

ELECTRIC AND MAGNETIC PROPERTIES OF $\text{Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4$ DOPED $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3$ CERAMICS PREPARED BY THE SOLID-STATE COMBUSTION METHOD

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Abstract

In this study, we synthesized $(1-x)((\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3)-x \text{ Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4$ [(1-x)BNBT-xNZLAF] ceramics with $x = 0, 0.05, 0.10, 0.15$, and 0.20 using the solid-state combustion method. The phase formation, microstructure, and dielectric, ferroelectric, ferromagnetic, and magnetoelectric properties were investigated. BNBT and NZLAF powders were calcined at 750°C and 900°C for 2 h., respectively. The calcined powders were then mixed in various ratios to produce (1-x)BNBT-xNZLAF composites, which were sintered at 1125°C for 2 h. X-ray diffraction analysis indicated that undoped BNBT ceramics exhibited a typical perovskite structure with coexisting rhombohedral (R) and tetragonal (T) phases. With increasing NZLAF content, the cubic spinel (C) phase emerged with the R and T perovskite phases. Rietveld refinement indicated a higher percentage of the C phase as x increased. When x increased from 0 to 0.10, the average grain size and density increased (from 1.0 to $9.8 \mu\text{m}$ and 5.45 to 5.61 g/cm^3 , respectively) followed by a drop at higher NZLAF levels. Doping BNBT ceramics with NZLAF resulted in a reduction in the dielectric constant (ϵ_r), dielectric loss ($\tan \delta$), and remnant polarization (P_r). The undoped BNBT ceramic showed a saturated P-E hysteresis loop, while NZLAF doped ceramics exhibited unsaturated P-E loops and increased leakage current. Magnetic measurements showed a transition from diamagnetic to ferromagnetic behavior with NZLAF doping. As x increased, the remnant magnetization (M_r), saturation magnetization (M_s), and magnetoelectric coefficient also increased ($M_r \approx 0.058 \text{ emu/g}$, $M_s \approx 3.594 \text{ emu/g}$, and $\text{ME}_{\text{coefficient}} \approx 2.64 \text{ mV/cm}\cdot\text{Oe}$ at $x=0.25$). These results suggest that NZLAF-doped BNBT ceramics hold potential for multiferroic material applications.

Keywords: BNBT; NZLAF; Combustion Method; Dielectric; Microstructure; Multiferroic

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Introduction

Multiferroic materials have gained significant research and technological interest due to their unique combination of properties. These materials exhibit more than one primary ferroic order parameter, such as ferromagnetic, ferroelectric, and ferroelastic, which allows them to respond to both electric and magnetic fields. This coupling of properties makes them candidates for a variety of applications, including memory devices, sensors, actuators, spintronics, transducers, and energy harvesting systems (Haque *et al.*, 2020; Li *et al.*, 2020). Despite their potential, challenges remain, such as producing multiferroicity at high temperature and also increasing magnetoelectric (ME) coupling. More research is needed to overcome these limitations and fully realize the potential capabilities of multiferroic materials. The literature suggests that two-phase composite multiferroic materials are widely studied and exhibit strong potential for high multiferroic performance. These composites combine electric and magnetic materials such as $\text{Ba}_{0.85}\text{Ca}_{0.15}\text{Zr}_{0.1}\text{Ti}_{0.9}\text{O}_3\text{-Ni}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ (BCTZ-NZF), $\text{SrTiO}_3\text{-MgFe}_2\text{O}_4$ (ST-MF), $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3\text{-NiFe}_2\text{O}_4$ (PZT-NF), and $\text{BaTiO}_3\text{-NiFe}_2\text{O}_4$ (BT-NF) (Sadik *et al.*, 2020; Bansal *et al.*, 2021; Matavž *et al.*, 2021; Chauhan *et al.*, 2023).

Lead-free $(1-x)\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-xBaTiO}_3$ ferroelectric ceramics have been widely studied (Li *et al.*, 2022; Yang *et al.*, 2022). These ceramics feature a perovskite (ABO_3) structure and exhibit impressive properties, including a high dielectric constant (ϵ_c), good piezoelectric constant (d_{33}), and high remnant polarization (P_r). These characteristics make them suitable for various electronic applications, such as multilayer capacitors (MLCCs), sensors, actuators, and transducers. Recently, B. Thatawong *et al.* (Thatawong *et al.*, 2022) investigated a $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3$ (BNBT) ceramic, prepared via the solid-state combustion method, calcined at 750°C for 2 h., and then sintered at 1150°C for 2 h. The ceramic showed high remnant polarization ($P_r \sim 38.6 \mu\text{C}/\text{cm}^2$), a good coercive electric field ($E_c \sim 38.9 \text{ kV}/\text{cm}$), and an excellent dielectric constant ($\epsilon_m = 5674$).

Ni-Zn spinel ferrites ($\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$) with the general formula AB_2O_4 are widely used to produce critical components for magnetic devices. Their applications include generators, transformers, and electromagnetic absorbers, owing to their excellent magnetic performance and good chemical stability (Datt and Abhyankar, 2016; Aggarwal and Narang, 2020). Recently, Ni-Zn spinel ferrites have been

modified to create materials with enhanced magnetic and electrical properties. For example, Li^+ doped soft magnetic ferrites (Kaiwen *et al.*, 2015) have shown potential for improving both soft magnetic and dielectric properties. Additionally, substituting Al^{3+} for Fe^{3+} can reduce dielectric losses and improve the magnetic coerciveness of materials (Lassoued *et al.*, 2019). Q. Ni *et al.* (Ni *et al.*, 2020) reported doping Li^+ and Al^{3+} into Ni-Zn spinel ferrite ($\text{Ni}_{0.5-x}\text{Zn}_{0.5-x}\text{Li}_x\text{Al}_x\text{Fe}_2\text{O}_4$) using the sol-gel auto combustion method. This system exhibited a low magnetic coercive field (62.93 Oe) and high saturated magnetization (110.95 emu/g) at $x = 0.025$ (NZLAF). Furthermore, the dielectric behaviors were improved by doping Li^+ and Al^{3+} .

BNBT and NZLAF powders can be prepared using various methods, including conventional solid-state reaction and the sol-gel process (Paul *et al.*, 2015; VS *et al.*, 2022). The conventional solid-state reaction is relatively simple, requires minimal equipment, and can be easily scaled up for large-scale production. However, it often requires high energy input due to the high processing temperatures and can result in larger particle sizes, which might not be suitable for all applications. The sol-gel process is a chemical method that involves transitioning from a liquid solution to a solid gel phase. This technique improves powder homogeneity and lower processing temperatures, but it involves complex chemical processes that require careful control of parameters to achieve the desired outcomes.

Therefore, researchers have developed a method known as solid-state combustion to synthesize high-quality ceramics (Kornphom *et al.*, 2021; Kornphom, 2022; Luangpangai *et al.*, 2024). The main benefit of this method is the use of energy released from fuels (e.g., glycine, urea, alanine, carbonylhydrazide, or citric acid), which accelerates the chemical reaction between raw materials and reduces the reaction temperature. Furthermore, during calcination, the diffusion of the compounds occurs in the liquid phase, which has a higher diffusion coefficient compared to the solid phase. This allows the components to mix more easily and produces a more uniform final compound. The powders obtained through this method exhibit high purity and homogeneity, while the resulting ceramics exhibit high density and good electrical and magnetic properties.

Therefore, in this work, we focused on fabricating $(1-x)[(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3] - x(\text{Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4)$ [(1-x)BNBT-xNZLAF] ceramics at $x = 0, 0.05, 0.10, 0.15$, and

0.20 by the solid-state combustion method and examined the effect of NZLAF content on the phase formation, microstructure, and the electric and magnetic properties of BNBT ceramics.

Experimental

The $(1-x)\text{BNBT}-x\text{NZLAF}$ ceramics, with $x=0, 0.05, 0.10, 0.15$, and 0.20 , were successfully synthesized using the solid-state combustion method with glycine as the fuel. High-purity grade reagents were used as raw materials, including $\text{Bi}(\text{NO}_3)_3 \cdot 5\text{H}_2\text{O}$ (> 98%, KemAus, AU), NaNO_3 (> 99%, Ajax, NZ), $\text{Ba}(\text{NO}_3)_2$ (> 99%, HiMedia, IN), TiO_2 (> 99%, KemAus, AU), NiO (> 99.99%, Merck, DE), $\text{Zn}(\text{NO}_3)_2 \cdot 6\text{H}_2\text{O}$ (98%, Ajax, NZ), Li_2O_3 (97%, Sigma-Aldrich, MX), $\text{Al}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (98%, Ajax, NZ), and $\text{Fe}(\text{NO}_3)_3 \cdot 9\text{H}_2\text{O}$ (> 99%, Ajax, NZ). The raw materials were weighed to the proper stoichiometric ratios for the different compositions of $(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3$ (BNBT) and $\text{Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4$ (NZLAF) and ball-milled in ethanol for 24 h. The suspensions were then dried at 130°C and sieved. The dried BNBT powders were mixed with glycine at a 1:1.11 ratio and calcined at 750°C for 2 h. (Thatawong *et al.*, 2022). Similarly, the dried NZLAF powders were mixed with glycine at a 1:2 ratio and calcined at 900°C for 2 h. (Sonchaopri *et al.*, 2023). The BNBT and the NZLAF calcined powders were combined to produce $(1-x)\text{BNBT}-x\text{NZLAF}$ composites in various compositions. The $(1-x)\text{BNBT}-x\text{NZLAF}$ powders were thoroughly mixed with a 3wt% polyvinyl alcohol (PVA) binder solution, followed by ball milling for 12 h. Afterwards, the powders were dried, sieved, and pressed into pellets with a diameter of ~ 15 mm and a thickness of ~ 1.5 mm using a hydraulic press at a pressure of 100 MPa. The PVA was removed by heating to 550°C for 15 min (Bhupaijit *et al.*, 2021; Klinbanmor *et al.*, 2023). Finally, the pure $(1-x)\text{BNBT}-x\text{NZLAF}$ pellets were sintered at 1125°C for 2 h., with a heating rate of $5^\circ\text{C}/\text{min}$ and allowed to cool naturally.

To investigate the properties of the $(1-x)\text{BNBT}-x\text{NZLAF}$ ceramics, X-ray diffraction (XRD, BRUKER, D2 Phaser) was used to analyze the phase structure. A Field Emission Scanning Electron Microscope (FESEM, Thermo Fisher Scientific model Apreo S) was used to examine the ceramics' surface morphologies and microstructure. The ceramics were polished with sandpaper and an alumina abrasive for the FESEM analysis, followed by thermal etching at 1025°C for 15 minutes (at a heating rate of $5^\circ\text{C}/\text{min}$, then cooled naturally). The average grain size of the ceramics was calculated

using the linear interception method on the FESEM images. The ceramics' microstructural components were investigated with an energy-dispersive X-ray spectrometer (EDS).

The densities of the ceramics were measured using the Archimedes method, with distilled water as the immersion medium. To measure the electrical properties, silver paste was applied to coat both sides of the ceramic samples, and then these were fired at 550°C for 15 minutes (with a heating rate of $5^\circ\text{C}/\text{min}$) and allowed to cool naturally. The frequency-dependence dielectric properties were measured using an LCR analyzer (HP4284A, Hewlett-Packard, Palo Alto, CA, USA) at room temperature, across a frequency range of 1 kHz-1 MHz. Polarization versus electric field (P-E) hysteresis loops were recorded using a computer-controlled modified Sawyer-Tower circuit at 1 Hz (Radiant, PLC2-The P-E loop). Magnetic properties were measured with a vibrating sample magnetometer (Versa Lab, Quantum Design) under an applied magnetic field of 10,000 Oe. The magnetoelectric coefficients were measured with a lock-in amplifier (Stanford Research System, model SR830).

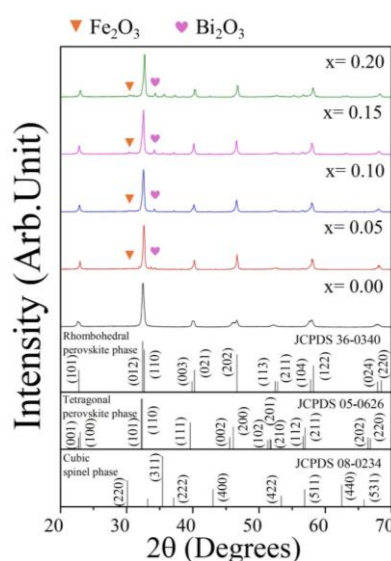


Figure 1. XRD patterns of $(1-x)\text{BNBT}-x\text{NZLAF}$ ceramics with $x = 0, 0.05, 0.10, 0.15$, and 0.20 in the 2θ range of $20\text{--}70^\circ$

Results and Discussion

Figure 1 shows the X-ray diffraction patterns of $(1-x)(\text{Bi}_{0.5}\text{Na}_{0.5})_{0.94}\text{Ba}_{0.06}\text{TiO}_3-x\text{Ni}_{0.475}\text{Zn}_{0.475}\text{Li}_{0.025}\text{Al}_{0.025}\text{Fe}_2\text{O}_4$ [$(1-x)\text{BNBT}-x\text{NZLAF}$] ceramics with $x = 0, 0.05, 0.10, 0.15$, and

0.20 at room temperature in the 2θ range of 20° – 70° . The results revealed that the (1-x)BNBT-xNZLAF ceramic at $x=0$ exhibited a pure perovskite structure. When NZLAF was added, perovskite, spinel, and secondary phases of Bi_2O_3 and Fe_2O_3 were observed in the ceramics. The XRD patterns were compared with JCPDS files No. 36-0340, 05-0626, and 08-0234, which correspond to rhombohedral perovskite (R), tetragonal perovskite (T), and cubic spinel (C) phases, respectively.

To examine the phase formation, Rietveld refinement was performed to analyze the ratio of R, T, and C phases of the (1-x)BNBT-xNZLAF ceramics by fitting the XRD data using the Fullprof program. The values for the cell parameters, lattice parameters, space group, and atomic positions were obtained from the corresponding reference patterns available in the Crystallography Open Database (COD). The phase structure models of $\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3$, BaTiO_3 , and $\text{Ni}_{0.5}\text{Zn}_{0.5}\text{Fe}_2\text{O}_4$ ceramics, with space groups of $R3c:H$, $P4bm$, and $Fd-3m:2$, respectively, were used as starting models to refine the ceramics' XRD data. The background was adjusted using the Chebyshev polynomial

function. The profile shapes were refined using the pseudo-Voigt function, a linear combination of Lorentzian and Gaussian functions (Sindhu *et al.*, 2012). The quality values used to evaluate how well the refinement fits the data are: R_p - the reliability of the profiles, R_{wp} - the reliability of the weighted patterns, R_{exp} - the expected residual, and χ^2 - the goodness of fit. The quality values were found to be $R_p \leq 29.60\%$, $R_{wp} \leq 29.50\%$, $R_{exp} \leq 17.17\%$, and $\chi^2 \leq 3.61$, as listed in Table 1, indicating good agreement between the observed and calculated intensity profiles for all ceramics.

Figure 2 shows the Rietveld refinement of the XRD patterns of the (1-x)BNBT-xNZLAF ceramics in the 2θ range of 20° to 70° . The results revealed that the ceramic with $x=0$ had coexisting R and T phases. The ceramics doped with NZLAF had coexisting R, T, and C phases. The percentage of the R phase increased from 30% to 67% as x rose from 0 to 0.15, then slightly decreased. At the same time, the T phase decreased from 70% to 20% as x increased. Moreover, the C phase increased from 6% to 19% as x increased from 0.05 to 0.20, as shown in Table 1

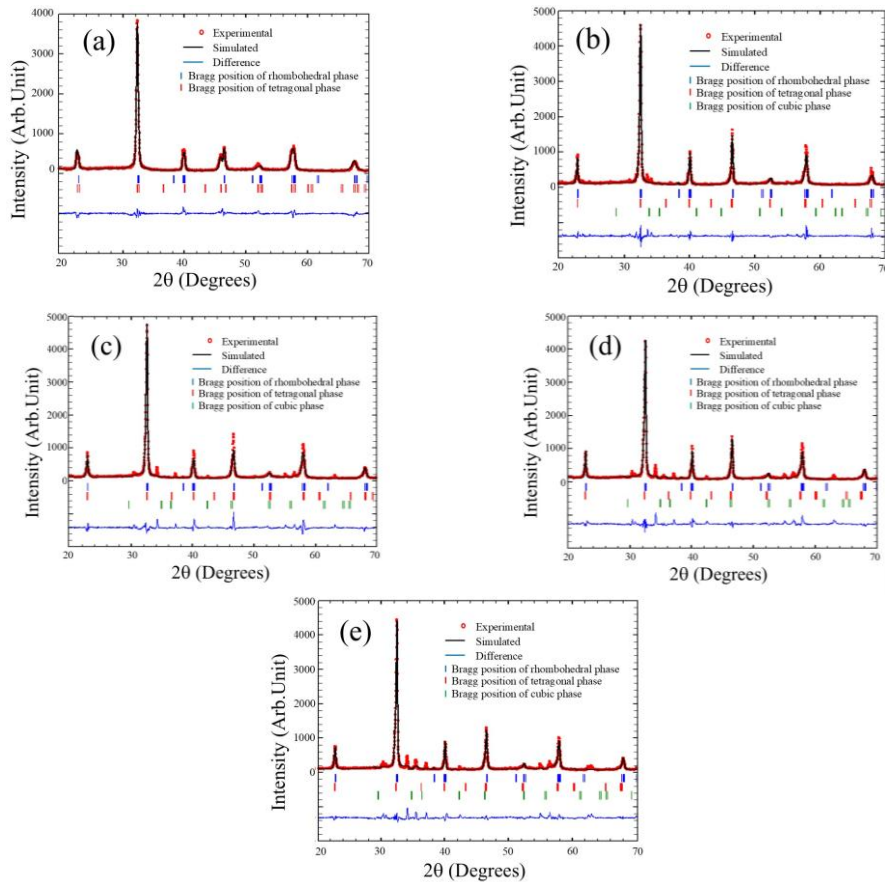


Figure 2. Rietveld refinement analysis for (1-x)BNBT-xNZLAF ceramics: (a) $x=0$, (b) $x=0.05$, (c) $x=0.10$, (d) $x=0.15$, and (e) $x=0.20$

Table 1. Goodness of fit, quality values, lattice parameters, atom information and phase percentage for (1-x)BNBT-xNZLAF ceramics with x = 0, 0.05, 0.10, 0.15 and 0.20

(1-x)BNBT- xNZLAF	Refine parameters	Phase structures	Profile parameters	Atoms information					The percentage of phase
				Label	x	y	z	Occ.	
x=0	$\chi^2=1.79$ $R_p=25.8\%$ $R_{wp}=23.0\%$ $R_{exp}=17.17\%$	<i>R3c:H</i>	a=5.51369Å	Bi1	0.0000	0.0000	0.2627	0.5443	30
			c=13.59782Å	Na1	0.0000	0.0000	0.2627	0.4977	
			u=0.48930	Ti1	0.0000	0.0000	0.0063	1.0628	
			v=0.00624	Ba1	0.0000	0.0000	0.2627	0.0895	
			w=0.00944	O1	0.1260	0.3360	0.0833	2.0019	
	<i>P4bm</i>	a=5.50036Å	Bi1	0.0000	0.5000	0.5450	0.3856	70	
		c=3.95175Å	Na1	0.0000	0.5000	0.5450	0.4446		
		u=0.59197	Ti1	0.0000	0.0000	0.0000	0.9593		
		v=0.40091	Ba1	0.0000	0.5000	0.5450	0.0319		
		w=0.00451	O1	0.0000	0.0000	0.5100	1.0432		
			O2	0.2710	0.2290	0.0150	1.0241		
x=0.05	$\chi^2=2.32$ $R_p=26.7\%$ $R_{wp}=25.1\%$ $R_{exp}=16.48\%$	<i>R3c:H</i>	a= 5.50811Å	Bi1	0.0000	0.0000	0.2627	0.4338	41
			c= 13.56801Å	Na1	0.0000	0.0000	0.2627	0.5031	
			u=0.21054	Ti1	0.0000	0.0000	0.0063	0.9250	
			v=0.06166	Ba1	0.0000	0.0000	0.2627	0.0290	
			w=0.01859	O1	0.1260	0.3360	0.0833	3.9090	
	<i>P4bm</i>	a= 5.53159Å	Bi1	0.0000	0.5000	0.5450	0.5418	53	
		c= 3.91637Å	Na1	0.0000	0.5000	0.5450	0.3287		
		u= 0.75212	Ti1	0.0000	0.0000	0.0000	1.2842		
		v= 0.15762	Ba1	0.0000	0.5000	0.5450	0.0666		
		w= 0.11037	O1	0.0000	0.0000	0.5100	2.2672		
			O2	0.2710	0.2290	0.0150	1.6240		
	<i>Fd-3m:2</i>	a=b=c=8.81398Å	O1	0.2500	0.2500	0.2500	1.2904	6	
		u= -2.56400	Fe1	0.1250	0.1250	0.1250	0.3593		
		v= 0.10157	Fe2	0.5000	0.5000	0.5000	0.4586		
		w= 2.48384	Ni1	0.1250	0.1250	0.1250	0.0234		
			Ni2	0.5000	0.5000	0.5000	0.0134		
			Zn1	0.1250	0.1250	0.1250	0.0450		
		Zn2	0.5000	0.5000	0.5000	0.0381			
		Li1	0.1250	0.1250	0.1250	0.0012			
		Li2	0.5000	0.5000	0.5000	0.0030			
		Al1	0.1250	0.1250	0.1250	0.0012			
		Al2	0.5000	0.5000	0.5000	0.0016			
x=0.10		$\chi^2=3.06$ $R_p=29.6\%$ $R_{wp}=28.2\%$ $R_{exp}=16.12\%$	<i>R3c:H</i>	a=5.49073Å	Bi1	0.0000	0.0000		0.2627
	c=13.52528Å			Na1	0.0000	0.0000	0.2627	0.4857	
	u=0.15560			Ti1	0.0000	0.0000	0.0063	0.8928	
	v=-0.11172			Ba1	0.0000	0.0000	0.2627	0.0518	
	w=0.03245			O1	0.1260	0.3360	0.0833	4.4536	
	<i>P4bm</i>	a=5.50534Å	Bi1	0.0000	0.5000	0.5450	0.4695	30	
		c=3.89865Å	Na1	0.0000	0.5000	0.5450	0.3392		
		u=-0.19122	Ti1	0.0000	0.0000	0.0000	1.2568		
		v=0.22758	Ba1	0.0000	0.5000	0.5450	0.0405		
		w=0.15112	O1	0.0000	0.0000	0.5100	1.6837		
			O2	0.2710	0.2290	0.0150	1.6935		
	<i>Fd-3m:2</i>	a=b=c=8.54935Å	O1	0.2500	0.2500	0.2500	1.0824	10	
		u=1.14797	Fe1	0.1250	0.1250	0.1250	0.4985		
		v=0.12664	Fe2	0.5000	0.5000	0.5000	0.3512		
		w=0.09243	Ni1	0.1250	0.1250	0.1250	0.0648		
			Ni2	0.5000	0.5000	0.5000	0.0410		
			Zn1	0.1250	0.1250	0.1250	0.0551		
		Zn2	0.5000	0.5000	0.5000	0.0420			
		Li1	0.1250	0.1250	0.1250	0.0150			
		Li2	0.5000	0.5000	0.5000	0.0045			
		Al1	0.1250	0.1250	0.1250	0.0054			
		Al2	0.5000	0.5000	0.5000	0.0045			
x=0.15		$\chi^2=3.24$ $R_p=28.7\%$ $R_{wp}=27.3\%$ $R_{exp}=15.19\%$	<i>R3c:H</i>	a=5.50905Å	Bi1	0.0000	0.0000		0.2627
	c=13.59328Å			Na1	0.0000	0.0000	0.2627	0.4981	
	u=0.34795			Ti1	0.0000	0.0000	0.0063	0.7929	
	v=-0.06767			Ba1	0.0000	0.0000	0.2627	0.0245	
	w=0.01735			O1	0.1260	0.3360	0.0833	4.1418	
	<i>P4bm</i>	a=5.61635Å	Bi1	0.0000	0.5000	0.5450	0.4523	19	
		c=3.90532Å	Na1	0.0000	0.5000	0.5450	0.3304		
		u=-0.19956	Ti1	0.0000	0.0000	0.0000	1.4653		
		v=0.16621	Ba1	0.0000	0.5000	0.5450	0.0513		
		w=0.09271	O1	0.0000	0.0000	0.5100	1.2380		
			O2	0.2710	0.2290	0.0150	1.4990		
	<i>Fd-3m:2</i>	a=b=c=8.55399Å	O1	0.2500	0.2500	0.2500	1.0052	14	
		u=0.60754	Fe1	0.1250	0.1250	0.1250	0.5798		
		v=-0.12983	Fe2	0.5000	0.5000	0.5000	0.5250		
		w=0.32259	Ni1	0.1250	0.1250	0.1250	0.0519		
			Ni2	0.5000	0.5000	0.5000	0.0513		
			Zn1	0.1250	0.1250	0.1250	0.0881		
		Zn2	0.5000	0.5000	0.5000	0.0971			
		Li1	0.1250	0.1250	0.1250	0.0039			
		Li2	0.5000	0.5000	0.5000	0.0049			
		Al1	0.1250	0.1250	0.1250	0.0063			
		Al2	0.5000	0.5000	0.5000	0.0052			

(1-x)BNBT- xNZLAF	Refine parameters	Phase structures	Profile parameters	Atoms information					The percentage of phase
				Label	x	y	z	Occ.	
x=0.20	$\chi^2=3.61$ $R_p=29.3\%$ $R_{wp}=29.5\%$ $R_{exp}=15.51\%$	<i>R3c:H</i>	a=5.48631Å	Bi1	0.0000	0.0000	0.2627	0.4657	61
			c=13.47527Å	Na1	0.0000	0.0000	0.2627	0.4778	
			u=0.36210	Ti1	0.0000	0.0000	0.0063	0.9376	
			v=-0.02769	Ba1	0.0000	0.0000	0.2627	0.0242	
			w=0.00989	O1	0.1260	0.3360	0.0833	2.9080	
		<i>P4bm</i>	a=5.53742Å	Bi1	0.0000	0.5000	0.5450	0.5045	20
			c=3.90427Å	Na1	0.0000	0.5000	0.5450	0.3663	
			u=1.27599	Ti1	0.0000	0.0000	0.0000	1.4869	
			v=-0.56668	Ba1	0.0000	0.5000	0.5450	0.0349	
			w=0.06330	O1	0.0000	0.0000	0.5100	2.1154	
				O2	0.2710	0.2290	0.0150	0.8334	
		<i>Fd-3m:2</i>	a=b=c=8.52036Å	O1	0.2500	0.2500	0.2500	0.7212	19
			u=4.43734	Fe1	0.1250	0.1250	0.1250	0.7150	
			v=0.01824	Fe2	0.5000	0.5000	0.5000	0.5702	
			w=-0.06432	Ni1	0.1250	0.1250	0.1250	0.0917	
				Ni2	0.5000	0.5000	0.5000	0.0913	
				Zn1	0.1250	0.1250	0.1250	0.0881	
				Zn2	0.5000	0.5000	0.5000	0.0971	
				Li1	0.1250	0.1250	0.1250	0.0082	
				Li2	0.5000	0.5000	0.5000	0.0063	
				Al1	0.1250	0.1250	0.1250	0.0026	
				Al2	0.5000	0.5000	0.5000	0.0042	

Figure 3 (a)-(e) displays the microstructure of the polished and thermally etched surfaces of (1-x)BNBT-xNZLAF ($x=0, 0.05, 0.10, 0.15$, and 0.20) multiferroic ceramics using a Field Emission Scanning Electron Microscope (FESEM). The grains of the ceramics exhibited a non-uniform distribution and a polygon shape for all samples. The undoped BNBT ceramic had small grains and low porosity (Figure 3 (a)). When NZLAF was doped into BNBT ceramics, the resulting samples exhibited two types of grains with different appearances: darker and lighter grains. The darker grains were found to form between the boundaries of the lighter grains. As x increased from 0.05 to 0.10 , the grain sizes of the lighter grains significantly increased, while the darker grains remained unchanged, and porosity decreased (Figure 3 (b)-(c)). This suggests that a small amount of doping promotes grain growth, likely due to oxygen vacancies created by the substitution of Fe^{3+} at the B-sites of the perovskite structure (acceptor doping), which enhances mass transfer and promotes grain growth during the sintering process (Cheng *et al.*, 2015). However, when x increased to 0.15 and 0.20 , the size of the lighter grains decreased, while the darker grains remained the same size (Figure 3 (d)-(e)). Furthermore, the number of darker grains increased when x increased, as seen in Figure 3 (b)-(e). An Energy Dispersive X-ray Spectrometer (EDS) was used to study the microstructural components of (1-x)BNBT-xNZLAF ceramics, as shown in Figure 3 ((a')-(e'), (b'')-(e'')). It can be observed that the main components of the lighter grains (spot ●) were Bi, Na, Ti, Ba, and O (BNBT), while the main components of the darker grains (spot ★) were Ni, Zn, Li, Al, Fe, and O (NZLAF) as listed in Table 2. This suggests that the lighter grains are ferroelectric grains, while the darker grains are ferrite grains. As

listed in Table 3, the average grain size and density of (1-x)BNBT-xNZLAF ceramics increased from 1.0 to $9.8 \mu\text{m}$ and 5.45 to 5.61 g/cm^3 , respectively, when x increased from 0 to 0.10 , and then decreased. This indicated that higher doping levels of NZLAF ($x \geq 0.15$) inhibited grain growth, which reduced average grain size and increased porosity.

Figure 4 shows the frequency dependence of the dielectric constant (ϵ) and dielectric loss ($\tan \delta$) of the (1-x)BNBT-xNZLAF ceramics with different x content ($x=0, 0.05, 0.10, 0.15$, and 0.20), measured at room temperature in the frequency range of $1 \text{ kHz} - 1 \text{ MHz}$. It was found that the dielectric constant (ϵ) for all samples decreased with increasing frequency (Figure 4 (a)). While the dielectric loss ($\tan \delta$) increased for pure BNBT samples, it decreased for NZLAF doped samples. The results can be explained by Koop's Theory (Koops, 1951), which shows the normal distribution behavior of dielectric materials. At low frequency, the grains in a material respond efficiently to conductivity and resistivity. In contrast, at higher frequencies, the charge distribution within the material slows down, leading to reduced conductivity and resistivity responses, which cause a decrease in the dielectric constant (ϵ) and dielectric loss ($\tan \delta$). However, for the pure BNBT sample, it was observed that the dielectric loss ($\tan \delta$) increased with higher frequency due to the polarization of space charges (Buchanan, 1991; Rayssi *et al.*, 2018). At 1 kHz , the dielectric constant (ϵ) of the ceramics decreased with increasing x , as shown in Table 3, while the dielectric loss tended to decrease from 0.046 to 0.017 when x increased from 0 to 0.10 . After that, the dielectric loss increased significantly due to the growing ferrite phase, which is attributed to its conductive nature (Sharma *et al.*, 2014)

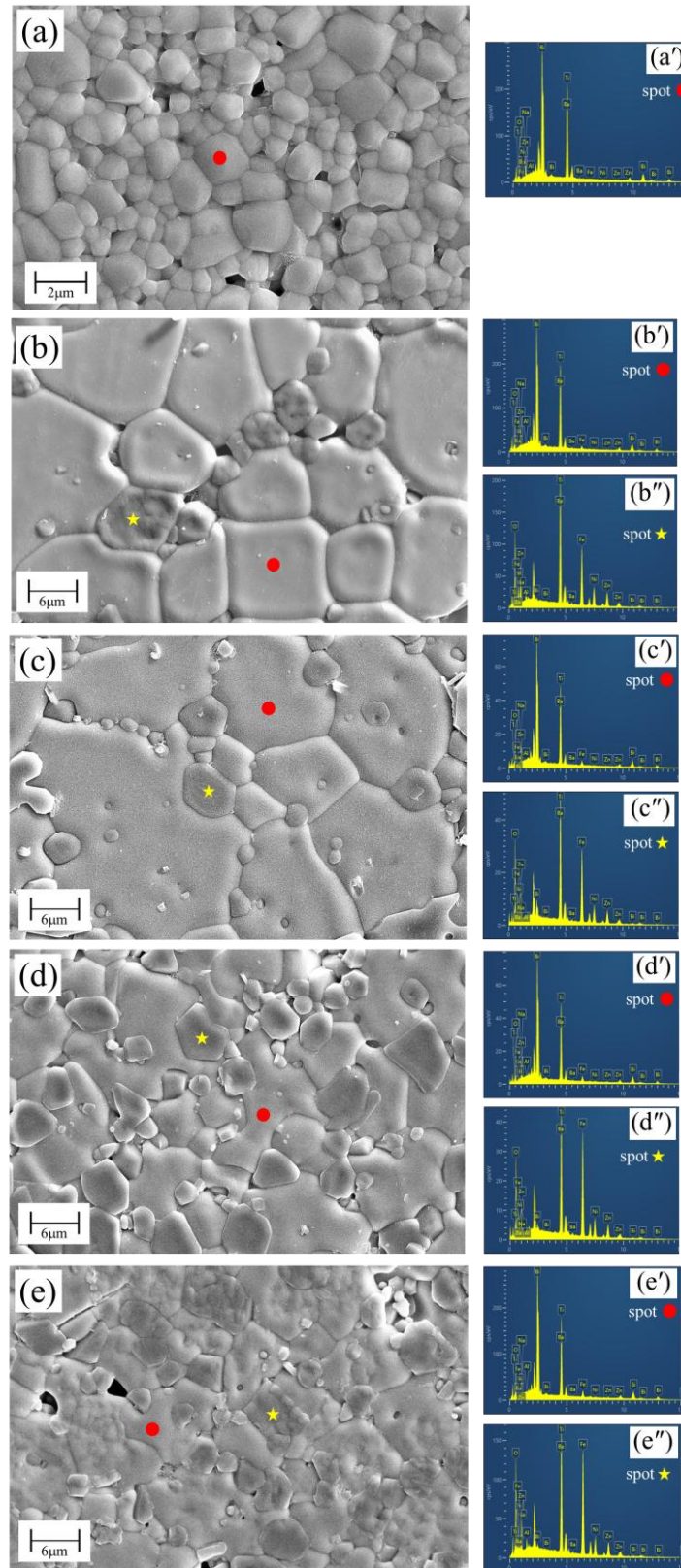


Figure 3. The microstructure and EDS spectra of (1-x)BNBT-xNZLAF ceramics: (a)-(a') $x=0$, (b)-(b'') $x=0.05$, (c)-(c'') $x=0.10$, (d)-(d'') $x=0.15$, and (e)-(e'') $x=0.20$

Table 2. EDS spectra analysis of (1-x)BNBT-xNZLAF ceramics for x=0-0.20

(1-x) BNBT-xNZLAF	Elements	spot •		spot ★	
		Wt%	σ	Wt%	σ
x=0	Bi	48.3	0.5	-	-
	Ti	22.9	0.3	-	-
	O	19.9	0.6	-	-
	Na	5.1	0.2	-	-
	Ba	3.6	0.4	-	-
	Fe	0	0	-	-
	Ni	0	0	-	-
	Al	0	0	-	-
	Zn	0	0	-	-
x=0.05	Bi	47.4	0.5	0.4	0.2
	Ti	21.6	0.3	19.2	0.2
	O	20.9	0.6	25.8	0.4
	Na	4.7	0.2	2.3	0.2
	Ba	1.3	0.4	10.7	0.3
	Fe	0.1	0.1	19.7	0.2
	Ni	0.1	0.1	9.2	0.2
	Al	0.1	0.1	1.4	0.1
	Zn	0.1	0.2	11.2	0.3
x=0.10	Bi	49.8	0.5	4.4	0.3
	Ti	21.4	0.3	17.6	0.2
	O	19.1	0.6	25.5	0.4
	Na	4.6	0.2	1.7	0.2
	Ba	3.0	0.4	10.9	0.3
	Fe	1.8	0.1	22.8	0.2
	Ni	0.1	0.2	7.7	0.2
	Al	0.0	0.1	0.3	0.1
	Zn	0.1	0.1	9.1	0.3
x=0.15	Bi	51.5	0.6	1.4	0.2
	Ti	22.1	0.3	15.8	0.2
	O	17.6	0.5	22.0	0.4
	Na	4.7	0.2	1.6	0.2
	Ba	0.9	0.4	11.2	0.1
	Fe	2.4	0.1	30.2	0.3
	Ni	0.4	0.1	8.2	0.2
	Al	0.0	0.1	0.3	0.1
	Zn	0.4	0.2	9.3	0.3
x=0.20	Bi	49.6	0.5	0.7	0.2
	Ti	20.8	0.3	15.0	0.2
	O	20.3	0.5	24.8	0.4
	Na	4.2	0.2	1.8	0.2
	Ba	1.2	0.4	10.7	0.3
	Fe	2.8	0.1	29.2	0.3
	Ni	0.4	0.1	7.5	0.2
	Al	0.1	0.1	1.0	0.1
	Zn	0.7	0.2	9.3	0.2

Table 3. Average grain size, density, dielectric, ferroelectric, magnetic, and magnetoelectric properties of the (1-x)BNBT-xNZLAF ceramics for x=0-0.20

(1-x) BNBT-xNZLAF	Average grain size (μm)	Density (g/cm^3)	ϵ at 1000 Hz	Tan δ at 1000 Hz	P_r ($\mu\text{C/cm}^2$)	P_{max} ($\mu\text{C/cm}^2$)	E_c (kV/cm)	M_r (emu/g)	M_s (emu/g)	H_c (Oe)	ME coefficient (mV/cmOe)
x=0	1.0	5.45	1561	0.046	33.6	38.3	33.8	-	-	-	1.17
x=0.05	7.4	5.52	660	0.052	33.0	23.7	48.9	0.0001	0.048	22	1.48
x=0.10	9.8	5.61	487	0.017	5.3	9.3	33.6	0.0005	0.049	62	2.04
x=0.15	4.3	5.28	334	0.101	2.6	6.2	26.1	0.004	0.418	55	2.06
x=0.20	3.9	5.06	298	0.173	2.7	4.7	33.2	0.058	3.594	16	2.64

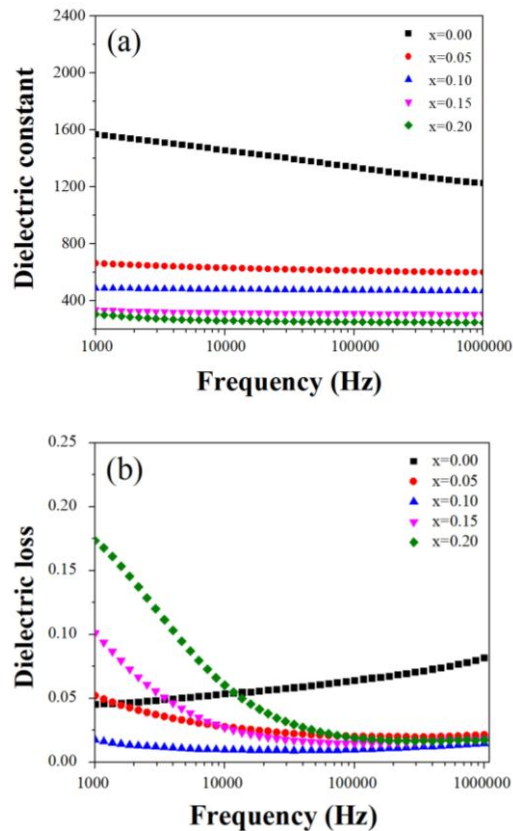


Figure 4. Frequency dependence of the dielectric constant and dielectric loss of the (1-x)BNBT-xNZLAF ceramics measured at room temperature: (a) $x=0$ (b) $x=0.05$ (c) $x=0.10$ (d) $x=0.15$, and (e) $x=0.20$

Figure 5 shows the polarization-electric field (P-E) hysteresis loops for (1-x)BNBT-xNZLAF ceramics under an applied field of 60 kV/cm at a frequency of 1 Hz. The pure BNBT ceramic exhibited a saturated hysteresis loop. While the NZLAF-doped ceramics displayed unsaturated loops and leakage current. As the NZLAF content increased, the remnant polarization (P_r) and polarization at the maximum electric field (P_{max}) decreased, from 33.6 to 2.7 $\mu\text{C}/\text{cm}^2$ and 38.3 to 4.7 $\mu\text{C}/\text{cm}^2$, respectively, as listed in Table 3. The coercive electric field (E_c) increased from 33.8 to 48.9 kV/cm when NZLAF increased from 0 to 0.05, but then slightly decreased at higher NZLAF content (Table 3). The results suggest that NZLAF doping into BNBT ceramics causes a deterioration in their ferroelectric properties, likely due to domain pinning by the ferromagnetic NZLAF phase, which limits the mobility of the domain walls in the ferroelectric phase (Jin *et al.*, 2014).

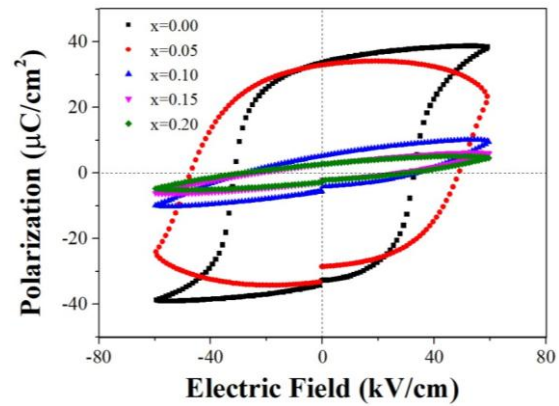


Figure 5. P-E hysteresis loops of (1-x)BNBT-xNZLAF ceramics with varying x content

Figure 6 (a)-(f) displays the magnetization versus magnetic field (M-H) hysteresis loops for (1-x)BNBT-xNZLAF ceramics with different x content, measured at room temperature under a maximum applied field of 10 kOe. For the pure BNBT ceramic (Figure 6 (a)), the M-H hysteresis loop showed a linear relationship, indicating diamagnetic behavior. In contrast, doping with NZLAF (Figure 6 (b)-(e)) resulted in the ceramics showing ferromagnetic properties. The remnant magnetization (M_r), saturation magnetization (M_s) and coercive magnetic field (H_c) are listed in Table 3. It was found that M_r increased from 0.0001 to 0.058 emu/g and M_s increased from 0.048 emu/g to 3.594 emu/g, as x increased from 0.05 to 0.20. These results suggest that NZLAF doping promotes the formation of a ferromagnetic phase.

Figure 7 represents the magnetoelectric coefficient values of (1-x)BNBT-xNZLAF ceramics with varying x content, measured under magnetic fields ranging from 0 to 5000 Oe. The results revealed an increase in the magnetoelectric coefficient as x increased, as shown in Table 3. The BNBT-xNZLAF with $x = 0.20$ exhibited the maximum magnetoelectric coefficient value (~ 2.64 mV/cm·Oe), which is higher compared to similar ceramics like $0.7\text{Bi}_{0.5}\text{Na}_{0.5}\text{TiO}_3\text{-}0.3\text{NiFe}_2\text{O}_4$ and $(\text{BaTiO}_3)_{0.7}\text{-(Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4)_{0.3}$ produced using other methods (Zhang *et al.*, 2013; Wang *et al.*, 2020). This study demonstrated that doping of NZLAF into BNBT ceramics enhances their potential as magnetoelectric materials, which are crucial for various electronic applications.

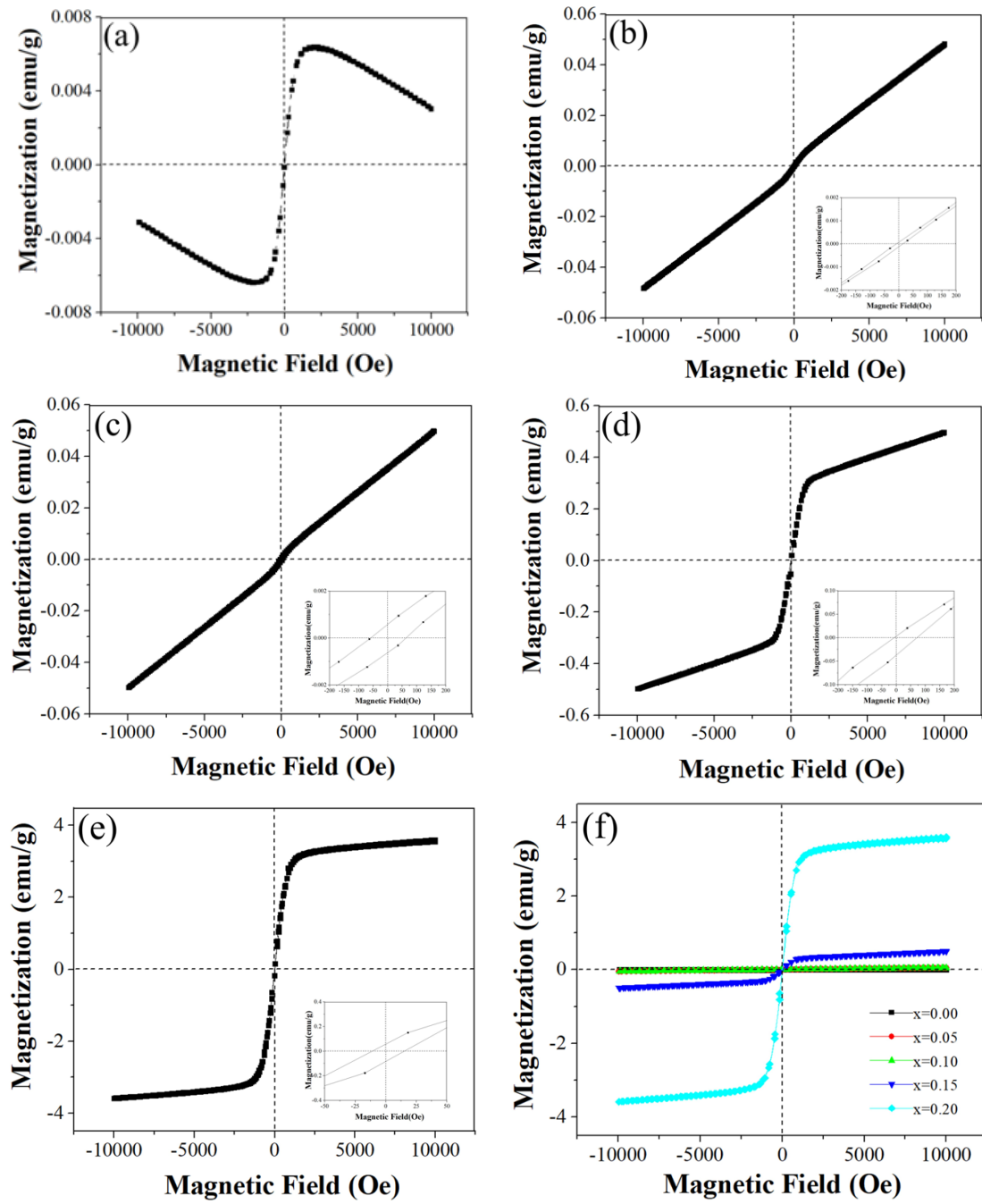


Figure 6. M-H hysteresis loops of $(1-x)\text{BNBT}-x\text{NZLAF}$ ceramics with: (a) $x=0$, (b) $x=0.05$, (c) $x=0.10$, (d) $x=0.15$, (e) $x=0.20$ and (f) the comparison of different x quantities

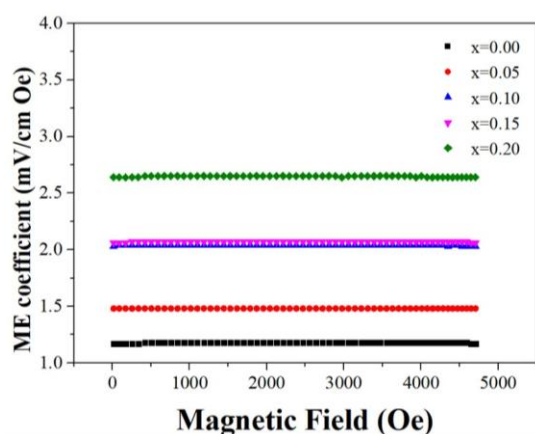


Figure 7. Magnetolectric properties of (1-x)BNBT-xNZLAF ceramics for x= 0-0.20

Conclusions

(1-x) (Bi_{0.5}Na_{0.5})_{0.94}Ba_{0.06}TiO₃-xNi_{0.475}Zn_{0.475}Li_{0.025}Al_{0.025}Fe₂O₄ [(1-x)BNBT-xNZLAF] ceramics were fabricated by the solid-state combustion method. Rietveld refinement revealed that the pure BNBT ceramic had coexisting rhombohedral (R) and tetragonal (T) perovskite phases. With NZLAF doping, the ceramics exhibited a combination of rhombohedral (R) perovskite, tetragonal (T) perovskite and cubic (C) spinel phases. As x increased, the percentage of the R phase decreased while the C phase increased. Small amounts of NZLAF doping enhanced grain growth due to oxygen vacancies, while higher doping levels resulted in decreased grain size and increased porosity due to growth limitations from excess NZLAF. The dielectric constant (ϵ) of (1-x)BNBT-xNZLAF ceramics decreased with increased x, due to the increased ferrite phase. NZLAF doped into BNBT ceramics also degraded the ferroelectric properties due to domain pinning by the ferromagnetic phase. Nevertheless, NZLAF-enhanced ceramics demonstrated improved magnetic and magnetolectric properties, exhibiting both ferromagnetic and ferroelectric characteristics. These properties make them promising candidates for magnetolectric materials in electronic devices.

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