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Characterization of (Mg0.5Zn0.5)TiO3 Ceramics As Material Candidates for Dielectric Resonator at Microwave Frequency

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ABSTRACT

$MgTiO_3$ ceramic is one of the dielectric materials used as a signal generator (dielectric resonator) in microwave telecommunication devices. This research is intended to explore the possibility of a new composition of $MgTiO_3$-based ceramic i.e. ($Mg_{0.5}Zn_{0.5}$)TiO_3 (abbreviated MZT05) as a dielectric resonator material. The aim was to characterize the resonant frequency of the ceramics on the microwaves (measured using spectrum analyzer) and the results were correlated to structure (X-Ray Diffraction, XRD), microstructure (Scanning Electron Microscope, SEM) and bulk density data (Archimedes method) of the ceramics. The work was begun by compacting the MZT05 crystalline powder in a cylindrical shape with a diameter of 5 mm using a uniaxial die press to produce tablets. The tablets were sintered at 1300 °C with variations holding times of 6, 8 and 10 h to produce ceramics. The structural data revealed that $MgTiO_3$ phase was identified as the main phase in the ceramics, i.e. (96.32-98.70) %molar. The bulk density increased with increasing sinter holding time, from 2.75 g/cm^3 (6 h), 2.84 g/cm^3 (8 h) to 2.99 g/cm^3 (10 h) due to the increase in grain size diameter from 919.75 nm (6 h), 1090.62 nm (8 h) to 1180.72 nm (10 h) accompanied by a decrease in the size of the pore diameter from 924.14 nm (6 h), 917.05 nm (8 h) to 800.22 nm (10 h). The ceramics produced resonant frequencies of 5.07–5.08 GHz which implies that the ceramics are proven to be potential candidates for dielectric resonator materials at microwave frequencies, especially at 5.07-5.08 GHz. The varying sinter holding time seems not to influence the resonant frequency of the ceramics because the variation holding times produce similar resonant frequencies.

Introduction

Microwave Dielectric Ceramics (MWDCs) have been widely reported as an important component of telecommunications systems, one of which is as a material signal generator (Dielectric Resonator, DR) [1]. The DR material was used to generate, stabilize signals and frequency filter in microwaves telecommunication devices [2]. The DR works on the same concept as a resonator cavity, in that microwaves kept inside the resonator will experience irregular variations in permittivity on the surface and the edge of the wall, causing the wave to oscillate on both sides of the resonator wall. The permittivity value changes from the

permittivity of vacuum, i.e. ε_o=8.85x10^{-12} F/m to the permittivity of the material [3,4]. The resonator will then form a standing wave that causes oscillatory motion with large amplitude at a certain microwave resonant frequency [5]. The microwave frequency usually used on the dielectric resonator is the transverse electric mode (TE), especially $TE_{01\delta}$ mode. $TE_{01\delta}$ mode was chosen because it has the lowest-order resonant level mode which allows this mode to operate without influencing the frequency of other higher-order modes [4,6]. The resonant frequency of this mode can be calculated based on Equation 1 [7].

$$f_0(GHz) = \frac{8.553}{\varepsilon_r{}^{1/2}\left(\frac{\pi}{4}D_r{}^2 L_r\right)^{1/3}} \quad (1)$$

Where ε_r is the dielectric constant of the DR material, *Dr* is the resonator diameter (mm) and *Lr* is the resonator height or thickness (mm).

The DR material has the characteristics of a high dielectric constant (ε_r), a high quality factor (Q_f) and a low temperature coefficient (τ_f) at resonant frequency [8]. The DR materials reported match these characteristics include $BaTiO_3$ dan $BaTi_4O_9$ [2], $Ba_2Ti_9O_{20}$ [9], $Bi_2(Li_{0.5}Ta_{1.5})O_7$-$TiO_2$ [10]. TEMEX [4] has manufactured DR materials such as E2000 and E5000. E2000 is a dielectric oxide material containing Zr, Sn and Ti and applied at frequencies range of 3-30 GHz. E5000 is a dielectric oxide material containing Ba, Sm and Ta and applied at frequencies range of 2-5 GHz. In this case, the composition of the E2000 and E5000 are not mentioned. Magnesium titanate ($MgTiO_3$) was reported as a dielectric material that meets the above characteristics, i.e. a high dielectric constant (ε_r~17), a high quality factor (Q_f~160000 at 7 GHz) and a low temperature coefficient (τ_f~50 ppm/^{o}C) [1,11,12].

Recently, Nilawati and Ermawati [13] reported $(Mg_{0.8}Zn_{0.2})TiO_3$+4 wt.% Bi_2O_3 ceramics as DR materials by varying sinter holding times of 4, 6 and 8 h. As DR materials, the ceramics produce the same resonant frequency16, i.e. 5.12 GHz due to variations in sinter holding time. Furthermore, Izza and Ermawati [14] reported $MgTiO_3$+2 wt.% Bi_2O_3 ceramics as DR materials. The ceramics have resonance frequencies of (5.0-5.2) GHz when used as DR materials due to variations in sinter holding times of 4, 6 and 8 h. Moreover, Shen et al. [15] characterized the $Mg_{0.95}Ni_{0.05}TiO_3$ ceramic and obtained ε_r~17.35; Q_f~192000 GHz and τ_f~47 ppm/^{o}C. However, Shen et al. have not reported the potential of this ceramic as a DR material.

In addition, Ermawati et al. [16] have reported the dielectric characterization of six different compositions of $(Mg_{1-x}Zn_x)TiO_3$-based ceramics (for *x*= 0; 0.1; 0.2; 0.3; 0.4 and 0.5). The ceramics were sintered at 1400 ^{o}C for 8 h. The structural characterization result of the ceramic for *x*=0.5, i.e. $(Mg_{0.5}Zn_{0.5})TiO_3$ ceramic revealed that the $MgTiO_3$ phase successfully formed as the main phase and accompanied by Zn_2TiO_4 as the secondary phase. Rostianbudi and Ermawati [17] reported the results of the characterization of the structure and bulk density of $(Mg_{0.5}Zn_{0.5})TiO_3$ ceramics with the addition of *x* wt.% Bi_2O_3 (for *x*=0, 1, 3, 5 and 7). The ceramics were sintered at 1000 ^{o}C for 2 h. The structural characterization result of the ceramic for *x*=0, i.e. $(Mg_{0.5}Zn_{0.5})TiO_3$+0 wt.% Bi_2O_3 ceramic consisting of 94.23% molar $MgTiO_3$ as the main phase and the remaining is rutile TiO_2. However, Ermawati et al. [16] and Rostianbudi and Ermawati [17] have also not reported the potential of $(Mg_{0.5}Zn_{0.5})TiO_3$ ceramics as DR materials.

This study is therefore intended to report the characterization of $(Mg_{0.5}Zn_{0.5})TiO_3$ ceramics (abbreviated MZT05) as DR materials at microwave frequencies. The ceramics were fabricated from MZT05 powder and sintered at 1300 ^oC with variation sinter holding times of 6, 8 and 10 h. The structure, microstructure and bulk density data of the ceramics were analyzed to discuss the resonance frequency of the ceramics as DR materials.

Experimental Method

MZT05 powder synthesized by Ermawati et al. [16] and calcined at 550 ^oC was used in this research to fabricate MZT05 ceramics. The calcined powder was identified containing $MgTiO_3$ as the main phase and TiO_2 rutile as an extra phase. Ermawati et al. [16] reported that using the 50 %mol Zn as a dopant agent to Mg^{2+} ions in MZT05 powder system was proven to lower the formation temperature of $MgTiO_3$ phase from 700 °C (without Zn^{2+} ions) to 550 °C (with the addition of Zn^{2+} ions). In this research, the MZT05 calcined powder was compacted into cylindrical tablets with a diameter of 5 mm using a hydraulic-hand press and a uniaxial die press. The compaction pressure of 2.5 MPa was applied for 10 seconds. The 2.5 MPa was chosen because of Izza and Ermawati [14] used the same pressure on $MgTiO_3$+2 wt.% Bi_2O_3 ceramics and produced the resonant frequencies of the DR materials at (5.0-5.2) GHz. In this work, the tablets were sintered at 1300 ^oC for 6, 8 and 10 h to produce ceramics using a furnace (BWS). The 1300 ^oC was chosen because Ermawati et al. [16] used the sintering temperature of 1400 ^oC for 8 h and obtained Zn_2TiO_4 as an extra phase. Meanwhile Rostianbudi and Ermawati [17] sintered the ceramics at 1000 ^oC for 2 h but did not produce single-phase $MgTiO_3$ yet. Gogoi et al. [18] chose 1300 ^oC to sinter $MgTiO_3$ ceramics and obtained single phase $MgTiO_3$. Based on that, the sintering temperature of 1300 ^oC was used in this research.

The structure analysis was carried out using x-ray diffraction (Bragg-Brentano Philips X'pert Diffractometer) with Cu-Kα radiation in the diffraction angle range (2θ) 15-70 ° and step size 0.02 ^{o}/min. Structural data analyzed using Match! software to identify the crystalline phases formed on the ceramics, then analyzed by the Rietveld method using Rietica software to obtain %molar, lattice parameter and unit cell volume of all identified phases belonging to the MZT05 ceramics. Bulk density was measured by the Archimedes method (Equation 2) using the Mettler Toledo Type ME 403 E Balance and Density Kit ME-DNY-43 integrated into the Hyperterminal software to input and read output data.

$$\rho = \frac{m_d}{m_w - m_a}\rho_a \quad (2)$$

Where ρ is bulk density of ceramic (g/cm^3), m_d is dry mass, m_w is wet mass, m_a is Archimedes mass and ρ_a is the density of aquadest (g/cm^3) as a medium.
Microstructure of a broken surface was characterized using a scanning electron microscope (SEM) FEI Inspect S50 operated at 20 kV and a magnification of 5000x. Diameter of grains and pores on the microstructure were measured using ImageJ software. Figure 1 demonstrates how to measure a pore diameter on the microstructure using ImageJ software.

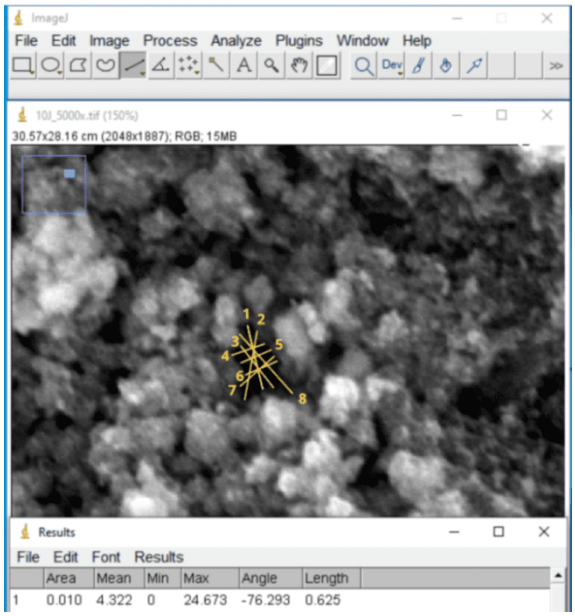


Figure 1. A demonstration of how a pore diameter of MZT05 microstructure was measured using ImageJ software. Considering that the shape of the pore is not symmetrical, the measurement must be repeated several (eight) times from different positions (No. 1 to 8) to obtain the average diameter value.

The resonant frequency of the ceramics as DR materials in the Dielectric Resonator Oscillator (DRO) circuit (see Figure 2) was measured using a spectrum analyzer (Keysight MXA Signal Analyzer N9020A). The spectrum analyzer operates within 3-12 GHz, a voltage of 9-12 V and an electric current of 100-200 mA using $TE_{01\delta}$ mode. In Figure 2, the DRO circuit consists of four primary components, i.e. A, B, C and D where A is a cable to connect between B and the spectrum analyzer; B (the square box) is the DRO circuit; C is (black and red) connectors to connect between B and power supply; D is MZT05 ceramic as a DR material.

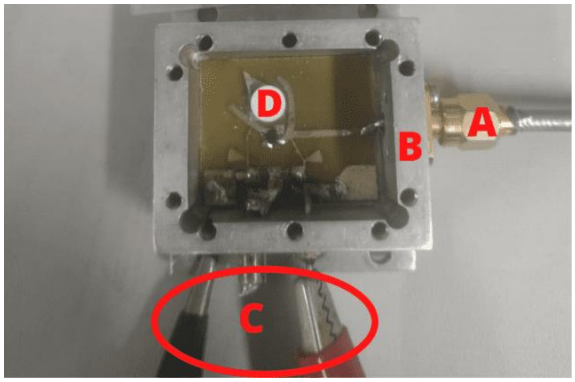


Figure 2. A demonstration on how the resonant frequency of MZT05 ceramic in the DRO circuit was set up and measured in this research.

Results and Discussion

1. Structure

Figure 3 shows of the XRD patterns of MZT05 ceramics measured using x-ray diffraction (Bragg-Brentano Philips X'pert Diffractometer) with Cu-Kα radiation in the diffraction angle range (2θ) of 15-70 ° and step size 0.02 °/min. Phase identification on the XRD patterns in Figure 3 was carried out using Match! software. Meanwhile, Figures 4a-c show the Rietveld refinement of the three XRD patterns in Figure 3 using Rietica software. Table 1 summarizes the output of the Rietveld refinement analysis in Figures 4a-c consisting of %molar, lattice parameter and unit cell volume of identified phases.

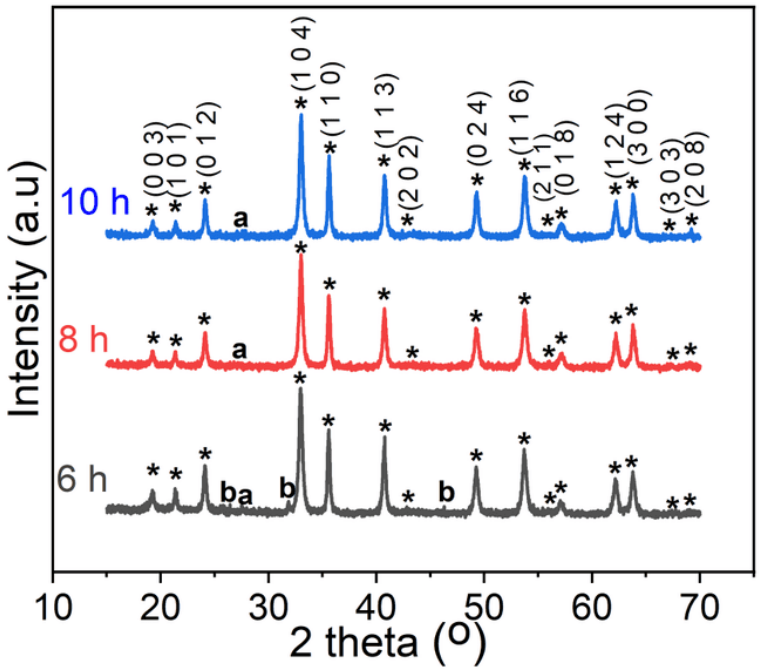
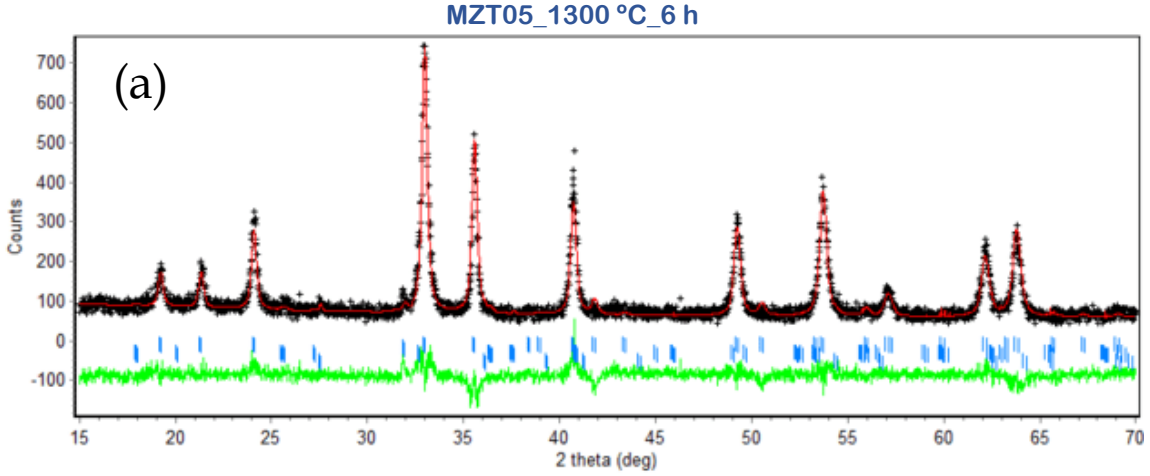


Figure 3. XRD patterns of MZT05 ceramics sintered at 1300 °C for 6, 8 and 10 h. * = $MgTiO_3$ (PDF No. 06-0494), a = TiO_2 *rutile* (PDF No. 21-1276), b = $MgTi_2O_5$ (PDF No. 35-0792)



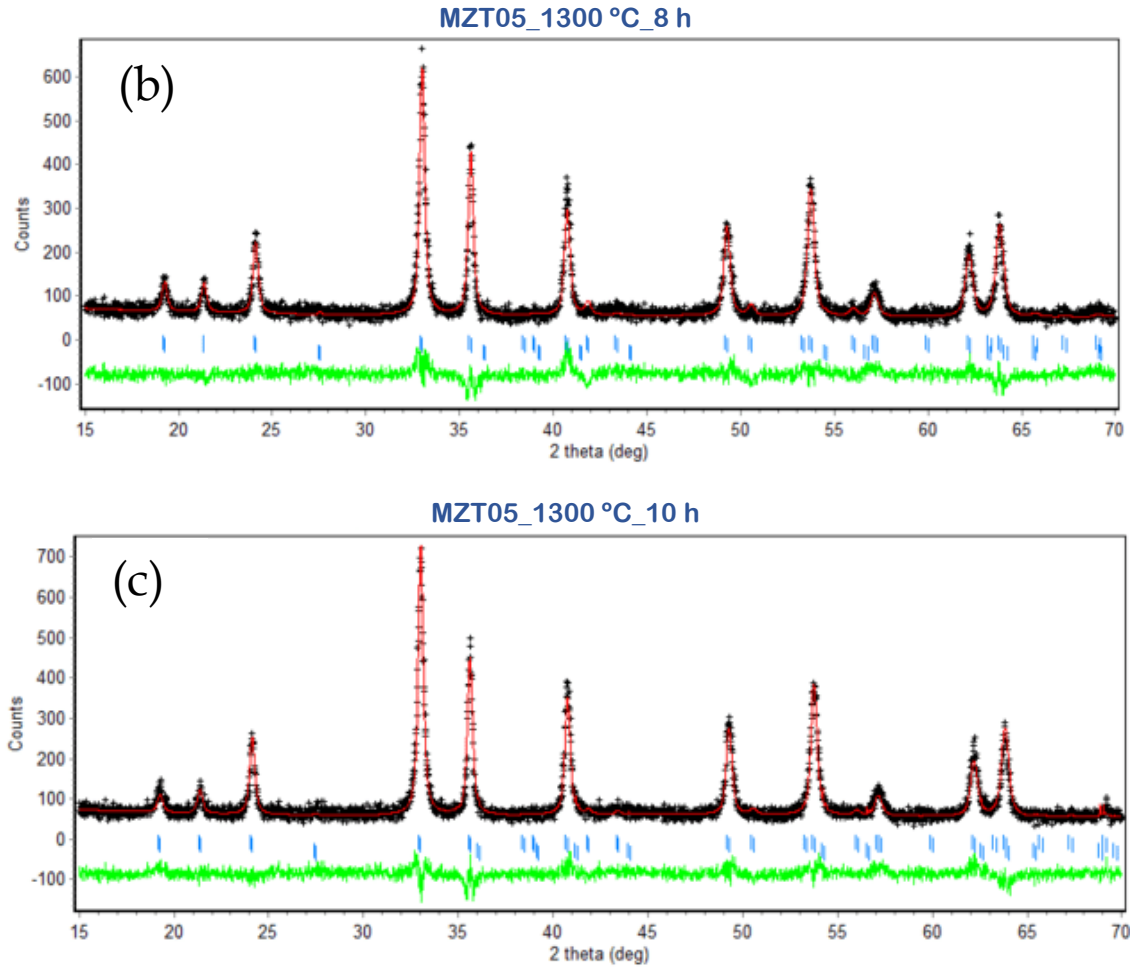


Figure 4. Rietveld refinement of XRD patterns in Fig. 3: (a) 6 h (FoM : R_p = 10.35; R_{wp} = 13.29; R_{exp} = 10.12; GoF = 1.72), (b) 8 h (FoM : R_p = 10.94; R_{wp} = 13.97; R_{exp} = 11.04; GoF = 1.60), (c) 10 h. (FoM : R_p = 10.37; R_{wp} = 13.16; R_{exp} = 10.74; GoF = 1.50)

Table 1. The output of Rietveld refinement analysis in Figure 4a-c.

Sinter holding time (h)	Phase	% Molar	Lattice Parameter (Å)		Unit Cell Volume (Å^3)
6	$MgTiO_3$	96.32±1.62	a=b c	5.06±0.00 13.90±0.00	308.34±0.06
	TiO_2	2.67±0.95	a=b c	4.59±0.00 2.96±0.00	62.68±0.05
	$MgTi_2O_5$	1.01±0.95	a b c	3.72±0.00 9.91±0.00 9.89±0.00	365.10±0.06
8	$MgTiO_3$	98.70±1.71	a=b c	5.06±0.00 13.89±0.00	307.96±0.06
	TiO_2	1.30±0.60	a=b c	4.59±0.00 2.94±0.00	62.20± 0.30
10	$MgTiO_3$	98.70±2.60	a=b c	5.06±0.00 13.90±0.00	308.30±0.05
	TiO_2	1.30±0.40	a=b c	4.60±0.00 2.97±0.00	63.10±0.01

In Figure 3, the peaks with (*) symbol correspond to peaks belonging to the $MgTiO_3$ phase (Powders Diffraction File, PDF No.06-0494). The peak with (a) symbol corresponds to the TiO_2 rutile peak (PDF No.21-1276) and the peaks with (b) symbol correspond to the $MgTi_2O_5$ peaks (PDF No.35-0792). The MZT05 ceramic with a sinter holding time of 6 h contained three phases (i.e. $MgTiO_3$, $MgTi_2O_5$ and TiO_2 rutile), whilethat of 8 and 10 h consist of $MgTiO_3$ and TiO_2 rutile phases only. Thus, the increase in sinter holding time (especially at 8 and 10 h) succeeded in removing the $MgTi_2O_5$ phase. Figure 3 also shows that $MgTiO_3$ are the dominant phase. Thus, fabrication of MZT05 ceramics carried out in this research was confirmed to produce $MgTiO_3$ as the main phase, i.e. (96.32-98.70) %molar (see Table 1). There is no peak containing Zn was also detected in Figure 3 which indicates that the Zn^{2+} ions has entered the Mg^{2+} ions sites in the ceramics system. As a result, the MZT05 ceramics are substitution solid solution because the radius of Mg^{2+} ions is similar with that of Zn^{2+} ions (see Figure 5 for the illustration of substitution solid solution). Ermawati et al. [16] reported the structural characterization of $(Mg_{1-x}Zn_x)TiO_3$-based ceramics (for x = 0; 0.1; 0.2; 0.3; 0.4 and 0.5), specifically for x = 0.5 (i.e. MZT05). The identified phase is $MgTiO_3$ as the main phase and accompanied by Zn_2TiO_4 phase. Ermawati et al. [16] also reported that Zn_2TiO_4 reduced the dielectric properties of ceramics from the host ceramic. Thus, the MZT05 ceramic structure in this study is better than that Ermawati et al. [16]. The XRD pattern in Figure 3 was further utilized to obtain the average crystallite size data of the MZT05 ceramics using the Scherrer Method. Equation (3) provides the Scherrer formula to calculate the crystallite size [19].

$$D = \frac{K\lambda}{\beta \cos\theta} \tag{3}$$

Where D is the average crystallite size of ceramic (nm), K is the Scherrer constant (0.94 for spherical crystallites with cubic symmetry), λ is wavelength of X-ray (0.15406 nm), β is full width at half maximum (radians), θ is diffraction angle or half of 2θ (^{o}).
The average crystallite size calculated from the peaks in Figure 3 increased from 20.62; 21.68 to 22.02 nm due to the increase in sinter holding time from 6, 8 to 10 h. The increase in sinter holding time caused the atomic diffusion rate to increase, thereby triggering increased crystal growth. The increase in crystal growth succeeded in increasing the crystallite size [20]. Sharon et al. [21] reported that $MgTiO_3$ ceramic sintered at 1300 °C for 4 h produced a larger average crystallite size than that in this present study, i.e. 70.44 nm.

In Figures 4a-c, the "+" symbol represents the measured peaks. The red line represents the calculated peaks patterns. The green line represents the difference height intensity between the measured and calculated peaks. The blue vertical line under the calculated and measured peaks indicates the Bragg peaks belonging to all identified phases. Figures of Merit (FoM) of the Rietvield refinement consisting of profile factor (R_P), weighted profile factor (R_{wp}), experimental factor (R_{exp}) are also shown. The FoM data are less than 20% with the Goodness of Fit (GoF) of less than 2% which indicates that the Rietveld refinement has been successful [22]. It is therefore the output analysis Rietveld refinement shown in Table 1 can be used for further analysis.

In Table 1, the %molar of $MgTiO_3$ phase in the three ceramics is (96.32-98.70) %molar, the remaining belong to TiO_2 rutile and $MgTi_2O_5$. According to Fang et al. [23], the $MgTi_2O_5$ phase was formed from $MgTiO_3$ and TiO_2, i.e. $MgTiO_3+TiO_2 \rightarrow MgTi_2O_5$. The increase in sinter holding time has increased the %molar of $MgTiO_3$ phase. As shown in Table 1, increasing the sinter holding time succeeded in removing the $MgTi_2O_5$ phase and reducing the %molar TiO_2 phase (especially for 8 and 10 h). The existence of an extra phase succeeded in reducing the %molar of the main phase $MgTiO_3$. In other words, the increase in sinter holding time was proven to increase the %molar of $MgTiO_3$ phase. The %molar of $MgTiO_3$ phase obtained in this research is better than that of Rostianbudi and Ermawati [17] (i.e. 94.23 %molar). As also seen in Table 1, the size of $MgTiO_3$'s lattice parameters a=b and c decreased by increasing sinter holding time from 6 to 8 h, i.e. from (a=b=5.06±0.00 and c=13.91±0.00) Å (for 6 h) to (a=b=5.06±0.00 and c=13.89±0.00) Å (for 8 h) and slightly increased to (a=b=5.06±0.00 and c=13.90±0.00) Å (for 10 h). The same trend was also detected in the unit cell volume; the value decreased by increasing holding time from 6 to 8, i.e. from (308.34±0.06) $Å^3$ (for 6 h) to (307.96±0.06) $Å^3$ (for 8 h) and slightly increased to (308.30±0.05) $Å^3$ (for 10 h). The lattice parameter and unit cell volume of $MgTiO_3$ phase obtained in this research were relatively bigger than that of Ermawati et al. [16] (the lattice parameter of a=b=4.98 and c=13.71 Å and the unit cell volume of 295.56 $Å^3$).

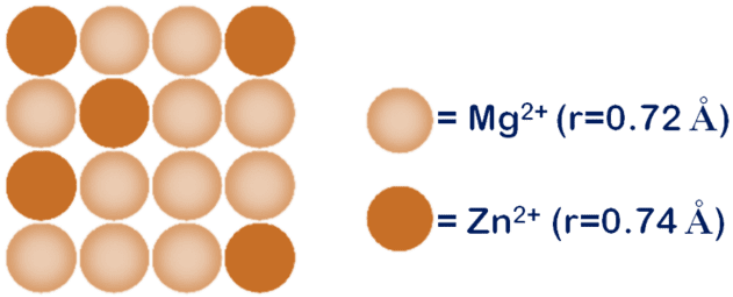


Figure 5. Illustration of a substitution solid solution in MZT05 ceramic (Source: Private Property)

In Figure 5, the array of circles illustrates the position of the Mg^{2+} ions in MZT05 ceramic structure. As previously mentioned, a fraction of Zn^{2+} ions were intentionally added as a dopant agent to lower the formation temperature of $MgTiO_3$ phase from 700 °C (without Zn^{2+} ions) to 550 °C (with the addition of Zn^{2+} ions). The Zn^{2+} ions was chosen due to several considerations, i.e. : 1) Zn^{2+} ions has a lower melting point i.e. 419 °C as compared to the formation temperature of $MgTiO_3$ phase [24]; 2) Zn^{2+} and Mg^{2+} ions both have the same coordination number (i.e. octahedral) so as to prevent any transfer (donor and acceptor) possibility of electrons between the ions that could interfere the structure of the ceramics; 3) the radius of the Zn^{2+} ions (r=0.74 Å) is similar to that of the Mg^{2+} ions (r=0.72 Å). This allows Zn^{2+} ions to partially replace of the Mg^{2+} ion sites and form a substitution solid solution as shown in Figure 5 [25,26]. Therefore, the dark brown circles in Figure 5 were occupied by Zn^{2+} ions, while the light brown circles were still occupied by Mg^{2+} ions. However, the Zn^{2+} ions has a relatively larger ionic radius than that of the Mg^{2+} ions. So Zn^{2+} ions still has a potential to increase the size of the lattice parameter and unit cell volume of $MgTiO_3$ phase in MZT05 ceramics (see Table 2).

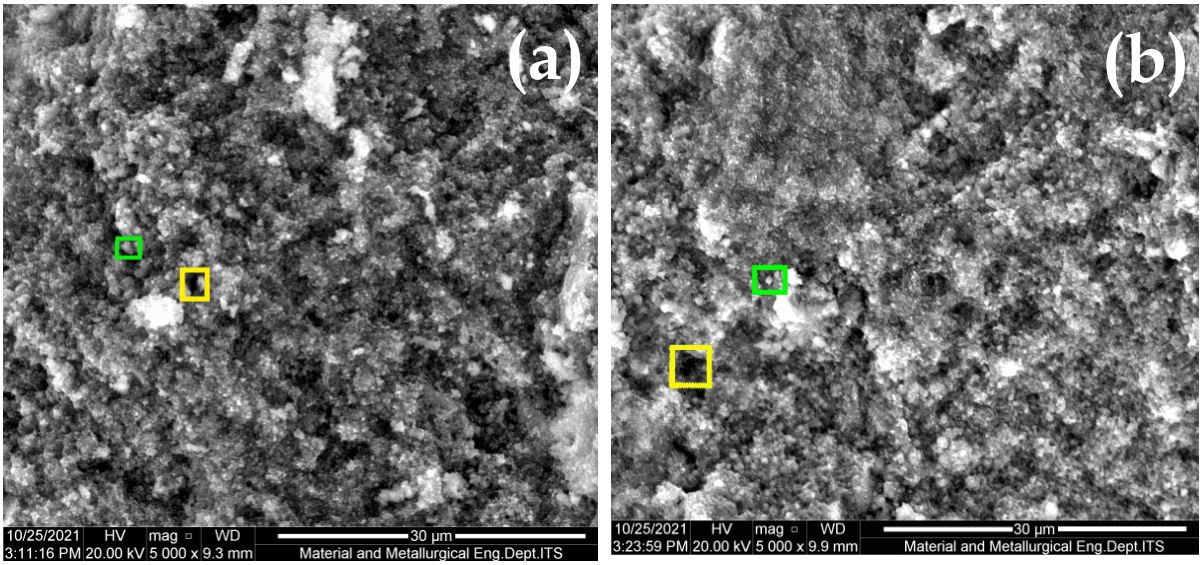
Table 2. A comparison of lattice parameter and unit cell volume size of $MgTiO_3$ phase between that in PDF No. 06-0494 database versus in MZT05 ceramics fabricated in this work.

Pure $MgTiO_3$ (PDF No. 06-0494 database)			MZT05 (fabricated in this work)			
Lattice Parameter (Å)		Unit Cell Volumes (Å^3)	Sinter holding time (h)	Lattice Parameter (Å)		Unit Cell Volume (Å^3)
a=b	c			a=b	c	
5.05	13.89	307.60	6	5.06±0.00	13.91±0.00	308.34±0.06
			8	5.06±0.00	13.89±0.00	307.96±0.06
			10	5.06±0.00	13.90±0.00	308.30±0.05

Based on Table 2, the increase in size of the lattice parameter and unit cell volume of $MgTiO_3$ phase was found in MZT05 ceramics as compared to that in the pure $MgTiO_3$. The data in Table 2 therefore confirmed the potential increase in size of the lattice parameter and unit cell volume of $MgTiO_3$ phase in MZT05 ceramics due to inclusion of Zn^{2+} ions as the dopant agent to Mg^{2+} ions [27,28].

2. Microstructure

Figure 6 shows the microstructure of the MZT05 ceramics each was taken from a broken surface of the ceramic and measured using scanning electron microscope (SEM) FEI Inspect S50 at 20 kV and a magnification of 5000x. Figure 7 shows the average diameter of grains and pores in Figures 6a-c measured using ImageJ software as seen in Figure 1.



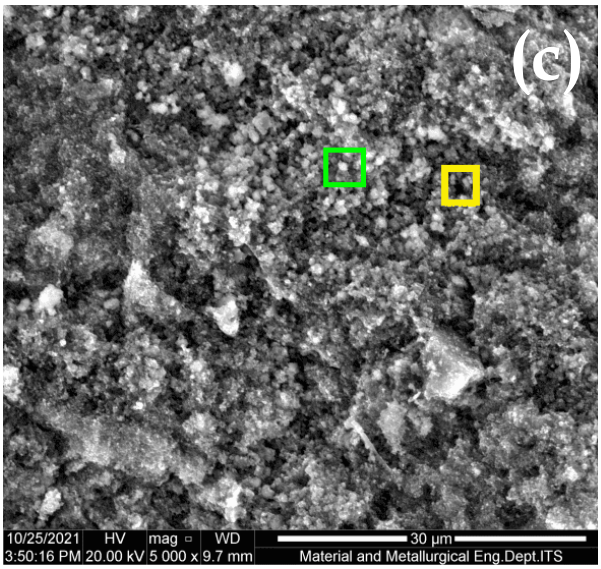


Figure 6. Microstructure of the broken surface of MZT05 ceramics for (a) 6, (b) 8, (c) 10 h at 5000x magnification.

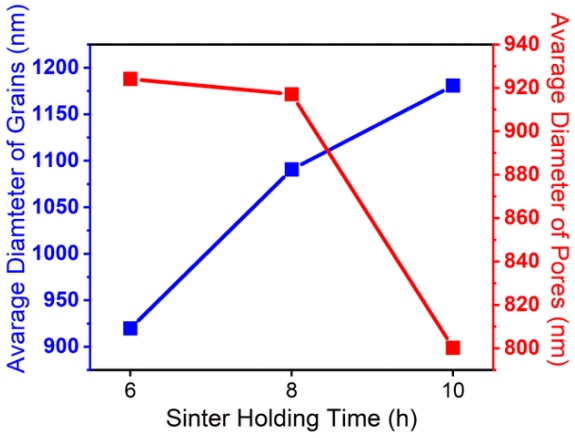


Figure 7. The average grains and pores diameters in MZT05 ceramics due to the three variations of sinter holding times and that was measured using ImageJ software.

In Figure 6, the microstructure appears solid consisting of grains (white and gray in the green box) and pores (black in the yellow box). The grains were not grown, instead it agglomerated to each other. Using the ImageJ software, grains and pores were measured and plotted as shown in Figure 7. As seen in Figure 7, the average grains diameter increased with increasing sinter holding times from 919.75 nm (6 h), 1090.62 nm (8 h) to 1180.72 nm (10 h). These results are inversely related to the average diameter of pores that reduced as sinter holding times increased, i.e. from 924.14 nm (6 h), 917.05 nm (8 h) to 800.22 nm (10 h). The increase in sinter holding time causes the grains to enlarge so that it can reduce the pores [29].

3. Bulk Density

Figure 8 shows bulk density of MTZ05 ceramics measured using the Archimedes method and Mettler Toledo Type ME 403 E Balance equipped with Density Kit ME-DNY-43 and Hyperterminal software.

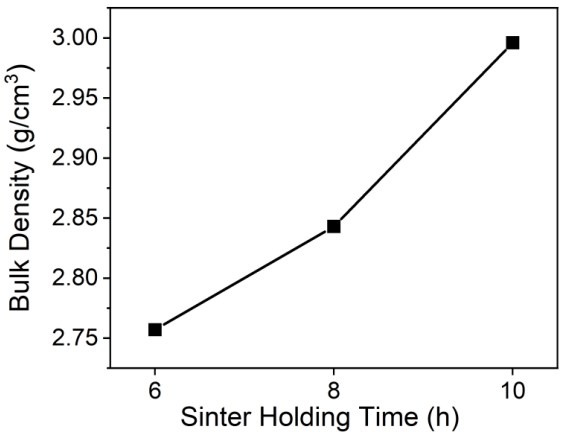


Figure 8. Bulk density of MZT05 ceramics as a function of sinter holding time

In Figure 8, the density of MZT05 ceramics increases with increasing sinter holding time, i.e. from 2.75 g/cm^3 (for 6 h), 2.84 g/cm^3 (for 8 h) to 2.99 g/cm^3 (for 10 h). The increase in bulk density was verified by microstructural data in Figures 6a-c and Figure 7. The increase in sinter holding time affect the bulk density of MZT05 ceramics. The increase in sinter holding time was proven to increase the grain size accompanied by a decrease in porosity (see Figure 7), so that the bulk density increased. Rostianbudi and Ermawati [17] reported the increase in bulk density occurred along the increase in the concentration of x wt.% Bi_2O_3, i.e. from 3.57 g/cm^3 (for x=0); 3.58 g/cm^3 (for x=1); 3.65 g/cm^3 (for x=3); 3.69 (for x=5) to 3.72 g/cm^3 (for x=7). Especially for the ceramic with x=0 (i.e. MZT05+0 wt.% Bi_2O_3), the bulk density of the ceramic is 3.57 g/cm^3. This bulk density value is much better than that measured in this present work. This might be related to the sintering temperature chosen by Rostianbudi and Ermawati, i.e. 1000 ^{o}C. The decrease in density due to sintering has been reported by Johan and Ramlan [30] on Na-β"-Al_2O_3 ceramics when the ceramics were sintered at 1150 ^{o}C for 6, 8 and 10 h. Johan and Ramlan reported that the density of Na-β"-Al_2O_3 ceramics decreased as sinter holding time increased. When the sintering holding time increased, the pore size increases as the gas required to strengthen the bonds between atoms burns and evaporates, causing the density to decrease. However, this explanation has not been confirmed yet for this work (i.e. MZT05 ceramics).

4. DRO Resonant Frequency

Figure 9 shows the resonant frequency on MZT05 ceramics as DR materials on DRO circuit and measured using spectrum analyzer at frequencies range of 3-12 GHz, a voltage of 9-12 V and an electric current of 100-200 mA with $TE_{01\delta}$ mode.

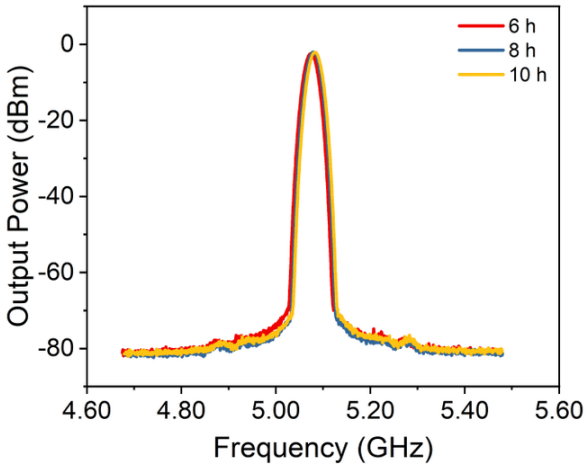


Figure 9. The resonant frequency of MZT05 ceramics as DR materials with three variations of sinter holding times.

As seen, the resonant frequency peaks of the three ceramics are located at the relatively similar position (overlapped to each other), i.e. at 5.07 GHz with output power of -2.41 dBm for (for 6 h), 5.08 GHz with output power of -2.01 dBm (for 8 h) and 5.08 GHz with output power of -2.07 dBm (for 10 h). The output power of ceramic (6 h) is the lowest as compared to the similar output power in the other two ceramics. This may be related to the fact that the %molar data

of the $MgTiO_3$ phase found in the 6-hour ceramic is the lowest as compared to the similar data in the other two ceramics, as shown in Table 1. The more %molar of $MgTiO_3$ phase found in the test ceramic, the greater the output power should be generated from the resonance curve because the resonance curve belongs to $Mg_{0.5}Zn_{0.5}TiO_3$ ceramic with the dominant phase is $MgTiO_3$. This fact shows the evidence that the three ceramics are capable of being applied as DR materials in microwave frequencies, especially in 5.07-5.08 GHz; the variation of sinter holding time does not seem to affect the performance of MZT05 ceramics as DR materials. Ermawati [31] reported $(Mg_{0.6}Zn_{0.4})TiO_3$'s performance as a DRO material at a frequency slightly lower than that in this study, i.e. 4.7 GHz. When the spectra in Figure 9 were fitted with a Gaussian curve, the Full Width at Half Maximum (FWHM) size of spectra in Figure 9 can be calculated (see Figures 10a-d).

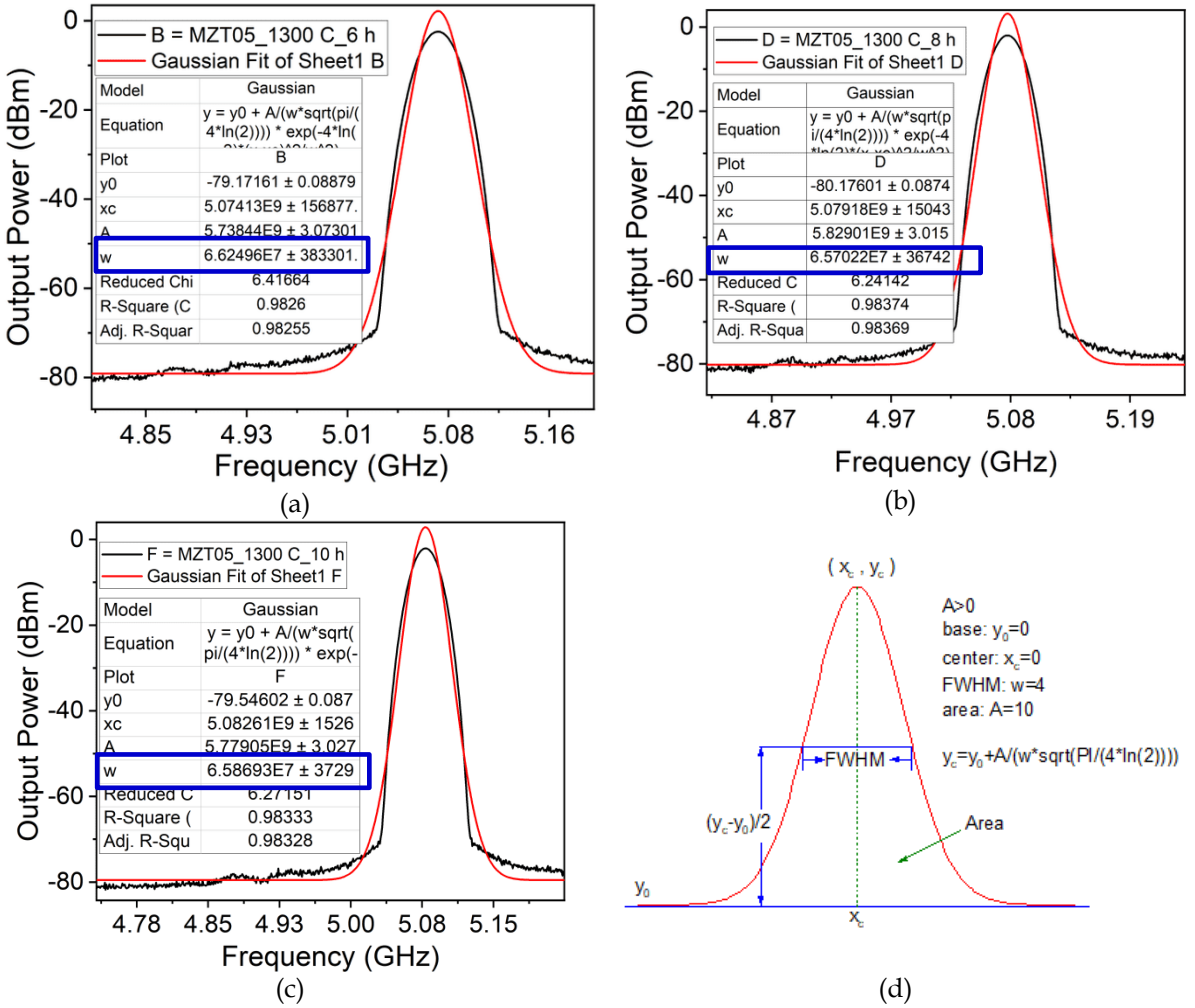


Figure 10. Fitting of the MZT05 ceramic resonance frequency curves to the Gaussian curve for (a) 6 h, (b) 8 h, (c) 10 h and (d) the theoretical Gaussian curve.

In Figure 10, the (w) symbol represents the FWHM width size. The FWHM size of MZT05 ceramics are relatively the same, i.e. 66.25 MHz (for 6 h); 65.70 MHz (for 8 h) and 65.87 MHz (for 10 h). Kadarosman and Ermawati [32] reported that $(Mg_{0.9}Zn_{0.1})TiO_3$+2 wt.% ceramics

exhibited the DR resonant frequencies at 5.08-5.12 GHz. The resonance frequency in [32] is similar to those measured in this work; however Kadarosman and Ermawati have not measured the FWHM size of the resonance frequency. The resonance frequency of the ceramics in Figure 9 were obtained due to the presence of $MgTiO_3$ as the main phase in all the sinter holding time, the similar microstructure with similar grains and pores diameter, as well as the bulk density that all suitably for DR material measurement.

Conclusion

The characterization of the structure, microstructure, bulk density and resonance frequency of MZT05 ceramics at 1300 ^oC by varying sinter holding times of 6, 8 and 10 h was completed. The structural analysis of MZT05 ceramics shows that the $MgTiO_3$ is the main phase. The microstructure of ceramics appears solid and bulk density increased with increasing sinter holding time due to an increase in grains diameter accompanied by a decrease in porosity. The bulk density and microstructure of MZT05 ceramics are affected by variations in sinter holding time. The MZT05 ceramics was confirmed capable to be used as a DR material in microwave frequencies, especially at 5.07-5.08 GHz with near-zero output power. The variations in sinter holding time produced relatively similar resonant frequencies.

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