

Variation of lattice parameter, average grain size, dielectric constant and saturation magnetization with different temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70$ PLZT composite by Co-precipitation method

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Abstract

Magnetoelectric composites containing $\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$ and $\text{Pb}_{0.93}\text{La}_{0.07}(\text{Zr}_{0.60}\text{Ti}_{0.40})\text{O}_3$ phases have been prepared by chemical co-precipitation method. The structure and morphology of the composites were examined by means of X-ray diffraction (XRD) and scanning electron microscopic (SEM). The XRD result showed that the composites consisted of spinel $\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$ and perovskite PLZT annealing at temperature higher than 700°C . The average grain size for the piezoelectric phase of the composite was found to increase the overall dielectric and ferroelectric properties of the composite increased with increasing in sintering temperature. Variation of dielectric constant (ϵ) and dielectric loss ($\tan\delta$) with frequency for $0.30\text{NZFO} + 0.70\text{PLZT}$ composite with annealed at different temperature showed dispersion in the low frequency range. The variation of dielectric constant with temperature reflects DPT type behaviour. The dielectric properties are strongly influenced by interface phenomena (Maxwell-Wagner) due to the local electrical inhomogeneity. At low frequency range, the composite showed the dielectric dispersion is associated with very high polarization. The peak value of dielectric constant for composite decreased with increasing its sintering temperature. coercivity, saturation magnetization and squareness have been found to vary with concentration of ferrite phase and annealing temperature due to the increase in crystallite size. The maximum value of the magnetoelectric conversion factor $(dE/dH)_{\text{max}}$ equal to 5.2 mV/cm.Oe is observed for $30\%\text{NZFO} - 70\%\text{PLZT}$ composites when sintered at 1100°C .

Key words : Magnetoelectric composites , chemical co-precipitation method

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تغايير معاملات الشبكة ، معدل الحجم الحبيبي ، ثابت العزل الكهربائي والإشباع
 $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70 \text{ PLZT}$ درجات الحرارة للمركب باختلاف المغناطيسي
بطريقة الترسيب الكيميائي

عبد السميع فوزي عبد العزيز

قسم الفيزياء – جامعة تكريت

الخلاصة :

المركبات الكهرومغناطيسية والتي تحتوي على الأطوار $\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$ و PLZT والمحضرة بطريقة الترسيب الكيميائي . شخص التركيب البلوري ومورفولوجية السطح للمركبات بواسطة التقنيات الأساسية حيود الأشعة السينية والمجهر الماسح الإلكتروني على التوالي . إن التشخيص بالأشعة السينية قد أظهر أن المركبات تحتوي على طور الفيرات والبيروفسكايت (وهذا الاسم مشتق من مادة معدنية تعرف باسم perovskite – وهي عبارة عن مادة صلبة خليط بين المعدن واللامعدن ولها توزيع ذري خاص) . عند تلدينها عند درجة حرارة أعلى من 700°C . معدل الحجم الحبيبي لطور البيروكترينك الموجود ضمن المركب سوف يزيد من ثابت العزل الكهربائي والخواص الفيروكهربائية للمركبات سوف تزداد بازدياد درجة حرارة التلدين . تغير ثابت العزل الكهربائي ومقدار الطاقة مع التردد لمركب $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ مع التلدين عند درجات حرارة مختلفة أظهرت إزاحة عند مدى التردد الواطي . التغير لثابت العزل الكهربائي مع درجة الحرارة بعكس نوع DPT . خواص ثابت العزل الكهربائي يظهر بواسطة ظاهرة (ماكسويل – ويكنر) لموقع الإلكترونات غير المتجانسة . عند مدى التردد الواطي . أظهر المركب إزاحة مع استقطاب عالي جدا . إن قيمة القمة لثابت العزل الكهربائي للمركب تقل مع زيادة درجة حرارة التسخين . المجال المغناطيسي للكاشف ومغناطيسية الإشباع مع نسبة المغناطيسية وحدت بأنها تتغير مع تركيز طور الفيريت ودرجة حرارة التلدين وبالتالي يؤدي إلى زيادة في حجم البلورة . القيمة العليا لمتجه معامل الكهرومغناطيسية يساوي لمركب عند درجة حرارة 1100°C

1. Introduction

The magnetoelectric effect in solids manifests itself as a change in the polarization (P) of a sample in a magnetic field (H) or a change in the magnetization (M) of a material in an electric field (E) [1]. This effect may occur only in the materials that simultaneously exhibit magnetic and electric ordering. Such materials include single phase crystalline multiferroic and artificially formed compounds containing ferromagnetic and ferroelectric phases. The composite can be considered as a new material with multiferroic properties. The magnetoelectric effect is extrinsic in this case since magnetoelectric effect is not exhibited by any of the constituent phases on their own. Such physical property of the composite is called a “product property” [2], which refers to an effect in one of the phases or sub- materials which is turn leads to a second effect in the other phase. The concept of “product property” in multiferroic composites was first proposed by the Philips Laboratory [3] using BaTiO_3 -

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Abdul Samee Flaws AbdulAziz,

CoFe_2O_4 as a model system. The multiferroic $\text{BaTiO}_3\text{-CoFe}_2\text{O}_4$ composite was synthesized by unidirectional solidification. Patankar et al. [4] have reported that the magnetoelectric composites containing $\text{CuFe}_{1.8}\text{Cr}_{0.2}\text{O}_4 + \text{Ba}_{0.8}\text{Pb}_{0.2}\text{Ti}_{0.8}\text{Zr}_{0.2}\text{O}_3$ phases have been prepared by sintering them at different sintering temperatures. They observed the sintering process brings about many Microstructural changes in the composite ceramics. The ME effect is a structural dependent property. The particle size for either phase of the composite was found to increase, whereas porosity decreases with increase in sintering temperature. Rashed et al. [5] have studied the effect of piezoelectric grain size on magnetoelectric effect on $\text{Pb}(\text{Zr}_{0.52}\text{Ti}_{0.48})\text{O}_3\text{-Ni}_{0.8}\text{Zn}_{0.2}\text{Fe}_2\text{O}_4$ particulate composites. They observed the grain size has significant effect on the piezoelectric, ferroelectric, and dielectric properties of the composite and hence influences the ME property. They were found grain size of around 100 nm show small magnitude of ME Coefficient (54.4 mV/cm.Oe) while those with grain size of 600 nm exhibit a value of 155 mV/cm.Oe. Manoj et al. [6] have studied the synthesis of nanocrystalline $(x)\text{CuFe}_2\text{O}_4 + (1-x)\text{BiFeO}_3$ magnetoelectric composite by citrate precursor method. The phase was found to be formed at 500 °C. They were found the magnetoelectric effect of the nanocomposites to be strongly depending on the magnetic bias and magnetic field frequency.

The present paper deals with the synthesis and characterization of magnetoelectric composites of $\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + \text{PLZT}$. In order to understand structural analysis, morphological, dielectric behaviour, ferroelectric properties, magnetic properties and ME effect exhibited by these composites.

2. Materials and Methods

The $\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$ sample was synthesized using Ni nitrate, Zn acetate and Fe nitrate as precursors, by dissolving them in distill water in the required mole proportion. The clear solution was co-precipitated with a 1 molar NaOH solution at fixed temperature of 60°C. Sample at this stage are refereed “as prepared”. The precipitate was filtered and then washed several times with distilled water until the pH of the filtered water became 7 (i.e., neutral). The filtrate was then dried at 100 °C over-night. The dried powder calcined at 700 °C for 6 h. The spinel cubic structure of the NZFO was confirmed by the X-ray diffraction (XRD) technique. The $\text{Pb}_{0.93}\text{La}_{0.07}(\text{Zr}_{0.60}\text{Ti}_{0.40})\text{O}_3$ [PLZT] sample inside morph tropic phase boundary (MPB) region was prepared using the chemical co-perception method, using of high purity(AR grade)lead acetate [$(\text{CH}_3\text{COO})\text{Pb} \cdot 3\text{H}_2\text{O}$], Lanthanum nitrate [$\text{LaN}_3\text{O}_9 \cdot 6\text{H}_2\text{O}$] and Zirconiumpropoxide $\text{Zr}(\text{OCH}_2\text{CH}_2\text{CH}_3)_4$. Firstly, [$(\text{CH}_3\text{COO}) \text{Pb} \cdot 3\text{H}_2\text{O}$] was separately dissolved in the acetic acid with constant stirring at room temperature. Lanthanum nitrate [$\text{LaN}_3\text{O}_9 \cdot 6\text{H}_2\text{O}$], Zirconiumpropoxide $\text{Zr}(\text{OCH}_2 \text{CH}_2\text{CH}_3)_4$, were separately dissolved in doubly distilled water with constant stirring at room temperature and then mixed together with an aqueous $\text{Pb}(\text{CH}_3\text{COO}) \cdot 6\text{H}_2\text{O}$ solution. Titanium tetra isopropoxide [$\text{C}_{12}\text{H}_{28}\text{O}_4\text{Ti}$] was separately dissolved in the acetic 25% and 75% ethanole and added drop by drop to the Pb-La-Zr solution with constant stirring. Finally, white turbid solution containing all the elements

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Abdul Samee Flaws AbdulAziz,

was obtained. This solution was precipitated with NaOH solution having pH=12 at 60 °C with constant stirring. The precipitate was filtered and then washed several times with distilled water until the pH of the filtered water became 7 (i.e., neutral). The filtrate was then dried at 100 °C for 12 h, resulted into white coloured powder. The dried powder calcined at 700 °C for 6 h. The tetragonal perovskite structure of the PLZT was confirmed by the X-ray diffraction technique. For preparation of composites, the NZFO phase and PLZT phase were mixed together in the proportions as 0.30NZFO + 0.70 PLZT composite. The magnetoelectric composites prepared by mixed the constituent phases were again milled in agate mortar for 2-3 h and mixed with 2-3 drops of polyvinyl alcohol as a binder. The constituents and composite powders were pressed into pellets (i.e. thickness = 2 to 3 mm and diameter = 10 mm) using hydraulic press by applying a pressure of 1 to 2 tonnes/inch for 5 min. The pellets were finally sintered at different temperature at 700, 900 and 1100 for 3h.

Structural characterization of the powders was carried out using X-ray powder diffraction (XRD) with a monochromatic CuK_α radiation. Scanning electron microscopy (SEM) was used to observed particle morphology and average particle size. The dielectric measurements were carried out using LCR meter bridge (Model HIOKY 3532 – 50) in the frequency range 100 Hz to 5 MHz from room temperature to 1000 °C. The polarization versus electric field measurements (P-E hysteresis loops) were carried out using a modified Sawyer- Tower circuit. The magnetization measurements of samples were carried out by using vibrating sample magnetometer (VSM; Model 7307, Lake Shore Cyrotronic, Westerville, OH) with a maximum magnetic field of 6 kOe. Magnetoelectric output (dE/dH) was measured as a function of static magnetic field for the present samples annealed at different temperatures. The composites samples has to be poled both electrically as well as magnetically because electric poling enhance the magnetostriction coefficient of the ferrite phase and magnetic poling enhance the piezoelectric coefficient of the ferroelectric phase [17-19]. Also magnetic poling was carried out at room temperature by placing the sample in a strong dc magnetic field of 4.5 K Oe.

3 Results and discussion

3. 1 Structural and morphological study of composites

Fig. 1 shows the x-ray diffraction pattern of pure Ni_{0.7}Zn_{0.3}Fe₂O₄ and PLZT phases sintered at 700°C for 6hrs, and the compositions of (y)Ni_{0.7}Zn_{0.3}Fe₂O₄ + (1-y) PLZT with y = 0.15, 0.30 and 0.45. The XRD pattern analysis the formation of spinel cubic – tetragonal perovskite mixed structure. The XRD lines observed at 2θ = 29.9, 35.2, 42.9, 53.1, 56.8, and 62.4° belongs to spinel cubic phase where as the XRD lines observed at 2θ = 21.9, 31.2, 38.4, 44.6, 50.4, 55.5, 65.1, and 73.7° belongs to perovskite tetragonal phase. Amongst there lines, the line observed at 2θ = 35.2° is characteristics of spinel NZFO phase where as the line observed at 2θ = 31.2° is characteristics of perovskite PLZT phase. The lattice parameters of pure Ni_{0.7}Zn_{0.3}Fe₂O₄: a = 8.39 Å; pure PLZT: a = b = 4.056 Å and c = 4.074 Å respectively.

Variation of lattice parameter, average grain size, dielectric constant and saturation magnetization with different temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite by Co-precipitation method

Abdul Samee Flaws AbdulAziz,

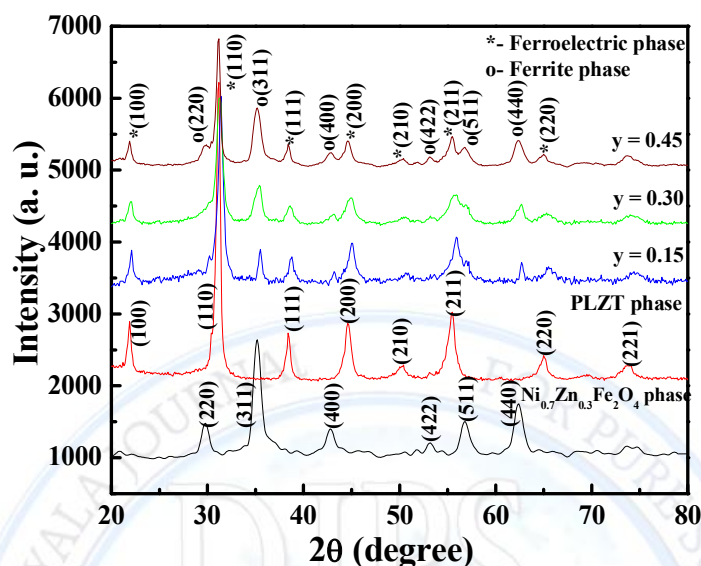


Fig. 1. XRD patterns of $(y)\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + (1-y)\text{ PLZT}$ composites where $y = 0.00, 0.15, 0.30, 0.45$ and 1 sintered at 700°C .

Fig. 2 shows the X-ray diffraction (XRD) patterns of the $0.30\text{NZFO} + 0.70\text{ PLZT}$ composites sintered at various temperatures ranging from 700 to 1100°C for 2 hrs. All diffraction lines observed corresponds to either spinel cubic or perovskite tetragonal phase with three doublets i.e. $(101)/(110), (111)/(111), (002)/(200)$, for 30 mole % NZFO + 70 mole % PLZT composites sintered at 900°C and 1100°C . When the sintered temperature increases further, the intensity and sharpness of peaks are further enhanced and d -spacing values of NZFO phase and PLZT phase correspond well with those JCPDS Card Nos. $46-0504$ and $8-234$ respectively, suggesting that the powder is largely crystallized into a mixed phase of spinel-perovskite structure without traces of any impurity phases. Furthermore, the increases in intensity of X-ray diffraction shows improved crystallinity of the materials and increase in the crystallite size as a function of temperature. The lattice parameters of $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite at different temperatures are given in Table 1. From the table, it can be concluded that the lattice parameters of two phases in the $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite sintering at different temperatures are slight variations to that of constituent phases. **Table 1 Lattice parameters of $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite annealed at different temperatures**

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Abdul Samee Flaws AbdulAziz,

Annealing temperatures (°C)	Lattice parameters of the phases (Å)			
	Ferrite	Ferroelectric		
	a	a	c	c/a
700	8.390	4.056	4.074	1.004
900	8.405	4.256	4.294	1.009
1100	8.420	4.327	4.034	1.01

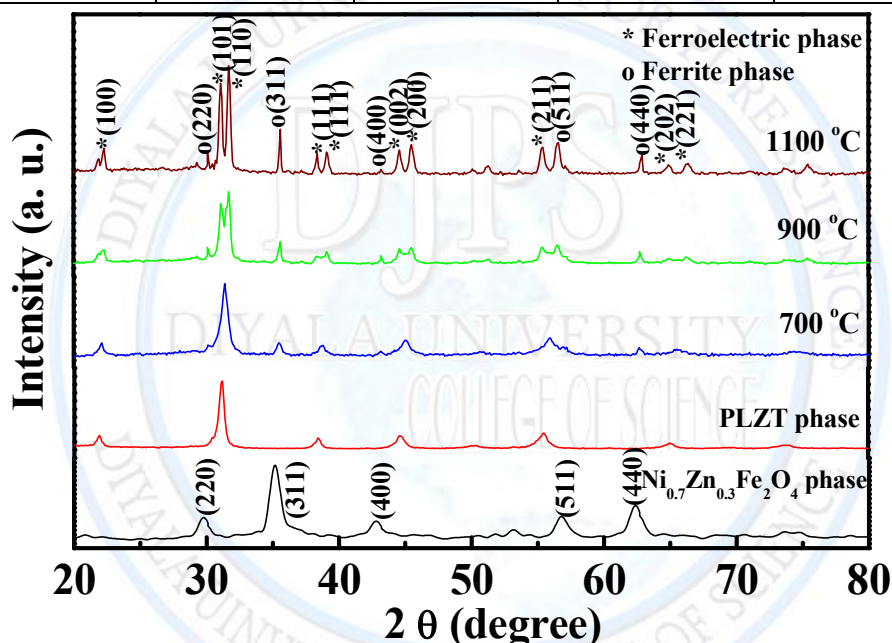


Fig. (2) XRD patterns of $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ powder annealed at different temperatures.

The scanning electron micrographs of $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite annealed at different temperatures are shown in Fig. 3(a-c). It shows two different phases distinctly, white for PLZT phase and dark grey for NZFO phase, which was latter confirmed by Energy-Dispersive X-ray Analysis (EDAX; not shown) studies. There are no unidentified phases. The grain size of PLZT phase present in $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite sintered different temperatures viz. 700 °C, 900 °C and 1100 °C was found to be 359nm, 471nm and 567nm respectively. In this chapter only the effect of sintering temperature on piezoelectric grain size is considered. Careful observations of the micrographs reveals that for PLZT are about ten times large than that of NZFO phase. The piezomagnetic material with small grain size and large boundary area helps in conduction, so that the piezomagnetic phase

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Abdul Samee Flaws AbdulAziz,

shows good electrical conductivity and dielectric property. The grain growth in composites is attributed to the presence of pore in the solid solution that migrates to grain boundaries. The presence of two phases in the composites indicates that grains of one of phases have higher probability of growing and hence they grow at the expense of small grains. The large grain size of ferroelectric phase as compared to ferrite phase can effectively reduce the leakage of electric charges induces by the chain formation of ferrite phase particles.

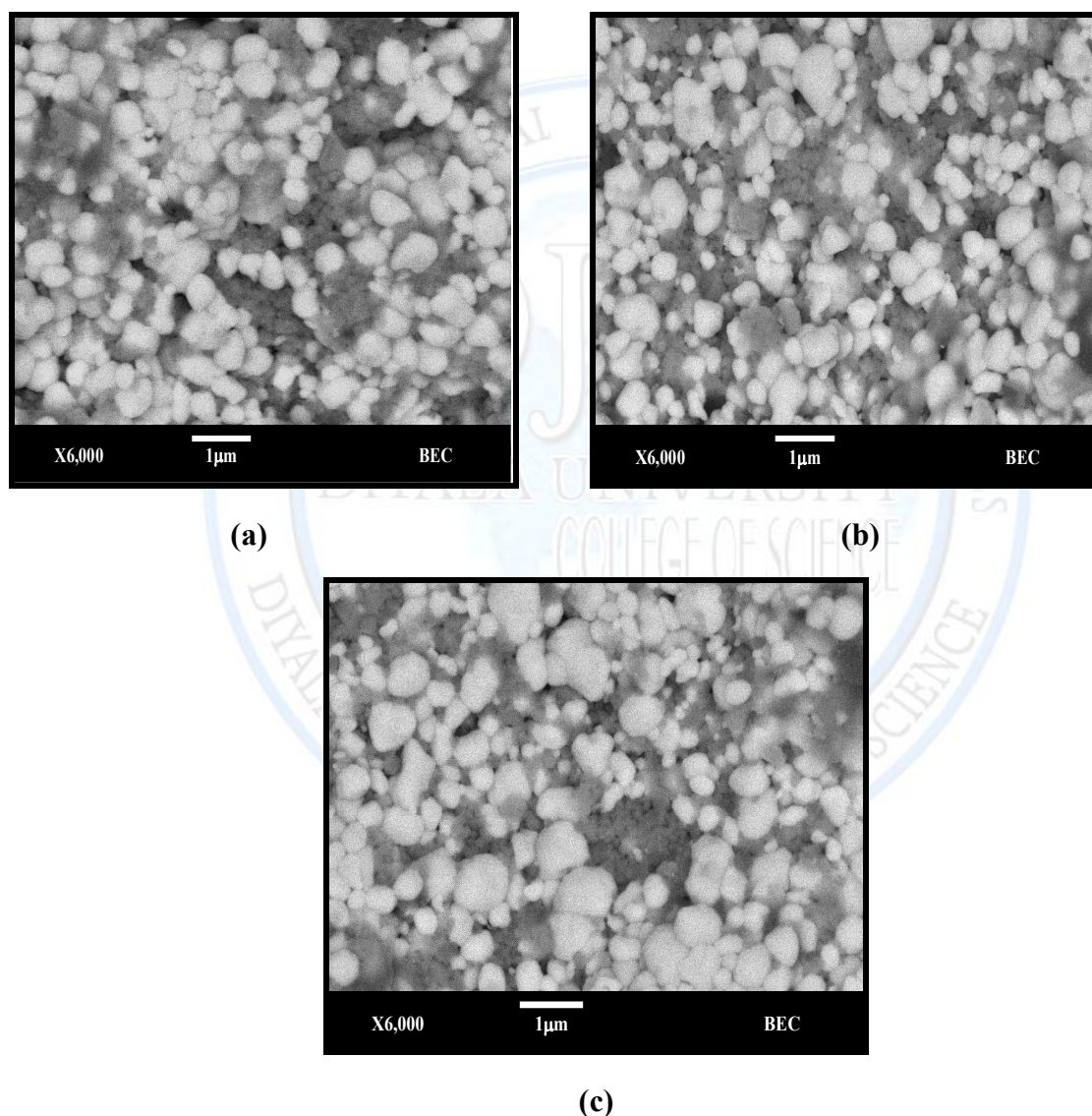


Fig. 3 SEM micrograph of $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70 \text{ PLZT}$ composite annealed at different temperatures (a) 700 °C, (b) 900 °C and (c) 1100 °C.

Variation of lattice parameter, average grain size, dielectric constant and saturation magnetization with different temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70$ PLZT composite by Co-precipitation method

Abdul Samee Flaws AbdulAziz,

3. 2 Dielectric properties

1) Frequency dependent variation

Variation of dielectric constant (ϵ') and loss tangent ($\tan\delta$) with frequency for the composites $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70$ PLZT composite sintered at various temperatures ranging from 700 to 1100 °C for 2 hours. It is clear that dielectric constant increases with increasing sintering temperature, indicating dispersion in the lower frequency range. Later on it attains almost constant value independent of frequency. All samples show dielectric dispersion with frequency is due to Maxwell–Wagner [7-8] type interfacial polarization in agreement with Koop's phenomenological theory [9]. High value of ϵ' at low frequency is explained on the basis of space charge polarization due to inhomogeneous dielectric structure. The inhomogeneities in the present system are impurities, porosity and grain structure. The higher sintering temperature in general lead to high bulk density, but lower electrical resistivity, higher dielectric constant and loss. At low frequency dipoles are able to follow the frequency of applied electric field whereas at high frequency dipoles are unable to follow the frequency of applied electric field, which explains the results observed in Fig. 4(a). However in case of composites the higher value of dielectric constant is ascribed to the fact that ferroelectric regions are surrounded by non-ferroelectric (i.e. ferrite) regions similar to the case relaxor ferroelectrics [10]. It is also observed that the dielectric constant increases with increase annealing temperature. Fig. 4(b) shows the variation of $\tan\delta$ with frequency for all the composites, which shows a similar dispersion as that of ϵ' . The maximum value of $\tan\delta$ is observed for the sample sintered at 700 °C as compared to the composites sintered at 900 and 1100 °C. This means the loss factor is observed to be reduced with increase sintered temperature. The maximum $\tan\delta$ is attributed to the fact that period of relaxation process is same as the period of applied field. When the relaxation time is large as compared to the period of the applied field, losses are small. Similarly, when relaxation process is rapid as compared to the frequency of the applied field, losses are small.

Variation of lattice parameter, average grain size, dielectric constant and saturation magnetization with different temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite by Co-precipitation method

Abdul Samee Flaws AbdulAziz,

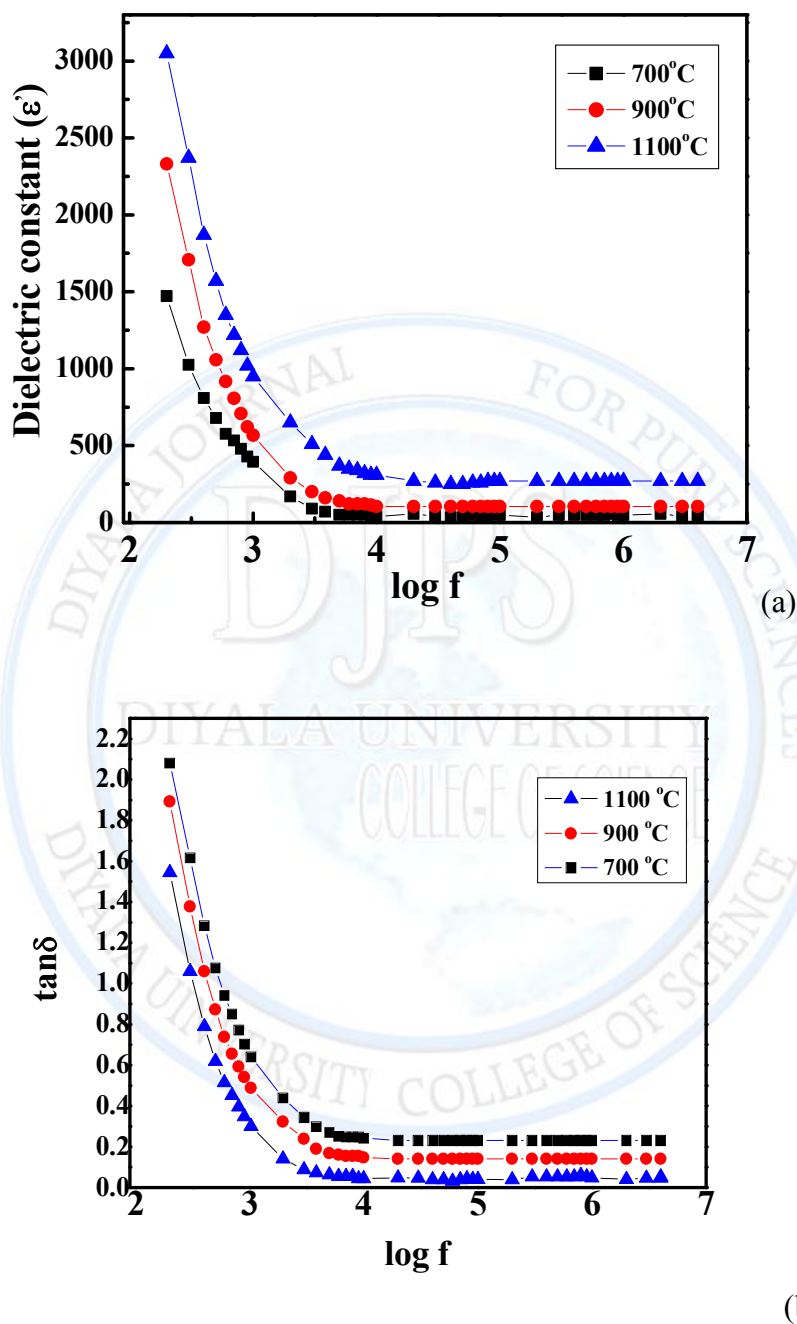


Fig.4 Variation of the (a) dielectric constant and (b) dielectric loss with frequency for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite annealed at different temperatures.

Fig. 5 (a–c) show the dielectric constant (ϵ') behavior as a function of temperature measured at 1 kHz, 10 kHz, and 100 kHz, and 1 MHz for the composites $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$

Variation of lattice parameter, average grain size, dielectric constant and saturation magnetization with different temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite by Co-precipitation method

Abdul Samee Flaws AbdulAziz,

+ 0.70PLZT sintered at various temperatures ranging from 700 to 1100 °C for 2 hours, respectively. The dielectric behaviour was found that as the grain size increases the dielectric constant increase and Curie temperature slightly increases. The transition temperature in present samples is 240 °C, 250 °C and 260 °C respectively, which are nearly equal to the transition temperature of PLZT phase. In the case of composites, the DPT type phase transitions is ascribed to the fact that ferroelectric regions are surrounded by non-ferroelectric regions (ferrite), similar to that reported earlier for different composites [11].

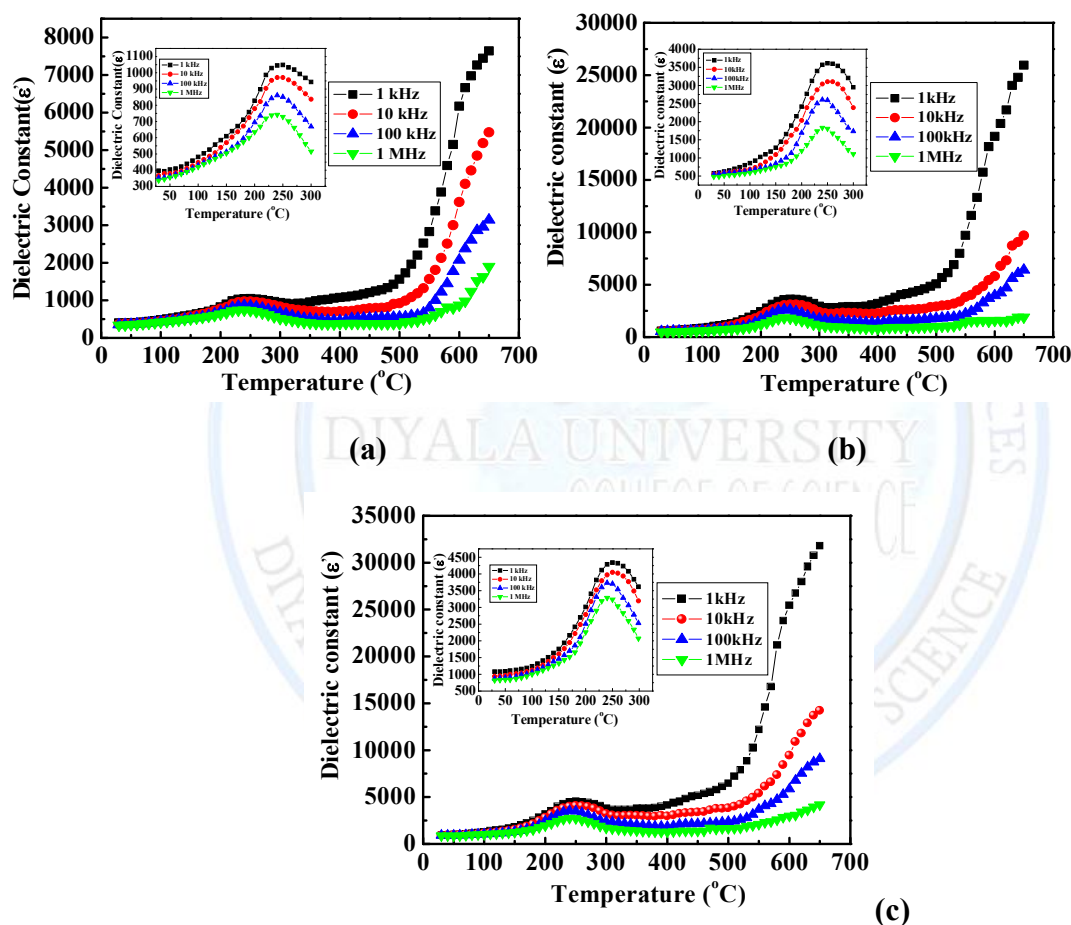


Fig.5 (a-c) Variation of the dielectric constant with temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite annealed at different temperatures (a) 700 °C, (b) 900 °C and (c) 1100 °C

Fig. 6 shows the variation of $\tan \delta$ as a function of temperature for the composites sintered at 1100 °C for 2 hours. The loss factor increases rapidly at high temperatures indicating the space charge conduction and is significantly smaller at high frequency of 100 kHz. Space charge conduction is related to the transport of defects such as oxygen vacancies to the

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Abdul Samee Flaws AbdulAziz,

dielectric-electrode interface [12-14]. In general, for smaller grain size the dielectric loss is lower because the grain boundary acts as pinning site for the domain wall movement [15-16]. From the data in Fig. 6, the $\tan\delta$ at 1 MHz and room temperature was found to be 0.641 for sample sintered at 700°C while 0.3 for sample sintered at 1100°C . According to this equation, dielectric loss would increase with decrease in relaxation time for a given frequency. Therefore, as the relaxation time decreases with increase in temperature, loss tangent increases. However, with further increase in temperature, $\tan\delta$ shows a decline after a certain maximum value. From the data in Fig. 6. An observed break in the plot is due to the instrumental limitation, as LCR bridge meter (model HIOKI 3532-50 LCR HiTESTER) cannot record value of $\tan\delta$ greater than 10. Similar results are reported in literatures [17-18].

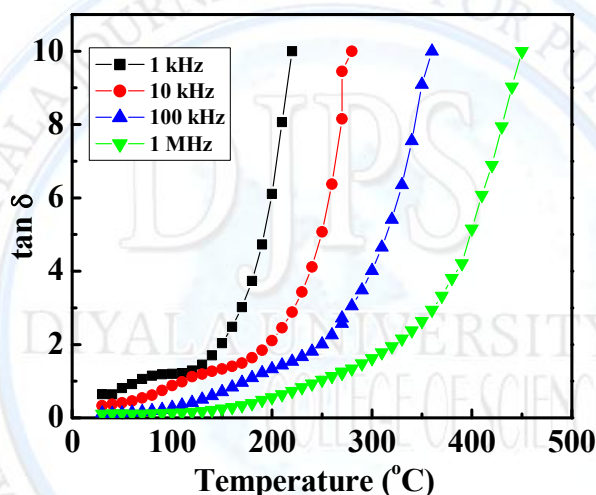


Fig. 6 Variation of the dielectric loss with temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite annealed at 1100°C .

3. 3 Ferroelectric properties

Fig. 7 shows P-E hysteresis of the $0.30\text{NZFO} + 0.70\text{ PLZT}$ samples annealed at various temperatures measured at room temperature. It was found that as the temperature increased the degree of loop squareness increasing indicates better homogeneity and uniformity of grain size nearly agrees with the surface morphological studies by SEM images. Also from the same figure we can see that as the annealing temperatures increases the remnant polarization (P_r) increases which indicate high internal polarizability, strain, electromechanical coupling and electrooptic activity. The decrease in coercivity with increasing in sintering temperature indicates grain growth at higher sintering temperature [19]. The saturation polarization (P_s) varies between $22.88\text{ }\mu\text{C}/\text{cm}^2$ and $27.63\text{ }\mu\text{C}/\text{cm}^2$, and remanent polarization (P_r) varies between $16.67\text{ }\mu\text{C}/\text{cm}^2$ and $20.13\text{ }\mu\text{C}/\text{cm}^2$ while the coercivity field (H_c) varied between

Variation of lattice parameter, average grain size, dielectric constant and saturation magnetization with different temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite by Co-precipitation method

Abdul Samee Flaws AbdulAziz,

20.75 kV/cm and 16.13 kV/cm. Similar behavior was observed in the dielectric constant versus temperature plot as shown in Fig 3(a-c). It was found that as the annealing temperature increased the dielectric constant increasing and Curie temperature slightly increasing.

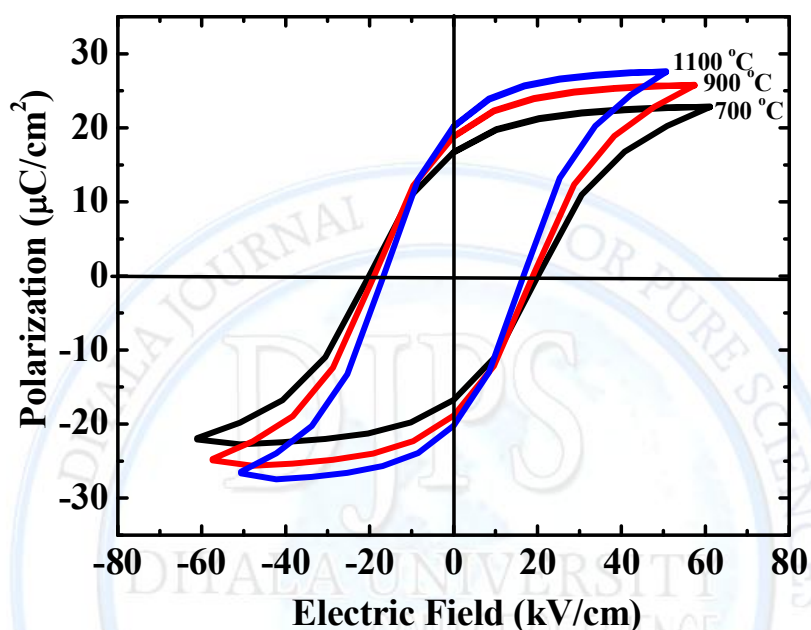


Fig. (7) P-E loops of $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite annealed at various temperatures measured at room temperature.

3. 4 Magnetic properties

Fig.(8) shows the isothermal magnetic field dependence of the magnetization for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite at different temperatures. The hysteresis loops of the composites shifts towards the magnetization axis with increasing temperature. The saturation magnetization (M_s) varied between 38.49 emu/gm and 76.15 emu/gm and the coercivity field (H_c) varied between 190.71 Oe and 441.43 Oe. The variation of saturation magnetization and coercivity field with sintering temperature for all samples is shown in Fig. (9). It is clear from Fig. (9) that the saturation magnetization and coercivity field of the samples depend on the sintering conditions and Microstructural changes brought by sintering process [20]. At higher sintering temperature, microstructure plays an important role in increasing magnetization. Since, grain growth occurs at higher sintering temperature, porosity of the samples decreases with increase sintering temperature [4]. In the present case, the grain size increases with increase sintering temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite and hence porosity decreases. The presence of pores break the magnetic circuits

**Variation of lattice parameter, average grain size, dielectric
constant and saturation magnetization with different temperature
for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite by Co-precipitation method**

Abdul Samee Flaws AbdulAziz,

between the grains, hence increase in porosity may reduces the net magnetization in bulk. The grain boundary area decreases with increase in grain size. Since the grain boundary is often associated with the pinning of domain walls, it follows that with increasing grain size, the pinning of domain wall motion decreases. Since coercivity H_c reflects amount and strength of pinning, we expect H_c to decrease with increase as grain size decreases. The results are in good agreement with the literature [21-23].

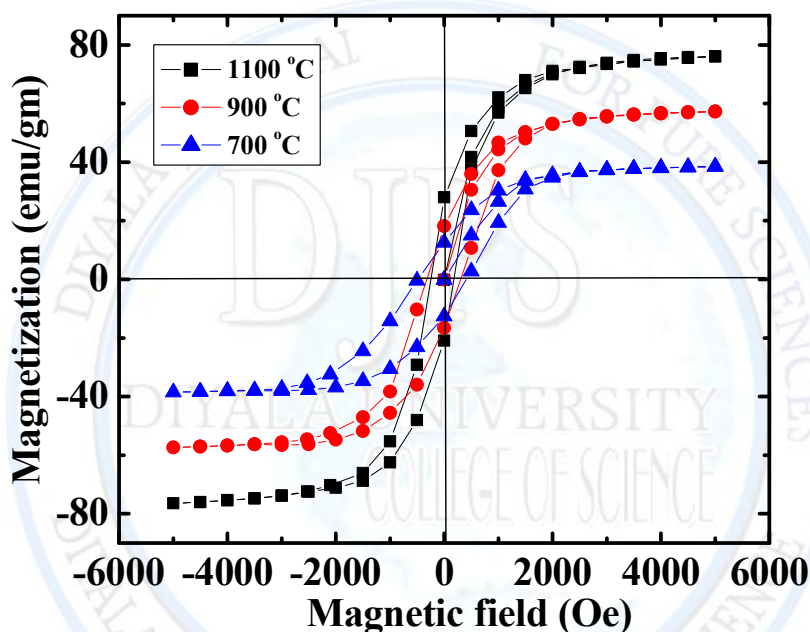


Fig.(8) Hysteresis loops of the $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composites annealed at various temperatures measured at room temperature

Variation of lattice parameter, average grain size, dielectric constant and saturation magnetization with different temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite by Co-precipitation method

Abdul Samee Flaws AbdulAziz,

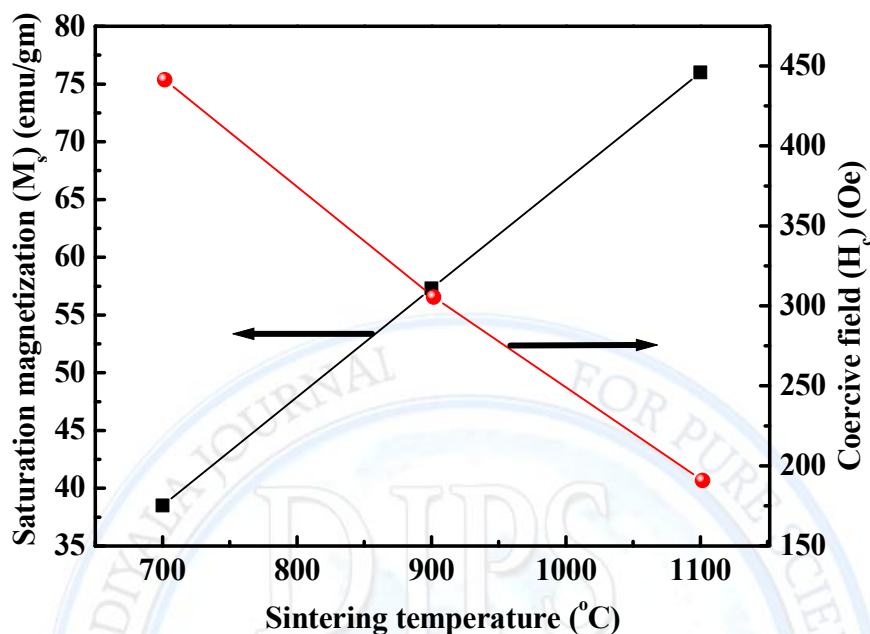


Fig. (9) Plots of saturation magnetization (M_s) and coercivity (H_c) of $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composites as a function of the annealing temperature.

3. 5 Magnetoelectric effect

The magnetoelectric effect in composites having ferrite and ferroelectric phase depends on the applied dc magnetic field, electrical resistivity, mole percentage of constituent phases, the relation between the particle size of individual phases and the efficiency of magnetoelectric conversion [24]. Hence in order to elucidate the relation between these parameters, measurements of magnetoelectric voltage coefficient are carried out with the variation of a applied dc magnetic field for all the samples.

It is known from the previous work [24], that there is an intimate relation between the piezoelectric grain size in the composite system and the magnetoelectric coefficient. In the present work, for a particular composite, the static ME output (dE/dH) increases with increasing piezoelectric grain size, which agrees with the earlier reported [5]. This is attributed to the fact that large grains can be polycrystalline and small ones cannot [25], hence, less effective in inducing a high piezoelectric coefficient. The variation of static magnetoelectric voltage coefficient at room temperature with applied dc magnetic field for $0.30\text{NZFO} + 0.70\text{PLZT}$ composite annealed at different temperatures is shown in Fig. (10). With increasing applied magnetic field we observed the strong magnetic field dependence of α_E up to 1140 Oe and after acquiring a maximum value, decrease. This initial rise in ME output is attributed to the enhancement in elastic interactions and confirmed by the hysteresis measurements. Therefore,

Variation of lattice parameter, average grain size, dielectric constant and saturation magnetization with different temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite by Co-precipitation method

Abdul Samee Flaws AbdulAziz,

high magnetization is useful to produce strong ME effect. In the spinels, the magnetization at a certain value of the magnetic field. Hence the strain produced in the ferrite phase would produce a constant electric field in the piezoelectric phase, there by decreasing the piezomagnetic coupling coefficient $q = \delta\lambda/\delta H$ [26]. For most ferrites (including $\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$), once the magnetostriction attains the saturative value, the q decreases and the piezomagnetic coupling gradually becomes weak, resulting in a decrease of the ME effect [27]. The maximum value $dc[(dE/dH)_H]$ of 4.08 mV/cm.Oe, 4.72 mV/cm.Oe and 5.17 mV/cm.Oe is observed in $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite sintered different temperatures viz. 700 °C, 900 °C and 1100 °C respectively.

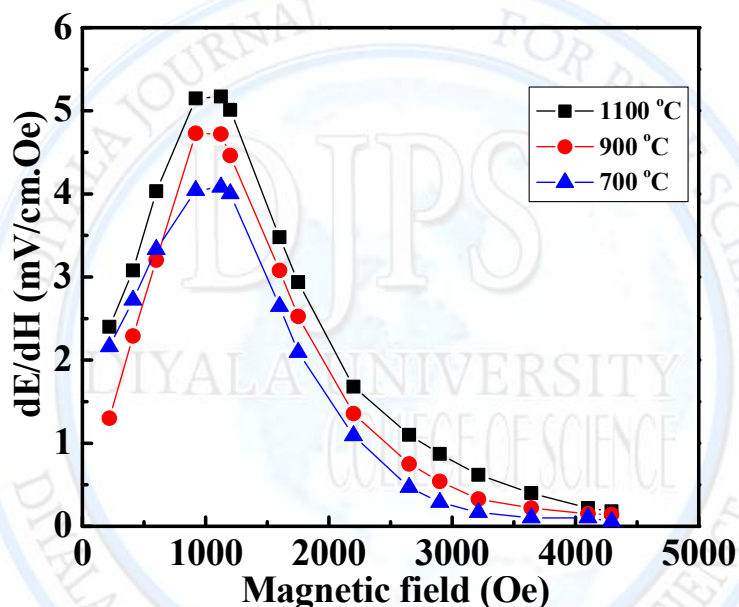


Fig. (10) Variation of ME voltage coefficient with applied magnetic field for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite annealed at different temperatures.

**Variation of lattice parameter, average grain size, dielectric
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Abdul Samee Flaws AbdulAziz,

4 Conclusions

The ME ceramic composites consisting of $\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4$ and PLZT as composites have been prepared by chemical method. Cubic spinel piezomagnetic phase and tetragonal perovskite piezoelectric phase formation was confirmed by XRD studies. There are no structural changes but slight variations in the lattice parameters of both the phases are observed for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{PLZT}$ composite annealed at different temperatures. SEM observation showed the particle size for PLZT is about ten times NZFO grain size. The variation of the dielectric constant and the dielectric loss with frequency showed dispersion in the low frequency range. All the samples exhibit strong magnetic characteristics. The coercivity and saturation magnetization have been found to vary with the sintering temperature due to the increase in crystallite size. Due to high magnetization and low coercivity these nanocomposites exhibited strong ME effect. Magnetoelectric measurements have shown that the ME effect in these nanocomposites strongly depends on applied magnetic field and sintering temperature.

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Variation of lattice parameter, average grain size, dielectric constant and saturation magnetization with different temperature for $0.30\text{Ni}_{0.7}\text{Zn}_{0.3}\text{Fe}_2\text{O}_4 + 0.70\text{ PLZT}$ composite by Co-precipitation method

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