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Dielectric Behavior and AC Conductivity of PVDF/MWCNT Composites: Role of Interfacial Polarization and β-Phase Formation

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Abstract

Poly(vinylidene fluoride) (PVDF) is a semi-crystalline polymer distinguished by its remarkable ferroelectric, piezoelectric, and pyroelectric characteristics, which are most pronounced in its electroactive β-phase. Embedding multi-walled carbon nanotubes (MWCNTs) within the PVDF matrix gives a viable route to enhance its dielectric, electrical, and thermal characteristics through interfacial polarization, β-phase induction, and micro-capacitor network formation. In this study, PVDF/MWCNT nanocomposites containing MWCNT concentrations of 0.5, 1, and 2 wt% were made-up using solution casting followed by hot pressing. The dielectric and electrical behaviors were examined within the frequency range of 0.5–10 kHz and temperature range up to 135 °C via impedance spectroscopy. The obtained results revealed that the dielectric constant exhibits an increasing trend with rising temperature as well as higher MWCNT content, reaching a value of approximately 60 at 3 kHz and 135 °C for the 2 wt% composite, primarily due to enhanced interfacial polarization and β-phase content.

Keywords: PVDF; MWCNT; Nanocomposites; Dielectric properties; Interfacial polarization; AC conductivity; Energy storage

1. Introduction

In the family of polymers, Poly(vinylidene fluoride) (PVDF) has attracted considerable research attention owing to its ferroelectric, pyroelectric, and piezoelectric properties, which make it durable and effective in a variety of technological settings. Poly(vinylidene fluoride) (PVDF) which is a semi-crystalline polymer known to crystallize into at least four polymorphic forms (α, β, γ, and δ), with the β-phase being particularly significant because to its favorable pyro- and piezoelectric properties [1, 2]. To improve the electrical properties of PVDF, more efficient strategies for promoting β-phase crystal formation in PVDF are essential. The β-phase of PVDF is typically not formed through melt crystallization but is generally achieved using various techniques such as tensile or uniaxial compressional deformation of the phase, blending with small amounts of poly(methyl methacrylate) (PMMA) or poly(o-methoxyaniline) (POMA), quenching followed by annealing [3,4], application of a strong electric field, or crystallization from solution under specific conditions [5, 6]. Among these methods, solution-grown β-phase formation using a single solvent is most commonly employed due to its simplicity and controllability. However, limited studies have focused on β-phase induction through solvent interactions in mixed solvent systems.

According to Ma, W. et al., 2008, variations in DMF content within mixed solvents markedly

affect the crystalline phase development of PVDF as well as the morphology of the resulting films. The incorporation of nanoparticles into polymers has garnered considerable attention owing to their outstanding mechanical strength, excellent electronic behavior, and unique optical characteristics. These features have led to a wide range of proposed applications, such as sensors, field emitters, and advanced electronic components. Poly(vinylidene fluoride) (PVDF) and its copolymers have gained significant attention due to their wide-ranging applicability in electromechanical systems. A variety of techniques have been investigated to enhance the dielectric constant and overall electrical performance of the PVDF matrix. The addition of functional fillers—including ceramics, semiconductors, metals, metal oxides, and carbon nanotubes (CNTs)—has proven effective in significantly improving the dielectric performance of PVDF-based composites. However, polymers heavily loaded with ceramics, semiconductors, or metals tend to lose their inherent flexibility. Moreover, the dielectric constants achieved in such polymer composites so far have generally remained limited to only a few hundred [7]. The outstanding electrical and dielectric properties of silica have been widely exploited in polymer systems. Additionally, encapsulated nanosilica has been synthesized within poly(methyl methacrylate) (PMMA), exhibiting distinct morphological characteristics [8]. Silica nanofillers have been found to markedly enhance the nucleation density and crystallization rate in poly(tetramethylene-terephthalamide). Conducting polymers notably polyaniline (PANI) and polypyrrole (PPY) are often employed as host matrices for the uniform dispersion and stable incorporation of inorganic nanoparticles. The most common approach for preparing conducting polymer nanocomposites containing oxide nanoparticles is the direct polymerization method. Recently, multi-walled carbon nanotubes (MWCNTs) have been functionalized with 3,4,5-trifluorobromobenzene (TFBB) to enhance the dispersion within the PVDF matrix. The enhanced interfacial interaction between TFP-functionalized MWCNTs and PVDF arises from the abundant fluorine groups on the TFP-MWCNT surface. Moreover, functionalization of MWCNTs with TFBB led to the development of a novel MWCNT/PVDF electroactive polymer composite (EAPC), which exhibited an exceptionally high dielectric permittivity ($\varepsilon \approx 8000$) beyond a critical concentration of TFP-MWCNTs, while maintaining the inherent flexibility of the polymer matrix [9]. Harmon et al. [10] developed conductive and transparent CNT–polymer composites by integrating sonication-assisted dispersion, in situ polymerization, dissolution, and film casting. In this process, CNTs were mixed with deinhibited methyl methacrylate (MMA) and ultrasonically dispersed. Dupire et al. [11] patented in 2001 a fabrication technique for reinforced polymers that achieved simultaneous orientation and uniform dispersion of polymer chains and carbon nanotubes by stretching the nanotube–polymer blend in both molten and solid states under high shear. Despite successful fabrication, the nanocomposites demonstrated only limited transparency within the visible region of the spectrum. Niu et al., 2003 described a method to produce mechanically strong and electrically conductive PVDF–CNT composites through two distinct fabrication techniques [12]. Using the solution approach, PVDF was first dissolved in a suitable solvent (e.g., acetone), followed by the incorporation of CNTs, which were dispersed through sonication. Subsequently, the mixture was precipitated with a nonsolvent, such as water, and then subjected to filtration and drying. Using the melt compounding approach, PVDF and CNTs were mixed at high temperatures, allowing the polymer to melt and disperse the nanotubes evenly throughout the matrix. The study revealed that composites prepared via the solution method exhibited superior electrical conductivity compared to those produced by melt compounding. Furthermore, the authors reported that these composites demonstrated higher conductivity than most previously known polymer-based composites.

CNTs are incorporated into PVDF and other electroactive polymers serves to enhance both

mechanical and electrical characteristics particularly conductivity and piezoelectricity, making them suitable for advanced electronic devices [13, 14, 15, 16]. These PVDF/CNT composites exhibit low weight and high flexibility, effectively merging the processability of PVDF with the exceptional electrical properties imparted by CNTs [13, 14]. Despite their potential, PVDF/CNT composites present certain processing challenges. CNTs often aggregate into clusters larger than a few hundred nanometers, resulting in stress concentrations in the polymer matrix that may cause early mechanical failure. Moreover, elevated CNT concentrations can complicate PVDF processing via standard melt-mixing methods, such as extrusion, injection molding, and blow molding [15]. The high cost of CNTs further limits their widespread use as fillers in polymer matrices [15, 16]. Overcoming these challenges remains a key focus in PVDF/CNT composite development.

Considerable research efforts in recent years have focused on nanocomposites formed from PVDF and carbon nanotubes (CNTs). Levi and colleagues [16] were early investigators of PVDF/CNT systems, fabricating nanocomposites of pure PVDF and its copolymers with single- and multi-walled carbon nanotubes (MWCNTs) through solution casting. The researchers achieved homogenous dispersion throughout the nanocomposites and demonstrated that CNT inclusion modifies the crystalline phase of PVDF. Owens and colleagues [17] fabricated SWNT/PVDF composites via evaporation of acetone suspensions containing PVDF and SWNTs, and examined the polymer–nanotube interactions using Raman and NMR techniques. Using melt blending in an internal mixer, Nam et al. [18] prepared PVDF/MWCNT nanocomposites and examined how their morphology influences physical properties. Similarly, Chen et al. [19] prepared PVDF/CNT nanocomposites through melt blending and examined their morphological and rheological, observing that the addition of CNT increased the complex viscosity, storage modulus, and loss modulus, while reducing damping factor.

Optimizing both performance and cost requires the development of PVDF/CNT composites with minimal CNT loading that nonetheless provide substantial improvements in properties. Studying the percolation threshold of such systems helps determine the optimal CNT concentration at which maximum electrical conductivity is achieved. Beyond this threshold, conductivity remains relatively constant, eliminating the need for additional CNT loading. Electrical conductivity measurements and rheological studies can be employed to investigate the percolation threshold, as morphological transitions due to nanotube network formation lead to pronounced changes in both electrical and rheological responses [20].

The introduction of MWCNTs into PVDF enhances the dielectric performance primarily through three mechanisms: (i) promotion of the electroactive β-phase due to the strong interfacial interactions between PVDF chains and CNT surfaces [21]; (ii) increased interfacial (Maxwell–Wagner–Sillars) polarization, resulting from charge accumulation at the CNT–polymer boundaries [22]; and (iii) formation of microcapacitor networks, where isolated conductive CNT domains are separated by the insulating polymer, leading to high permittivity without direct electrical percolation [23]. However, the improvement is highly dependent on the dispersion quality, filler loading, and surface functionalization of the CNTs.

Experimental studies report that well-dispersed MWCNTs can increase the dielectric constant of PVDF from ~10 to over 70 at 1 kHz, with acceptable dielectric loss values when filler loading remains below the percolation threshold (~1–3 wt%) [24]. For instance, Lin *et al.* [25] achieved a dielectric constant of 74.6 at 1 kHz for a 5 wt% MWCNT/PVDF three-layer composite, while maintaining moderate dielectric losses. Functionalized MWCNTs and electrospun aligned fibers have shown improved β-phase crystallinity and reduced leakage current [26], whereas core–shell and hybrid approaches combining MWCNTs with ceramic or MXene fillers further balance

permittivity enhancement and dielectric breakdown strength [27, 28].

Despite the notable progress, several challenges remain, including increased dielectric loss and reduced breakdown strength near the percolation threshold [29]. Current research focuses on optimizing CNT surface treatment, filler hybridization, and multilayer or gradient architectures to maintain high permittivity with low losses [30, 31]. Emerging trends also emphasize energy-storage parameters—such as recoverable energy density and charge–discharge efficiency rather than permittivity alone, to better evaluate the practical potential of PVDF/MWCNT systems [32].

This work aims to explore the dielectric properties of PVDF/MWCNT nanocomposites, providing insights into the effects of filler concentration (wt %) and temperature on dielectric performance across a range of frequencies. It is possible to get insight into how the presence of carbon nanomaterials impacts the resulting dielectric properties of the MWCNT-PVDF composites. In addition, we make an effort to provide comprehensive experimental data that might aid in improving knowledge of the network structure and electrical characteristics of MWCNT - PVDF composites for use in heating devices and/or conducting composites. Our prior research on the activation energy and dielectric characteristics of carbon black-epoxy composites at various temperatures, as well as the analysis of the effects of low weight % filler on the dielectric properties of MCWNT-epoxy nanocomposites, are both complemented by this work [33, 34]

2. Experimental details

2.1 Materials

Poly(vinylidene fluoride) (PVDF) is a semi-crystalline polymer characterized by its high dielectric permittivity, excellent thermal stability, and remarkable chemical resistance. As a result, PVDF is widely utilized in both industrial and biomedical fields for the fabrication of capacitors, transducers, and sensors. In present studies Poly (vinylidene fluoride) of average Mw ~180,000 by GPC, average Mn ~71,000, beads or pellets of Sigma Aldrich Company is used with the dielectric constant of 8.0 - 9.5 at 100 Hz (ASTM D 150). Industrial-grade multi-walled carbon nanotubes (MWCNTs, type 1205YJ) with a purity exceeding 95%, outer diameter of 10–20 nm, inner diameter of 5–10 nm, length of 10–30 μm, specific surface area of 180–230 m^2/g, and bulk density of 0.04–0.05 g/cm^3 were procured from Nanostructured & Amorphous Materials, Inc., USA.

Composite preparation

Poly Vinylidene Fluoride (PVDF) polymer was dissolved in N, N-Dimethylacetamide (DMAc) at 100°C. Care was taken to disperse MWCNT uniformly through the composite. Without any further purification, a measured quantity of MWCNTs was ultrasonically dispersed in the same organic solvent, N, N-Dimethylacetamide (DMAc), for up to 2 hours to obtain a stable suspension. Subsequently, this MWCNT suspension was added to the PVDF solution, followed by additional stirring and ultrasonic treatment for 30 minutes to ensure uniform dispersion. Films were prepared by casting this solution on glass substrates. The films were etched after the evaporation of the solvent. Afterwards, suitable amount of film was taken in a pellet die of 10mm diameter and consequently molded by hot pressing at about 125°C and 10 MPa. Different composite samples of pure PVDF, 0.5 wt%, 1 wt%, and 2 wt% Multiwall carbon nanotube PVDF composite were prepared. The final samples with a disk-shape of 10mm in diameter and around 2mm thick were obtained. Using the above mentioned preparation method, samples of different wt % were prepared and used for further study. For electrical measurements, silver paste was used to paint the electrodes onto the sample surface. The AC electrical properties of the samples were then characterized using a Wayne Kerr 6500B impedance analyzer.

3. Characterization

Dielectric measurements

The dielectric measurements were carried out using Wayne Kerr 6500B impedance analyzer in the frequency range 500Hz to 10kHz for pellet samples using a solid sample holder Wayne Kerr

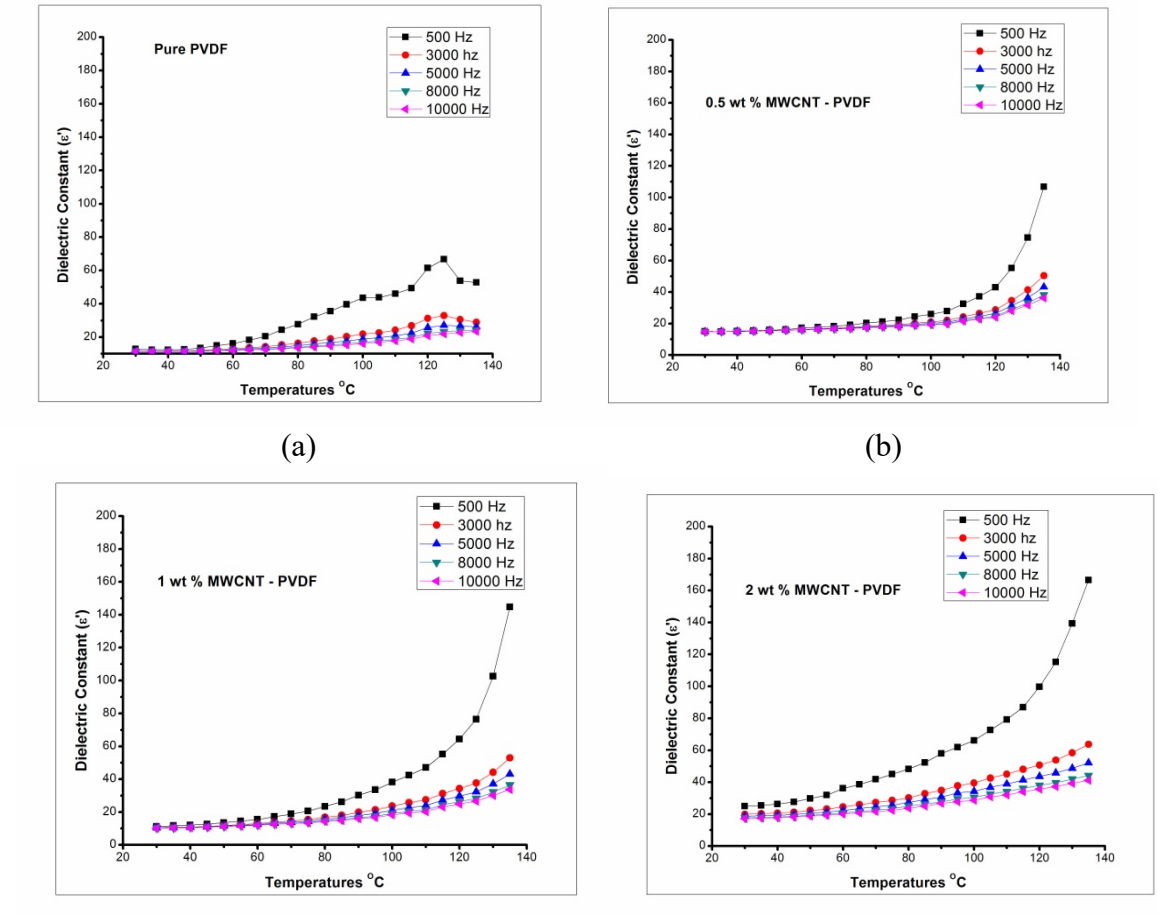
(TF-1000). A high-temperature furnace is connected with the impedance analyzer through RS-232 cable and the impedance analyzer with the desktop computer through a GPIB cable (National Instruments). The frequency dependence of the real and imaginary components of permittivity was thoroughly investigated across a range of temperatures.

The term dielectric constant (ε') refers to the measure of the reduction in Coulombic interactions between ion pairs within polymer electrolytes. Consequently, the dielectric properties of a material or composite provide valuable insights into the ionic and molecular interactions within the polymer electrolyte, as well as a deeper understanding of ion transport behavior. Increases in ions are always implied by increases in the dielectric constant [33]. It is detailed elsewhere how to measure the epoxy composite's dielectric properties [33, 34].

4. Results and Discussions:

4.1 Dielectric Constant (ε′)

To observe an effect of temperature, frequency and filler concentration on dielectric constant, its variation as a function of frequency and filler concentration is plotted at different temperatures. Figures 4.1(a–d) illustrates the variation of the dielectric constant (ε') with temperature (T) for (a) pure PVDF, (b) 0.5 wt%, (c) 1 wt%, and (d) 2 wt% multi-walled carbon nanotube (MWCNT)/PVDF composites, measured at frequencies of 0.5, 3, 5, 8, and 10 kHz, respectively



(a) (b)

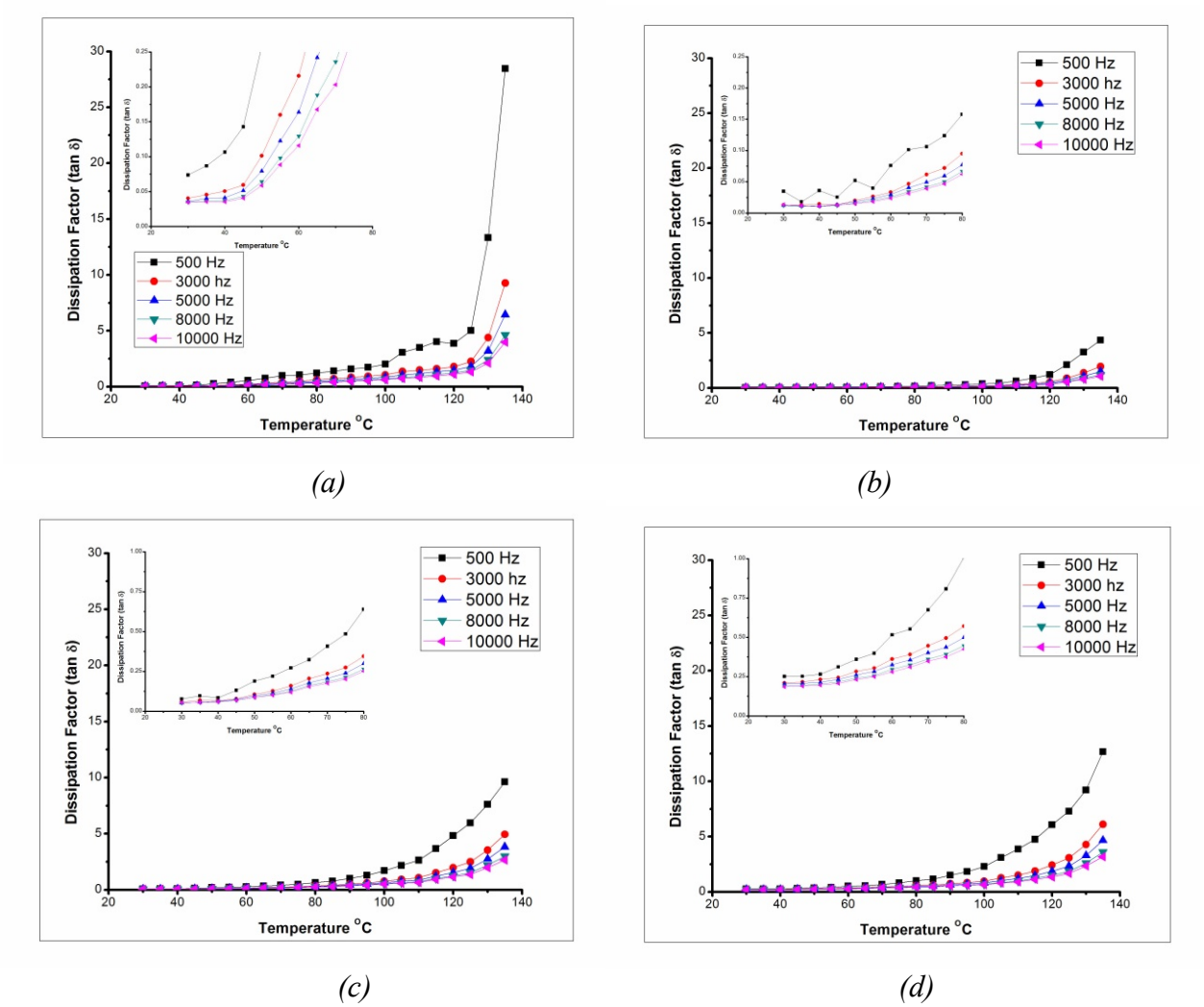
(c) (d)

Figures 4.1(a–d) depicts the variation of the dielectric constant (ε') with temperature (T) for (a) pure PVDF, (b) 0.5 wt%, (c) 1 wt%, and (d) 2 wt% multi-walled carbon nanotube (MWCNT)/PVDF composites, measured at frequencies of 0.5, 3, 5, 8, and 10 kHz, respectively.

For pure PVDF, the dielectric constant decreases with increasing frequency. This trend becomes even more pronounced in nanocomposites with higher MWCNT loadings. Electrons, having a much smaller mass than ions, respond more quickly to a changing electric field. For electric fields oscillating at very high frequencies, such as those of light, only electronic polarization can occur. At lower frequencies, relative displacement of positive and negative ions becomes possible. The orientation of permanent dipoles, which involves molecular rotation, can occur only under relatively slow oscillations, typically in the MHz range or lower.

4.2 Dissipation Factor (tan δ)

Figures 4.2 (a-d) shows the variation of dissipation factor (tan δ) with different temperatures (T) for four samples i.e. (a) pure PVDF, (b) 0.5 wt%, (c) 1 wt% and (d) 2 wt% Multiwall carbon nanotube - PVDF composite at 0.5, 3, 5, 8 and 10 kHz respectively.

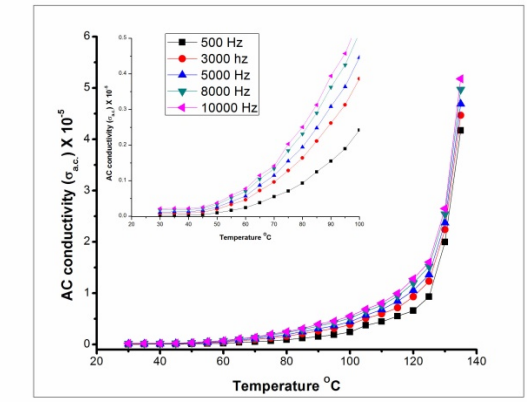


Figures 4.2 (a-d) Variation of dielectric dissipation factor (tan δ) with different temperatures (T) for four samples i.e. (a) pure PVDF, (b) 0.5 wt%, (c) 1 wt% and (d) 2 wt% Multiwall Carbon nanotube - PVDF composite at 0.5, 3, 5, 8 and 10 kHz respectively.

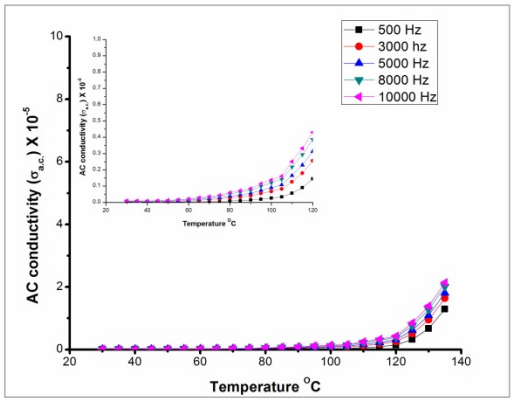
As a common characteristic for all four samples, dissipation factor increases with temperature and decreases with frequency. The smallest values of tan δ are obtained for 0.5 wt% MWCNT loaded PVDF composite.

4.3 AC Conductivity (σ_{ac})

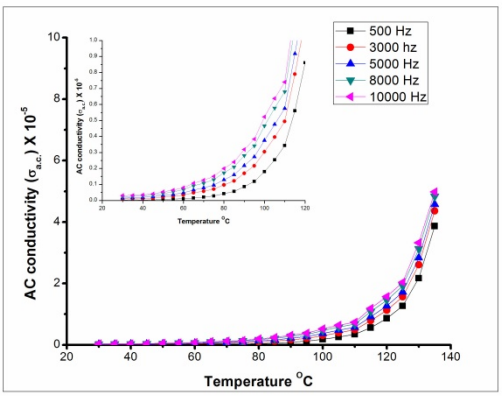
Figures 4.3 (a-d) show the plots of AC conductivity (σ_{ac}) with different temperatures (T) for four samples i.e. (a) pure PVDF, (b) 0.5 wt%, (c) 1 wt% and (d) 2 wt% Multiwall Carbon nanotube - PVDF composite at 0.5, 3, 5, 8 and 10 kHz respectively. Value of AC conductivity increases with the increase in temperature but a reverse trend is followed with the frequency. With the increase in frequency, the AC conductivity value decreases.



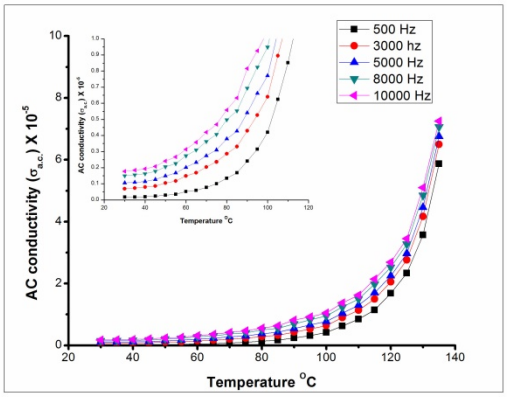
(a)



(b)



(c)



(d)

Figures 4.3 (a-d) Change in AC conductivity (σ_{ac}) with different temperatures (T) for four different samples i.e. (a) pure PVDF, (b) 0.5 wt%, (c) 1 wt% and (d) 2 wt% Multiwall Carbon nanotube - PVDF composite at 0.5, 3, 5, 8 and 10 kHz respectively.

The dielectric constant was observed to increase with rising MWCNT content, reaching approximately ∼60 at 3 kHz and 135 °C for a composite containing 2 wt% MWCNT. The dielectric constant (ε') of the 2 wt% MWCNT–PVDF composite is noticeably greater than that of the other MWCNT–PVDF composite samples. The observed increase is likely due to interfacial polarization in the MWCNT-filled heterogeneous PVDF system and the creation of multiple mini-capacitive regions within the PVDF matrix. The data show that the dielectric permittivity and loss increased with increasing temperature because of an induction of polarization at high temperatures [35].

MWCNTs, as conductive fillers with a high aspect ratio, effectively enhance the dielectric constant. Additionally, interfacial polarization,

which typically occurs at the interface between the polymer and conductive fillers, further contributes to the significant increase in dielectric constant.

At low filler concentrations, the dielectric constant (ε') remains very close to the ε' of the pure PVDF, since the fillers are dispersed individually or are present as small clusters in the matrix. If the concentration of MWCNTs is large enough to establish a percolating network via agglomerate connections, the resulting composite exhibits conductivity. However, a lower dielectric constant (ε') is obtained, when compared with samples of other wt% MWCNT-PVDF composites. According to Pan et al., the conductivity in polymer composites containing MWCNTs is largely dictated by the establishment of a percolating conductive network. A uniform dispersion of MWCNTs, resulting from strong polymer-nanotube interactions, contributes minimally to electrical conductivity enhancement but plays a significant role in mechanical reinforcement [36]. The fact that the composites have the different content of MWCNTs exhibit visibly different dielectric constant (ε') reveals the dispersion state of MWCNTs in polymer matrix [20]. The observed differences are likely attributable to variations in the aspect ratio of the MWCNTs and the mixing conditions employed [37]. In the β-phase of PVDF, the dipoles align additively, resulting in a network of dipoles that enhances spontaneous polarization, thereby facilitating the conduction process [38]. The emergence of the β-phase can be promoted through various techniques, one of which is melt blending with MWCNTs [16].

Upon heating the PVDF sample to 100 °C, the crystallinity of the PVDF sample increased, beyond α-relaxation temperature. This promotes the conversion from the α-phase to the β-phase through conformers motion without significant crystal deformation, resulting in an observed increase in β-phase content. This behavior may be attributed to the fact that, at the annealing temperature of 100 °C, the material's viscosity decreases sufficiently to allow molecular mobility but remains high enough to inhibit crystal orientation. The increased chain mobility facilitates the recognition of conformer structures, enabling active chain motion and reorientation within the crystalline region through trans–gauche conformational exchange in the β-phase. The β-phase exhibits a faster crystallization rate than the other phase of PVDF. The β-phase structure consists of extended trans-zigzag chains linked by skew bonds or alternating gauche–trans conformations.

We infer that the elevated permittivity of the composite arises from the high aspect ratio of the incorporated MWCNTs. The elevated β-phase crystallinity can be attributed to the high aspect ratio of CNTs, whose zigzag carbon arrangement matches favorably with the all-trans chain conformation of PVDF. The enhanced dielectric behavior of the PVDF/MWCNT composite system can also be attributed to the change in α-phase content and the interfacial polarization between PVDF and MWCNTs arising from nanotube entanglement. Additionally, this improvement can be explained by the mini-capacitor principle, wherein the dispersed MWCNTs act as conductive regions separated by the insulating polymer matrix, forming numerous mini-capacitors within the composite. Within the PVDF matrix, MWCNTs act as conductive domains separated by the insulating polymer, effectively forming a network of mini-capacitors. The number of these capacitive junctions increases with higher MWCNT loading, facilitated by COOH functional groups on the nanotube surfaces that promote interfacial polarization. As the inter-tube distance diminishes with increasing concentration, the capacitance of these mini-capacitors correspondingly increases [39].

For a significant increase in electrical conductivity, MWCNTs must be positioned sufficiently close to establish a continuous conductive network, which promotes efficient electron tunneling or hopping process. In the molten polymer state, however, nanotube–polymer interactions restrict chain mobility and interfere with polymer relaxation, thereby requiring a more densely packed nanotube network to reach the electrical percolation threshold. Once this threshold

is surpassed, MWCNT–MWCNT interactions predominate over PVDF–MWCNT interactions, reducing the effective contact area between polymer chains and nanotubes. The strong van der Waals attraction among CNTs favors nanotube agglomeration and limits their wetting by the polymer. This phenomenon underscores the need for chemical functionalization of CNTs to improve dispersion, interfacial adhesion, and ultimately the mechanical performance of PVDF-based nanocomposites [20, 40].

Figure 4.1 (d) demonstrates that at 2 wt% loading, MWCNT-based PVDF nanocomposites display a distinct enhancement in conductivity, suggesting a semiconducting nature of the sample [41]. This enhancement can be understood through the tunneling conduction mechanism (TCM), in which electron transport occurs via tunneling between neighboring MWCNTs. The carboxylic groups present on the surface of MWCNTs partially suppress tunneling current, making the tunneling process more difficult and resulting in a moderate increase in conductivity. As the concentration of CNTs rises, the interparticle distance decreases, facilitating tunneling and thereby improving conductivity [42]. At sufficiently high MWCNT contents, the composite exhibits frequency-independent conductivity, indicating electron-dominated charge transport. Overall, the electrical conductivity is strongly dependent on MWCNT loading: at lower concentrations, it increases with frequency, while at higher concentrations it approaches DC behavior and loses dielectric characteristics. These results align well with previously reported studies [43, 44].

A continuous conductive network of nanotubes can only be established when the MWCNT loading reaches the percolation threshold. However, aggregation of nanotubes reduces the density of random interconnections, even at identical filler concentrations. Therefore, for the same MWCNT content, a well-dispersed system promotes the formation of a more compact and efficient conductive network, which can be detected through rheological measurements. Consequently, applying high shear intensity and prolonged mixing facilitates homogeneous dispersion of MWCNTs within the PVDF matrix, thereby enabling the formation of an effective conductive network more readily [37].

With increasing frequency, the dipoles experience reduced time for orientation in response to the applied electric field. Consequently, both the dielectric constant and loss factor decrease for all composites containing filler concentrations above the percolation threshold. The dielectric constant and loss factor both increase with higher filler loading at a fixed frequency, which can be attributed mainly to space-charge interfacial polarization. Each CNT particle embedded in the insulating polymer matrix can be treated as a nanocapacitor. With higher CNT loading, the population of these nanocapacitors increases, resulting in enhanced net capacitance and a corresponding rise in the dielectric loss [23, 45].

Figures 4.2 (a-d) shows the variation of dissipation factor (tan δ) with different temperatures (T) for (a) pure PVDF, (b) 0.5 wt%, (c) 1 wt% and (d) 2 wt% Multiwall Carbon nanotube - PVDF composite at 0.5, 3, 5, 8 and 10 kHz respectively. The data values show that the value of dissipation factor (tan δ) increases with the increase in temperature and decreases with an increase in frequency. It is clear from the Figures 4.2 that the dielectric loss is relatively low for 0.5 wt% MWCNT – PVDF sample, while those with higher wt% samples lead to much more pronounced increase in tan δ values. The increased dielectric loss is related to the increase in electrical conductivity due to the effect of the electrically conductive MWNTs. The strong dielectric loss of the composites indicates that the addition of MWNTs increases the concentration of the mobile charges in the composites. A higher loss tangent reflects greater energy dissipation, primarily attributed to the establishment of a conductive network within the composite. Interestingly, the tan δ values show minimal dependence on frequency within the 0.5 kHz–10 kHz range, a characteristic advantageous for electrical and electronic device applications. Moreover, an increase in CNT

concentration leads to a consistent rise in tan δ across all frequencies studied.

For the PVDF - MWCNT composites, the change in AC conductivity with frequency provides information about the overall connectivity of the conducting network. Figures 4.3 (a–d) illustrates the variation of AC conductivity (σ_{ac}) with different temperatures (T) for (a) pure PVDF, (b) 0.5 wt%, (c) 1 wt%, and (d) 2 wt% MWCNT–PVDF composites at 0.5, 3, 5, 8, and 10 kHz, respectively. As observed, σ_{ac} increases steadily with temperature, confirming a negative coefficient of resistance behavior. The inset plots depict the variation of conductivity up to 120 °C, demonstrating that higher MWCNT loading enhances the overall conductivity of the composite. This behavior can be attributed to the increased segmental mobility of polymer chains at elevated temperatures, which facilitates charge transport through the composite network. Conductivity values increases as the frequency increases. Inset curves of Figures 4.3 (a-d) shows that the AC conductivity is almost constant from 30°C to 45°C for all measured frequencies. With the increase in temperature a constant value of AC conductivity is obtain for Pure, 0.5wt% and 1wt% MWCNT – PVDF samples, which starts increasing after 45°C up to 135°C. In 2 wt% MWCNT – PVDF composite sample AC conductivity starts increasing from initial temperature.

Conclusion:

In this work, PVDF/MWCNT nanocomposites with varying nanotube loadings were successfully synthesized via solution casting and hot pressing to investigate their dielectric and electrical properties. The incorporation of MWCNTs was found to significantly influence the dielectric behavior of PVDF through interfacial polarization, enhanced β-phase formation, and micro-capacitor network effects. The dielectric constant increased steadily with both temperature and filler concentration, reaching approximately 60 at 3 kHz and 135 °C for the 2 wt% composite, indicating improved dipolar and interfacial polarization. The dielectric loss (tan δ) showed a frequency-dependent decrease, while the lowest loss was observed for the 0.5 wt% sample, signifying optimal dispersion and minimized conductive pathways. The AC conductivity exhibited thermally activated behavior, rising with both temperature and filler loading, suggesting the onset of near-percolative charge transport at higher CNT concentrations.

Overall, the synergistic interaction between PVDF and MWCNTs leads to a substantial enhancement of dielectric and electrical properties without severe deterioration in loss characteristics when the filler content is optimized. These results demonstrate that controlled incorporation of MWCNTs can effectively tailor the dielectric response of PVDF for use in flexible electronics, high-energy-density capacitors, and electroactive devices. Future work may focus on optimizing filler functionalization, multilayer architectures, and hybrid nanofiller systems to further improve breakdown strength and energy-storage efficiency.

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Declaration: All authors declare that they have no conflicts of interest.
My research did not involve human participants.

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