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Synthesis and characterization of (y) Ni_{1-x}Cd_xFe₂O₄ and (1-y) Ba_{0.8}Sr_{0.2}TiO₃ Magnetoelectric composites prepared through combustion method

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Abstract

The magnetoelectric composites of ferrite and ferroelectric phase with [y] $Ni_{1-x}Cd_xFe_2O_4$ and [1-y] $Ba_{0.8}Sr_{0.2}TiO_3$ (x =0.2, 0.4, 0.6, y =15%, 30%, 45%) were synthesized by self-propagating auto combustion method. The formation of spinel phase for ferrite and pervoskite phase for ferroelectrics were confirmed by X- ray diffraction. The surface morphology was studied by scanning electron microscopy with average grain size about 180nm. The dispersion of dielectric constant with frequency shows the maxwel-wegner type interfacial polarization. The magnetic properties were analyzed by the B-H hysteresis curve. The static value of magnetoelectric conversion factor (dE/dH) was measured as a function of applied magnetic field. Also the dependence of ME coefficient with resistivity is discussed.

Keywords: Composites, XRD, SEM, Resistivity, Hysteresis, ME effect etc.

Introduction

The magneto electric composites are of great interest as transducers that transform the changes in the magnetic field into electric voltage The ME effect can be realized in composite based on product properties. Suitable combination of piezomagnetic and piezoelectric phase can yield desired ME property. The properties of composite materials not only depend on the properties of their individual parents but also their morphology and interfacial characteristics. The magnetoelectric materials that contain both electric permittivity and magnetic permeability greater than unity (magneto-dielectrics) are currently being investigated for microwave applications [1]. Magnetoelectric laminate composites of piezoelectric-magnetostrictive

materials were investigated highest magnetoelectric voltage coefficient of 4.68 V/cm [2]. The single domain particles of the ferrite phase together with large lattice distortion serve as better candidates in piezomagnetic effect and hence enhance the ME output [3-5]. The effect of grain size on piezoelectric, dielectric and ferroelectric properties has been widely studied in literature [6-10]

Now days some interesting results were developed related to Hysteresis measurements to determine saturation magnetization (M_s), magnetic moment (μ) and the variations of saturation magnetization and magnetic moment were interpreted in terms of the composition of the samples [11]. The attractive ME response were produced for the ceramics of NiFe₂O₄ (NFO) and lead zirconate titanate (PZT) by in situ processing based on sol-gel method [6].

There are various methods used for synthesis of magnetoelectric composites such as sol-gel method [12], weight chemical method [13, 14] and ceramic method [15-17] etc. Among these, the self-propagating auto combustion route is widely used. This method is ease of cost and gives the fine nature of the ferrite as well as ferroelectric particles. In the present communication we report on preparation, structural and magnetic properties of [y] $Ni_{1-x}Cd_xFe_2O_4$ and [1-y] $Ba_{0.8}Sr_{0.2}TiO_3$ (x =0.2, 0.4, 0.6, y =15%, 30%, 45%) magnetoelectric composites.

Materials and Methods

Experimental procedure

The high purity AR grade Ni (NO₃)₂ 6H₂0, Cd (NO₃)₂ 4H₂O, Fe (NO₃) 9H₂O were used to prepare Ni_{0.8}Cd_{0.2}Fe₂O₄. The raw material is heated to evolve NO₂, CO₂ and H₂O at 800C. The 10% PVA and sucrose were added to the nitrates solution. The sucrose acts as a fuel for the combustion purpose and that of PVA is to form polymer regions with the metal ions. The dried and fluffy gel is burnt in self-propagating combustion manner at about 200^oC. The prepared ash was calcined about 600^oC for 6 hours. On the other hand same procedure is followed to make Ba_{0.8}Sr_{0.2}TiO₃ by using Ba (NO3)₂, Sr (NO₃)₂ and Ti (NO₃)₂ as precursor material. The prepared ash was calcined about 1000^oC for 10 hours. The mixture of (y) Ni_{1-x}Cd_xFe₂O₄ + (1-y) Ba_{0.8}Sr_{0.2}TiO₃ (x =0.2, 0.4, 0.6 and y = 15%, 30% and 45%) were uniaxially pressed into pellets of thickness of 2-3 mm and diameter 10 mm using hydraulic press and annealed at 950^oC for 9 hours.

Characterization

The crystalline phase of the samples was determined by XRD (Model- PW-1037) with Cu-k α radiation. The microstructures were characterized by SEM (Model- JEOL JSM 6360). The resistivity measurements were carried out by the two probe method in the temperature range of 40° C to 700° C and was calculated as,

$$\rho = R \pi r^2 / t \tag{1}$$

Where ρ = resistivity of samples, r is the radius of the sample and t is the thickness of the sample. The room temperature saturation magnetization was carried out by using high field hysteresis loop tracer designed by TIFR, Mumbai.

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The ME output of the samples was measured as a function of dc magnetic field. The electric poling was carried in an electric field of 3KV/cm during cooling of the sample from 30 to 40^{0} C above the Tc of ferroelectric phase to room temperature. The composites were magnetically poled by applying dc magnetic field of 1KOe at room temperature. The static ME voltage coefficient was calculated by the change in ME output with respect to dc bias magnetic field.

Results and discussion

3.1) Structural analysis

Fig.1 shows XRD patterns for (y) $Ni_{1-x}Cd_xFe_2O_4 + (1-y) Ba_{0.8}Sr_{0.2}TiO_3$ (x =0.2, 0.4, 0.6 and y = 15%, 30% and 45%) composites.



Fig.1 a) XRD pattern for (y) $Ni_{0.8}Cd_{0.2}Fe_2O_4+$ (1-y) $Ba_{0.8}Sr_{0.2}TiO_3$ composite (y = 15%, 30%, 45%)

No secondary peaks were found other than spinel and pervoskite structure. The intensity of BST peak (100) decreases with increase in ferrite content. The intensity of ferrite peaks increases with

Cd content y	Lattice parameter (Å)			
	Ferrite phase	Ferroelectric phase		
	a	а	с	c/a
15%	8.342	3.982	4.144	1.04
30%	8.342	3.971	4.107	1.03
45%	8.346	3.961	4.079	1.02
15%	8.370	3.989	4.114	1.03
30%	8.373	3.976	4.072	1.02
45%	8.376	3.973	4.056	1.01
15%	8.381	3.996	4.128	1.03
30%	8.387	3.982	4.081	1.02
45%	8.384	3.894	4.065	1.04
	t y 15% 30% 45% 15% 30% 45% 15% 30% 45%	t y Ferrite phase a 15% 8.342 30% 8.342 45% 8.346 15% 8.370 30% 8.373 45% 8.376 15% 8.381 30% 8.387 45% 8.384	t y Ferrite phase Ferror a a a 15% 8.342 3.982 30% 8.342 3.971 45% 8.346 3.961 15% 8.370 3.989 30% 8.373 3.976 45% 8.376 3.973 15% 8.376 3.973 30% 8.373 3.976 45% 8.376 3.973 15% 8.381 3.996 30% 8.381 3.996 30% 8.381 3.982 45% 8.384 3.894	Lattice parameter (Å)Ferrite phaseFerroelectric phaseaac15%8.3423.9824.14430%8.3423.9714.10745%8.3463.9614.07915%8.3703.9894.11430%8.3733.9764.07245%8.3763.9734.05615%8.3813.9964.12830%8.3873.9824.08145%8.3843.8944.065

increase in its content in composites. In addition, the dependence of lattice parameter for ferrite and ferroelectric phase in composite shown in table 1 below.

3.2) Scanning Electron Microscopy

Fig. 2 shows the scanning electron micrographs for (y) $Ni_{1-x}Cd_xFe_2O_4 + (1-y)$ Ba_{0.8}Sr_{0.2}TiO₃ (x =0.2, 0.4, 0.6 and y = 15%, 30% and 45%) composite.

The grain size plays an important role to create best performance of the composite. The grain size decreases with increases in cadmium content in composite reveal the increase in grain boundary. This would be responsible to increase resistivity and magnetocrystalline anisotropy of the composites.



Fig. 2 SEM micrograph for (y) $Ni_{1-x}Cd_xFe_2O_4+$ (1-y) $Ba_{0.8}Sr_{0.2}TiO_3$ composite (x = 0.2, 0.4, 0.6 and y = 15%, 30%, 45%)



Fig.3.) variation of Resistivity with temperature for (y) $Ni_{1-x}Cd_xFe_2O_4+$ (1-y) $Ba_{0.8}Sr_{0.2}TiO_3$ composite (x = 0.2, 0.4, 0.6)

3.3) Resistivity dependence

The temperature dependent resistivity measurements were carried out for (y) $Ni_{1-x}Cd_xFe_2O_4$ and (1-y) $Ba_{0.8}Sr_{0.2}TiO_3$ (x = 0.2, 0.4, 0.6) samples by the two probe method in the range of $40^{0}C$ to $700^{0}C$ as shown in Fig.3.

When cadmium added into the ferrite phase in the composite then there is increase in resistivity of composite. Linear decrease in resistivity with temperature shows semi conducting nature of the samples. Transition from Para to Ferro occurs after crossing the ferroelectric temperature.

4.1) Magnetic characterization

Fig. 4 shows the B-H curve for saturation magnetization with ferrite content in the composite. The interaction between two magnetic ions through non-magnetic ion leads to super exchange interaction produces large surface spin order [18].the substitution of cadmium leads to increase Fe^{3+} ions in B-site, which create more magnetization on B- site, compare to A site.





The increase in cadmium content in composite increases magnetocrystalline anisotropy decreases the domain wall energy, which produces high coercive field.

4.2) Magnetoelectric measurements

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Fig. 5 shows the variation of magnetoelectric voltage coefficient with applied magnetic field for (y) $Ni_{1-x}Cd_xFe_2O_4$ and (1-y) $Ba_{0.8}Sr_{0.2}TiO_3$ (x= 0.2, 0.4, 0.6 and y = 15%, 30% and 45%) samples. Magnetoelectric conversion is the result of piezomagnetic strain in the spinal phase, which create piezoelectric charge in ferroelectric phase and latter would depend upon magnetic field.



Fig.5 a) Variation of magnetoelectric coefficient with Magnetic field for(y) $Ni_{0.8}Cd_{0.2}Fe_2O_4 + (1-y) Ba_{0.8}Sr_{0.2}TiO_3$ composite (y = 15%, 30%, 45%)

Fig.5 b) Variation of magnetoelectric coefficient with Magnetic field for(y) $Ni_{0.8}Cd_{0.4}Fe_2O_4+$ (1-y) $Ba_{0.8}Sr_{0.2}TiO_3$ composite (y = 15%, 30%, 45%)

Fig.5 c) Variation of magnetoelectric coefficient with Magnetic field for(y) $Ni_{0.8}Cd_{0.6}Fe_2O_4+$ (1-y) $Ba_{0.8}Sr_{0.2}TiO_3$ composite (y = 15%, 30%, 45%)

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The decrease of Tc for ferroelectric phase affects low value of dE/dH. The further increase in dE/dH is due to increase of resistive cadmium content in the composite. The maximum value of ME coefficient (588μ V/cm) is observed for (45%) Ni_{0.8}Cd_{0.6}Fe₂O₄ and (55%) Ba_{0.8}Sr_{0.2}TiO₃.The high value of resistivity prevents the piezoelectric phase leakage. in composite. The increase in ME coefficient is due to connection between highly resistive ferrite phase (Table.1) with ferroelectric phase.



Fig.6 shows variation of magnetoelectric voltage coefficient and resistivity with Cd content

Also the dependence of magnetoelectric coefficient and resistivity with cadmium content is studied from fig. 5 above It is clear that addition of non-magnetic cadmium into the matrix produce high resistivity that would increase the magnetoelectric output.

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References

- [1] M. C. Golt, J. Thermoplastic Composite Materials, Vol. 22, No. 6, (2009) 551-567.
- [2] Jungho Ryu, Alfredo Vázquez Carazo, Kenji Uchino and Hyoun-Ee Kim, *Jpn. J. Appl. Phys.* 40 (2001) 4948-4951.
- [3] K.K. Patankar, V.L. Mathe, R.N. Patil and B.K. Chougule, *Mater. Chem. and Phys.*, Vol.96, Issues 2-3, (**2006**), 197-200.
- [4] R. S. Devan et al, *Smart Mater. Struct.* 15 (2006) 1877-1881.
- [5] R S Devan et al, J. Phys. D: Appl. Phys. 41 (2008) 105010 (8pp).

- [6] B.S. Kang, D.G. Choi, S.K. Choi, K. Kor. Phys. Soc. (1998) 32:S232.
- [7] H. T. Martirena, J.C. Burfoot J. Phys. C: Solid State Physics (1974) 7:3182.
- [8] S. Choudhury, Y.L. Li, C. Krill, L.Q. Chen, Act. Mater. (2007) 55:1415.
- [9] H.M. Duiker, P.L. Beale Phys. Rev. B 41(1) (1990) 490.
- [10] V. Sundar, N. Kim, C.A. Randall, R. Yimnirun, R.E. Newnham, Proceeding of the 10th IEEE international symposium on application of ferroelectrics (1996) 935.
- [11] B.K. Bammannavar, G.N. Chavan, L.R. Naik and B.K. Chougule, *Mater. Chem. Phys.* Vol.117, Issue 1(**2009**) 46-50.
- [12] Dandan Wu et al J. Phys. D: Appl. Phys. 40 (2007) 5002-5005.
- [13] S.V. Suryanarayana Bull. Mater. Sci., 1.7 (7) (1994) 1259.
- [14] A. C. F. M. Costa, E. Tortella, M. R. Morelli, R. H. G.A. Kiminami J. Mag. and Mag. Mater. 256 (2003) 174-182.
- [15] J.V. Boomgaard, R. A. J. Born J. Mater. Sci., 13 (1978) 1538.
- [16] V. M. Laletin Sovt. Techn. Phys. Lett. 18 (8) (1992) 484.
- [17] J.V. Boomgaard, A. M. J. G. Van Run and J. V. Suchetelene, Ferroelectrics, 10 (1976) 295.
- [18] L.K. Leung, B.J. Ewans and A.H. Morrish Phys. Rev. B8 (1973) 29.