it turns out that the ceramic is proven to be a DRO element. This means that the miniaturization requirement stated earlier has been fulfilled by this MZT04 ceramic. This achievement was obviously due to the advantageous dielectric characteristics, the presence of single phase MgTiO$_3$ and the relatively high bulk density of the ceramic as discussed in the previous sections.
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Figure 3. The frequency response and the transmitted power of MZT04+2 wt.% Bi₂O₃ ceramics as DR elements with two different thicknesses in the DRO circuit

4. Conclusion
The work to demonstrate the response of MZT04+2 wt.% Bi₂O₃ ceramic system as a dielectric resonator element in a dielectric resonator oscillator circuit operating at C-band frequencies has been completed. The ceramics with two different thicknesses displayed the positive response at 4.69973 GHz and 4.71933 GHz with near zero output power. This finding suggests that the ceramic is a proficient DRO candidate for C-band frequency applications.

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