Lightweight, ferroelectric-ferromagnetic polymer- nanocomposites for field sensor applications

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Abstract

The coexistence of ferroelectric and magnetic order parameters in multiferroic materials opens up a host of new collective properties. In particular, the magnetoelectric (ME) effect namely the induction of electric polarization by a magnetic field or magnetization by an electric field has been widely studied in multiferroics. PVDF is a ferroelectric polymer that has attracted considerable research interest for sensing applications. Compared to traditional ceramic composites, the polymer-ceramic nanocomposites offer inherent advantages, including easy processing, mechanical flexibility and the ability to be moulded into complicated configurations for advanced devices with reduced volume and weight. Additionally, polymer composites generally exhibit superior ME coefficients, attributable to the improved displacement transfer capability of the flexible polymer matrix. In this report, free-standing, flexible and lightweight polymer–ceramic nanocomposite thin films with a fixed weight percentage of ferromagnetic CoFe\textsubscript{2}O\textsubscript{4} nanoparticles have been fabricated using a solution casting technique. The structural, microstructural, and electrical properties of the composite have been characterized by standard experimental techniques. The structural and chemical analyses prove a homogeneous dispersion of the fillers in the microstructure of the composite. The electrical response investigated by impedance spectroscopy reveals the contributions of grains and grain boundaries to the whole impedance of the composites. The ac conductivity as a function of frequency obeys Jonscher’s power law. The improved magnetoelectric properties suggest promising applications in multifunctional devices, including field sensor applications.

Key word: Magnetoelectric Polymer Nanocomposite, PVDF, Dielectric properties, Electrical properties.

1. Introduction:

Magnetoelectrics (ME) is a special class of multiferroics (MF) which are well-known for combining two mutually exclusive order parameters, i.e., ferroelectric and ferromagnetic order initially thought to be two independent phenomena that can separately scripted data within an exclusive multiferroic bit but later found intrinsically coupled to each other.[1] This coupling permits the data to be written electrically and read magnetically which is the basis of FeRAM and MRAM.[2] The ME can further be split into two categories; (a) single phase ME which involves a direct coupling of ferroelectric and ferromagnetic ordering and (b) multiphase ME composite which shows a strain mediated coupling. Unfortunately, the chemical antagonistic nature and interactive exclusiveness creates a big problem in combining both order parameters in a single phase. So, they are less in number. [1-2] In order to overcome the obstruction of use of single-phase multiferroics let us examine developments in composite or multi-phase multiferroic (MF) materials. Composite MF generally comprises of alternating layers of piezoelectric and magnetostrictive materials; which experiences a configuration change under the application of electric or magnetic field, respectively. The materials experiences mechanical coupling. Because neither the piezoelectric nor magnetostrictive material alone has magnetoelectric properties, the magnetoelectric response is referred to as a product property ($\alpha_{ME}$). [3] In maximum single phase MF systems $\alpha_{ME}$ is less than 20 mV/(cm Oe) which is regarded as...
inadequate for smart device implication. [4] Beside this, wide imbalance of transition temperatures in single-phase ME, they are rare in number; which shows ambient temperature ME characteristics.[5] So, few MEs can be practically applicable at a temperatures like ≈ 10 K, which badly restricts the prototyping of devices.

Though ME coefficients gained in ceramic MF composites is more than single phase systems [6] but they may insubstantial & limited by deleterious reactions at the interface, following small resistivity with huge loss, hindering for inclusion into devices. [7] Beside the above drawbacks, ceramic composites possess complication like being extravagant, dense and brittle, which results breakdown in the course of operation. [8] And hence are not glamorous from a technical view point. A current way to have extreme flexible, portable, light weight and non-breakable ME system is to adopt polymer-based nanocomposites which can resolve all the previous difficulties [9-10]. If we compare ceramic based ME composites with polymer based one, the latter one can be comfortably manufacture via low heat processing technique into a numerous forms, like thin sheets or moulded structures, and can display enhance mechanical characteristics along with spectacular flexibility, biocompatibility making them potential candidates for wearable and implantable electronics for human bodies. There are three leading category of ME polymer-based composites can we have i.e., (a) laminated composites, (b) polymer as a binder, and (c) nanocomposites.

Semi crystalline polymer poly(vinylidene fluoride) (PVDF)[11] is a good ferroelectric polymer[12] but it’s ferroelectric order parameters not such much enough as like ceramics and single crystal to integrate in device in its virgin form [13-14]. So, it is very essence to improve the ferroelectricity in PVDF to trigger its electroactive phases for real device application which strictly depends upon the processing condition. [15] PVDF possesses four vital phases namely α, β, δ, and γ. normally, the α form of PVDF dominates owing to its good thermal reliability under ambient environment. The molecular chain conformation in α phase is tg-tg (t = trans, g = gauche) has no ferroelectric properties. The δ phase of PVDF may be simply realized through poling of α form at ambient condition under a low electric field in the order of MV/m. But the inclination of C-F dipoles at an angle to the molecular axis, the perpendicular and parallel components of the dipole moments to the chain axis are as low as 1.20 and 1.02D, respectively, leads to a poor response of ferroelectricity in δ form. [16]. similarly, γ phase exhibits a minor ferroelectric property which is resulted owing to its low dipole moment. The electro active β- phase is mostly studied owing to all-trans (t t t t) planar zigzag chain conformation having largest dipole moment per chain, resulting maximum P, with noticeable ferroelectric responses .[13] So, it is crucial to enhance the content of β phase in PVDF which is the fundamental aspect to materialize its ferroelectric performance in technological applications. There are many simple economical ways to enhance β-content in PVDF which is solely processing condition dependent for different application as reported by various reports [17-33]. β - phase may be gained via solvent casting technique but when the material crystallized below 70⁰C results high porosity[34]. As a result poor mechanical and electrical features of the developed materials hinder for application. Hence it is very essence how to get materials having good electrical and mechanical properties that can be integrate in devices in more economical way. In this line an innovative solution casting technique is very popular where we can shape a polymer composite depending upon our requirement. A quite contemporary and fascinating method is the nucleation of the electroactive form of the polymer via incorporation of ferrite nanoparticles into PVDF.[35-38] Low ferrite nanoparticles loading are generally used as magnetostrictive phase in ME composites and are attractive for both elemental and industrial view point. So, the physical characteristics of PVDF not only depend on fabrication process but also energetically governed by the existence of nanoparticles in crystallization kinetics [39]. From the previous report it has been observed that the growth kinetics for the nucleation is higher for
ferrites like Ni0.5Zn0.5Fe2O4 (NZFO) and CoFe2O4 (CFO) nanoparticle as compared to NiFe2O4 (NFO) nanoparticles. Similarly the crystallization rate in ME composites with PVDF with ferrite follows the order NFO > CFO > NZFO. Elevated activation energies of the nanocomposites in comparison to PVDF counterpart may be ascribed to the constraint motion of polymer chains resulted from the existence of ferrite nanoparticles.[40] Lower loading (i.e. 5% by weight) of CFO nanoparticles in PVDF became effective for nucleation of polymer for electroactive β phase i.e., about 84% in the nanocomposite and which further can be enhanced to 95% as compared to 73% in the pure PVDF. [34] The β-form of PVDF seems to have highest values of 47.1 following poling and 78.5% following stretching, in the PVDF-CFO nanocomposite films; which infers that different physical conditions results different value of β content. Nanofillers are quite advantageous to deliver a physical barrier which assists to preserve parallel dipoles of the β-form and concurrently plays the obstructive character of conductors to instigate electrical percolation.[41] Similarly ferrites nanoparticles in PVDF enhances the tunneling electroresistances to 450% @10K and 100% in ambient condition room temperature (RT), which is quite excessive than those of the pure PVDF based device 70% @10K and 7% at RT in organic multiferroic tunnel junction (OMFTJ).[18] So, keeping all these in mind we have processed a very innovative, cost effective method of solution casting to fabricate free standing, flexible, light weight polymer nanocomposite with low filler loading of ferromagnetic CFO in ferroelectric PVDF to ignite the electro active phases in the composite for making suitable it for device application.

2. Materials and Methods:-

The CoFe2O4 within the range of 5-15 nm has been used for the fabrication of polymer nanocomposite which has been discussed elsewhere. [42] The ferroelectric polymer (PVDF) and ferromagnetic CoFe2O4 was taken for the configuration of composite 0.95PVDF-0.05CoFe2O4 by weight percentage using high precession microbalance. Initially desired quantity of PVDF granules (supplied by M/S-HiMedia Lab., India) was dissolved in completely N-methyl-2-pyrrolidone (NMP) of M/s Loba Chemie Co. Ltd at 100°C using Remi magnetic stirrer (Model-2MLH) at 250 RPM for nearly 90 minutes. Polar solvent NMP has been used to conserve the polarity of the polymer in the solution. Phase pure ferromagnetic CoFe2O4 nanoparticles was mixed slowly under constant stirring to the above solution for 240 minutes at a RPM of 300. The resulted homogenous suspension poured on a glass substrate of the micrometer controlled laboratory designed solution caster and permitted it to dry in the air for a few days in desiccators to remove the solvent trace in ambient condition. The thin film (here after it is named as polymer nanocomposite) was then peeled off from the glass substrate for different experimental measurement. The schematic representation of fabrication process of polymer nanocomposite thin film has been represented in Figure1. Phase analysis of the fabricated polymer nanocomposite was performed by XRD of Bruker; Model-D8Advance, using CuKα radiation. The surface morphology polymer nanocomposite thin film was examined via SEM of ZEISS and topography by AFM of Bruker; model- Multimode 8.[43-45 ] Prior to SEM experiment magnetron sputtering was employed to coat the thin film by gold. Prior to examine the ferroelectric nature of the thin film pooling was performed via corona discharge method. P-E loop tracer of M/S-Radiant Technology, USA, has been used to investigate the ferroelectric nature of the thin film at room temperature at different applied frequency. Superconducting Quantum Interference Device (EverCool SQUID VSM DC magnetometer Quantum Design, USA) used to analyse the magnetic nature of the thin film at an optimised condition of 3T. To record electrical parameters high purity silver-paste was used which was act as an electrode on the thin film in a wide range of frequency (100Hz-1MHz) and temperatures (25-120°C) using impedance analyzer of Keysight technologies (Model E4980A ). ME measurement was carried out by a ME set up supplied by M/S-Marine India being assembled with a
lock in amplifier of Stanford Research (Model SR830). A DC magnetic field of 0.7T was generated by an electromagnet while AC magnetic field was generated by a Helmholtz coil consisting of 200 turns of radius of 2.5cm. For ME measurement of the polymer nanocomposite thin film was placed in the magnetic field with its faces perpendicular to the field direction. The ME output voltage developed over the thin film was studied using the above lock-in amplifier as ME output voltage.

3. Results and Discussion

3.1. Crystal Phase investigation:-

Figure 2 presents the XRD pattern of CoFe$_2$O$_4$ nano particle whose phase has been represented as (*) in the combined plot of PVDF thin film along with XRD pattern of 0.95PVDF-0.05CoFe$_2$O$_4$ thin film. Though there are four phases [34] in PVDF but we able to detect α, β, and γ phase as α phase can be converted to δ phase with the application of external polarizing field. Hence it is not noticeable. From the figure it is found that the 0.95PVDF-0.05CoFe$_2$O$_4$ consists of all the diffraction peaks of PVDF along with CoFe$_2$O$_4$. The strongest peak which appears at 2θ = 20.4°(110) is corresponds to β phase and it makes distinct from other phases. Whereas the peak appears in the vicinity of 18° corresponds to α and γ allows us to differentiate it from β.[35] The diffraction peak near 2θ = 39.3 could be assigned to (002) plane of α phase and 2θ=36.2 related to (200) plane of γ phase. Since the filler load is very low hence the diffraction peaks of PVDF is found to be dominating. However the β content in 0.95PVDF-0.05CoFe$_2$O$_4$ thin film is more than that of PVDF. In 0.95PVDF-0.05CoFe$_2$O$_4$ thin film, the highly crystalline peaks of CoFe$_2$O$_4$(CFO) appear in the higher diffraction angle. With the introduction of CoFe$_2$O$_4$, the broad peak having peak maxima at 2θ=19.99° manage to shift to a higher angle of 20.4°, owing to the emergence of β phase. From this representation it confirms that CFO nanoparticles may be used as an effective β-phase nucleation factor for PVDF [35]. Having solely charged -ve surfaces, the introduced CFO nanoparticles in PVDF matrix may attract incomplete +ve CH$_2$ bonds of the PVDF by ion-dipole interaction. Subsequently, the chains of PVDF aligned themselves on the surface of CFO nanoparticles, in turns favourably generates all-trans β phase. Beside this, the attached nanoparticles creates a “swell” in the adjacent polymer leads to the expansion of the chains of PVDF, being beneficial for evolution of the β form of PVDF. So, it can be concluded that the electroactive β phase can be enhanced in PVDF via low loading of ferrite nanoparticle as already discussed in the introduction part with the help of innovative economical solution casting technique.

3.2 Morphological Study:

Figure 3(b) represents the SEM micrograph of 0.95PVDF-0.05CoFe$_2$O$_4$. From the micrograph of the thin film it is observed that the nanoparticles are dispersed arbitrarily within the PVDF matrix. It was observed that the composite comprising of ceramic particles mixed with CFO and PVDF have more homogenous distribution of particles with less porosity as compared to pure PVDF as seen from figure 3(a). Which indicates the nanoparticles facilitated the dispersion in polymer matrix.[46]

Generally PVDF film possesses a spherulitics microstructure where the polymer chains are self-connected into a continuous network having clear boundaries. But in this case the incorporation of ferromagnetic CFO nanoparticles has a vital role on the spherulitics structure of the nanocomposite thin film. With the introduction of CFO nano particles the nucleation process will be restricted as well as growth kinetics of the spherulitics.[43] As a result, the size of the spherulitics is less than the spherulitics of PVDF. The microstructure, in conjunction with the structural analysis, concludes that CFO nano-particles are well dispersion through the polymer matrix and expands the nucleation process of β-phase of PVDF. The good dispersion and homogenous packing of ceramic filler is likely to results in superior dielectric, ferroelectric, magnetic and magnetolectric properties.
In order to have knowledge on the dimensional aspect of filler which has been introduced in the 0.95PVDF-0.05CoFe₂O₄ thin film we have conducted the HRTEM experiment of phase formed ferromagnetic CFO powders. Figure 3(c-d) represents the HRTEM micrographs along with its SAED pattern respectively. The bright field image of the specimen represents spherical shaped nano-particles having uniform dimension in the range of 10-15nm are uniformly distributed without any agglomeration which ensures the nano nature. The distinct but diffused rings of the SAED pattern is a clear indication of polycrystalline system at nano level. Surface morphology further analysed with the assistance of Atomic Force Microscopy (AFM)[44-45], which are presented in figure 2(e-f). Figure 3(e) and (f) represents the 2D and 3D AFM picture of 0.95PVDF-0.05CoFe₂O₄ respectively. The bright spots signify the surface areas of the film having maximum height, while the dark spots signify to valleys. The AFM image of 0.95PVDF-0.05CoFe₂O₄ confirms the spherulitic size within the range 42.7 nm as CFO nanoparticles obstruct the growth kinetics of spherulites and leads to a increase in β-content in the composites.[46] Beside this it is found that the conducted fabrication method is effective to increase the electroactive β-phase in the composite.

3.3 Dielectric Study:

Figure 4(a) represents the frequency dependence of relative permittivity of 0.95PVDF-0.05CoFe₂O₄ thin film over a broad frequency range of 10² Hz to 10⁶ Hz at suitable temperature. With rise in applied frequency the relative permittivity decreases which is resulted from the reduction in εᵣ of CFO nano-particles as per Maxwell–Wagner-Sillar (MWS) scheme of interfacial polarization & space charge polarization. MWS interfacial polarization along with space charge polarization primarily works in lower frequency domain owing to its long relaxation period. The εᵣ value of thin film is greater than PVDF (which is generally less than 10) is encouraging for capacitive energy device fabrication. The εᵣ of a composite thin film could be easily described by effective medium theory. As per that theory, the depolarization factor is greatly depends on aspect ratio of nano-fillers in the nano-composite [44]. Hence, the outstanding enhancement in the dielectric performance of the nano-composite can be ascribed to nano-ceramic having immense aspect ratios, which are quite effective in amplifying εᵣ of composites at lower concentration [47]. The loss tangent also follows the same rule like εᵣ response with frequency which is represented in Figure 4(b). At low temperature, the segmental motion of the chain is almost freezes which results decrease in εᵣ (quite noticeable from the figure). With rise in temperature, the intermolecular attraction within polymer chains is broken which intensify the thermal agitation. So, higher temperature facilitates the smooth locomotion of various dipoles, as a result these dipoles are able to follow the variation of frequency. As polymer matrix swells with temperature leading to rise in interfacial polarization being another reason of increase in εᵣ with temperature [48]. εᵣ Seems to be non responsive at higher temperatures as constrained polymer chain restricts the evolution of electrical polarization. For that reason the value of εᵣ is less as compared to ceramic counterpart but more than pure PVDF. The increase in εᵣ could be due to the large amount of charges that gathered at the interfaces of the two phases owing to the distinction between the ferrite particles and PVDF matrix resulting interfacial polarization[48].The loss tangent value remains always less than 1 as seen from figure 4(b) indicating suitability of thin film for device application over a broad range of temperature.

3.4 Impedance Analysis

Figure 5(a-b) shows the alteration of both Z’(real component of impedance) and Z” (imaginary component of impedance) with frequency at suitable temperature of 0.95PVDF-0.05CoFe₂O₄ of the thin film. As observed from the figure Z’ has gigantic value in lower frequency domain which
decreases slowly with rise in frequency, and attains a constant value at higher frequencies and gradually moves towards higher frequency domain with increase in temperature. The value of \( Z' \) coincides in the higher frequency domain irrespective of the applied temperatures. The decrease trend of \( Z' \) with increase in applied temperature and frequency could be ascribed to the rise in ac conductivity in the thin film. The coincidence of \( Z' \) in higher-frequency region irrespective of temperature recommends probability of release of space charge owing to diminish of barrier characteristics of the thin film. The appearance of \( Z' \) maxima could be correlated to the nature and strength of electrical relaxation process in the thin film. As evident from the figure the curves shows broader as well as symmetric peak which recommends the relaxation happens over a broad frequency domain. This symmetric nature of peaks could be designated to different relaxation processes and deviated from ideal Debye characteristics[42,49-50]. Similar trends being observed for \( Z'' \) case with the variation of frequency as evident from the figure 5(b). With rise in temperature, \( Z''_{\text{max}} \) moves to higher frequency domain where the space charge has comparatively lesser time period to relax three and hence recombination becomes quicker. As a result, in the higher frequency domain, the space charge polarization shrinks leading to merger of curves there.

Figure 5(c) represents the \( Z' \) vs. \( Z'' \) plot i.e., Nyquist plot of 0.95PVDF-0.05CoFe\(_2\)O\(_4\) at few suitable temperatures. Generally Nyquist plots consist of single semicircular arc which are primarily because of contribution from grains (bulk) and relaxation owing to Debye. But in the present case the existence of non–semicircular, non-symmetric semi-circle recommends the presence of non-Debye type of relaxation in the thin film [49] which is due to existence of dispersed phase elements. This non-ideal nature of relaxation may be emerged from various factors like orientation of grains, grain size distribution, boundaries of grains, distribution of atomic defects, and phenomena associated stress–strain in the thin film. The declining value of bulk resistance(\( R_{b} \)) with increase in temperature recommends the existence of negative temperature coefficient of resistance (NTCR), as like semiconductors [50-51]. The semi-circles spectra of impedance possess a distinguished peak at a definite frequency generally designated as relaxation frequency (\( f_{r} \)) which could be enunciated as \( \omega_{r} R_{b}C_{b} = 1 \) and hence \( f_{r} = 1/2\pi R_{b}C_{b} \).

3.5 Conductivity Analysis

Figure 6 (a and b) represents the variation of \( \sigma_{ac} \) with frequency as well as temperature respectively. As evident from the figure that the conductivity spectra include two regions specifically as low-frequency plateau region and the high-frequency dispersion region. \( \sigma_{ac} \) can be found from the plot by extra plotting the low-frequency plateau to x-axis or simply we can think it as frequency independent conductivity. Johnshcer’s power has been used to investigate the high-frequency dispersion region [42]. As per power law rule if the exponent ‘\( n \)’ < 1, then the backward hopping is quite slow than the site relaxation which leads to a translational movement of Co\(^{2+}\) ions of CFO in the PVDF matrix and if ‘\( n \)’ is >1, the backward hopping is more rapid than site relaxation time. Here, it is observed from non linear fitting that ‘\( n \)’< 1, confirms the backward hopping is slower than the site relaxation. [50] Based on jump relaxation model [52], in lower frequency domain, ions jumps from one site to its neighbouring vacant site, where as at higher frequency domain, due to the little time interval, the possibility of ions to fall back to its original site increases and more hopping of ions is accountable for higher conductivity in low frequency domain.

The thermally triggered transport phenomena of the thin film obey Arrhenius relation. As observed from the figure 6(b) \( \sigma_{ac} \) decreases with rise in temperature up to a certain temperature, and then displays a rising tendency with rise in temperature affirms the NTCR characteristics of the composite. Beside this there is a distinct proclivity of dispersion in conductivity spectra and all curves of various
frequencies try to merge at elevated temperatures which could be ascribed to the recombination of released space charge at high temperature [53]. With the assistance of free-volume model [54] the rising tendency of conductivity with temperature may be well understood. As per that model with rise in temperature, the polymer may swell/expand comfortably results in easy generation of free volume for ions to move.

3.6 M-H study

Figure 7 represents the M-H hysteresis 0.95PVDF-0.05CoFe$_2$O$_4$ over a magnetic field sweeping of ±30kOe at an ambient temperature. The saturation magnetization ($M_s$) is found to be 4emu/gm and remnant magnetization ($M_r$) is nearly 2 emu/gm which is quiet encouraging for a polymer thin film to be integrated in devices. However, the $M_s$ of CFO($> 100[42]$) is greater than the $M_s$ of the fabricated thin film due to the presence of higher volume fraction of non-magnetic PVDF phase [55-56]. There are two probable reason of decrease of $M_s$ in the thin film as compared to CFO. First reason is the non-magnetic PVDF polymer coating layer, which can be predicted as a magnetic dead layer on the surface of ceramic influencing the value of $M_s$ owing to the quenching of magnetic surface moment. Second reason may be ascribed to the magnetization of the film corresponds to the volume fraction of CFO nano-particles as per the relation $M_s=\phi M_m$, where $\phi$ the volume fraction of the particles and $M_m$ is the saturation of the single particle. [57]

3.7 P-E study

Figure 8 shows the ambient temperature P-E hysteresis of 0.95PVDF-0.05CoFe$_2$O$_4$ at different frequencies. As evident from the figure that $P_r$ (remnant polarization), $E_c$ (coercive field )and $P_s$ (saturation polarization) declines with rise in applied frequency. The improvement in higher polarization in polymer/ferrites thin film composite for low loading content has already been described by others [58-62]. Two important responses may be concluded based on that case. First one is CFO nano-particles may brings in surplus free carriers which is needed to balance the polarization domain, and second one is the nano-particles may serves as a heterogeneous nucleation centre for ferroelectric domains during the polarization [63]. Beside these large interfacial regions in the composite nano-fillers may stimulates the exchange coupling effect via dipolar interface layer results in esteem polarization.[64] The good ferroelectric response of 0.95PVDF-0.05CoFe$_2$O$_4$ thin film closely corresponds to the equal distribution of low concentrated CFO in PVDF matrix. [25,26] From the literature it is found that ferromagnetic CFO interacts with homopolymer matrix of PVDF to favours crystallization of electroactive β-phase, in comparison to α-phase. [65] Similarly, low nanofiller density in present case which is 5wt% in polymer matrix may support settings of polar conformations results in rise the ferroelectricity. So, in this way, the presence of small quantities of the magnetostrictive phase in the composite remarkably upgrades the polarization, demonstrating promising candidate for ambient temperature ferroelectric application.

3.8. Magneto-electric effect study

There are numerous experimental methods are available for examine ME output either in the form of voltage or charge. Here we have adopted the dynamic method. The detailed experimentation has already been discussed in the materials and method section and same method has already been reported by my group elsewhere [50] with mathematical calculation. The magneto-electric coefficient ($\alpha_{ME}$) of 0.95PVDF-0.05CoFe$_2$O$_4$ composites at room temperature represented in figure 9. As observed from the plots, that $\alpha_{ME}$ initially rises with rise in the DC magnetic field, reaches an optimum and then follows a declining trend with further rise in in magnetic field.[66] This nature is connected with piezomagnetic coefficients.[8,67], which is termed as M–E coupling coefficient (first
order differentiation of magnetostriction on the applied field. For the fabricated system the maximum $\alpha_{\text{ME}}$ is found to be 220 mV/come (figure 8(a) and reconfirmed from figure 8(b) over a single quadrant cycle) is quite suitable for designing an ME sensor. The enhancement in the ME coefficient could be related to the rise of mechanical deformation in the magnetostrictive phase [68].

3.9 Device prototyping for ME field sensor

For extensive judgement of ME response of the developed 0.95PVDF-0.05CoFe$_2$O$_4$ ME multiferroic components a prototype has been developed as like other report in the similar fashion of experimentation [67]. To estimate the execution of developed prototype 0.95PVDF-0.05CoFe$_2$O$_4$ sensor, a real validation setup has been developed by creating artificial ME field surrounding. When the EM field interact with 0.95PVDF-0.05CoFe$_2$O$_4$ ME sensor, under resonance frequency it gets strained resulting a voltage output at the electrodes. For the current system the maximum output voltage is 98mV. Figure 10 represents the variation of sensed voltage with respect to physical distance from the axis of the coil under artificial EM field surroundings. The output voltage emerges across the sensor device decreases with the rise in physical separation which may be due to depletion in magnetic field intensity with rise in distance which confirms the magnetic field sensing ability of the fabricated 0.95PVDF-0.05CoFe$_2$O$_4$ field sensor.

4. Conclusion

Free standing, light weight 0.95PVDF-0.05CoFe$_2$O$_4$ nano-composite thin films has been fabricated via solution casting technique having $\alpha$, $\beta$ and $\gamma$-phases in them by introducing ferromagnetic CFO in ferroelectric PVDF. The HRTEM micrograph of the prepared ceramic is found to be in nano-regime with less agglomeration. The structural, microstructural, and electrical properties of the composite have been characterized by standard experimental techniques reveals the presence of electroactive $\beta$ phase as a major phase. The structural and chemical analyses prove a homogeneous dispersion of the fillers in the microstructure of the composite with the conclusion of smaller spherulitics size owing to the introduction of ferrite nanoparticle which hinders the growth kinetics of spherulitics being confirmed by AFM topography study. The electrical response investigated by impedance spectroscopy reveals the contributions of grains and grain boundaries to the whole impedance of the composites. The ac conductivity as a function of frequency obeys Jonscher’s power law. The improved magnetoelectric properties suggest promising applications in multifunctional devices, including field sensor applications.

Acknowledgement: Author acknowledge CRF, IIT Kharagpur for SEM, HRTEM and SQUID experiments and Prof. R.N.P Choudhary, Multifunctional Materials Research Laboratory of Siksha O Anusandhan, for extending the support of ME experiment.

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