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A Study of Effective Polymer Hybrid Nanocomposites for Electronics Applications of Emi Shielding

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Abstract. The development of radar absorbing materials is a well-researched area with applications in electronics and stealth technology. In this work, two-phase and three-phase systems were produced in which a nano-conducting and magnetic charge were embedded in an insulating polymer matrix. Nickel spinel ferrite was synthesizes using a wet chemical route with a magnetic character, while the graphene nano-filler is a conductive part, both were built into an insulating polymer matrix. Various concentrations were produced for the two-phase system (PVDF polyvinylidene fluoride/graphene) and a selected nano-filler concentration, spinel ferrite, which was incorporated by upto 30% of the weight. These two-phase and three-phase hybrid systems were fabricated using the solution casting route and exemplified using Wide Angle X-ray Diffraction (WAXD), Scanning Electron Microscope (SEM), Impedance Analyzer, Final Tensile Tester (IAFTT) and Network Analyzer Vector (VNA). A summary is presented with recommendations for setting up instruments related to the laboratory.

Keywords: radar absorbing materials, two-phase, three-phase, PVDF (polyvinylidene flouride), nano material composite

Introduction

The rapid development in the field of electronics and telecommunications has exacerbated the challenge of electromagnetic interference (EMI), as it can affect the performance of precise electronic components. Common examples include distortion of radio/television signals, interference in communication devices such as cell phones, restrictions or bottlenecks in sensitive electrical equipment. On the other hand, electromagnetic radiation has increased electromagnetic pollution that can seriously harm human health in various ways. Research has also been conducted into radiation damage caused by electromagnetic waves influencing living species. Electromagnetic waves consist of coupled electrical and magnetic components (Butcher, 2016). Therefore, materials with good electrical and magnetic properties have been used for shielding against electromagnetic radiation. Metals have good conductivity and a good magnetic character, which makes them suitable for this application (Chung, 2000). However, due to the sensitivity to the environment, the processing and the high mass/volume ratio of the metal are not suitable materials for such an application. On the other hand, conductive and magnetic films made from polymer-based nanocomposites find their place in EMI shielding because of their low cost, ease of processing, low mass/volume ratios, flexibility and others. (Chavhan and Wankhade, 2020), new materials like hybrid composite materials are being paid attention to in industrial applications. Recent research work indicates that the hybrid fiber-reinforced polymer composites have many applications in different areas because, it has good mechanical properties like specific strength, low weight and high resistance to corrosion. In (Katheria, 2022) polymer composites are excellent candidates to prevent breakdown of electronic equipment which help increase the electrical conductivity. Here, specifically thermo-plastic elastometer (TPE) nanocomposites are used in the shielding of electromagnetic interference (Wang *et al*., 2016).

The Radar Absorbing Materials (RAMs) also demonstrate the application in defense mainly on the surface of jets, armed vehicles, and safe houses to make it undetectable by radar (stealth technology) (Das *et al*., 2015). The researchers are continuously developing such nanocomposite polymer films to meet commercial requirements with high conductive and magnetic properties and to attenuate EM radiation (Chung, 2000) (Kumar *et al*., 2016), PVDF (Polyvinylidene fluoride) has an excellent combination of mechanical, thermal,

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chemical and piezoelectric properties (Chen *et al.*, 2016; Zhang *et al.*, 2009). It is suitable to be used as waterproof and transparent films can be made with polymers. These properties of PVDF make it interesting for scientists and researchers to use PVDF as a matrix for polymer composites. Further, PVDF is used when high purity is the primary concern as PVDF is a type of plastic used in applications where maximum purity is paramount or in areas where contact with solvents, acids and bases is required. Compared to another fluoropolymer, PVDF is considered a low-density polymer like poly-tetrafluoroethylene (Heap, 2016). The PVDF-based composites are being used in many industrial and scientific applications, such as semi-conductor composites, sensor based applications and electrolytes for lithium-ion batteries (Mishra *et al.*, 2016; Xu *et al.*, 2011).

Nickel ferrite synthesis *via* co-precipitation method. Various hybrid PVDF-based nanocomposites have been developed to increase EMI shielding performance. (Joseph *et al.*, 2017) investigated the increase in the shielding performance (SE) from 0.5 dB to 55 dB (Decibel) by increasing the concentration of graphite flakes from 0% by volume to 71%, reinforced in a PVDF matrix at 15.5 GHz (Gigahertz). (Wang *et al.*, 2016) examined and prepared multi-walled CNT (MWCNT)/PVDF nanocomposites (7% by weight) and achieved an SE value of 33.23 dB at 9.6 GHz. (Eswaraiah *et al.*, 2011) reported an EMI SE from functionalized graphene (f-G)/PVDF nanocomposites, 28.5 dB reported for 7.1% by weight on exposure to f-G over a frequency range from 0.9 to 8 GHz. (Joseph *et al.* 2017) reported the addition of silver (Ag) in PVDF/BT nanoparticle composites as microwave absorbing materials with an SE of 26.25 dB at 10.2 GHz. (Sharma *et al.*, 2014) described that carbon nanotubes (CNT)/PVDF composites have a shielding value of 10.1 dB, which was then increased to 31 dB by increasing the CNT value in the PVDF matrix in various frequency bands to 3% by weight (Maaz *et al.*, 2009) and worked on the “synthesis of nickel ferrite nanoparticles using the co-precipitation” method (Mostofa *et al.*, 2016). They synthesized nickel ferrite nanoparticles from iron and nickel chloride salts with NaOH as a precipitant and oleic acid as a surfactant to prevent agglomeration. The researchers calcined the nanoparticles at various temperatures between 600 and 1000 °C but the results were good for 10 h at 600 °C. The higher the temperature of the calcination process, the larger the particle size and the wider the particle size range obtained with this chemical process. The peaks shown in the XRD diagram confirmed the nickel ferrite nanoparticles as in Fig. 1. SEM and TEM confirmed the particle size of nickel ferrite to be 8 to 28 nm at a calcination temperature of 600 °C and 45 to 70 nm at 1000 °C.

Morphology of ABS/graphene nanocomposites. The morphology of the composite explained by (Eswaraiah *et al.*, 2011). It can be viewed in the illustrated SEM images shown in Fig. 2.

Figure 2 shows the fracture surfaces of ABS composite films that contain different weights of C18 graphene weight (Okamoto *et al.*, 2016; Okamoto and *at el.*,

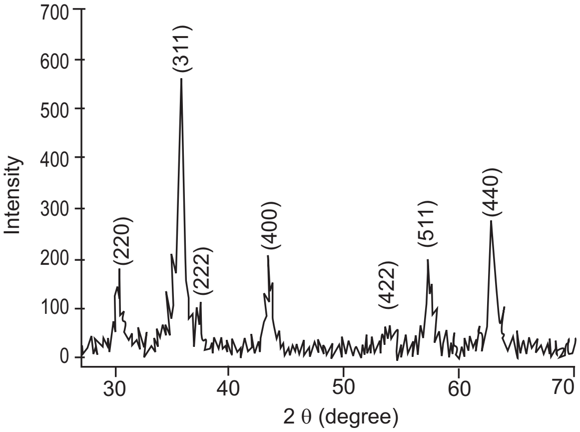

Fig. 1. XRD Pattern.

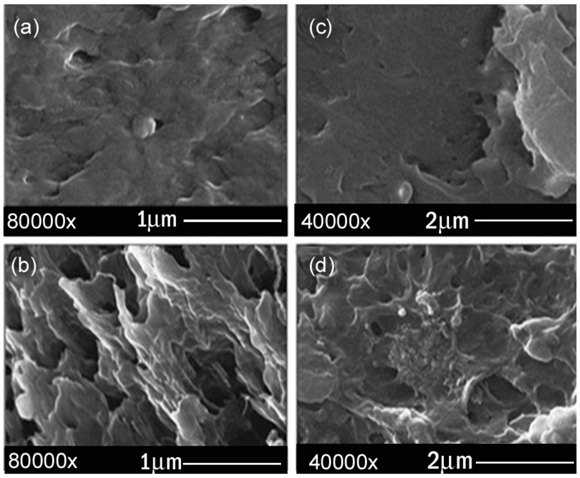

Fig. 2. SEM images of ABS nanocomposites containing **(a)** 0 (pure ABS), **(b)** 1, **(c)** 2 and **(d)** 3 wt. % C18-graphene.

2009). The graph shows the morphology of the platelet orientation distribution. Since there is a difference in the dispersion densities of graphene and ABS polymer, the graphene dispersions are observed in the SEM images. Figure 2 (b-d) shows good dispersion of graphene in ABS at different levels of magnification. It can also be seen that graphene remains in the composite as straight and rigid platelets, indicating that graphene is very rigid. According to (Oaki *et al*., 2011) nano-composites are made from ABS using exfoliated graphene. GO was first synthesized from graphite, then a thermal peeling process was used to obtain exfoliated graphite sheets. SEM images of natural graphite flake and exfoliated graphene are shown in Fig. 3, SEM images of a nano-ABS composite material using exfoliated graphene, manufactured. The use of the solution casting method is demonstrated in Fig. 3c.

PVDF/graphene nanocomposites. The Boehm *et al*. (1962) investigated the impact of lowered graphene oxide on the microstructure, the ferroelectric properties and the deformation of PVDF induced by the electric field. A simple solution casting process was used to produce nanocomposite films. The researchers investigated that by adding RGO to PVDF, the polar phase of PVDF with high pyroelectric and piezoelectric properties, the β phase of PVDF was increased and strengthened. However, this phase decreases after a certain percentage of GERD in the PVDF. It has also been observed that the dielectric properties increase with increasing RGO content in PVDF. The piezoelectric and ferroelectric properties of pristine PVDF were increased several times by increasing RGO to a certain level. Subsequently, it shows a decrease in these properties due to the declination of the β phase (Rahman and Chung, 2013).

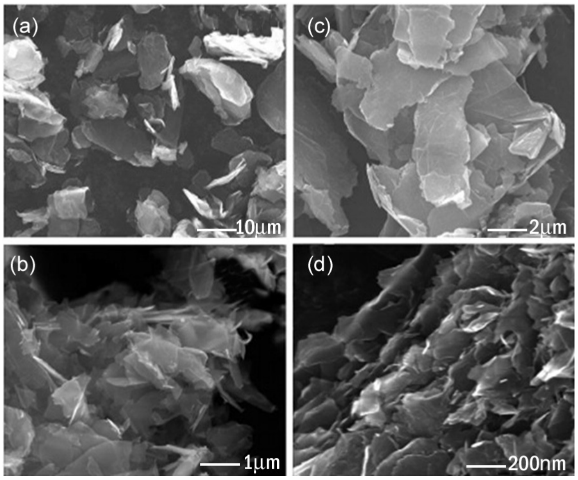

Fig. 3. **(a)** SEM images of natural flake graphite at 800X, **(b)** graphite agglomerates at 3000X, **(c)** exfoliated graphite at 1000X and **(d)** separated sheets of exfoliated graphite at 12000X.

PVDF/ZnO nano-composites. Radha Manohar Aepuru and her coworkers worked on radial zinc oxide and PVDF nanocomposite. The ZnO nanoparticles were produced by a simple "solvothermal process" and the PVDF-ZnO films were made by solution melting. Researchers use dimethylacetamide (DMA) as a solvent for PVDF. As in Fig. 4, it was observed that by adding ZnO to PVDF, the crystalline phases α and γ disappear and β increases. This is due to the incorporation of nanofillers into the PVDF chains, which shrink the chain and reduces the crystallinity of the PVDF matrix, leading to polar crystalline phase due to non-polar phases. It was investigated whether the tensile potency and the extension at break decreased with escalating ZnO content, as has also been shown in other publications. This is due to the weakening of the interaction between PVDF and Nano fillers (Zhao *et al*., 2017). Increase in stress concentration leads to agglomeration phenomena. The researchers find that these dielectric properties increase with a higher percentage of ZnO due to the formation of a pseudo-conductive network not reported in the previous literature (Aepuru *et al*., 2015).

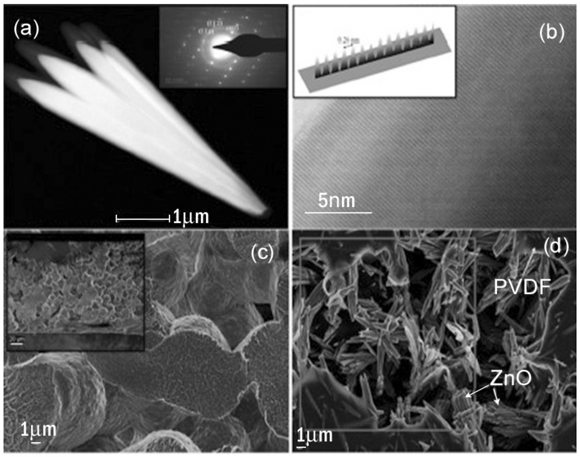

Fig. 4. **(a)** TEM images of 3-D flower-like ZnO **(b)** surface profile of ZnO **(c)** PVDF **(d)** PVDF-RZnO nanocomposite film with 50 wt.%.

PVDF/Cu Nanoparticles-based nano-composites. Arranz and his colleagues worked on a nanocomposite based on PVD-Cu nanoparticles to analyze the effectiveness in protecting against electromagnetic interference. They used a melt processing technique in an extruder to make nanocomposite films. It was reported that the β and α phases of PVDF increased with the escalating incorporation of nanofillers into the PVDF matrix. This unique existence of the α-phase was confirmed by XRD and FTIR analysis. The SEM images also show that a homogeneous dispersion of nanofillers was obtained and by increasing the Cu (Copper) nanoparticles, more aggregates of the Cu nanoparticle content were obtained and a more fibrous structure was obtained. The thermal properties confirmed the formation of different phases in the XRD peaks. The micro hardness value also increases by increasing the percentage of CU nanoparticles, which also corroborated the homogeneous dispersion of the PVDF matrix whitening nano-fillers. The results of the EMI shielding efficiency show that increasing the Cu nano-particles increases this property due to the high conductivity and dielectric constant of the nano films, which is the key factor in reducing electromagnetic interference. 20% of the nanofibers bleach the PVDF matrix and have an SE value of approx 70 dB. In any case, numerous publications show that an SE of 30 dB causes 99% attenuation of the electromagnetic radiation, which is achieved by 10% by weight of the Cu nanoparticle content (Arranz-Andrés *et al*., 2012).

PVDF/Barium titanate particles-based nanocomposites. Nina Joseph and her colleagues investigated the effect of BT particles (barium titanate) on the PVDF polymer matrix for EMI shielding, they made BT particles in the micrometer and nanometer range. The micrometer-sized BT particles were made by a conventional solid-state ceramic process. The nanocomposite films were synthesized by solution casting. From the XRD peaks, it was observed that the tetragonal $BATiO_3$ phase formed along this path. The nanoscale BT particles were examined to show a greater agglomeration than the micrometer-sized BT particles and this agglomerate limits the higher loading of the polymer composite with nanofillers.

They investigated various properties of nanocomposite films that indicate improved dielectric constant by increasing the volume percentage of nanoscale and micro-sized particles. This increase in the efficiency of EMI shielding is due to the increased electrical conductivity of composite foils (Joseph *et al*., 2013).

Material and Methods

Problem statement. The development in the area of materials for electronic and telecommunication industries has increased the problem of electromagnetic interference (EMI), which may affect the performance of precise electronic components and may affect human health in certain manners. Therefore, it is needed to protect the relevant equipment against EMI. The main cause of EMI is related to environmental concerns and therefore a relevant system of shielding is required.

Research objectives. The current research objective is to produce EMI observing materials at the local level to accomplish commercial requirement so that the performance of precise electronic equipment can be improved. These materials also find their usage in defense, on the surface of jets, armed vehicles and safe houses to make these untraceable to radar. And for this purpose, we want to create a materials corner in the R&D Lab comprised of materials testing equipment like a tensile testing machine, scanning electron microscope, dielectric testing facility and X-ray diffraction machine so that these necessary tests can be easily performed.

Research scope. The intended study covers the comparison of the characteristics of PVDF/graphene nanocomposite and PVDF/Graphene/ferrite hybrid nanocomposite for EMI shielding application. Firstly, study works on lab level. Then it will be tested in a particular organization.

Polymer nanocomposites synthetized through solution compounding method. Considering the simplicity and versatility of the solution compounding method, nano-

Table 1. Materials, manufacturer and specification

Material	Manufacturer
Iron nitrate	Sigma Aldrich
Nickel nitrate	Sigma Aldrich
Polyvinylidene fluoride (PVDF)	Sigma Aldrich (Mn=71,000 and Mw= 180,000)
N,N Dimethyl form amide (DMF)	Sigma Aldrich
Graphene	-
Sodium hydroxide (NaOH)	-

composites are found to be easy to manufacture and effective as well. This method is a solvent based system, where the polymer is dissolved in a particular solvent and nano-filler is dispersed through ultra-sonication in resulting solution. Ultra-sonication allows the filler to be homogenously dispersed in the polymer matrix. In this method, shear force induced in the polymer- filler solution is quite lower as compared to melt processing method. By using ultra-sonication in this techniques, polymer-filler can be effectively loaded with very low viscosity than a polymer melt method, which has an advantage over the other synthesized methods such as the melt blending method. After homogenous mixing of polymer-filler in solution, the nanocomposites are molded and casted in desired appropriate shape by evapourating the solvent the electrical conducting nanocomposites fabricated through solution casting method has relatively low percolation threshold. However, the major problem with this technique is its limitation for large-scale production. In the following section, the raw materials used are discussed. Table 1 gives an overview of the raw materials used.

Preparation of nickel ferrite nano particles. Nickel ferrite nanoparticles were prepared *via* a chemical reaction route, in which nitrates of nickel and iron were used in the form of salts, as the starting materials. Firstly, the salts of nickel and iron nitrate were dissolved separately in distilled water under magnetic stirring (400 RPM) corresponding to the molarities of 0.1 M and 0.2 M, respectively. After the formation of simple salt solutions, both were mixed together and heated to a temperature 80-85 °C (Maaz *et al*., 2009). In order to attain a pH value of 7.0, the precipitate was washed with distilled water for several times. Finally, the precipitate was washed with ethanol in order to avoid agglomeration. The extracted precipitates were then, dried in an oven at 65 °C and calcined in an electric furnace at a temperature of 850 °C for 6 h, followed by crushing and grinding processes in a mortar. Several experiments were performed with different calcination temperatures, ranging from 600 to 880 °C. Best results were achieved at 850 °C calcination temperature.

Polymer based nanocomposites. Nanocomposites are multiphase materials structurally comprised of a matrix phase and a reinforcement phase. Reinforcement phase particles are in the dimension of about or less than 100 nanometers (nm) or simply it can be defined as a material having nanoscale repeat distances between different phases, matrix and filler phase, that make up the entire material. These both phases combine together to confer mechanical, thermal, optical, electrochemical and catalytic properties of the nano composites. There is a different method used for preparation of polymer based nanocomposites.

Solution casting method. The simplest and most used method for the preparation of nano fillers based polymer composites is solution casting method. But the limitation of this process is that polymers that can dissolve by themselves in a particular solvent can be utilized only. In this technique, nano-fillers are dispersed in a suspension of polymer in solution form by energetic excitement, in such cases sonication, magnetic stirring or mechanical stirring is utilized in most cases. Then solvent is allowed to evapourate in an environment depending upon the nature of the solvent in order to cast nanocomposites film.

Melt mixing method. Thermoplastic polymers are mostly manufactured by this melt mixing method. Polymeric melt is made under high temperatures and then shear mixing is applied in order to homogenize the nano-fillers within the polymeric matrix. A variety of shapes for the output design can be produced by the processes such as compression/injection molding or using an extruder at high temperature and pressure.

***In-situ* polymerization.** In situ polymerization method the dispersion of nano-fillers is achieved in between polymerization of the monomers. In this process a stronger and a sharp interface between polymeric matrix and nano-fillers can be achieved because as the polymer is in its early growth stages it can result in good interactions with the nano-fillers as compared their formation after polymerization.

Preparation of pure polymer film. Polymer films were prepared by solution casting method. In this method, pure polymer pellets were dissolved in N, N, dimethyl formamide (DMF)/1 g PVDF/20 mL DMF, and kept on magnetic stirring at 35 °C in order to dissolve the polymer in DMF solvent. Magnetic stirring was continued until clear solution appeared. Then the solution was casted in Petri dish and dried in oven at temperature 80 °C and 300 mL bar pressure.

Preparation of PVDF nanocomposites films. PVDF-graphene films were prepared by solution casting route. In brief, PVDF in DMF solution were prepared as discussed earlier (1 g PVDF/20 mL DMF). Then graphene solution in DMF (100 mL/g of graphene in 1 mL of DMF) was prepared and ultra-sonicated (1 cycle) for 10 min, followed by mixing in PVDF-DMF solution under magnetic stirring at 35 °C and 500 rpm. Keep this solution for 20-24 h under stirring at 35 °C. After that, the solution was again ultra-sonicated for 15-20 min in order to achieve homogenous dispersion. Cast the solution in Petri dish and dried in vacuum oven at 80 °C and 300 m bar pressure. After 12 h drying, composite film with throughout dispersion was obtained.

Preparation of PVDF-graphene hybrid nanocomposites films. The same route was employed for PVDF-graphene-spinal ferrite film. The only difference was that mechanical stirring was employed in the place of magnetic stirring for the three-component system. PVDF-graphene solution was prepared as discussed above. Then spinal ferrite nano particles were the first ultra-sonicated (1 cycle) in DMF for 15 min, then mixed in PVDF-graphene solution under mechanical stirring for 4 h and then again ultra-sonicated the hybrid solution. The suspension was then poured in Petri dish for drying at the same condition as discussed earlier.

Number of samples prepared. The Table 2 shows the percentage of PVDF weightage which varies from 95 to 100 and it also shows percentage graphene weightage from 0 to 5 with respect to PVDF.

Table 2. Samples prepared of binary nanocomposites system

PVDF (wt. %)	Graphene (wt. %)
100	0
99.99	0.1
99.97	0.3
99.95	0.5
99	1
97	3
95	5

Table 3. Samples prepared of tertiary nanocomposites system

PVDF (wt. %)	Graphene (wt. %)	Nickel ferrite (wt. %)
97	3	0
92	3	5
82	3	15
67	3	30

Table 3 shows the percentage of PVDF weightage which varies from 67 to 97. Not only it shows percentage graphane weightage which is constant 3 but also the percentage of nickleferrite with respect of PVDF and graphane.

Results and Discussions

Morphological and structural analysis. At the highest Cu content absence of detectable metallic domains of large size across the film is noticed (Aepuru *et al*., 2015; Arranz-Andrés *et al*., 2012) reported that uniform dispersion was achieved of flower like RZnO in PVDF polymer as a matrix, suggesting that solution casting route is an efficient way to disperse nano-fillers in to the polymer matrix and he examined that formation of network like structure at PVDF having 50 wt.% RZnO. (Aepuru *et al*., 2015). SEM images of fractured surfaces for the nanocomposite films were obtained in order to examine the influence of graphene on the morphology and microstructure of nanocomposite films. Figure 5 shows the rough surfaces for PVDF with different weight percentage of graphene from 0.1 weight percent to 5 weight percent. It was observed that graphene was well dispersed in PVDF matrix from 0.1 weight percent to 0.5 weight percent without clustering of graphene but above 0.5 weight percent, partial clustering of graphene occurs and clustering increasing when we increased the graphene but even at high weight percentage of graphene of detectable graphene domain of large size is absent.

Due to such clustering, proper network of graphene (percolation) was achieved whiten the PVDF matrix which is responsible for boosting dielectric and other

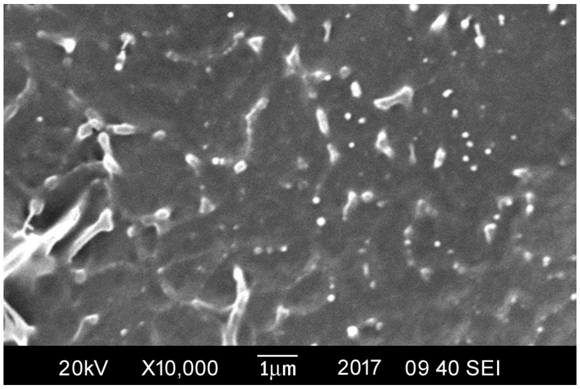

Fig. 5. SEM images of PVDF with different weight percentage of graphene.

properties. Such percolation is helpful for EMI shielding effectiveness. It was noticed that by increasing the graphene content within the PVDF matrix, the more fibrous is the structure achieved. Figure 6 shows the WAXD pattern of $NiFe_2O_4$ nanoparticles. Face centered cubic structure of $NiFe_2O_4$ was confirmed through sharp peaks observed in XRD pattern. It was examined that no secondary phase peaks were noticed. The sharp peaks appearing at 30.2°, 35.7°, 43.4°, 53.8°, 57.4° and on 63° corresponds to (220), (311), (400), (422), (511) and (440) planes of spinal ferrite crystal lattice as discussed earlier by Shanmugavel *et al*. (2015). These values were matched with the standard JCPDS data. Similar results were shown by (Ali and Noorani, 2013) at a temperature ranges from 500 °C to 1000 °C. Researcher showed that by increasing the temperature, size of nanoparticles and crystallinity also goes to increasing. In addition, the samples which were heated at 400 and 500 °C showed super paramagnetic property but at this temperature some unstable phases also exist (Ali and Noori, 2013).

An interesting feature of PVDF is its polymorphism (existence at different crystalline structures). PVDF exists in minimum four different crystalline phases (α, β, γ and δ) depending upon the crystallization condition (Arranz-Andrés *et al*., 2012). XRD measurement was taken at 2θ from 5° to examine the crystalline phases in semi-amorphous PVDF and its nanocomposite films Fig. 6. Pristine PVDF show four main diffractions (Arranz-Andrés *et al*., 2013) corresponds to the following Miller indices: (100), (020), (021) and (110), whereas nanocomposite films show five main diffractions, one extra peak compared to pristine PVDF is assigned to graphene, which appears at 26.53°, while (100), (020), (110) and (021) reflections assigned to α-PVDF and β-PVDF respectively (Arranz-Andrés *et al*., 2012). It was examined that α phase decreases by increasing graphene upto 3-5 weight percent increasing. Some agglomeration may have occurring at this graphene concentration. The conditions for nanocomposite films prepared were such that it yielded a microstructure of α phase predominantly but β phase also appeared. The XRD pattern exhibit intense crystalline peaks with increasing graphene concentration. It was observed that β phase increases by increasing graphene, which is the polar phase of PVDF, as compared to other crystalline phases present in PVDF. The broadening of peaks was observed as graphene are incorporated to the PVDF matrix. These characteristics showed that although the crystalline character is kept in the nanocomposite films but some changes are occurring within the PVDF crystalline regions because of the incorporation of graphene content.

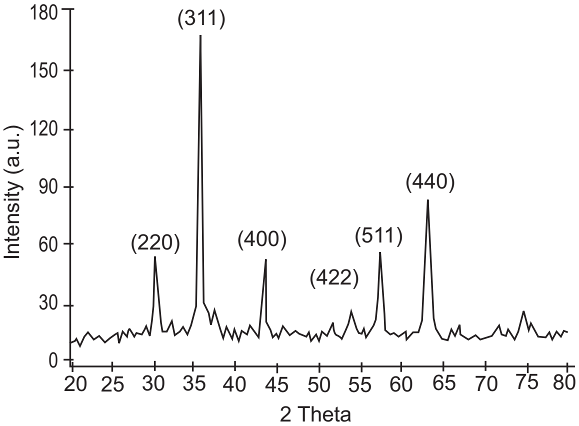

Fig. 6. XRD spectrum of $NiFe_2O_4$ based graphene nanocomposite with different weight percentage.

Thermal properties. ***Differential scanning calorimetry.*** Information about crystalline structure can also be deduced from calorimetric results. Melting and crystallization process, respectively, for the different composition of samples is shown in Figure. Endothermic peaks attribute to the melting process of the PVDF crystallites and its nano composite is displayed (Marega and Marigo, 2003). In the first heating curves, it was observed that melting peak temperatures shifted to lower temperatures in a smaller degree. It is supposed that slightly shifting to the lower temperatures of the nanocomposites samples with higher weight percentage of graphene content is linked to the presence of thin crystals formed during the melt crystallization at 10 °C as compared to the pristine PVDF (Novoselov *et al*., 2004). It was observed that cooling curves promoted to high temperature with increased concentration of graphene, this is because of addition of graphene which acts as a heterogeneous nucleation sites for growth, leading to a faster crystallization process (Sánchez *et al*., 2015). As mentioned earlier, that sometimes two melting peaks are shown in PVDF and its nano composite (Fig. 7) (Marega and Marigo, 2003) which is seemed to be merged in each other, belonging to α and β phases of PVDF. The melting peak at lower temperature around 170 °C is due to the melting of α phase crystal, while at high temperature melting peak at around 180 °C corresponds to β phase of PVDF (Marega and Marigo, 2003). It was examined from the DSC curves that β phase is present in pristine PVDF and then decreasing in lower concentration of graphene and maximum β phase was observed in three weight percent of graphene and after that again decreasing as briefly discussed in WAXD discussion. It was also observed that broader melting endotherms were observed by increasing graphene and nickel ferrite content and that are caused by α phase broader distribution in thickness formed in the nano composite films. Graphene which acts as a heterogeneous nucleation sites for growth, leading to a faster crystallization process (Sánchez *et al.*, 2015).

Dielectric properties of nano composites. As our main application to target EMI shielding effectiveness, dialectic properties are one of the primary requirements to attenuate electro-magnetic radiations (Rahman and Chung, 2013) reported that dielectric constant as well as loss and conductivity improved considerably. Researchers therefore conclude on the conductor-insulator transition which they purported to have occurred at 0.5 wt.%. It is well known that graphene show good conductivity, so we expect that by homogenously dispersing graphene in PVDF as a matrix will attenuate the EM radiations. Dielectric properties were measured by impedance analyzer. Dielectric constant, dielectric loss, AC conductivity and tan δ verses frequency curves of graphene with different weight percentage in PVDF at room temperature are shown in Fig. 8. The high dielectric constant at lower frequency

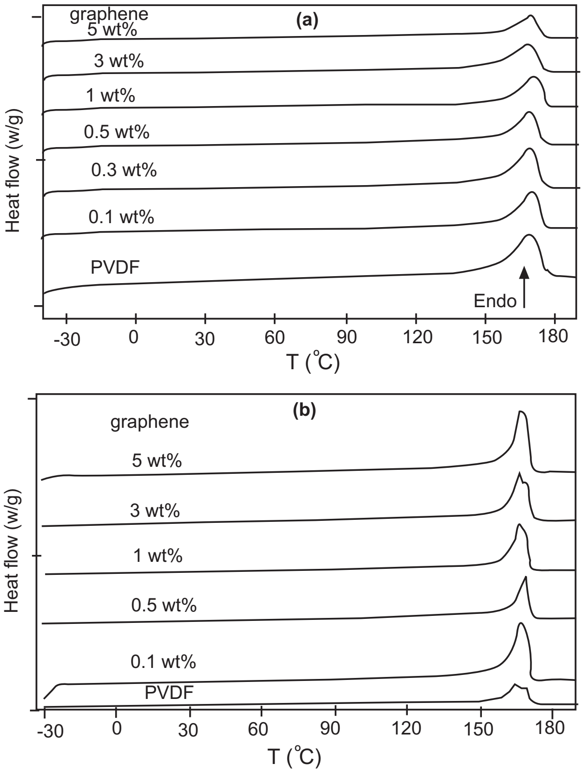

Fig. 7. **(a)** First heat **(b)** second heat curves as a function of temperature obtained from DSC.

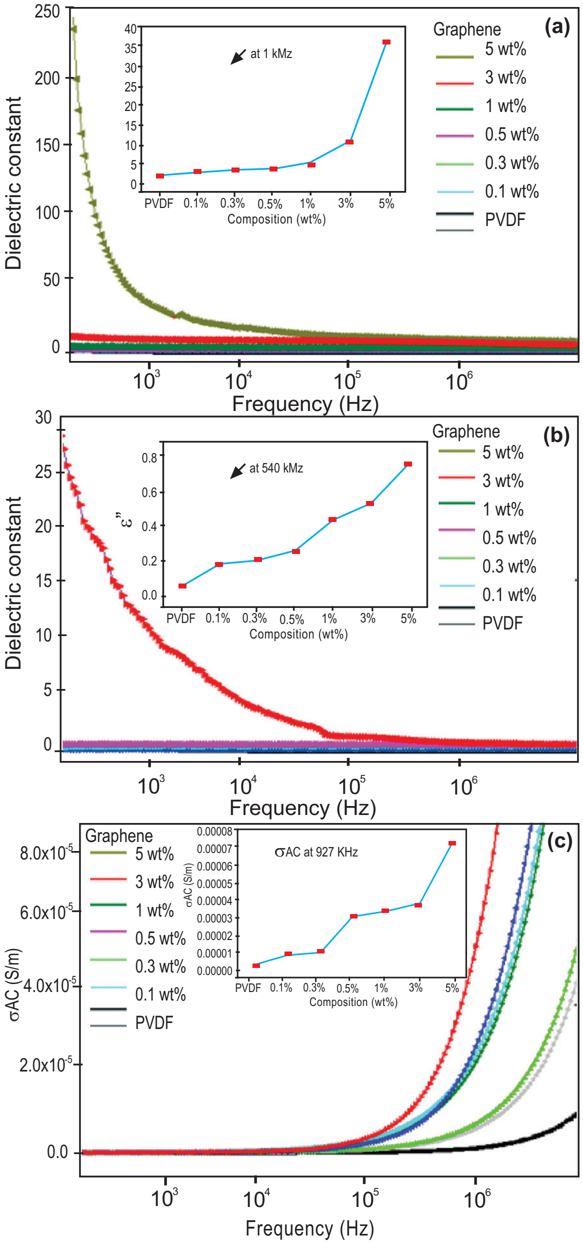

Fig. 8. **(a)** Dielectric constant **(b)** dielectric loss and **(c)** conductivity as a function of frequency.

was assigned to Maxwell-Wagnar-Sillars interfacial polarization, because of heterogeneous conductivity of graphene and PVDF (Rahman and Chung, 2013). It was observed from the curves that by increasing the graphene, dielectric properties increases continuously, but the values increased drastically at 3 and 5 weight percent of graphene. Therefore, we resolve that insulator conductor transition happened at 3 weight percent and this sudden increase in dielectric constant at 3.

Mechanical properties. Mechanical performance is very important factor for nanocomposite in order to insure their using in practical application with advanced properties (conductive, magnetic, antimicrobial, electronic etc). None materials will be marketed/ commercialized, if these necessities are not achievable, autonomously of their goodness of particular functionality. Homogenously dispersion of nano-fillers within the PVDF matrix and the collaboration of nano-fillers and PVDF matrix are the key factor in the resulting mechanical behaviour and they are critical parameters to obtain the strong mechanical reinforcement effect (Arranz-Andrés *et al*., 2013). Figure 9 shows the stress-strain graphs of pure PVDF and of different weight percentage of nano-fillers within the PVDF matrix. The specimen prepared for the tensile test was cut from solution casted films of dimensions (10 mm width, 20 mm gauge length and 0.22 average thicknesses of specimens) according to ASTM D6287 standards at a crosshead speed of 2 mm/min (ASTM D882) (Waheed *et al*., 2016). Average values were taken from five samples and tests were performed at room temperature for all the specimens. The figure also shows that by increasing the weight percentage of graphene, stiffness value increases which is calculated from the slop of the elastic region. Tensile strength also increases up to 0.5 weight percent of graphene and then decreases slightly as weight percentage increases (Fig. 9) due to the agglomeration of graphene within the PVDF matrix. Young modulus (Fig. 9) increasing and strain percentage (Fig. 9) is decreasing on increasing the graphene weight percent. Pristine PVDF shows a presence of graphene is basically restricted the chain mobility in PVDF matrix.

Crystallinity also increasing by increasing the weight percentage of graphene by homogenously dispersion of graphene which shows the PVDF-graphene interaction which leads to improvement in young modulus and to change in deformation mechanism with respect to pristine PVDF, as PVDF is semi-crystalline polymer. These distinctions in the mechanical response are also identified to the elongation at break and tensile strength.

Presence of graphene is basically restricted the chain mobility in PVDF matrix. Crystallinity also increasing by increasing the weight percentage of graphene by homogenously dispersion of graphene which shows the PVDF-graphene interaction which leads to improvement in young modulus and to change in deformation mechanism with respect to pristine PVDF, as PVDF is semi-crystalline polymer. These distinctions in the mechanical response are also identified to the elongation at break and tensile strength. The stress-strain curve for different weight percentage of nickel ferrite, as three weight percent graphene is constant in all these ferrite compositions is shown in Figure. It is clear from curves that by increasing the weight percentage of nickel ferrite, mechanical properties decrease *i.e* tensile strength, stiffness, modulus elongation at break etc. The decrease in mechanical properties by increasing the weight

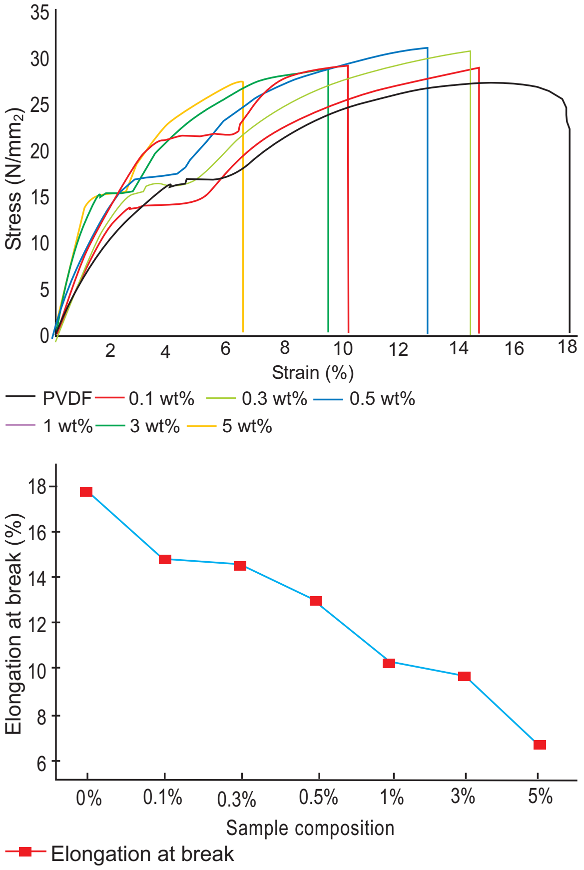

Fig. 9. Stress-strain curves and elongation at break graph of pristine PVDF and PVDF-graphene nano composites at room temperature.

percentage of nickel ferrite is basically a restriction of polymer chains within the system and by agglomeration phenomena.

EMI shielding of PVDF based nanocomposites. The property for preventing the effect of external elements resulting in the term shielding effectiveness (SE). Here SE from signal loss after transmission has been calculated for the synchrotron radiation (3.06 EHz). SE (units of decibels) is defined as the ratio of radiation transmitted in presence of a given material, E0 and without the material present, E1

$SE\ (dB) = 20\ \log E_o/El$

Here, 28 dB of SE was reported for 7 weight% loading of f-G over a frequency range Eswaraiah *et al*. (2011) studied EMI SE of functionalized graphene (f-G)/PVDF nano from 1-8 GHz. This can be implemented for different samples.

EMI shielding effectiveness was measured by vector network analyzer (VNA). Results emanating varying composition of samples is shown in Fig. 10. Pure polymer shows very small shielding effectiveness due to the dipoles generation in their chemical bond. Figure indicating that ~ - 22 dB is obtained for 3 weight percent loading of graphene, producing a conductive network within the polymer matrix. When the filler concentration is increasing with spinal ferrite along with these graphene Nano-fillers then EMI further increases to ~ -28 dB indicating the magnetic dipoles along with conductive network are obtained. As we further increase the filler concentration then EMI Shielding effectiveness decreases pointing toward the formation of agglomeration of nano fillers, which decreases the overall properties of EMI Shielding effectiveness. Further work on complex nano fillers combination is under process which may show better results than this.

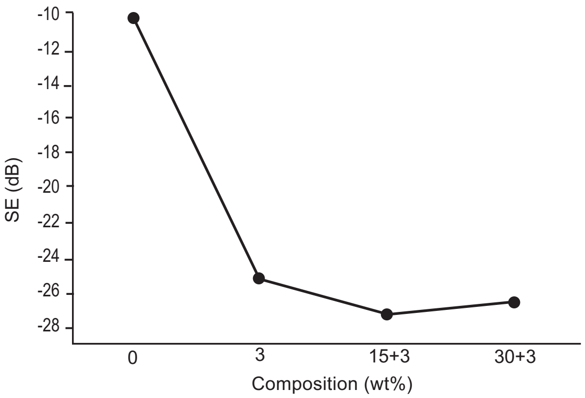

Fig. 10. EMI shielding effectiveness at 5.7 GHz.

Conclusion

Graphene and other 2D material have outstanding electrical and mechanical properties. One of the interesting and promising properties of graphene is its conductivity (25000 cm²/volt-sec *i.e*. 200 times of silicon). Nickel spinal ferrite nanoparticles exhibit magnetic properties having cubic crystal structure. PVDF is a semi-amorphous polymer, which easily dissolved in N, N dimethyl formamid (DMF). It is having insulative character with glass transition temperature -40 °C and melting temperature is 170 °C. By combining such materials together, would give good properties such as dielectric, thermal, EMI shielding and mechanical properties.

Initially polymer nanocomposite were made with graphene as a conductive nano-filler and PVDF as insulative polymer matrix and five different weight percentages of such compositions were prepared. It was observed that by increasing graphene concentration mechanical properties were enhanced up (*i.e*. ultimate tensile strength, modulus) to 0.5 weight percentages and after that ultimate tensile strength decreases, while modulus increases in the case of 1, 3 and 5 weight percent. Similarly, dielectric properties were enhanced by increasing weight percentage of graphene because of its conductive nature.

Among the five different weight percentage, one of them (97-3) for PVDF and graphene were selected for further addition of spinal ferrite. It was observed that by incorporated spinal ferrite mechanical properties decreases but dielectric properties enormously increases. So, such hybrid systems can be of wonderful choice for attenuating EM radiations and can be used for stealth technology.

Future recommendations. Innovative changes occurred in the world regarding the research in the field of nanocomposite materials. To make progress in research and development field we should provide some basic facility on university and college level to the students. For this type of research activity regarding composite materials, nano-material metal and non-metal, I will recommend the following equipment in our university lab, which requires financial commitment from outside sources.

Conflict of Interest. The authors declare that they have no conflict of interest.

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