

ENHANCED FERROMAGNETIC PROPERTIES OF SPINEL $\text{Ni}_{0.25}\text{Mn}_{0.25}\text{Cu}_{0.15}\text{Zn}_{0.35}\text{Fe}_2\text{O}_4$ CERAMICS SYNTHESIZED VIA THE SOLID-STATE COMBUSTION

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Abstract

$\text{Ni}_{0.25}\text{Mn}_{0.25}\text{Cu}_{0.15}\text{Zn}_{0.35}\text{Fe}_2\text{O}_4$ (NMCZF) soft ferrite ceramics were successfully synthesized using the solid-state combustion. The samples were calcined at temperatures ranging from 900 to 1,000°C for 2 h and sintered between 1,100 and 1,250°C for 5 h. A pure cubic spinel-based ferrite structure was observed in the NMCZF powder calcined at 1,000°C for 2 h. The phase structure, microstructure, dielectric, and ferromagnetic properties of the NMCZF ceramics were investigated. Field emission scanning electron microscopy (FESEM) showed that the average grain size increased with increasing sintering temperature. The measured density of the NMCZF ceramics increased from 5.01 g/cm³ to 5.14 g/cm³ as the sintering temperature increased from 1,100°C to 1,150°C but slightly decreased to 5.09 g/cm³ and 5.07 g/cm³ at temperatures of 1,200°C and 1,250°C, respectively. The frequency-dependent dielectric properties were measured across the 100 Hz to 1 MHz frequency range at room temperature. Results showed that the dielectric constant and dielectric loss were frequency-dependent for all samples. The dielectric constant (ϵ) at 100 Hz increased from 354 to 860 with higher sintering temperatures. Under a magnetic field of 15 kOe, the NMCZF ceramics exhibited a typical ferromagnetic response with saturated magnetization. The maximum saturation magnetization (M_s) of 95.2 emu/g and the minimum coercive field (H_c) of 5.3 Oe was observed at a sintering temperature of 1,150°C.

Keywords: Combustion; Dielectric; Ferromagnetic; NMCZF; Spinel

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Introduction

Magnetic materials are necessary in the development of computers and other electronic devices, particularly in applications such as Hard Disk Drives (HDD). Among these materials, ferrites are a type of ceramic magnetic material known for their good ferromagnetic properties and high electrical resistance. These properties make ferrites suitable for many industries such as computing, electronics, and medicine. Ferrites are found in many products, including capacitors, storage equipment, speakers, telephones, televisions, and motors. Ferrite ceramics with a spinel structure are commonly represented by the formula AB_2O_4 with examples including $MnFe_2O_4$ (MFO) (Penchal Reddy *et al.*, 2015), $ZnFe_2O_4$ (ZFO) (Mekap *et al.*, 2013), $CoFe_2O_4$ (CFO) (Cernea *et al.*, 2016) or $NiZnFe_2O_4$ (NZF) (de Biasi and Carvalho, 2014). These materials exhibit high physical and chemical stability (Deka and Joy, 2008).

Among the different types of cubic spinel ferrites, nickel zinc ferrite ($NiZnFe_2O_4$: NZF) is known for its good electrical and magnetic properties. This due to the NZF ceramics having high saturation magnetization (M_s) and low coercivity (H_c). Bhise *et al.* (1991) reported on the effects of Mn substitution in NZF using conventional ceramic processing methods. Sattar *et al.* (2005) investigated how Mn ion substitution affected the functional characteristics of NZF prepared via a conventional solid-state reaction method, while Shirsath *et al.* (2010) reviewed its impact on the electromagnetic properties of $Ni_{0.2}Mn_{0.3}Zn_{0.5}Fe_2O_4$ (NMZF). They reported that substituting Mn ions significantly affected the physical properties of NZF ceramics, increasing both the lattice parameters and average grain size. This results in higher saturation magnetization (M_s) and initial permeability, although it also causes an increase in coercivity (H_c).

Additionally, Cu doping has been shown to tune magnetic properties, which could be used for applications in multilayer chip inductors and electromagnetic interference filters (Krishnaveni *et al.*, 2006; Hadi *et al.*, 2021). Kothapally *et al.* (2023) investigated the effect of Co and Cu substitution in NZF films prepared by the sol-gel method, finding that Cu doping reduced the coercivity (H_c). Ahad and Akther Hossain, (2024) prepared NZF ferrite ceramics with partial Mn substituting for Ni, described by the chemical formula $Ni_{0.25}Mn_{0.25}Cu_{0.15}Zn_{0.35}Fe_2O_4$ (NMCZF), using the sol-gel auto combustion technique. This research found that NMCZF ceramics exhibited a notably high initial magnetic

permeability ($\mu_i' = 315$), while both coercivity and Néel temperature decreased while the grain size increased with Mn substitution.

Although NMZF and NMCZF prepared by the sol-gel method has been extensively investigated (Sun *et al.*, 2008; Sukata and Mezinskis, 2012; Huang *et al.*, 2015), the synthesis of ferrite materials continues to be explored using different preparation methods and firing temperatures, leading to a diverse range of magnetic properties. The solid-state combustion technique is an interesting method due to its ability to produce high density materials. It produces ultra-fine powders with excellent properties and a narrow particle size distribution (Hwang *et al.*, 2004) and has not yet been reported for the preparation of NMCZF ceramic ferrites.

This study investigated the effect of firing temperatures on $Ni_{0.25}Mn_{0.25}Cu_{0.15}Zn_{0.35}Fe_2O_4$ (NMCZF) ferrite ceramics. The samples were calcined in a temperature range of 900 to 1,000°C using the solid-state combustion method with glycine as the fuel. After calcination, the ceramics were sintered between 1,100 to 1,250°C for 5 h. The phase structure, microstructure, electrical and ferromagnetic properties of the ceramic were investigated.

Materials and Method

NMCZF ceramics were synthesized by the solid-state combustion technique using glycine as a fuel. Raw materials of NiO (99.9%, Merck, Germany), $MnCO_3$ (99.9%, Aldrich, USA), CuO (98.0%, Fluka, USA), $Zn(NO_3)_2 \cdot 6H_2O$ (98.0%, Ajax, Australia) and $Fe(NO_3)_3 \cdot 9H_2O$ (98.0%, Kemaus, Australia) were weighed according to their stoichiometric ratios. The powders were combined using ball milling with alcohol for 24 h, then dried at approximately 120°C and mixed with glycine in a 1:1.11 molar ratio. The powders were calcined between 900 and 1,000 °C for 2 h with a heating rate of 5°C/min. The calcined powders were ball milled with Polyvinyl Butyral (PVB) as a binder to improve the flexibility and integrity of the green pellets for 12 h, then dried at 120°C. The powders were pressed into pellets at a pressure of 80 MPa with a diameter of approximately 15 mm and were 1.2 to 1.4 mm thick. The pellets were sintered between 1,100°C and 1,250 °C for 5 h with a heating rate of 5°C/min.

The phase structure of the NMCZF samples was detected using X-ray diffraction (XRD, Bruker,

D2 Phaser). The phase composition of each sample was quantified via Rietveld refinement. The microstructure was analyzed using scanning electron microscopy (SEM, Thermo Fisher Scientific, Apreo S) after the ceramic surfaces were thermally etched at a temperature 100°C lower than the sintering temperature for 20 min. The grain size was measured by the Scopephoto program with a comparison of the real scale from the SEM image. The average grain size was determined by the linear intersection method. The density of the sintered ceramics was measured using Archimedes' method. Dielectric properties were measured at room temperature across a 100 Hz to 1MHz frequency range using an LCR meter (Hewlett-Packard, HP4284A). Magnetic properties were measured under an applied magnetic field of 15,000 Oe using a vibrating sample magnetometer (Versa Lab, Quantum Design).

Results and Discussion

Figure 1 shows the XRD patterns of NMCZF powders calcined at different temperatures ranging between 900 and 1,000 °C for 2 h, over the 2θ range of 10°-70°. At calcination temperatures between 900°C and 975°C, X-ray diffraction patterns displayed peaks corresponding to Fe₂O₃ impurities (♦) identified using JCPDS No. 01-1053. As the calcination temperature increased to 1,000°C, the NMCZF powders formed a pure cubic spinel phase (JCPDS No. 08-0234). Increased calcination temperatures resulted in higher purity of the spinel phase, as illustrated in Figure 1. The purity percentage of the spinel phase was calculated using the following equation:

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

where I_{NMCZF} is the intensity of the main perovskite peak, and $I_{\text{Fe}_2\text{O}_3}$ is the intensity of the main Fe₂O₃

impurity peak. The results showed that as the calcination temperature increased from 900 to 1,000°C, the purity of the NMCZF phase rose from 97.4% to 100%. Therefore, the optimal conditions for preparing pure NMCZF powder are a calcination temperature of 1,000°C for 2 h with a heating rate of 5 °C/min. In ferrites, the vibrations of cations bonded to oxygen are detected as infrared (IR) absorption bands in the Fourier-transform infrared (FTIR) spectra, which can confirm the spinel structure and identify different functional groups within the sample. Figure 2 shows the room temperature FTIR spectra for NMCZF powders

calcined at temperatures between 900 and 1,000°C for 2 h, over a spectral range of 400-1,500 cm⁻¹. The results revealed two distinct absorption bands below 1,000 cm⁻¹, characteristic of spinel ferrites, and a C-O stretching band above 1,000 cm⁻¹. The FTIR spectra displayed fundamental metal-oxygen (M-O) absorption bands below 600 cm⁻¹,

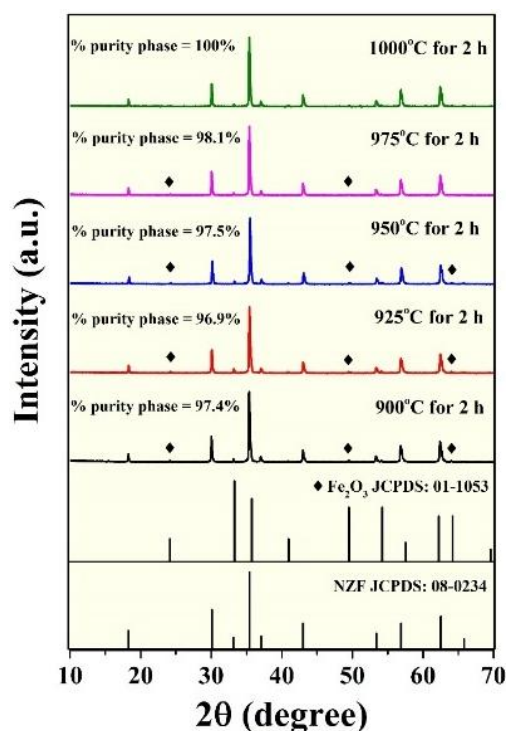


Figure 1. The NZF cubic spinel (JCPDS 08-0234) and Fe₂O₃ (JCPDS 01-1053) phases with the X-ray diffraction patterns of NMCZF powders with various calcination temperatures between 900 and 1,000 °C for 2 h in the 2θ range of 10°-70°

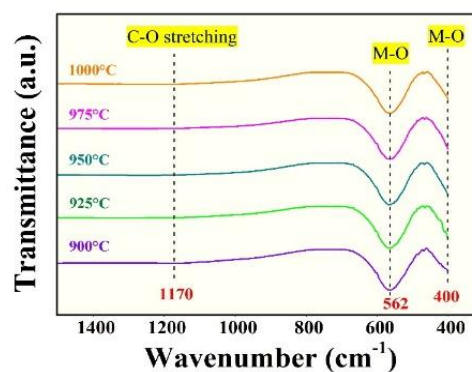


Figure 2. FTIR spectra of NMCZF powders with various calcination temperatures between 900°C and 1,000°C for 2 h

indicating the formation of the spinel ferrite structure with two sublattices, tetrahedral (A-sites voids) and octahedral (B-sites voids), which corresponds to $M \leftrightarrow O$ stretching vibrations of the spinel (Waldron, 1955). The two M-O absorption bands were observed at 400 cm^{-1} and 562 cm^{-1} . Waldron assigned the high-frequency band to the intrinsic M-O vibrations of the tetrahedral complexes and the low-frequency band to the M-O vibrations of octahedral complexes (Waldron, 1955). The vibration mode of the tetrahedral complex is normally higher than that of the octahedral complex, which is associated with the shorter bond length of oxygen-metal ions in the tetrahedral site than the octahedral site. This result matches well with XRD data, confirming the formation of spinel NMCZF powder. The presence of C-O stretching bands in the $1,000$ to $1,300\text{ cm}^{-1}$ region was also observed and is typically associated with oxygen-containing functional groups such as carboxylic acids, esters, and ethers.

The NMCZF powder with the highest purity, produced at a calcination temperature of $1,000^\circ\text{C}$ for 2 h, was pressed into pellets and sintered at $1,100^\circ\text{C}$, $1,150^\circ\text{C}$, $1,200^\circ\text{C}$ and $1,250^\circ\text{C}$ for 5 h. The phase composition of these sintered samples was characterized using X-ray diffraction. The NMCZF ceramics exhibited a pure cubic spinel structure with no secondary phases, matching the reference standard JCPDS no. 08-0234 (Figure 3). All compositions exhibited single peaks with no splitting, indicating a cubic spinel structure for all sintering temperatures. The room temperature FTIR spectra of NMCZF ceramics in the 400 - $1,500\text{ cm}^{-1}$ range are shown in Figure 4.

The scanning electron micrographs of NMCZF powders with their particle size distributions for various calcination temperatures are presented in Figure 5(a)-(e). The microstructure of the NMCZF powders were agglomerated and had a polyhedron shape. The particle size distributions of the NMCZF powders became narrower with increasing calcination temperatures. The average particle size of the NMCZF powders decreased from 0.45 to $0.29\text{ }\mu\text{m}$ as the calcination temperature increased from 900°C to $1,000^\circ\text{C}$, as shown in Table 1. As the firing temperature increased, the mixed powders decompose and release gases (e.g., CO_2), leading to a reduction in particle size, similar to the trend observed in $\text{SrFe}_{12}\text{O}_{19}$ and $\text{BaFe}_{12}\text{O}_{19}$ hexaferrite powders (Hlosta *et al.*, 2024).

The FTIR spectra detected two strong M-O absorption bands at 400 cm^{-1} and 551 cm^{-1} for all samples. The absorption bands below 600 cm^{-1} verify the XRD results, indicating the formation of the cubic spinel phase. A structural model based on $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$, featuring a cubic spinel phase with

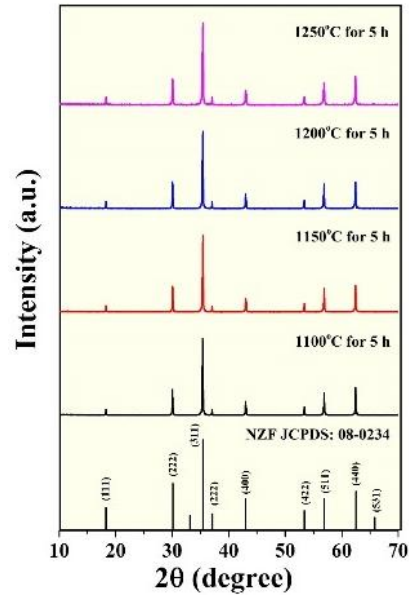


Figure 3. The NZF cubic spinel (JCPDS 08-0234) and X-ray diffraction patterns of NMCZF ceramics synthesized with sintering temperatures of $1,100^\circ\text{C}$, $1,150^\circ\text{C}$, $1,200^\circ\text{C}$, and $1,250^\circ\text{C}$ for 5 h in the 2θ range of 10° - 70°

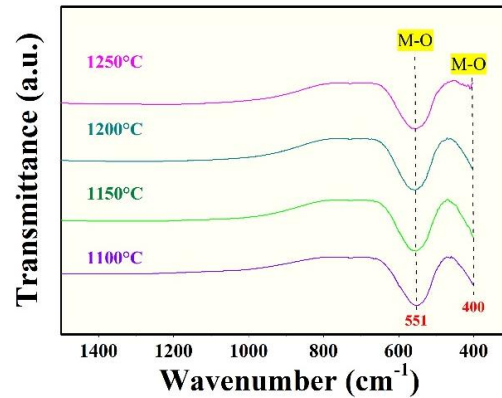


Figure 4. FTIR spectra of NMCZF ceramics with different sintering temperatures

Table 1. The average particle size of NMCZF powders

Calcination temperatures	Average particle size (μm)
900°C	0.45
925°C	0.40
950°C	0.37
975°C	0.36
$1,000^\circ\text{C}$	0.29

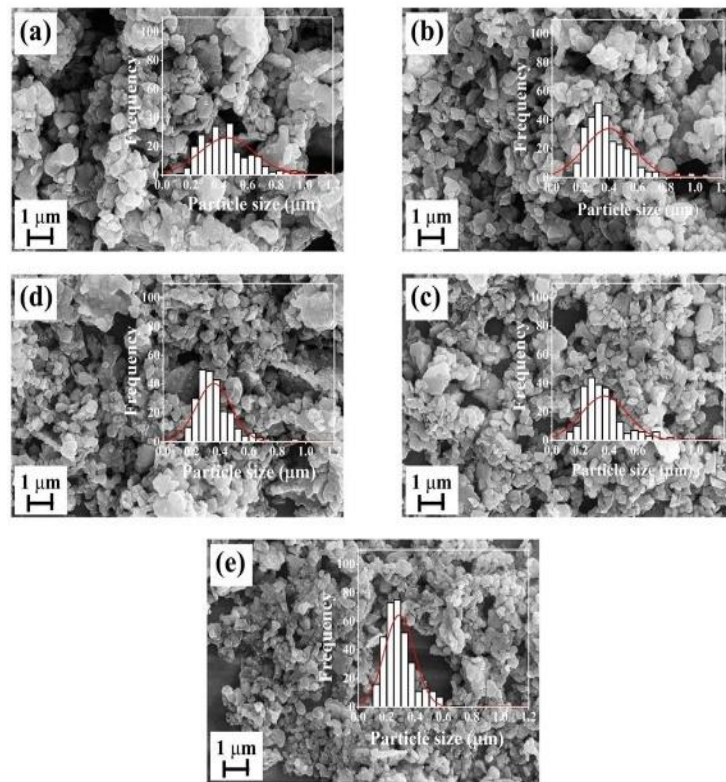


Figure 5. The scanning electron micrographs of NMCZF powders and particle size distributions with calcination temperatures of: (a) 900 °C, (b) 925 °C, (c) 950 °C, (d) 975 °C, and (e) 1,000 °C for 2 h

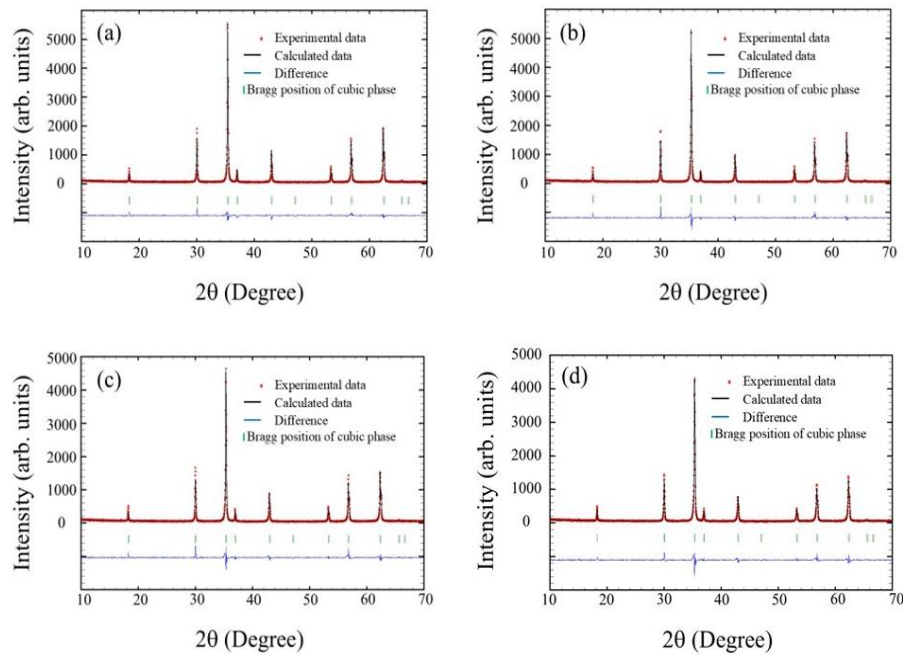


Figure 6. Final plots of the Rietveld refinement, showing the experimental (dots) and calculated (line) intensities of NMCZF ceramics in the range 10–70° with sintering temperatures of (a) 1,100 °C, (b) 1,150 °C, (c) 1,200 °C, and (d) 1,250 °C

Table 2. Goodness-of-fit parameters, lattice parameters, and atomic information for the NMCZF ceramics

Temperature	Goodness of Fit	Phase	Profile parameters		Atomic information				
					Label	x	y	z	Occ.
1,100°C	$\chi^2 = 1.643$	Fd3m	a	8.41228 Å	Fe1	0.125	0.125	0.125	0.46020
	R _p = 11.8 %		u	0.080252	Fe2	0.5	0.5	0.5	0.99263
	R _{wp} = 13.5 %		v	-0.057077	Ni1	0.125	0.125	0.125	0.35510
	R _{exp} = 10.6 %		w	0.018347	Ni2	0.5	0.5	0.5	0.59153
				Zn1	0.125	0.125	0.125	0.40983	
				Zn2	0.5	0.5	0.5	0.98183	
				Mn1	0.125	0.125	0.125	0.25361	
				Mn2	0.5	0.5	0.5	0.26567	
				Cu1	0.125	0.125	0.125	0.04132	
				Cu2	0.5	0.5	0.5	0.21207	
		O1	0.25	0.25	0.25	6.14378			
1,150°C	$\chi^2 = 2.085$	Fd3m	a	8.41758 Å	Fe1	0.125	0.125	0.125	0.46020
	R _p = 14.2 %		u	0.077068	Fe2	0.5	0.5	0.5	0.99263
	R _{wp} = 15.5 %		v	-0.052949	Ni1	0.125	0.125	0.125	0.35510
	R _{exp} = 10.7 %		w	0.018278	Ni2	0.5	0.5	0.5	0.59153
				Zn1	0.125	0.125	0.125	0.40983	
				Zn2	0.5	0.5	0.5	0.98183	
				Mn1	0.125	0.125	0.125	0.25361	
				Mn2	0.5	0.5	0.5	0.26567	
				Cu1	0.125	0.125	0.125	0.04132	
				Cu2	0.5	0.5	0.5	0.21207	
		O1	0.25	0.25	0.25	6.27659			
1,200°C	$\chi^2 = 2.023$	Fd3m	a	8.41950 Å	Fe1	0.125	0.125	0.125	0.46020
	R _p = 15.6 %		u	0.072954	Fe2	0.5	0.5	0.5	0.99263
	R _{wp} = 16.6 %		v	-0.045171	Ni1	0.125	0.125	0.125	0.35510
	R _{exp} = 11.7 %		w	0.017741	Ni2	0.5	0.5	0.5	0.59153
				Zn1	0.125	0.125	0.125	0.40983	
				Zn2	0.5	0.5	0.5	0.98183	
				Mn1	0.125	0.125	0.125	0.25361	
				Mn2	0.5	0.5	0.5	0.26567	
				Cu1	0.125	0.125	0.125	0.04132	
				Cu2	0.5	0.5	0.5	0.21207	
		O1	0.25	0.25	0.25	6.36358			
1,250°C	$\chi^2 = 1.804$	Fd3m	a	8.42134 Å	Fe1	0.125	0.125	0.125	0.45773
	R _p = 15.6 %		u	0.025705	Fe2	0.5	0.5	0.5	0.98981
	R _{wp} = 16.5 %		v	0.026965	Ni1	0.125	0.125	0.125	0.35510
	R _{exp} = 12.3 %		w	0.004902	Ni2	0.5	0.5	0.5	0.59153
				Zn1	0.125	0.125	0.125	0.40983	
				Zn2	0.5	0.5	0.5	0.98183	
				Mn1	0.125	0.125	0.125	0.25353	
				Mn2	0.5	0.5	0.5	0.26819	
				Cu1	0.125	0.125	0.125	0.01744	
				Cu2	0.5	0.5	0.5	0.19315	
		O1	0.25	0.25	0.25	6.60522			

an Fd3m space group, was used to refine the XRD data of the NMCZF ceramics with the Fullprof program (Figure 6). The initial lattice parameters (a, b, c), bond angles (α , β , γ), atomic information, and occupancy rates were taken from the Crystallography Open Database (COD). A pseudo-Voigt function was used to fit the XRD peaks, and a Chebyshev polynomial function was used for the background. The refinement quality was evaluated by the parameters: predicted residual (R_{exp}), profile reliability (R_p), weighted pattern reliability (R_{wp}), and goodness of fit (χ^2). Fitting parameters of $R_p \leq 15.6\%$, $R_{wp} \leq 16.6\%$, $R_{exp} \leq 12.3\%$, and $\chi^2 \leq 2.08$ (Table 2) were found in this study and indicated that the observed XRD patterns

corresponded well with the computed XRD patterns for all ceramics. The lattice parameters slightly increased from 8.4123 Å to 8.4213 Å when the sintering temperature increased, as shown in Table 2.

The scanning electron micrographs of the thermally etched NMCZF ceramics and their grain size distributions for various sintering temperatures are presented in Figure 7(a)-(d). The results showed that the grains of the NMCZF ceramics had a polyhedral grain shape and exhibited random growth. At the sintering temperature of 1,100°C, it was observed that the surface of the ceramics exhibited a significant amount of porosity, and grain boundaries were unclear (Figure 7 (a)).

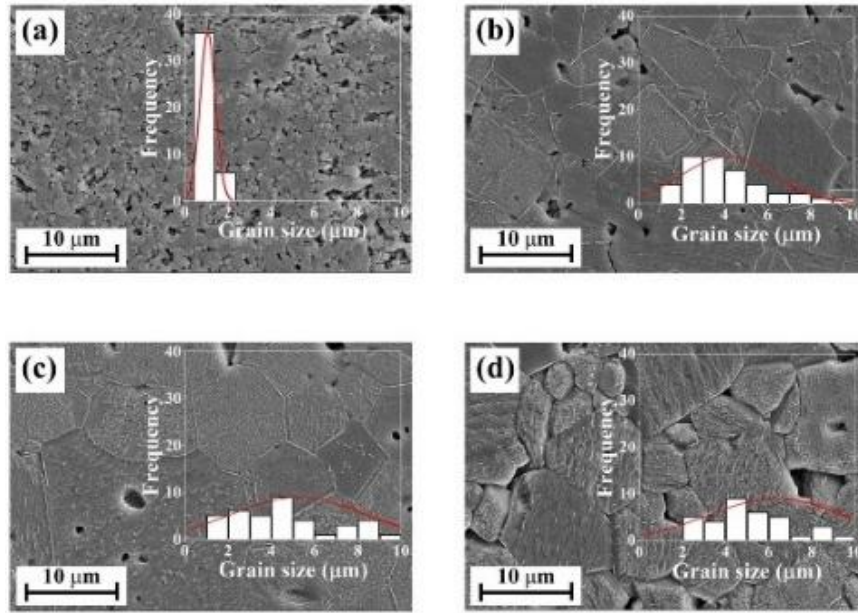


Figure 7. The scanning electron micrographs and grain size distributions of the NMCZF ceramics with sintering temperatures of (a) 1,100°C, (b) 1,150°C, (c) 1,200°C, and (d) 1,250°C

Table 3. The average grain size, density, dielectric, and ferromagnetic properties of NMCZF ceramics with different sintering temperatures

Sintering temperatures	D (μm)	Density (g/cm ³)	Dielectric constant (ε) at 10 ² Hz	Maximum dielectric loss (tanδ)	Frequency at tanδ (Hz)	M _r (emu/g)	M _s (emu/g)	H _c (Oe)	K
1,100°C	1.0	5.01	354	9.3	1.0×10 ²	1.9	72.9	19.0	692
1,150°C	3.6	5.14	571	9.5	4.5×10 ²	0.5	95.2	5.3	252
1,200°C	5.6	5.09	800	7.6	7.3×10 ²	0.8	71.5	8.9	318
1,250°C	6.4	5.07	860	7.6	8.0×10 ²	2.2	70.1	23.1	809

As the sintering temperature increased (as shown in Figure 7(b)-(d)), the ceramic's porosity decreased, the grains became significantly larger, and the grain boundaries became clearer. The average grain size (D) increased from 1.0 μm to 6.4 μm as the sintering temperature increased, as shown in Table 3. This indicates that higher sintering temperatures lead to significant grain growth, a common phenomenon due to enhanced atomic diffusion at elevated temperatures (Shen *et al.*, 2009). In general, the evaporation of Na²⁺ cations in NZF ceramics occurs during the high-temperature sintering process. This results in oxygen vacancies and promotes the formation of a more liquid phase, facilitating mass transfer and enhancing grain growth (He and Ma, 2000).

The dielectric constant (ε) and dielectric loss (δ) for the NMCZF ceramics produced with sintering temperatures of 1,100°C, 1,150°C, 1,200°C, and 1,250°C for 5 h were measured. The room temperature measurements were made

in the 100 Hz to 1 MHz frequency range, as shown in Figure 8. As shown in Figure 8(a), an increase in sintering temperature revealed a frequency dependent dielectric constant. The dielectric constant decreased with increased frequency, indicating the normal behavior of ferrites with a spinel structure. The decrease in the dielectric constant suggests an increase in Fe²⁺ ions, which results from electron exchange between Fe²⁺ and Fe³⁺ at the grain boundaries, leading to an increase in polarization (Ahmed *et al.*, 2013). The dielectric constant (ε) at 100 Hz increased from 354 to 860 when the sintering temperature increased, as shown in Table 3. Depending on the frequency, the maximum dielectric loss (tanδ) ranged between 7.6 and 9.5. The dielectric loss (Figure 8(b)) had relaxation peaks in all samples. These relaxation peaks can indicate a relatively high mobility of charge carriers (Singh *et al.*, 2011). Higher sintering temperatures improved the densification of the material, reduced the porosity, and increased

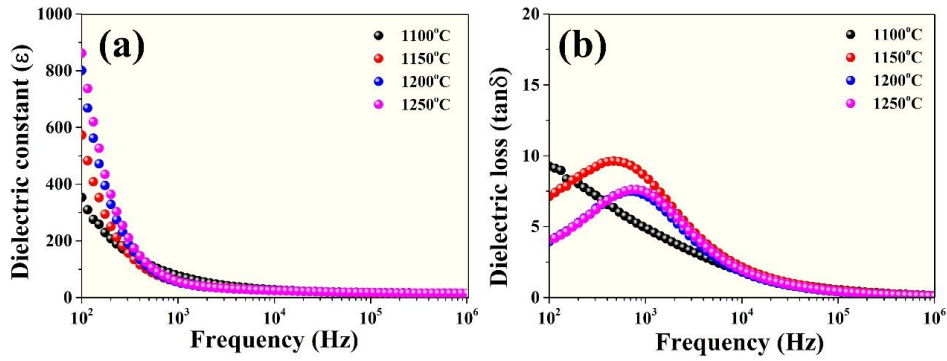


Figure 8. The dielectric constant (ϵ) and dielectric loss (δ) of NMCZF ceramics with sintering temperatures of 1,100 °C, 1,150 °C, 1,200 °C, and 1,250 °C for 5 h measured at room temperature in the frequency range of 100 Hz to 1 MHz

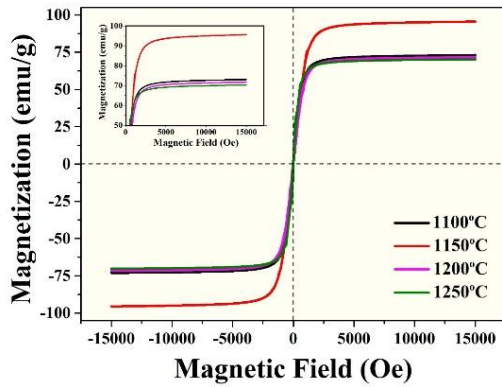


Figure 9. (a) M–H and magnified M–H curves of NMCZF ceramics produced with sintering temperatures between 1,100 °C and 1,250 °C for 5 h measured in a magnetic field of 15 kOe

the grain size. This resulted in fewer defects, less charge accumulation, and higher electrical conductivity (Nishkala *et al.*, 2024).

The magnetic properties of NMCZF ceramics synthesized with various sintering temperatures are shown in Figure 9(a). All samples exhibited saturated M–H loops, indicating that the NMCZF ceramics displayed ferromagnetic behavior. The magnetization (M_s) increased from 72.9 emu/g to 95.2 emu/g as the sintering temperature increased from 1,100 °C to 1,150 °C, then decreased to 71.5 emu/g and 70.1 emu/g at 1,200 °C and 1,250 °C, respectively, as shown in Table 3. The coercive field H_c values reduced from 19.0 to 5.3 Oe when the sintering temperature increased from 1,100 °C to 1,150 °C and then rose slightly (Table 3). The increase in M_s at the sintering temperature of 1,150 °C was due to the ceramic having

the maximum density with the least porosity between the grains. The crystal structure was complete, allowing for smooth domain wall movement, which stimulates domain rotation and creates optimal conditions for the magnetization process. The relationship of the magneto crystalline anisotropy constant (K) and the H_c is determined by the equation $H_c = 2K/M_s$, according to the Stoner–Wohlfarth model (Zhang *et al.*, 2011). A high H_c should produce a high K value. In this study, the K value continuously decreased when the sintering temperature increased to 1,150 °C, and then the K value increased, as seen in Table 3. It was believed that the lowest H_c , obtained from the sample sintered at 1,150 °C, was due to the large grain size, the high density, and the lowest magneto crystalline anisotropy (Suzuki *et al.*, 2008; Zhang *et al.*, 2021; Castle *et al.*, 2017). When compared with similar research, the NMCZF ceramics synthesized via solid-state combustion technique in this study obtained M_s and H_c values ($M_s \sim 95.2$ emu/g and $H_c \sim 5.3$ Oe) better than the previously researched $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ ceramic ($M_s \sim 58$ emu/g and $H_c \sim 92$ Oe (Wani *et al.*, 2023)), $\text{Ni}_{0.2}\text{Mn}_{0.3}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ ($M_s \sim 64$ emu/g and $H_c \sim 22$ Oe (Shirsath *et al.*, 2010)) and $\text{Ni}_{0.25}\text{Mn}_{0.25}\text{Cu}_{0.15}\text{Zn}_{0.35}\text{Fe}_2\text{O}_4$ ($M_s \sim 55$ emu/g and $H_c \sim 5.2$ Oe (Ahad and Akther Hossain, 2024)). This indicates that the synthesis of NMCZF ceramics via the solid-state combustion technique has the potential to enhance its soft ferromagnetic properties

Conclusions

The results of this study demonstrate the successful synthesis of NMCZF ferrite ceramics using

the solid-state combustion technique, with the optimal conditions being a calcination temperature of 1,000°C for 2 h and a sintering temperature of 1,150°C for 5 h. The obtained powders exhibited a narrow particle size distribution with an average particle size of 0.29 μm . As the sintering temperatures increased, a rapid increase in the average grain size, from 1.0 μm to 6.4 μm , while the measured density increased from 5.01 g/cm³ to 5.14 g/cm³ before dropping to 5.07 g/cm³, was seen in the NMCZF ceramics. The dielectric properties of the NMCZF ceramics showed a frequency dependent behavior, characteristic of typical ferrites with a spinel structure. All ceramic samples exhibited saturated M–H loops, confirming that the NMCZF ceramics exhibit ferromagnetic behavior with small H_c . At a sintering temperature of 1,150°C, the M_s was observed to be the highest at 95.2 emu/g, while H_c was the lowest at 5.3 Oe, making it an excellent soft ferrite material suitable for applications in magnetic sensors, gas sensors, and energy systems.

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