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Investigations on (y)$Ni_{0.8}Zn_{0.2}Fe_2O_4$ (NZF) + (1-y)$Ba_{0.9}Sr_{0.10}La_{0.001}Ti_{0.96}O_3$ Composites

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ABSTRACT
In the present paper, the properties of composites with composition (y)$Ni_{0.8}Zn_{0.2}Fe_2O_4$ (NZF) + (1-y)$Ba_{0.9}Sr_{0.10}La_{0.001}Ti_{0.96}O_3$(y=0.00 and 0.15) is presented. Solid state route was used to prepare the samples. Presence of both phases was confirmed by XRD pattern. Dielectric properties were measured with variation of frequency and temperature. Ferromagnetic and ferroelectric properties were also studied for composite samples.

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Introduction

Studies on ferroelectric and ferromagnetic materials are technologically important research subjects, which led to new discoveries and many useful device applications because single phase materials have their limiting use in practical devices because magnetoelectric (ME) effect is considerably weak even at low temperature [1-2]. The alternatives are magnetoelectric composites because they exhibit large value of ME coefficient. Ferrite and ferroelectric are constituent phases of these composites. They can be exploited as waveguides, sensors, phase shifter, modulators and memory devices etc[3-6]. Limited data is available in the literature “on investigation of dielectric and ferroelectric properties of ferroelectromagnetic composites”. In the present work, the addition of Sr in barium titanate can shift the Curie temperature towards room temperature and hence increase the room temperature dielectric constant. La substitution can further decrease the T_c which results in increase the room temperature dielectric constant. The Saturation magnetization of nickel ferrite increases with zinc content hence its magnetoelectric coefficient increases. MnO_2 was added to minimize the hopping mechanism $Fe^{2+} \leftrightarrow Fe^{3+}$ which results in an increase the resistivity of the samples[7-9]. In nickel ferrite, conductivity is mainly due to hopping between $Fe^{2+} \leftrightarrow Fe^{3+}$ In this paper, we present the detailed report on the properties of the prepared composite. They can be exploited as waveguides, sensors, phase shifter, modulators and memory devices etc [10].

Experimental

The individual phases (ferrite and ferroelectric) were prepared by conventional solid state reaction method. AR grade NiO, ZnO, Fe_2O_3 were used as raw materials for synthesis of ferrite phase. The mixing process was carried out by ball-milling using zirconia balls and distilled water as milling media. The slurry was dried and calcined in alumina crucible at 1000°C for 4 hrs. To the calcined powder, a small amount (0.5% by weight) of MnO_2 was added, ball milled and recalcined at 1000°C for 4 hours. The recalcined powder was then ball milled again and dried. The ferroelectric phase was prepared using the same route by using AR grade $BaCO_3$, $SrCO_3$,La_2O_3 and TiO_2 as raw materials and calcined at 1100°C. The composite was prepared by mixing 15% by wt. (NZF) phase to the (BSLT) (85% by wt.) phase. The mixing process was carried out by ball-milling using zirconia balls and distilled water as milling media. After drying, small amount of diluted PVA was added as binder and the pellets having 2-3mm thickness and 15 mm diameter were pressed using the uniaxial hydraulic press. The pellets were sintered at 1325°C for 4hrs with constant heating rate of 5°C/min. The structural characterization of the samples was carried out by using X-ray diffractometer using Cu Kα radiation (λ=1.541Å). for electrical measurement the samples were then coated with silver paste and heated in an oven at 400°C for 1hr to ensure good ohmic contacts. The dielectric measurements were carried out as a function of frequency at different temperatures by using 4263B LCR meter interfaced to PC and programmable temperature chamber. P-E hysteresis loops were recorded at 20Hz for all the samples.

Results and Discussion

XRD pattern for constituent phases and their composite are shown in figure1. It can be observed that intensity of ferrite phase peak is very small because of low concentration of ferrite phase in composite sample. Calculated values of various parameters for all samples are given in table 1. To understand the microstructure of the

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Table 1: Structural & dielectric parameters

	Ferroelectric			Ferrite	d_{exp}	d_{th}	Porosity	ε_{RT}	$\tan\delta_{RT}$	ε_{max}	$\tan\delta_{max}$
y	a(Å)	c(Å)	c/a	a (Å)	(g/cc)		(%)	at (35°C,100 kHz)		(at100kHz)	
0.00	3.9873	4.0027	1.0038	-	5.28	5.99	11.85	4305	0.068	17548	0.162
0.15	3.9880	4.0057	1.0044	8.3461	5.18	5.99	13.53	2393	0.039	3108	0.059

samples, SEM micrographs were recorded. SEM micrographs for both of the samples are shown in figure 2. It can be observed that the grain size of composite sample is small as compared to pure ferroelectric phase.

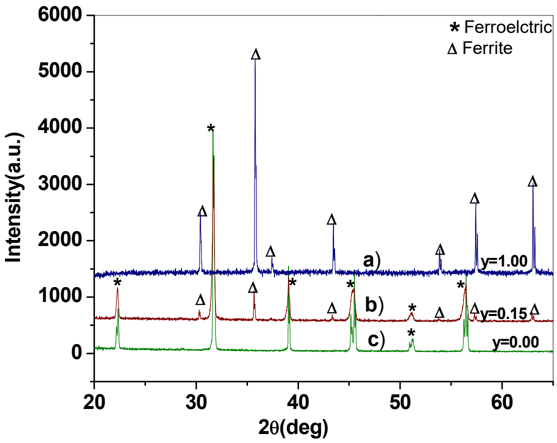
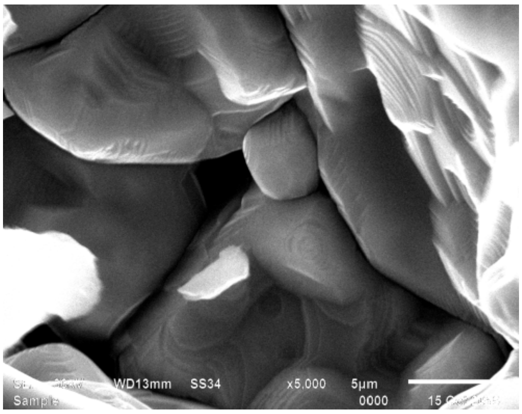
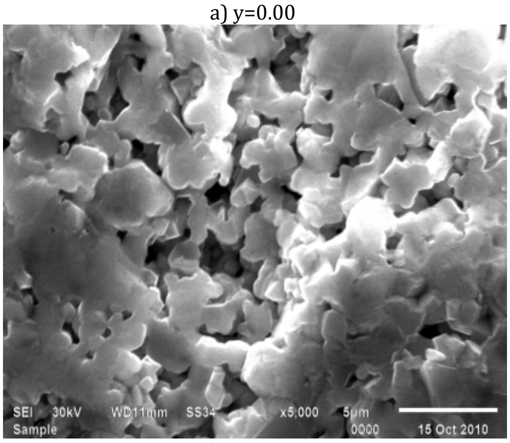


Figure 1: XRD pattern for a) Ferrite phase b) Composite c) Ferroelectric phase



a) y=0.00



b) y=0.15

Figure 2: SEM micrographs a) ferroelectric phase b) composite phase

At three different frequencies, the graph between dielectric constant and temperature is shown in figure 3. From figure it is observed that there are maxima for dielectric constant at a particular temperature called Curie temperature. All dielectric parameters for all samples are given in table1. From Fig.3 it can be observed that the dielectric constant for composite samples is less as compared to ferroelectric phase and also broadening of the peak is observed in composite samples. That may be due to incorporation of ferrite phase in pure ferroelectric phase that decrease the ferroelectric properties of the composites samples [11-12].

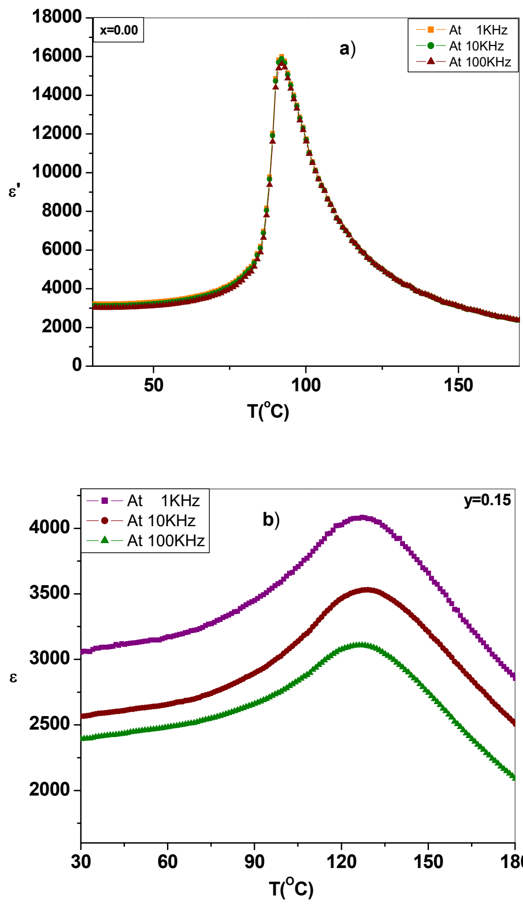


Figure 3: variation of dielectric constant with temperature for a) ferroelectric b) composite

At room temperature, the polarization- field (P-E) hysteresis loops and M-H hysteresis loop for composite samples are shown in figure 4. Presence of both loops indicating that composites are spontaneously polarized as well as spontaneously magnetization. The various ferroelectric parameters and magnetic parameters like) for all samples are shown in table 2. It is observed that as the content of the ferrite phase is added, all ferroelectric parameters decreases.

Table 2: All ferroelectric and ferromagnetic parameters

Y	P_r (μC/cm²)	E_c (kV)	M_s(emu/g) Unpoled	P_s(μC/cm²)
0.00	11.54	3.2	-	25.02
0.15	2.93	3.95	6.53	11.35

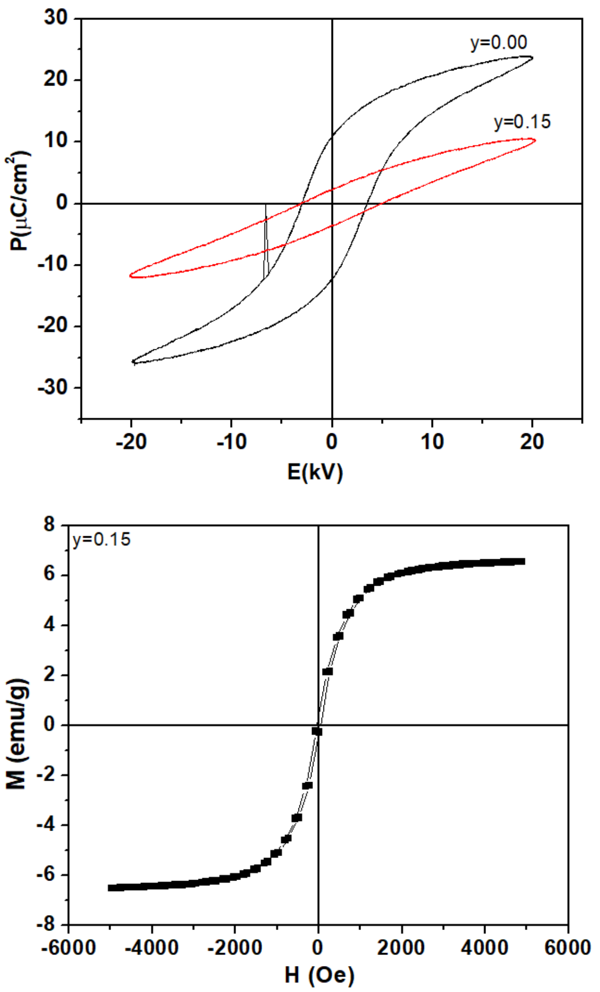


Figure 4: Ferroelectric and Ferromagnetic hysteresis loops for all samples

Conclusions

Ferroelectric rich ME composites have been prepared successfully by conventional solid state method. The dielectric peak for composite samples get broadened and depressed with addition of ferrite content. The observation of both P-E and M-H hysteresis loops in composite samples confirm the coexistence of ferroelectric and magnetic ordering in composites. Increase in P_r and P_s with increase in ferroelectric content and increase in M_r and M_s with increase in ferrite content shows that composites obey rule of mixtures.

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