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Structural, dielectric and magnetic properties of 0.3CoFe₂O₄-0.7BaTiO₃-PVDF composite film

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ABSTRACT

Structural, Dielectric and magnetic properties 0.3 CoFe₂O₄–0.7 BaTiO₃–PVDF (polyvinylidene fluride) composite film with different concentration of PVDF: 20, 30 and 40 wt% are reported here for the first time. The structural analysis was carried out using X-Ray diffraction technique, which indicates cubic spinel structure for ferrite phase CoFe₂O₄ (CFO) and tetragonal structure for ferroelectric phase BaTiO₃ (BT). The average grain size was observed to be (~106 nm, 30 nm and 26 nm) for 20%, 30% and 40% addition of PVDF by using AFM analysis. The dielectric constant variation with temperature at three fixed frequencies (1 kHz, 50 kHz and 100 kHz) was studied and it was found that the dielectric constant and dielectric loss decrease with increasing amount of polyvinylidene fluride. The values of ac conductivity for 0.3CoFe₂O₄ – 0.7BaTiO₃ –PVDF composite film were found to decrease with increasing concentration of PVDF. The ferroelectric hysteresis loops also indicate that the value of polarization decreases with the addition of PVDF and the value of remnant polarization for 20% PVDF was found to be $0.5286 \ \mu\text{C/cm}^2$. The magnetocapacitance of $0.3 \ \text{CoFe}_2O_4$ – $0.7 \ \text{BaTiO}_3$ – $0.3 \ \text{PVDF}$ was found higher for this composition. Copyright © 2014 VBRI press.

Keywords: Composite film; dielectric properties; ferroelectric; multiferroic.



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carrier, he has published more than 140 papers in SCI Journals and is an author of one contributory chapter in a book of International level in the field of materials science and technology. His major research interests are Functional Nanomaterials, Multiferroic, Smart Sensors and Biomaterials. His recent research interest is focused on designing and development of smart materials for device applications. He is an active member and leader in the Materials Engineering community. He contributes regularly to a number of professional societies (e.g., MRSI, and Discussion Group), journals and universities.

Introduction

Multiferroics are attracting interest and provoking much research activity driven by profound physics of these materials due to the coexistence and coupling of ferroelectric and magnetic orders and the potential applications in novel multifunctional devices such as sensors, transducers, memories and spintronics [1]. The magnetoelectric effect observed in most of the single phase multiferroic materials is very small, which is likely too weak for any practical application. So nowadays magnetoelectric (ME) composites made by combining piezoelectric and magnetic substances together have drawn significant interest in recent years. ME effect in composites is due to induced strain in the piezomagnetic phase by the application of magnetic field that is mechanically coupled to induce stress in the piezoelectric phase that results in the generation of induced voltage. The ME effect in composites can be written as:

ME effect = $\frac{magnetic}{mechanical} \times \frac{mechanical}{electric}$, or

 $\frac{electric}{mechanical} \times \frac{mechanical}{magnetic}$

The multiferroic composite was reported in 1970s, consisting of a piezoelectric phase BaTiO₃ (BT) and ferromagnetic phase CoFe₂O₄ (CFO) [2]. CFO is known to have one of the largest magnetostriction among magnetic materials and a high Neel temperature (520 °C), and it is a perfect choice as ferromagnetic phase and BT has been considered to be an important ferroelectric material for tunable microwave devices because of its high dielectric constant and low dielectric loss. BT has been utilized as an excellent dielectric material in electronic industry but BaTiO₃ ceramic needs to be sintered at a high temperature and that process consumes vast power. In addition, the ceramic materials have lots of disadvantages, such as poor flexibility which are not well suited for fabrication of electronic devices. То address these issues, ceramic/polymer composites have been evaluated as candidate dielectric materials for new type of capacitors, since the combination of polymer and ceramic may give the composites some advantages from both sides by a careful design and preparation [3]. Many efforts have gone into the development of BaTiO₃/polymer composites [4-6], which combine the advantages of polymers such as flexibility, ease of processing and high dielectric constant with the good piezoelectric characteristics of BaTiO₃. With the addition of polymer we want to enhance the ME coupling and here we have taken PVDF polymer because it is a semicrystalline polymer with one of the largest pyro and piezoelectric properties among the polymers [7].

Moreover polymers are flexible, easy to process with low processing temperature, and possess high dielectric breakdown field. In the last few years, the potential of electro active polymers has been recognized for biomedical applications due to its ability to convert mechanical, thermal, or magnetic signals into electrical ones. In this sense, these materials can be used as smart scaffolds to stimulate cell growth and compatibility and in biosensors, mechanical sensors, and actuators [8]. Polyvinylidene fluoride (PVDF) works as matrix to improve the magnetoelectric sensitivity in composites [9]. Barium titanate polymer composites are of great interest as capacitor materials because they exhibit high permittivity. In past few year several efforts have been made by making composite with PVDF to reduce dielectric loss such as in PVDF – La_{0.7}Sr_{0.3}MnO₃ [10], BiFeO₃-PVDF [11], PVDF and nickel (Ni) composites [12], Hot-Pressed BiFeO3-PVDF Composite films [13], PVDF-Fe₃O₄ Composite films [14], PVDF-BaCa₂Fe₁₆O₂₇ composite thick films [15], PVDF-TrFE/CoFe₂O₄ ME composites [16], BaTiO₃-CoFe₂O₄-polyvinylidene fluoride composite [17], $(Bi_{0.5}Na_{0.5})_{0.94}Ba_{0.06}TiO_3$ -PVDF 0-3 composites [18]. Dielectric and magnetic properties of ferrite/polyvinylidene fluride nanocomposites have been reported in the work of P. Martins et al [19]. We have already prepared the composite of $0.3CoFe_2O_4$ –0.7BaTiO₃ and we found that there is less ME effect and high value of dielectric loss. In addition to enhance the ME effect and reduce the dielectric loss we choose 0.3CoFe₂O₄ -0.7BaTiO₃-PVDF with different wt% of PVDF. When the two phases of BT and CFO are dispersed into the polymer matrix, due to the improved displacement transfer capability of flexible polymer, the Magneto-electric coupling of BT-CFO composite improves by an order of magnitude. Embedding

the ceramic powders in a polymer matrix reduces the porosity and provides a ductile matrix to blind the two brittle constitute together [17]. With the addition of PVDF we have found low value of dielectric loss in the composite and enhanced value of the magnetocapacitance. In this paper we report the synthesis and characterization of composite film of 0.30CoFe₂O₄ -0.70 BaTiO₃-PVDF with 0, 20, 30 and 40 wt% of PVDF. We have studied the phase formation, dielectric, ferroelectric, magnetic properties of the synthesized material.

Experimental

The composite of $0.3\text{CoFe}_2\text{O}_4$ -0.7BaTiO₃ was prepared through a solid state reaction method. For ferrite phase stoichiometric amounts of CoO (Cobalt (II) Oxide, A.R., 98.5%, Himedia, India) and Fe₂O₃ (Iron (III) Oxide, 98%, Himedia, India) were weighed and mixed thoroughly in an acetone media using an agate mortar for 5-6 h. The well mixed powder was then dried and the resultant mixture was then calcined at 1100 °C for 6 h in air. After calcination, powders were again ground in an agate mortar to get a homogeneous fine powder of the desired compound.

The same route was used for synthesize the ferroelectric phase taking amounts of BaCO₃ (NLT 97.0%, Himedia, India) and TiO₂ (NLT 98%, Himedia, India) in stoichoimetric ratio. These powders were weighed and mixed thoroughly in acetone media for 5-6 h. The mixture was then calcined for 2 h at 1000 °C in air. The ferroelectric-ferrite composites having specific formula 0.3CoFe₂O₄-0.7BaTiO₃ were prepared by mixing CFO and BT in an acetone medium. Calcined powder of 0.3CoFe₂O₄ and 0.7BaTiO₃ and polyvinylidene fluride (PVDF) (Nalgene, Alpha Aesar) were grinded for (~ 4 h) with different weight ratio of polymer which are 20%, 30%, 40%. Obtained powders were pressed for 20 min at 180 °C in a hot polymer press with a pressure of ~7-8 tones and cooled to room temperature. After cooling down to room temperature, the sample was obtained in regular thin sheet (~ 0.16 mm) form. Uniform composite films were coated with silver to measure the dielectric properties.



Fig. 1. X-ray diffraction patterns for 0.30 $CoFe_2O_4$ –0.70 $BaTiO_3$ –PVDF composite films with 0, 20, 30 and 40 wt% of PVDF where (*) denote the ferroelectric phase and (+) denote the ferrite phase.

The structural analysis of the composite films was carried out with the help of an X-ray diffractometer (Bruker D8 Advance) with Cu (K_a) (λ =1.5402 Å) radiation. Atomic force microscopy (AFM) was used to check the roughness and as well as grain size in composite films. The dielectric measurements were done using an automated HIOKI 3532-50 Hi tester LCR meter in the temperature range 30-160 °C at 1 KHz, 50 KHz and 100 KHz. The microstructure analysis was carried out using field emission scanning electron microscope (FESEM). A Magnetocapacitance study was carried out using a Wayne Kerr 6500 high frequency LCR meter along with a magnetic field up to 8 kOe (provided by Marine India Electr. Pvt. Ltd). The magnetic properties of composite films were determined using a Vibrating Sample magnetometer (VSM) with magnetic field in the range of (-10 kOe \leq H \leq 10 kOe).



Fig. 2. (a) Atomic Force Microscopy (AFM) photographs for grain size (b) 3D image of roughness and (c) Average Particle size distribution respectively for composite film of $0.30 \text{ CoFe}_2O_4 - 0.70 \text{ BaTiO}_3 - 20 \text{ wt}$ % of PVDF.

Results and discussion

Structural analysis

Fig. 1 shows the X-ray diffraction (XRD) patterns of $0.3\text{CoFe}_2\text{O}_4$ –0.7BaTiO₃–PVDF composite films with 0, 20, 30 and 40 wt% of PVDF. All the XRD peaks could be identified for both the spinel (ferrite) and perovskite (ferroelectric) phases. At higher composition (PVDF= 40 wt %) we found some amorphosity in composite which is due to the amorphous nature of PVDF. Generally a polymer is neither fully crystalline nor fully amorphous. X-Ray diffraction pattern of all the compositions with PVDF has a sharp peaks superimposed on a broad amorphous hump as shown in **Fig. 1**.

AFM analysis

The grain size and roughness of the film (~106 nm and 73 nm respectively for $0.3\text{CoFe}_2\text{O}_4 - 0.7\text{BaTiO}_3 -20$ wt % of PVDF composite film was estimated by using AFM. The particle distribution and surface topography are shown in **Fig 2(a-c)**. In AFM it was found that CFO-BT particles are embedded in longe range order fibre matrix.

Dielectric properties

Fig. 3 shows the variation of the dielectric constant and dielectric loss with temperature at 1 kHz. Dielectric constant (\mathcal{E}) decreases with increase in the concentration of PVDF, but increases with increasing temperature. In the dielectric curve we observed one peak of BaTiO₃, which shows its transition temperature ~ 120 °C and we can't see here the transition temperature of CoFe₂O₄ because PVDF has low melting point around 177 °C and the measurement was restricted up to 140 °C. The suppression of the Curie temperature can be caused by two reasons. Firstly, porous structure of thick film can result in a weak dependence of dielectric constant on temperature [**20**].



Fig. 3. Variation of (a) dielectric constant (\mathcal{E}), (b) dielectric loss with temperature at 1 kHz for 0.30 CoFe₂O₄ - 0.70 BaTiO₃ -PVDF composites film with 0, 20, 30 and 40 wt% of PVDF.

Secondly, the dielectric peak of the film could be covered by the contribution of conduction mechanism from CoFe₂O₄ phase [21]. It is found that the dielectric loss also decreases with increasing amount of PVDF. With increasing temperature, the effect of charge polarization is enhanced, giving rise to higher dielectric constant values. Therefore, the high dielectric constant measured at high temperature does not mainly arise from the ionic polarization, but more significantly from the polarization of space charges [11]. The dielectric properties of polycrystalline ferrite composites arise mainly from the interfacial polarization and the intrinsic electric dipole polarization. The dielectric constant increases from about 684 to 1106 with increasing temperature at 1 kHz in the thick film of $0.2\text{CoFe}_2\text{O}_4 - 0.8\text{BaTiO}_3$ in the work of Zhang et al. and the value of dielectric constant and dielectric loss with 50% of BT with PVDF composite film was found to be ~ 55 and ~0.2, respectively in the work of Lin et al. [22]. It is found that with increasing wt% of PVDF the dielectric constant and dielectric loss decreases. From the dielectric graph it is observed that in the composite film there is a diffuse type of phase transition. The effective dielectric permittivity of composite materials depends not only on the dielectric permittivity of the polymer and the filler, and the volume fraction of the filler, but also on the dielectric permittivity of the interphase region. The dielectric constant and dielectric loss of CoFe₂O₄/PVDF with 0.08 volume fraction of CoFe₂O₄ at 1 kHz was found to be (~14) and (~0.5) respectively at 77 °C in the work of Martins et al [19]. In our work we found high value of dielectric constant with the addition of BaTiO₃ because BT is a ferroelectric material and PVDF has the polar nature, so the presence of both materials increases the dielectric constant.

Microstructural study

The surface morphology of pure and composite films was studied by using FE-SEM in **Fig. 4(a)-(d)**. The FE-SEM images of the composite films clearly reveal that CFO-BT grains are homogenously embedded in polymer matrix, as a result porosity decreased. CFO-BT grains acts as filler in the polymer matrix of PVDF.



Fig. 4. FESEM micrographs of $0.30 \text{ CoFe}_2O_4 - 0.70 \text{ BaTiO}_3 - \text{PVDF}$ composites film with 0, 20, 30 and 40 wt% of PVDF.

AC conductivity

The variation of ac conductivity with temperature of 0.30 $CoFe_2O_4 - 0.70 BaTiO_3 -PVDF$ composite films at 100 kHz is shown in **Fig. 5**. The values of ac conductivity for 0.30 $CoFe_2O_4 - 0.70 BaTiO_3 -PVDF$ composite films with different concentration of PVDF, were calculated to be 8.63×10^{-5} , 6.69×10^{-5} , 3.91×10^{-5} , $1.59 \times 10^{-5} \text{ ohm}^{-1} \text{ M}^{-1}$, respectively at 35 °C. These values are found to be greater than that of composite film of BiFeO₃ -PVDF [**11**].



Fig. 5. Variation of the ac conductivity with temperature for 0.30 $CoFe_2O_4 - 0.70 BaTiO_3 - PVDF$ with 0, 20, 30 and 40 wt% of PVDF.

The main contribution of electrical conductivity at high temperatures generally originates from thermally activated oxygen vacancies. The temperature dependence conductivity follows the Arrhenius equation $\sigma = \sigma_0 \exp(-E_a/k_B T)$, where σ_0 is the pre-

exponential factor, E_a is the activation energy per charge carrier, k_B is the Boltzmann's constant and T (K) is the absolute temperature [23, 24]. From these values it is clear that with the addition of PVDF conductivity decreases and it is similar to the conductivity properties of PVDF. The activation energy of composites film is found to be 0.192 eV, 0.073 eV, 0.127 eV and 0.135 eV for 0.3CoFe₂O₄ – 0.7BaTiO₃–PVDF with 0, 20, 30 and 40 wt % of PVDF.



Fig. 6. (a) Room temperature magnetic hysteresis loops of 0.30 $CoFe_2O_4$ – 0.70 $BaTiO_3$ –PVDF composites film with 0, 20, 30 and 40 wt% of PVDF (b) Variation of Saturation magnetization (2M_s) and Remnant Magnetization (2M_r) with different PVDF concentration.

Table 1. Data on remnant magnetization $(2M_r)$, Saturation magnetization $(2M_s)$ and coercitivity $(2H_c)$ of .30 CoFe₂O₄ – .70 BaTiO₃ –PVDF composites film with 0, 20, 30 and 40 wt% of PVDF.

Composition	Remnant	Saturation	Coercitivity
with PVDF wt%	Magnetization (2M _r) emu/gm	Magnetization (2M _s) emu/gm	(2H _c) kOe
0	22.36	52.87	2.24
20	20.04	43.59	1.64
30	15.77	34.98	2.28
40	11.25	28.33	2.28

Magnetic properties

The magnetic properties of composites films of 0.3CoFe₂O₄ - 0.7BaTiO₃-PVDF with 0, 20, 30 and 40 wt% of PVDF analysed at room temperature using a VSM with an applied field of -10 kOe \leq H \leq 10kOe are shown in **Fig. 6**. At room temperature the $CoFe_2O_4$ behaves as a ferromagnet with blocked magnetic moment with the particle, and it develops a hysteresis loop with coercivity of 0.27 T and reaches a saturation magnetic moment of 60 emu/gm at magnetic field of 10 T [15]. It is found that with the addition of PVDF powder in the composite of $0.30 \text{ CoFe}_2O_4 - 0.70$ BaTiO₃, the values of magnetic saturation and remnant magnetization decreases. It may be due to the presence of PVDF. Since PVDF acts as non-magnetic phase so it decreases the magnetization. Saturation magnetization values of the ferrite particles within the polymer matrix fit well to that value when the loops are normalized with the concentration of magnetic particles in the composites. The shape of the measured loops demonstrates that magnetic particles are randomly oriented within the polymer matrix [16]. The value of saturation magnetization in the thick film of PVDF/BaCa₂Fe₁₆O₂₇ is found to be 1.3 emu/gm.

Magnetization (M) versus field (H) loop for 70% $(0.75CoFe_2O_4 - 0.25 \text{ BaTiO}_3)$ -30% PVDF taken at 300 K has been reported by Gupta et al. and the value of M_s was found to be 37.2 emu/gm [**17**]. The values of remnant polarization (2M_r), saturation polarization (2M_s) and coercive field (2H_c) are shown in **Table 1**.

Capacitance and polarization measurements

Fig. 7(a) shows the variation of capacitance with magnetic field. Due to the effective mechanical interaction between CFO and BT particles through the PVDF matrix, a stress is experienced by the BT particles. The stress on the piezoelectric BT phase thus induces ferroelectric polarization/charge across the sample and as a result the capacitance of the sample is set to increase [13]. With the addition of PVDF in composite, there is enhancement in magnetocapacitance value because PVDF acts as good coupler so it enhances the coupling between ferroelectric and ferromagnetic phases. That's why these values increase with increasing wt % of PVDF. Capacitance (C) and dielectric loss (tan δ) versus magnetic field (H) upto 6 kOe were measured at 1 kHz for composite films with different concentration of PVDF are shown in figure 7(a). The value of capacitance is found to be 52 pF for 40 wt% of PVDF at a magnetic field of 5 kOe. It may be due to the effective mechanical interaction between CFO and BT particles through the PVDF matrix, a stress is experienced by the BT particles. The stress generated on the piezoelectric BT phase induces ferroelectric polarization in the sample and as a result the value of capacitance is increased [17]. Fig. 7(b) shows values of saturation polarization and remnant polarization for composites film of $0.3C_0Fe_2O_4 - 0.7$ BaTiO₃ –PVDF with different concentration of PVDF; 0, 20, 30, 40 wt % which indicates the ferroelectric behaviour of the composites film. These obtained values of saturation polarization and remnant polarization is less than that of the thick film of $.20 \text{ CoFe}_2\text{O}_4 - .80 \text{ BaTiO}_3$ [25].



Fig. 7. The graph demonstrates (a) the variation of Capacitance at 1 kHz for $0.30 \text{ CoFe}_2O_4 - 0.70 \text{ BaTiO}_3 - \text{PVDF}$ composites film with 0, 20, 30 and 40 wt% of PVDF (b) Variation of remnant polarization (2P_r) and Coercive field (2E_c) with different PVDF concentration.

Magnetodielectric studies

Fig. 8 shows the variation of the magnetocapacitance with magnetic field at 1 kHz frequency with the applied external magnetic field (0-8 kOe). The magnetoelectric coupling can be measured indirectly by recording the change in

dielectric constant with magnetic field, the resulting effects are described using terms 'magnetocapacitance'. In the presence of magnetic field the material (multiferroic) will be strained, due to the coupling between the magnetic and ferroelectric domain. As a result, the dielectric behaviour will be modified. The Magnetocapacitance is defined as:

$$MC = \frac{\varepsilon(H) - \varepsilon(0)}{\varepsilon(0)} \times 100$$
(1)



Fig. 8. The graph demonstrates the variation of Magnetocapacitance with magnetic field at 1 kHz for $0.30 \text{ CoFe}_{2}O_4 - 0.70 \text{ BaTi}O_3 - \text{PVDF}$ composites film with 0, 20, 30 and 40 wt% of PVDF.

The numerical value of magnetocapacitance is found to be ~ 2.2% at a magnetic field of 8 T for 30 wt% of PVDF which is the highest among these compositions. This shows the presence of coupling effect at room temperature, which may have been induced by magnetoelectric coupling. The change of the dielectric constant under the variation of magnetic field can be induced by magnetostriction effect, which occurs due to the change in lattice parameters on applying magnetic field [26]. When the magnetic field is applied, magnetic particles change their shape due to magnetostriction and the strain thus produced passes to the piezoelectric phase through the polymer matrix and results into generation of induced charge. The difference of the dielectric constant $\Delta \varepsilon$ below T_M will be proportional to square of magnetic-order parameter: $\Delta \varepsilon \sim \gamma M^2$, where M is magnetization. The sign of $\Delta \varepsilon$ depends on the sign of the constant magnetoelectric interaction γ , and it can be either positive or negative [27]. The variation of $\Delta \varepsilon / \varepsilon(0)$ as a function of the square of magnetization (M^2) is shown in Inset of Fig. 9 for $0.3CoFe_2O_4-0.7BaTiO_3$ -20 wt% PVDF shows the variation of M² with the applied magnetic field. It can be seen that $\Delta \varepsilon / \varepsilon(0)$ changes linearly with M², in the inset of Fig. 9. Thus, $\Delta \varepsilon / \varepsilon(0)$ versus M² can also be expressed by $\Delta \varepsilon \sim \gamma M^2$ for composite film of 0.30CoFe₂O₄ - 0.70BaTiO₃ - 20 wt % PVDF. Linear fitting with this formula give the value of $\gamma = 1.32 \times 10^{-3}$ (emu/g)⁻² which is slightly greater than in the composite of 0.3NiFe₂O₄-0.7BiFeO₃ [28].

0.1





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ndard Error 0.01788 4.43076E-5 y = a + b*x Valu 0.04067 cept 0.0 0.00132 lope -0.1 500 ε(H)–ε(0)/ε(0)(*10⁻² -0.2 -0.3 -0.4 Magnetic field(kOe) -0.5 -0.6 -0.7 600 100 200 300 400 500 Magnetization² (emu/gm)²

Fig. 9. Variation of magnetocapacitance with (magnetization) 2 for 0.30 CoFe₂O₄-0.70 BaTiO₃-20 wt% of PVDF. Inset shows variation of M^2 with magnetic field.

Conclusion

In summary, different composite films of 0.30CoFe₂O₄ -0.70BaTiO₃ –PVDF with different concentration of PVDF; 0, 20, 30, 40 wt % were prepared by a hot press method for the study of dielectric and magnetic properties. From X-Ray diffraction pattern it was found that the plots of all composite films have sharp peaks superimposed on a broad amorphous hump. The values of dielectric loss decreased with the addition of PVDF. It was found that values of remnant magnetization $(2M_r)$ and saturation magnetization (2M_s) decreases than those of without the addition of PVDF powder. Porous structure of the film shows a low polarization value. It is found that the $0.30 \text{ CoFe}_2\text{O}_4 - 0.70$ $BaTiO_3 - 0.30$ PVDF shows the enhanced value of magnetocapacitance. The value of magnetoelectric coupling coefficient was found to be 1.32×10^{-3} (emu/g)⁻² for 0.30CoFe₂O₄ - 0.70BaTiO₃ -20 wt % PVDF. It will be useful for applications in transducers, pyroelectrics, piezoelectrics, and some actuators. Magnetoelectric coupling of the thick film compared to bulk material indicates that the composite thick film has a good opportunity to evaluate their potential application in multifunctional devices.

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