

OPTIMUM FIRING TEMPERATURES FOR FABRICATION OF SOFT FERROMAGNETIC $\text{Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4$ CERAMICS VIA THE SOLID-STATE COMBUSTION TECHNIQUE

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Abstract

The optimum conditions for fabricating $\text{Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4$ (NZLAF) ceramics prepared by the solid-state combustion technique with glycine as fuel were investigated. The NZLAF samples were calcined and sintered in the temperature range of 850-1,050°C (in 50°C increments) for 2 h and 1,150-1,350°C (in 50°C increments) for 4 h, respectively. A pure cubic spinel phase was obtained in the NZLAF powders calcined above 900°C for 2 h, as well as in all of the NZLAF ceramics. Rietveld refinement analysis indicated that the lattice parameter a of the NZLAF ceramics decreased from 8.3803 to 8.3663 Å when the sintering temperature increased from 1,150 to 1,300°C, then increased. The grain morphology of the NZLAF ceramics exhibited a polygonal grain shape, and the average grain size increased with increasing sintering temperatures. The NZLAF ceramic produced with the optimum firing condition (1,300°C for 4 h) showed good crystalline and morphology, the highest density (5.31 g/cm³), the highest dielectric constant at 1 kHz ($\epsilon \sim 130,987$) and good ferromagnetic properties ($M_s \sim 82.40$ emu/g, $M_r \sim 0.05$ emu/g, and $H_c \sim 3.65$ Oe).

Keywords: Dielectric; Ferromagnetic; NZLAF; Solid-State Combustion

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Introduction

Spinel ferrites are a class of magnetic materials with the general formula $M\text{Fe}_2\text{O}_4$, where 'M' represents a metal, such as manganese (Mn), cobalt (Co), nickel (Ni), or copper (Cu) (Liandi *et al.*, 2023). These materials exhibit ferrimagnetic properties and possess unique magnetic, electrical, and catalytic characteristics. Consequently, these materials have a wide range of applications, such as catalysis, magnetic storage, and sensors (Salih and Mahmood, 2023).

Nickel ferrite (NiFe_2O_4) is an important soft magnetic material with diverse applications. It has a spinel structure, where nickel ions (Ni^{2+}) occupy octahedral sites, while iron ions (Fe^{3+}) occupy both octahedral and tetrahedral sites (Bharati *et al.*, 2020). Nickel ferrite exhibits typical ferrimagnetic properties, including relatively high saturation magnetization and low energy losses. These characteristics make nickel ferrite useful in applications such as magnetic storage, microwave devices, and magnetic resonance imaging (MRI) (Nejati and Zabihi, 2012).

Recently, several researchers have successfully developed high-performance nickel ferrite ceramics. Vepulanont *et al.* (2021) synthesized NiFe_2O_4 ceramics using a combustion method. They found that the ceramic sintered at $1,200^\circ\text{C}$ exhibited the highest saturation magnetization ($M_s \sim 62.84 \text{ emu/g}$) and the lowest coercive field ($H_c \sim 31.18 \text{ Oe}$). To improve the ferromagnetic properties of nickel ferrite ceramics, Premkumar *et al.* (2019) substituted Zn into $\text{Ni}_{(1-x)}\text{Zn}_x\text{Fe}_2\text{O}_4$ ceramics with $x = 0.2, 0.3$ and 0.4 using the solid-oxide route. The maximum M_s of 64.57 emu/g and low $H_c \sim 7.92 \text{ Oe}$ were obtained when $x = 0.4$. Moreover, Ni *et al.* (2020) successfully increased the M_s of $\text{Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4$ (NZLAF) ceramics from 103.00 to 110.95 emu/g by increasing the sintering temperature from $1,000$ to $1,200^\circ\text{C}$, using the sol-gel auto combustion method. Increasing the sintering temperature resulted in larger grains and more domain walls, which increased wall mobility leading to enhanced magnetization and higher M_s .

Spinel ferrite ceramics, such as $\text{Co}_{0.6}\text{Zn}_{0.4}\text{Fe}_{1.7}\text{Mn}_{0.3}\text{O}_4$ and $\text{Ni}_{0.6}\text{Zn}_{0.4}\text{Fe}_2\text{O}_4$, have been successfully synthesized using a solid-state combustion technique (Thawong *et al.*, 2019; Sonchaopri *et al.*, 2023). This method relies on a heat-releasing combustion reaction, in which a solid precursor is mixed with a fuel (e.g., urea, glycine, alanine, carbonylhydrazide, or citric acid) (Yotthuan *et al.*, 2020). The energy released from the fuel reaction can reduce the firing temperature

and dwell time while also producing ceramics with high purity, high density, and good electrical and magnetic properties (Kornphom *et al.*, 2016).

Therefore, this research focuses on investigation of the optimum firing temperatures of NZLAF ceramics using the solid-state combustion technique. The effects of firing temperatures on the phase structure, microstructure, and electrical and magnetic properties were also studied.

Materials and Method

$\text{Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4$ (NZLAF) ceramics were synthesized via the solid-state combustion technique using glycine as fuel. Raw materials of NiO (99.00%; Merck, Ger), ZnO (99.00%; Merck, Ger), Li_2O (99.00%; Aldrich, UK), $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (98.00%; Ajax, Aust), and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (98.00%; Kemaus, Aust) were weighed in stoichiometric proportions and mixed using a ball-milling method for 24 h, with ethyl alcohol as the grinding media. The suspensions were dried, and the powders were ground using an agate mortar and then sieved into a fine powder. The powders were mixed with glycine ($\text{C}_2\text{H}_5\text{NO}_2$; Kemaus, Aust) at a ratio of 1:2 in an agate mortar. The mixed powders were calcined at temperatures ranging from 850 – $1,050^\circ\text{C}$ for 2 h, with a heating rate of 5°C/min .

The calcined powders were then mixed with a 3% polyvinyl butyral (PVB) solution and ground in an agate mortar until the ethanol evaporated. The calcined powders were pressed into disks with a diameter of 10 mm and a thickness of 1 mm under a pressure of 80 MPa. The pellets were sintered between $1,150$ and $1,350^\circ\text{C}$ for 4 h, using a heating rate of 5°C/min in covered alumina crucibles.

The phase formation of the calcined powders and sintered ceramics was investigated using X-ray diffraction (XRD, Bruker, D2 Phaser) and the Rietveld refinement method. A Field Emission Scanning Electron Microscope (FESEM, Thermo Scientific Apreo S) was used to examine surface morphology. To prepare FESEM samples, the ceramics were polished with sandpaper and alumina abrasive, then thermally etched at a temperature 100°C below the sintering temperature for 20 min to reveal their grain boundaries. The particle and average grain size were determined using the mean linear intercept technique. The density of the sintered ceramics was measured using Archimedes method with distilled water as the medium.

For dielectric property measurements, the ceramics were coated with silver paste on

both sides and then fired at 550°C for 30 min. The dielectric properties of the samples were measured using an LCR analyzer (HP4284A, Hewlett-Packard, Palo Alto, CA, USA) at room temperature across a frequency range of 1 kHz to 1 MHz. The ferromagnetic properties were measured with a vibrating sample magnetometer (Versa Lab, Quantum Design) with an applied field of 10,000 Oe.

Results and Discussion

Figure 1 presents the X-ray diffraction patterns of NZLAF powders calcined at different temperatures, measured at room temperature in the 2θ range of 20° to 70° . The XRD results showed a spinel phase (JCPDS 008-0234) in all samples. A secondary phase of Fe_2O_3 (JCPDS 072-0469) was observed at the lowest calcination temperature (850°C). A pure spinel structure was obtained by calcining samples at temperatures between 900 and 1,050°C. The phase purity was determined by the relative quantities of the spinel and impurity phases, measured by the dominant XRD peak intensities of the spinel (I_{max}) and impurity ($I_{\text{Fe}_2\text{O}_3}$) phases. The phase purity was calculated using the following equation:

$$\% \text{purity phase} = \frac{I_{\text{max}}}{I_{\text{max}} + I_{\text{Fe}_2\text{O}_3}} \times 100 \quad (1)$$

It was observed that the purity phase of the powder calcined at 850°C was 98.1%, while a pure spinel phase was found in powders calcined above 900°C for 2 h. At lower temperatures,

the calcination process may not be sufficient to complete the reaction necessary to form the desired phase, resulting in the presence of unreacted or partially reacted materials, which can lead to the formation of a secondary phase (Singh *et al.*, 2009).

Figure 2(a-e) show the FESEM morphology and particle size distribution of NZLAF powders calcined between 850 and 1,050°C for 2 h. ImageJ software was used to calculate the particle size distributions. The powders exhibited a roughly spherical morphology with an agglomerated form. As the calcination temperature increased,

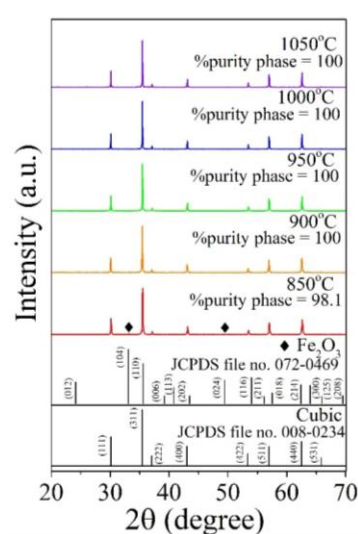


Figure 1. XRD patterns of NZLAF powders calcined at temperatures between 850°C and 1,050°C for 2 h, with 2θ values ranging from 20° to 70°

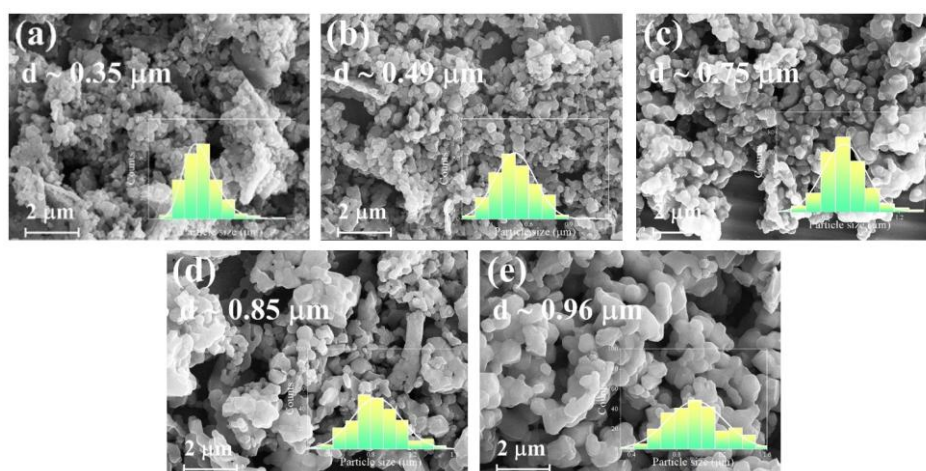


Figure 2. FESEM images and particle size distribution of NZLAF powders calcined at (a) 850, (b) 900, (c) 950, (d) 1,000, and (e) 1,050°C for 2 h

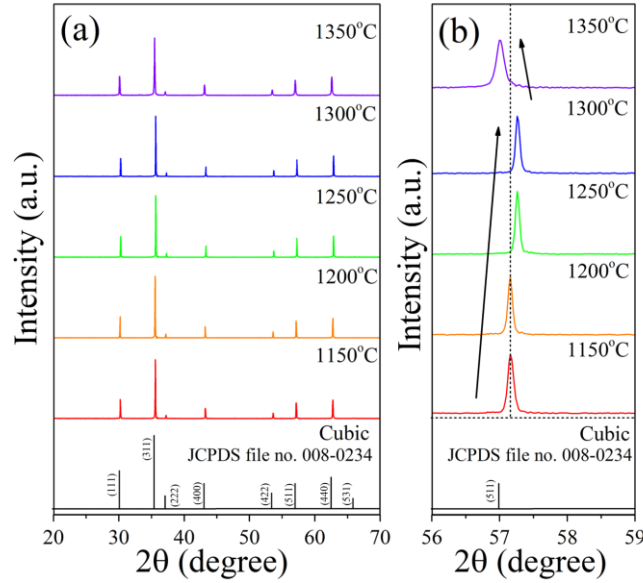


Figure 3. XRD patterns of NZLAF ceramics sintered in the range of 1,150°C and 1,350 °C for 4 h with 2θ from: (a) 20° to 70° and (b) 56° to 59°

the particle size distribution became broader, and the average particle size (d) of the calcined NZLAF powders increased from 0.35 to 0.96 μm , as shown in Figure 2(a-e). At higher temperatures, particles tend to stick together or agglomerate, resulting in larger overall particle sizes (Yin *et al.*, 2021).

The pure NZLAF powder calcined at 900°C was pressed into disks and sintered with temperatures of 1,150, 1,200, 1,250, 1,300, and 1,350°C for 4 h. X-ray diffraction patterns of the NZLAF sintered ceramics are shown in Figure 3. The XRD patterns exhibited a single spinel phase in all samples, and no secondary phase was detected in the XRD data (Figure 3(a)). To determine the phase formation, the XRD patterns of all samples in the 2θ range of 56°-59° were expanded and are shown in Figure 3(b). Normally, the cubic spinel phase (JCPDS file no. 008-0234) exhibits characteristic single peaks in the 2θ range of 20° to 70°. The phase formation of all specimens showed a single peak around 57°, indicating the cubic phase, as shown in Figure 3(b). In addition, the diffraction peak around 57° shifted to a higher angle as the sintering temperature increased from 1,150°C to 1,300°C, and then shifted to a lower angle at a sintering temperature of 1,350°C. This indicated that the unit cell size of the samples initially decreased and then increased.

To confirm the phase and evaluate the lattice parameters of NZLAF ceramics, Rietveld refinement was performed on all the ceramics using the FullProf

program to fit the XRD data (Figure 4(a-e)). The Crystallography Open Database (COD) was used for reference values of cell parameters, lattice constants, space groups, and atomic positions. A structural model based on $\text{NiZnFe}_2\text{O}_4$ in the cubic phase (C) and space group $\text{Fd}\bar{3}\text{m}$ was used as the starting model for refining the XRD data of the NZLAF ceramics. A Chebyshev polynomial function was used to refine the background, while the pseudo-Voigt function was used to fit the diffraction peaks. (Marques *et al.*, 2009; Yansen *et al.*, 2015). To verify the accuracy of the Rietveld refinement, various parameters were calculated, including the profile reliability (R_p), the weighed pattern reliability (R_{wp}), the expected residual (R_{exp}), and the goodness of fit (χ^2), as follows: (Young and Wiles, 1980)

$$R_{wp} = \left[\frac{\sum_i w_i |y_{oi} - y_{ci}|}{\sum_i w_i y_{oi}^2} \right]^{1/2} \quad (2)$$

$$R_p = \frac{\sum_i w_i |y_{oi} - y_{ci}|}{\sum_i y_{oi}} \quad (3)$$

$$R_{exp} = \left[\frac{n - p}{\sum_i w_i y_{oi}} \right]^{1/2} \quad (4)$$

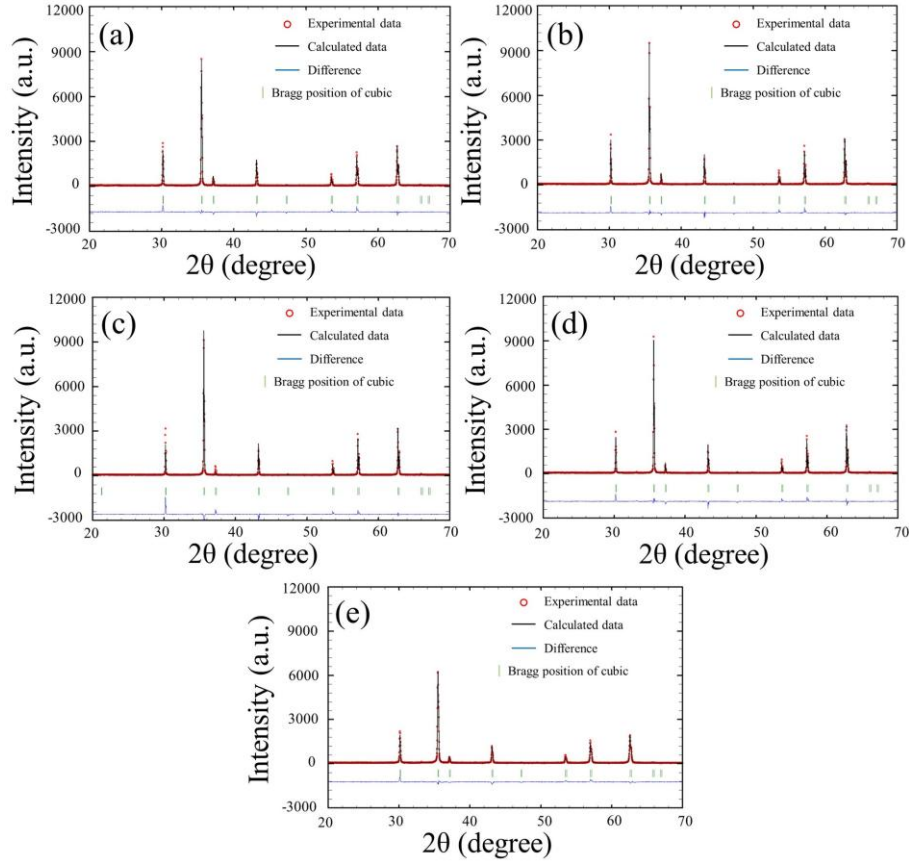


Figure 4. Rietveld refinement of XRD patterns for NZLAF ceramics sintered at (a) 1,150, (b) 1,200, (c) 1,250, (d) 1,300, and (e) 1,350°C for 4 h

$$\chi^2 = \left[\frac{R_{wp}}{R_{exp}} \right]^2 \quad (5)$$

where y_{oi} is the experimental intensities, y_{ci} is the calculated intensities, and $w_i = (1/y_{oi})$ is the weight of the experimental observations. All parameters obtained from the Rietveld refinement are listed in Table 1. $R_p \leq 22.0\%$, $R_{wp} \leq 20.2\%$, $R_{exp} \leq 15.6\%$ and $\chi^2 \leq 1.69$ were obtained from the fitting procedure, confirming good agreement between the experimental and calculated intensities for all of the NZLAF ceramics. The fitting results confirmed that all ceramics had a pure spinel structure with a cubic phase. The lattice parameter, a , and unit cell volume of the NZLAF ceramics decreased from 8.3803 to 8.3662 Å and from 588.47 to 585.54 Å³, respectively, as the sintering temperature increased from 1,150 to 1,300°C. At a sintering temperature of 1,350°C, the lattice parameter and unit cell volume increased to 8.3878 Å and 590.11 Å³, respectively. The increased

atomic mobility at higher sintering temperatures (up to 1,300°C) leads to greater atomic packing, causing a decrease in the unit cell volume as the crystal structure becomes denser (Low *et al.*, 2018). At the highest sintering temperature (1,350°C) though, the higher movement rates of atoms in the material, increasing defect formation, increasing the lattice parameter and the unit cell volume (Li, 1959).

Figure 5(a-e) display FESEM images of the surface morphology of NZLAF ceramics sintered at different temperatures after polishing and thermal etching. All images revealed a polygonal grain shape with a broad grain size distribution for all samples. As the sintering temperatures increased from 1,150 to 1,300°C, the average grain size (D) increased, with more distinct grain boundaries and reduced porosity. However, at 1,350°C, the grain surface had a layered appearance, and pores emerged along the grain boundaries. The average grain size increased from 1.75 to 8.50 μm as the sintering temperature increased, as seen in Figure 5(a-e). Higher sintering temperature increase

Table 1. Refined structural parameters for NZLAF ceramics sintered at various temperatures

Sintering temperature (°C)	Goodness of fit	Volume cell (Å ³)	Profile parameters	Atoms information				
				Label	x	y	z	Occ.
1,150	$\chi^2 = 1.59$ $R_p = 18.9\%$ $R_{wp} = 18.4\%$ $R_{exp} = 14.6\%$	588.47	a = 8.3803 Å b = 8.3803 Å c = 8.3803 Å u = 0.038262 v = -0.014358 w = 0.006738	Fe1	0.12500	0.12500	0.12500	0.72195
				Fe2	0.50000	0.50000	0.50000	1.19448
				Zn1	0.12500	0.12500	0.12500	0.11437
				Zn2	0.50000	0.50000	0.50000	0.36449
				Ni1	0.12500	0.12500	0.12500	0.09089
				Ni2	0.50000	0.50000	0.50000	0.37190
				O1	0.25000	0.25000	0.25000	3.86634
				Li1	0.12500	0.12500	0.12500	0.02570
				Al1	0.12500	0.12500	0.12500	0.02561
1,200	$\chi^2 = 1.69$ $R_p = 19.9\%$ $R_{wp} = 19.4\%$ $R_{exp} = 14.9\%$	586.45	a = 8.3707 Å b = 8.3707 Å c = 8.3707 Å u = 0.003831 v = 0.008950 w = 0.000591	Fe1	0.12500	0.12500	0.12500	0.75409
				Fe2	0.50000	0.50000	0.50000	1.25273
				Zn1	0.12500	0.12500	0.12500	0.11440
				Zn2	0.50000	0.50000	0.50000	0.36203
				Ni1	0.12500	0.12500	0.12500	0.09531
				Ni2	0.50000	0.50000	0.50000	0.38144
				O1	0.25000	0.25000	0.25000	3.90944
				Li1	0.12500	0.12500	0.12500	0.02498
				Al1	0.12500	0.12500	0.12500	0.02484
1,250	$\chi^2 = 1.55$ $R_p = 20.2\%$ $R_{wp} = 19.0\%$ $R_{exp} = 15.2\%$	586.05	a = 8.3687 Å b = 8.3687 Å c = 8.3687 Å u = 0.013804 v = -0.006465 w = 0.004470	Fe1	0.12500	0.12500	0.12500	0.72780
				Fe2	0.50000	0.50000	0.50000	1.18219
				Zn1	0.12500	0.12500	0.12500	0.11528
				Zn2	0.50000	0.50000	0.50000	0.36283
				Ni1	0.12500	0.12500	0.12500	0.10161
				Ni2	0.50000	0.50000	0.50000	0.38942
				O1	0.25000	0.25000	0.25000	3.86126
				Li1	0.12500	0.12500	0.12500	0.02543
				Al1	0.12500	0.12500	0.12500	0.02481
1,300	$\chi^2 = 1.68$ $R_p = 22.0\%$ $R_{wp} = 20.2\%$ $R_{exp} = 15.6\%$	585.54	a = 8.3662 Å b = 8.3662 Å c = 8.3662 Å u = 0.004975 v = 0.001199 w = 0.002816	Fe1	0.12500	0.12500	0.12500	0.75265
				Fe2	0.50000	0.50000	0.50000	1.24703
				Zn1	0.12500	0.12500	0.12500	0.11451
				Zn2	0.50000	0.50000	0.50000	0.36461
				Ni1	0.12500	0.12500	0.12500	0.09535
				Ni2	0.50000	0.50000	0.50000	0.38034
				O1	0.25000	0.25000	0.25000	3.95404
				Li1	0.12500	0.12500	0.12500	0.02660
				Al1	0.12500	0.12500	0.12500	0.02526
1,350	$\chi^2 = 1.26$ $R_p = 15.8\%$ $R_{wp} = 16.3\%$ $R_{exp} = 14.5\%$	590.11	a = 8.3877 Å b = 8.3877 Å c = 8.3877 Å u = 0.002571 v = 0.033575 w = 0.000033	Fe1	0.12500	0.12500	0.12500	0.77529
				Fe2	0.50000	0.50000	0.50000	1.24855
				Zn1	0.12500	0.12500	0.12500	0.11383
				Zn2	0.50000	0.50000	0.50000	0.36197
				Ni1	0.12500	0.12500	0.12500	0.09571
				Ni2	0.50000	0.50000	0.50000	0.38071
				O1	0.25000	0.25000	0.25000	4.08630
				Li1	0.12500	0.12500	0.12500	0.02560
				Al1	0.12500	0.12500	0.12500	0.02572

atomic diffusing, leading to grain growth as smaller grains merge with larger ones, and atoms move from regions of high curvature in smaller grains to lower curvature in larger grains, increasing the average grain size (Buatip *et al.*, 2018).

The experimental density (ρ_{ex}), theoretical density (ρ_{th}) and relative density ($R.D.$) of the NZLAF samples with different sintering temperatures are listed in Table 2. The $R.D.$ and ρ_{th} were calculated from the following equations (Srinivas *et al.*, 2016; Sun *et al.*, 2018; Ni *et al.*, 2020):

$$R.D. = \frac{\rho_{ex}}{\rho_{th}} \quad (6)$$

$$\rho_{th} = \frac{8M}{N_A V_{cell}} \quad (7)$$

where M represents molecular weight, N_A is Avogadro's number, and V_{cell} is the unit cell volume. The ρ_{ex} , ρ_{th} and $R.D.$ of the ceramic increased from 5.09 to 5.31 g/cm³, 5.29 to 5.32 g/cm³, and 96.26%

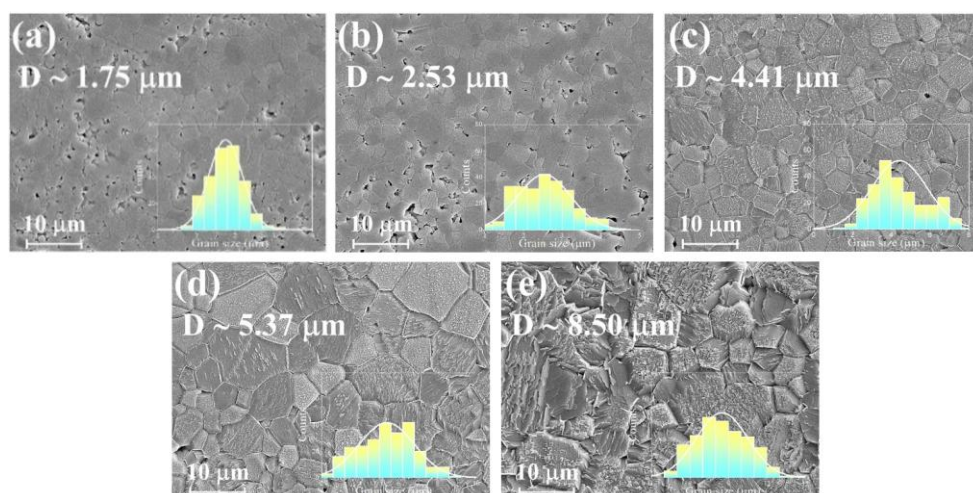


Figure 5. FESEM images and grain size distribution of NZLAF ceramics sintered at (a) 1,150, (b) 1,200, (c) 1,250, (d) 1,300, and (e) 1,350°C for 4 h

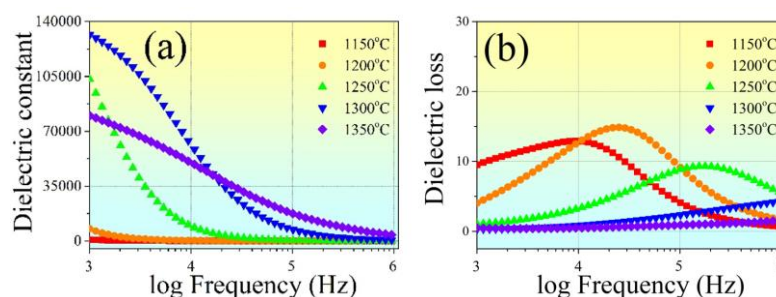


Figure 6. Room-temperature frequency dependence of (a) dielectric constant and (b) dielectric loss of NZLAF ceramics sintered at various temperatures

Table 2. Experimental density, theoretical density, relative density, dielectric and ferromagnetic properties of NZLAF ceramics

Sintering temperatures (°C)	Experimental density (g/cm ³)	Theoretical density (g/cm ³)	Relative density (%)	Dielectric constant (ϵ) at frequency 1 kHz	Dielectric loss ($\tan\delta$) at frequency 1 kHz	M_s (emu/g)	M_r (emu/g)	H_c (Oe)
1150	5.09	5.29	96.26	2169	9.48	77.18	10.81	40.24
1200	5.22	5.31	96.88	8793	3.82	78.91	6.16	6.97
1250	5.25	5.31	97.48	103627	1.29	79.18	1.25	9.70
1300	5.31	5.32	98.61	131000	0.54	82.40	0.05	3.65
1350	5.12	5.28	95.05	78761	0.69	79.18	0.77	20.73

to 98.61%, respectively, when the sintering temperature increased from 1,150 to 1,300°C, and then decreased. The changes in the density are attributed to changes in porosity and the formation of the liquid phase during sintering (Yothuan *et al.*, 2015). The decrease in density at higher sintering temperatures was likely due to the evaporation of alkaline elements, which created pores in the ceramics, leading to increased porosity in the ceramics (Mathrmool *et al.*, 2017). The *R.D.* of

NZLAF samples synthesized using the solid-state combustion method was higher than those prepared using the sol-gel auto-combustion technique (89.81%) (Ni *et al.*, 2020).

Figures 6(a-b) show the log frequency dependence of the dielectric constant (ϵ) and dielectric loss ($\tan\delta$) of NZLAF ceramics sintered with various temperatures from 1,150 to 1,350°C, measured at room temperature. The dielectric constant (ϵ) decreased with increasing frequency,

as shown in Figure 6(a). As the sintering temperature increased from 1,150 to 1,300°C, the ε at 1 kHz increased while the dielectric loss ($\tan\delta$) decreased from 9.48 to 0.54, as seen in Table 2. The $\tan\delta$ peak shifted to higher frequencies

conductivity resulting from the porous structure at the grain boundaries and the reduced density (Wu *et al.*, 2012; Chootin and Bongkarn, 2017; Kornphom *et al.*, 2018). The dielectric loss peak shifted to higher frequencies with increasing

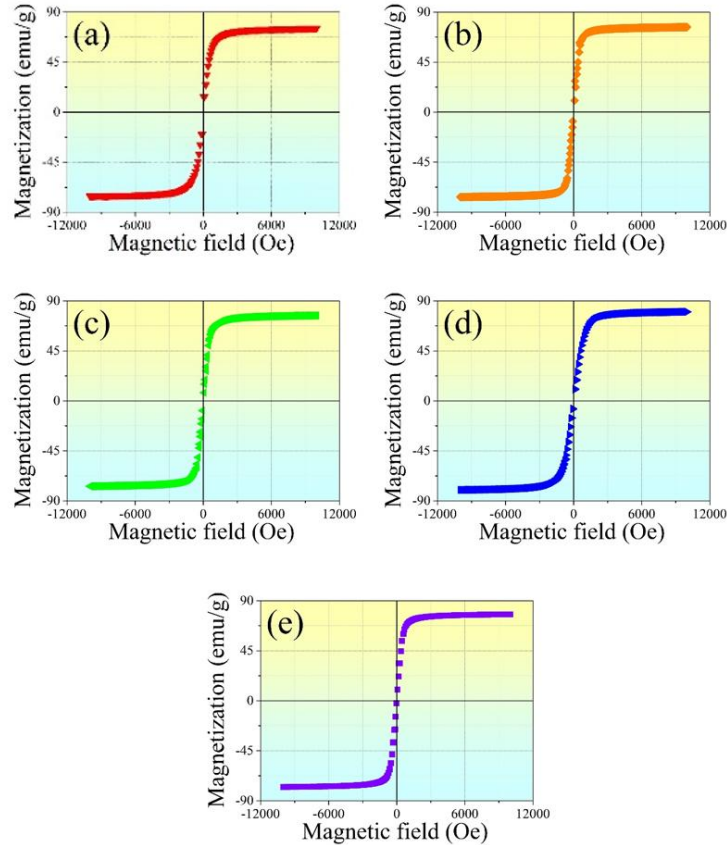


Figure 7. Ferromagnetic (M-H) hysteresis loops of NZLAF ceramics sintered at the temperatures of (a) 1,150, (b) 1,200, (c) 1,250, (d) 1,300, and (e) 1,350 °C for 4

as the sintering temperature increased. The ε decreased with increasing frequency because the polarization mechanism within the material cannot respond quickly to rapidly changing electric fields (Thawong *et al.*, 2019; Bansal *et al.*, 2021). This variation in dielectric behavior corresponds to the Maxwell-Wagner (interfacial polarization) type of heterogeneous systems (Thawong *et al.*, 2019). The increase in dielectric constant with rising sintering temperatures was consistent with the increase in grain size and the reduction in porosity (Mishra and Kumar, 2012). The increase in grain size leads to an increase in domain size, which further enhances polarization (Cao and Randall, 1996; Mishra and Kumar, 2012). The decrease in dielectric constant at the highest sintering temperature was due to the increased electrical

sintering temperature due to the relaxation phenomenon. This phenomenon is caused by the increased transition of $\text{Fe}^{3+} \leftrightarrow \text{Fe}^{2+}$ at B-sites (Ni *et al.*, 2020), where the increasing temperature enhances electron hopping between Fe^{2+} and Fe^{3+} , leading to the shift in the dielectric loss peak (Ni *et al.*, 2020; Bansal *et al.*, 2021).

Figure 7 (a-e) show the room temperature M-H loops of NZLAF ceramics sintered in the temperature range of 1,150-1,350°C, using a magnetic field of 10,000 Oe. The M-H loops exhibited a slim and saturated hysteresis loop, which is a typical soft magnetic characteristic and is consistent with previous research (Ni *et al.*, 2020). The saturation magnetization (M_s), remanence magnetization (M_r), and coercive field (H_c) of the NZLAF ceramics are shown in Table 2. It was found

that when the sintering temperature increased from 1,150 to 1,300°C, M_s increased from 77.18 to 82.40 emu/g, while M_r decreased from 10.81 to 0.05 emu/g. H_c tended to decrease from 40.24 to 3.65 Oe with increasing sintering temperatures. In contrast, M_s decreased while M_r and H_c increased when the sintering temperature was increased to 1,350°C. From the experimental results above, it can be seen that as the sintering temperatures increased from 1,150 to 1,300°C, the average grain size and density also increased. This results in a larger magnetic moment per unit volume, leading to an increase in the M_s value. However, the decrease in M_s at the sintering temperature of 1,350°C was due to the decrease in density (Rahman *et al.*, 2014; Nasrin *et al.*, 2019). Compared to the sol-gel auto-combustion method, the M_s value of NZLAF ceramics obtained in this study is lower. However, there was a significant decrease in M_r and H_c from 14 to 0.05 emu/g and 62.93 to 3.65 Oe, respectively, indicating desirable soft magnetic characteristics (Ni *et al.*, 2020).

Conclusions

In summary, the effect of the firing temperatures on the phase formation, microstructure, dielectric, and ferromagnetic properties of lead-free $\text{Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4$ (NZLAF) ceramics prepared via the solid-state combustion technique was investigated. The single spinel phase was achieved at a calcined temperature of 900°C for 2 h in air. The XRD of all NZLAF ceramics exhibited a cubic spinel phase, and lattice parameters decreased with an increasing sintering temperature up to 1,300 °C, which was confirmed by the Rietveld refinement technique. The optimal experimental density, relative density, dielectric properties and ferromagnetic properties were obtained from the sample sintered at 1,300°C for 4 h. The solid-state combustion technique can produce good-quality soft ferromagnetic ceramics with simpler preparation steps and a shorter dwell time.

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