# "Synthesis and Structural Characterization of (1-x) $NiFe_2O_4$ + (x) $KNbO_3$ Magneto electric Composites"

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**Abstract**

The spinel ferrite($NiFe_2O_4$) and perovskite potassium niobate ($KNbO_3$) as co-existing phases have been grown by solid state reaction over wide region compositions of the $NiFe_2O_4$+$KNbO_3$ system. The conformation of phase formation of $NiFe_2O_4$+$KNbO_3$ magnetelectric composites was confirmed by X-ray diffraction (XRD).The lattice constant, X- ray density, Bulk density, Porosity such structural analysis of both the phases and their composites were estimate. The scanning electron microscopy (SEM) used to study the microstructure of the composite.

***Keyword:*** *X-ray diffraction, Bulk density, Porosity, Scanning electron microscopy (SEM).*

**Introduction:**

The magneto-electric composites with piezoelectric and magneto-strictive material are of interest because of their applications in many fields. Magnetoelectric (ME) materials are capable of converting a magnetic field in to the electric field and vice versa. Essentially, ME effect properties originate when the materials conceives simultaneously both peizo-electric (PE) and peizo-magnetic (PM) properties either in single phase or composite phase system. The primary ME materials becomes magnetized when placed in electric field and electrically polarized when placed in magnetic field. In the secondary effect, the permeability or permittivity change is accepted. Magnetoelectric (ME) materials posses two or more iron based properties like polarization, magnetization, or strain as a result of electric field, magnetic field, and stress [1–2]. The interrelation between two spontaneous effects viz. ferroelectricity and ferromagnetism allows magnetic control of ferroelectric properties and vice versa. Recently, laminated ME composites synthesized by using piezoelectric and magneto-strictive materials have gained attention because they exhibit superior ME response [3–5]. The laminates are

generally fabricated by sandwiching and bonding piezoelectric plate/disk/fibers between two layers of magneto-strictive plates/disks/foils [6–8].

The ME effect is a property of ME composites, which is absent in their individual constituent phases [9]. Such magneto-electric composites are widely used in many applications like radio-electronic device, optoelectronic, microelectronic, transducers, etc. [10, 11]. The ME effect occurs due to the interaction between the magnetic and electric dipoles [12].

Literature survey on ME composite material does not contain any systematic work on $NiFe_2O_4 + KNbO_3$ ME composite and hence it was decided to carry out a systematic investigations of structural properties of $NiFe_2O_4 + KNbO_3$ ME composite with a view to understand the magneto-electric behaviour and their co-relation with the other properties.

In the present work, an attempt is made to prepare ME composite materials of nickel ferrites ($NiFe_2O_4$) and potassium niobate ($KNbO_3$) composite with varying composition by standard ceramic technique. The structural characterization studied by X-ray diffractions and S.E.M and to investigate magneto-electric effect.

**Preparation of ME composites:**

ME composites with compositions (1-x) $NiFe_2O_4$+(x) $KNbO_3$ with (x= 0.0, 0.2, 0.4, 0.6, 0.8and 1 mole %) was prepared by solid state reaction method. Fine powders of nickel ferrite ($NiFe_2O_4$) and potassium niobate ($KNbO_3$) were mixed thoroughly in molar proportion and ground for about 3 hours, 2 wt. % polyvinyl alcohol was as a binder in the mixed powders. The mixed powder is then press into pellets of thickness of around 2-3 mm and diameter 10mm using a hydraulic press. A pressure of 5 ton/cm$^2$ was applied. The palletized samples were sintered at 1050$^o$ C for 24 hours in a programmable furnace. The pellets were lastly furnace cooled to room temperature.

**Characterization techniques:**

The prepared ME composite of (1-x) $NiFe_2O_4$+(x) $KNbO_3$ were characterized by using X-ray diffractometer (Philips Model PW1710). The X-ray diffraction patterns were recorded in the 2θ range of 20$^o$ to 80$^o$ using $CuK_\alpha$ radiation. The XRD patterns are taken at room temperature. The x-ray diffraction (XRD) pattern of the composite phase containing20% $NiFe_2O_4$+80%$KNbO_3$ is as shown in Fig.1.

**Result and Discussion:**

***Structural Characteristics:***

Magneto-electric composites of nickel ferrite ($NiFe_2O_4$) and potassium niobate ($KNbO_3$) were characterized by X-ray diffraction method. Figure1. X-ray diffraction patterns of composite (1-x) $NiFe_2O_4$+(x) $KNbO_3$ for x=0.0, 0.2, 0.4, 0.6, 0.8 and 1.0. All the XRD patterns of the composites under investigation show sharp and intense reflections, all the reflections were indexed using powder-X program. The Inter-planer spacing'd' values slightly changing as percentage of ferroelectric ($KNbO_3$) increases. The inter-planer spacing values are used to calculate the lattice constant of the samples.

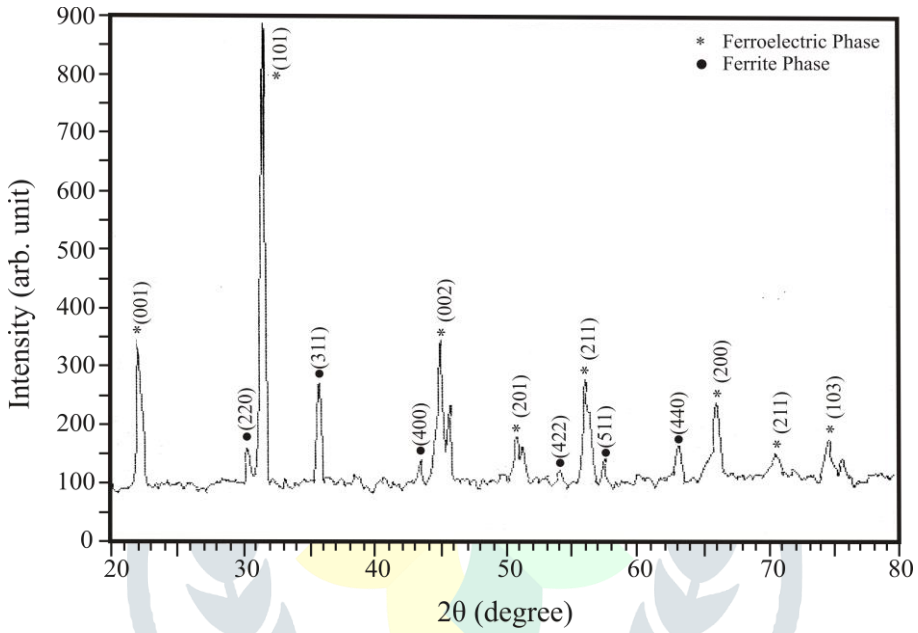


***Fig.1. XRD Patterns of (1-x) $NiFe_2O_4$ +(x) $KNbO_3$ (x = 0.8)***

It is reveled from the XRD patterns that the composite consist of two separate phases which corresponds to ferrite phase ($NiFe_2O_4$) and ferroelectric phase ($KNbO_3$). No extra peaks other than the cubic spinel structure of ferrite and tetragonal structure of ferroelectric is seen in the XRD pattern. A close examination of XRD patterns indicate that the intensity of the most intense peak (101) of a $KNbO_3$ increases with increase in $KNbO_3$ percentage, whereas the intensity of the most intense peak (311) of ferrite ($NiFe_2O_4$) phase decreases with decreasing percentage of ferrite. The XRD patterns of composite material under investigation are as per the ASTM (74-2081 for $NiFe_2O_4$, 71-0945 for $KNbO_3$) data.

The XRD patterns were used to determine Lattice constant of ferrite phase and ferroelectric phase. The XRD pattern shows only the $NiFe_2O_4$ phase for x = 0.0 and for x = 1.0, it shows ferroelectric phase. The value of lattice constant obtained from XRD data are given in the Table 1. Since XRD analysis revealed that nickel

ferrite possess cubic spinel structure. XRD data was used to obtain lattice parameter of ferrite phase. The values of lattice parameter 'a' for x =0.0, i.e. for pure ferrite phase is in close agreement with the reported values [13]. The lattice constant for other composition slightly changes with the addition of ferroelectric content $KNbO_3$. The lattice constant of ferroelectric phase was also calculated using XRD data. The values of lattice parameter 'a' and 'c' for ferroelectric phase are almost same for all the compositions. The X-ray density of ferrite phase, ferroelectric phase and composite material was calculated using the value of molecular weight and volume of the samples. The values of X-ray density for ferrite phase, ferroelectric phase and composite phase are given in Table 1. It is observed from table that the X-ray density of ferrite phase decreases with an addition of $KNbO_3$. Similarly the X-ray density of ferroelectric phase and ME composite phase also decreases with the addition of KNbO3.The decreases in X-ray density of ferrite phase is related with the decrease in mass overtakes the decrease in volume of the ferrite phase. The decrease in X-ray density of ferroelectric phase is related with the molecular weight and volume of the ferroelectric phase. The molecular weight of ferroelectric phase increases the addition of $KNbO_3$ at the same time volume also increases with the addition of ferroelectric phase. The decrease in X-ray density of ferroelectric phase is due to the predominance of increase in volume over the increase in molecular weight. The molecular weight of composite material goes on decreasing with the addition of $KNbO_3$ similarly volume of composite also decreases with addition of $KNbO_3$. Thus both mass and volume of composite decreases with increase in $KNbO_3$, the decrease in mass (molecular weight) overtakes the decrease in volume and hence the X-ray density of composite decreases with addition of $KNbO_3$.

**Table 1: Structural data for (1-x) $NIFe_2O_4$+(x) $KNbO_3$ (x=0.0-1.0)**

| Comp. 'x' | Lattice parameter (Å) | | X-ray density '$d_x$' ($gm/cm^3$) | Bulk density 'd' ($gm/cm^3$) | Porosity P (%) | Particle size 't' (Å) | Average grain size (μm) |
|---|---|---|---|---|---|---|---|
| | Ferrite | Ferroelect. | | | | | |
| $CuFe_2O_4$ | a = 8.353 | ------ ----- | 5.340 | 4.225 | 20.884 | 280 | ---- |
| 0.0 | a = 8.348 | a =3.955 c = 4.066 | 5.207 | 3.505 | 32.692 | 291 | 2.14 |
| 0.2 | a = 8.343 | a =3.980 c = 4.092 | 5.113 | 3.396 | 33.833 | 277 | 2.17 |
| 0.4 | a = 8.335 | a = 3.982 c = 4.081 | 4.914 | 3.637 | 25.983 | 221 | 2.38 |
| 0.6 | a = 8.320 | a = 3.987 c = 4.082 | 4.755 | 3.569 | 24.929 | 218 | 2.62 |
| 0.8 | ------ ----- | a = 3.989 c = 4.084 | 4.598 | 3.584 | 22.049 | 218 | 3.1 |

The bulk density for ferrite phase, ferroelectric phase and their composites was obtained by Archimedes liquid immersion method and the values are presented in Table 1.

The percentage porosity 'P' was also determined for each composition 'x'. Table 1 indicates the large value of porosity.

The scanning electron microscopy is one of the powerful techniques used to analyze the microstructure of the composite. Fig.2. (a, b, c and d) shows series of SEM micrograph of the (1-x) $NiFe_2O_4$+(x) $KNbO_3$ with x =0.2, 0.4, 0.6 and 0.8. The average grain size was calculated and listed in Table 1. The grain size is found to increase with increasing $KNbO_3$ content. The grain size for (1-x) $NiFe_2O_4$+(x) $KNbO_3$ with x =0.2, 0.4, 0.6 and 0.8 is lies between 3.1 -2.14 μm. The grain size of composites is found to decrease with increasing ferrite content. This may due to the fact that the average grain size of the ferrite phase is smaller size of the ferroelectric phase and the composites are prepared in ferroelectric rich region.

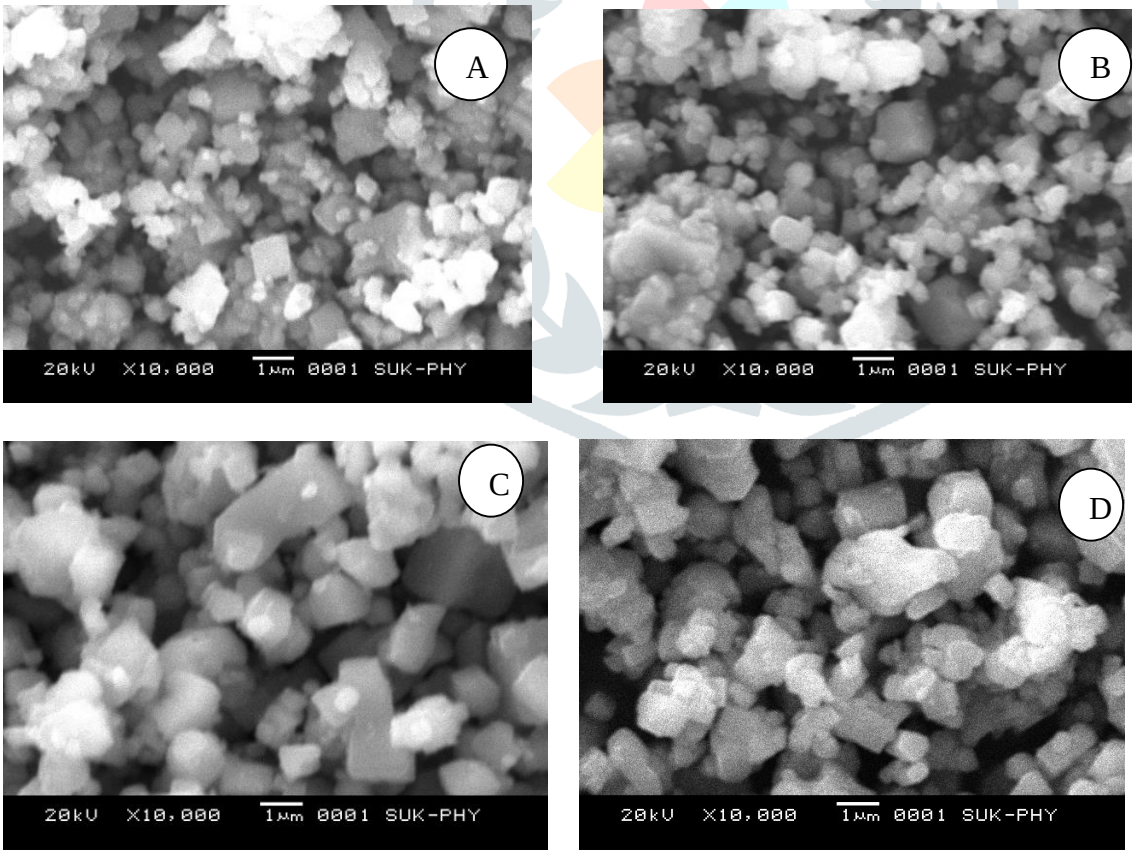


***Fig. 2: SEM micrographs of (1-x) Ni $Fe_2O_4$ + (x) $KNbO_3$ With (A) x= 0.2, (B) x = 0.4, (C) x= 0.6 , (D) x = 0.8***

**Conclusions:**

The composite material of (1-x) $NiFe_2O_4$ + (x) $KNbO_3$ was successfully synthesized by standard ceramic technique. The low anisotropy favours magnetization mechanism in the crystal form such as domain wall moment and domain rotation which are the key factors for magneto- mechanical coupling, that give rise to the ME effect. Soft ferrite i.e. Nickel ferrite ($NiFe_2O_4$) have low anisotropy and good magnetic properties which makes it a promising candidate for composite material. Perovskite is a family name of a group of materials of which potassium niobate ($KNbO_3$) is one of the members. The crystal structure of potassium niobate ($KNbO_3$) is a tetragonal at room temperature the Curie temperature of potassium niobate ($KNbO_3$) is $435^0$C.The structural parameters like lattice constant, X-ray density, bulk density, and porosity derived in the present study closely agrees with reported values. Well defined grains appeared in the SEM photographs. Average grain size derived from SEM images gives grain size.

**References:**

[1] Wang J., Zheng H., Lofland S. E., Ma Z., Ardabili L. M., (2004) Science 303:661.

[2] Ederer C., Spaldin N.; (2004) Nat Mater 3:849.

[3] Eerenstein W., Mathur N. D.; Scott JF Nature 442 (2006) 759.

[4] Hur N., Park S., Sharma P. A.; Ahn JS, Guha S, Cheong SW; Nature 429(2004) 392.

[5] Ryu J., Priya S., Uchino K., Viehland D., Kim H.; J Korean Ceram Soc 39(2002) 813

[6] Ryu J., Priya S., Uchino K., Kim H; J. Electroceram 8(2002) 107.

[7] Srinivasan G., Rasmussen E., Levin B., Hayes R.; Phys Rev B 65 (2002) 134402.

[8] Dong S., Zhai J., Li J. Viehland D.; J Appl Phys 88(2006)082907

[9] Nan C-W; Phys Rev B 50(1994)6082

[10] Van Suchetelene; Philips J (1972) Res Rep 27:28

[11] Schmid H.; Bull Mater Sci 17(7) (1994)1411

[12] Suryanarayana S. V.; Bull Mater Sci 17(7) (1994)1259

[13] Manish Srivastava, Animesh K. Ojha, Chaubey, A. Materny; J. Alloy. Comp. 481 (2009) 515.