

Investigating the Effect of Crystallinity on PVDF-PMMA Polymer Blend and Analyzing its Dielectric and Piezoelectric Performance

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ABSTRACT

In this study, a binary polymer blend using Polyvinylidene fluoride (PVDF) and Polymethyl methacrylate (PMMA) were prepared by simple solution casting method. PVDF having good dielectric and piezoelectric properties are in huge demand for applications like energy storage, sensors, actuators. To examine the changes in PVDF's properties resulting from polymer blending, three different blends were prepared using 20%, 30% and 40% PMMA. Using XRD and FTIR the changes in α (α) and β (β) phases of PVDF for different PMMA percentages were analyzed. It was seen that, β -phase of the blend increases up to 30% PMMA and starts to decrease after that and for 30% PMMA the β -phase content was higher than other samples. From FTIR spectrum, the calculated highest and lowest β -phase content was 63.6% and 53.3% for 30% and 20% PMMA respectively. The piezoelectric coefficient (d_{33} value) was higher for high PVDF content and decreased as the PMMA content increased. With 20% PMMA, the d_{33} value was 14.8 pC/N. Maximum dielectric constant was observed for 30% PMMA sample and with the PMMA percentage increment, lower dielectric loss was observed.

Keywords: PVDF, PMMA, Blend, Crystallinity, Dielectric



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1. Introduction

In recent years, instead of developing entirely new polymers, researchers frequently enhance material properties by blending existing polymers or incorporating fillers into a host matrix. Nowadays polymer blending is considered one of the simplest and most cost-effective strategies for developing advanced materials with enhanced performance and lower cost [1]. By combining different polymers, it enables the creation of new properties through the synergistic interaction of the components. However, while certain blends achieve complete miscibility at the molecular scale, many remain immiscible because of the low entropy of mixing, often leading to phase-separated structures [2].

In our everyday lives, materials like Polyvinylidene fluoride (PVDF) are quietly working behind the scenes such as sensing motion, storing energy, or even capturing vibrations. PVDF's piezoelectric strength arises from its β -phase, a highly polarized arrangement where molecular chains adopt an all-trans (TTTT) conformation, resulting in uniformly aligned dipole moments under mechanical stress [3]. PVDF is found in four crystalline forms (α , β , γ , and δ), but its strong ferroelectricity arises mainly from the highly polar β -phase, where dipoles align in the same direction [1, 4]. Recent studies have demonstrated that advanced processing methods, such as folding-hot-pressing, can yield PVDF films with up to 97.5% β -phase content and improved piezoelectric constants while maintaining mechanical flexibility, making them well suited for wearable devices [5]. Similarly, incorporating functional fillers, including BaTiO₃ nanoparticles or metal phosphate-based particles, has been

shown to further enhance β -phase crystallinity, dielectric response, and energy-harvesting efficiency [6]. Collectively, these advancements highlight PVDF as an excellent candidate for flexible sensors, energy-harvesting systems, and next-generation smart devices [7]. However, pristine PVDF is predominantly obtained in the non-polar α -phase, as the β -phase is thermodynamically unstable, and its direct production remains costly [8]. Blends using PVDF-PEO resulted in maximum ionic conductivity of $9.334 \times 10^{-5} \text{ S cm}^{-1}$ [9]. Also electrochemical stability above 5.0V was observed [10]. PVDF-PVC/PET blends showed improvement in the β (β) phase content, increasing up to 80% [11]. Other blends using PS, HDPE, LDPE, PMMA, PP were also studied to understand the change in properties such as α and β phases, crystallinity, electrical behavior and the effect of stretching, heat treatment, layer formation etc. [12-15].

The challenge is not simply to make PVDF based devices, but to stabilize and amplify the β -phase in formats that are thin, flexible, cost effective and manufacturable. To overcome these limitations and develop a cost-effective material that retains PVDF's electroactive properties, blending with oxygen-containing polymers such as PMMA has been explored [16]. Blending PVDF with poly(methyl methacrylate) (PMMA) is a versatile strategy that not only balances mechanical toughness and optical clarity but also influences phase formation [17]. A classic study by Gregorio and Souza showed that small additions of PMMA around 10–15 wt.%, facilitate the nucleation of the β -phase during solution crystallization, thus boosting PVDF's electroactive

behavior and more recently, Hussain *et al.* showed that PVDF blends with tailored PMMA copolymers promote β -phase crystallization and significantly enhance ferroelectric performance [18-21]. Recent progress shows that formulation and processing controlled from solution casting and stretching to additive engineering can push β -phase fractions toward the practical limits, while preserving mechanical compliance required for high energy storage functions [22]. Experimental investigation shows that a solvent-cast PVDF/PMMA (85:15 w/w) blend achieves superior sensing performance, generating ~ 31.4 mV under a bending strain of 0.0075 which is over four times higher than pure PVDF (~ 7.6 mV) due to its enhanced dielectric constant [23]. On the dielectric front, blending tactics such as in situ polymerization of PMMA within a PVDF matrix have achieved remarkable improvements in energy storage performance demonstrated by energy densities around 8 J/cm³ and efficiency around 80% between 30 and 90°C [24]. While PVDF remains one of the most promising electroactive polymers, the strategic blending with PMMA has been consistently shown to enhance β -phase nucleation, improve dielectric constant, and stabilize piezoelectric performance and reduce cost. Recent studies confirm that such blends deliver not only higher crystalline alignment but also improved breakdown strength, energy density, and voltage output compared to neat PVDF [25]. Against this backdrop, the present work seeks to systematically investigate how PVDF/PMMA blends can maximize β -phase content while optimizing dielectric and piezoelectric performance aiming to highlight PVDF/PMMA blends as scalable and application-ready materials for flexible sensors, energy harvesters, and next-generation soft electronics.

2. Methodology

This research work involved simple solution casting method to synthesis PVDF-PMMA blend. In this work beta phase PVDF pellets, PMMA and N, N-Dimethylformamide (DMF) as solvent was used. Firstly, the polymers were dried for 24h at 70°C. After drying, the polymers ratio of PVDF:PMMA was taken as 80:20, 70:30, 60:40. For each sample 5 gm of polymer and 50 ml of DMF were taken. Then the polymers were poured into DMF and stirred vigorously for 8h at 50°C. After stirring when the solution became clear and no visible polymer particles were seen then the solution was then transferred to a petri dish for drying. Complete miscibility of the two polymers was noticed. The blend was dried for 48h at 50°C in the oven. Here the synthesis process involves very low temperature as higher temperature reduces β phase of PVDF. After drying, the final solid polymer blend using 20%, 30% and 40% PMMA were formed shown in Table 1.

Table 1 Composition of three polymer blends

Samples	S1	S2	S3
Composition	PVDF-PMMA (80:20)	PVDF-PMMA (70:30)	PVDF-PMMA (60:40)

3. Characterization Techniques

For characterization and phase analysis X-ray Diffraction (XRD) and Fourier-transform infrared spectroscopy (FTIR) was used. XRD was done using Shimadzu LabX XRD-6100 diffractometer with continuous

scanning at a rate of 2°/min where 2θ values ranging from 10° to 80°. The FTIR spectrum was acquired with a resolution of 2 cm⁻¹, using 30 co-added scans at wavenumbers ranging from 500 cm⁻¹ to 4000 cm⁻¹ using Shimadzu IRTracer-100. The piezoelectric coefficient (d_{33}) was measured using YE2730A meter. Dielectric measurements were taken for calculating dielectric constant and loss of the samples using Hioki IM3590 Chemical Impedance Analyzer.

3.1 X-Ray Diffraction (XRD) Analysis

XRD spectrum of the three polymer blends are shown in Fig.1. From various literatures, the XRD of neat PVDF sample displayed intense diffraction peaks at 2θ values of 17.2°, 18.4°, 20.0° associated with the (100), (020), and (110) planes, confirming the presence of the α -phase [26]. The PMMA curve exhibits no distinct peak, with a broad peak at $2\theta=13.6^\circ$ signifying its amorphous characteristics. Diffraction peaks at 2θ values of 17.66°, 18.22°, 19.90° and 26.28° associated with the (100), (020), (110), and (021) planes, represent the alpha phase of PVDF [27, 28]. Prominent peaks at $2\theta=20.01^\circ$, 20.3°, 20.6°, and 23.16°, corresponding to the (110), (020) and (021) planes, are ascribed to the beta phase [29, 30]. The peak at $2\theta=22.80^\circ$ associated with the (101) plane signifies the gamma phase of PVDF [31, 32].

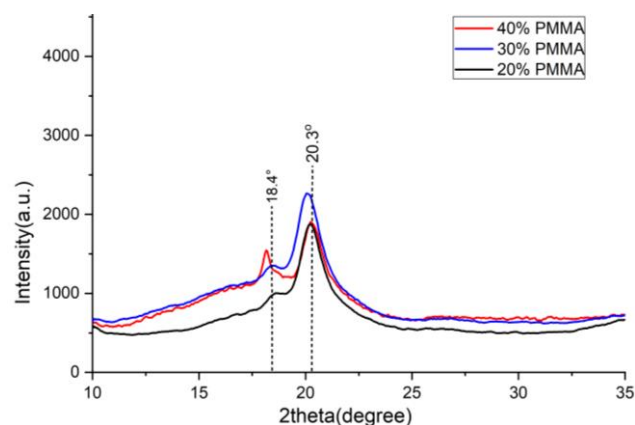


Fig.1 XRD spectra of 20%, 30%, 40% PMMA sample

In Fig.1 it is seen that, strongest peak at $2\theta=20.3^\circ$ is seen for all three samples corresponding to (110) planes. Here it represents the beta phase of PVDF. In addition, a weak peak around $2\theta=18.4^\circ$, assigned to the (020) plane, signifies the α -phase of PVDF. From the XRD it is seen that, the peak intensity representing beta phase is seen to be strongest for 30% PMMA sample which aligns well with the research work done by A. Yousefi [11]. Also, it is noticed that the peak intensity associated with the α -phase rises with increasing PMMA content. The peak achieves highest intensity for 40% PMMA sample. This can be due to the interaction between the polar carbonyl groups (C=O) of PMMA with PVDF's CH₂/CF₂ dipoles which favors the all-trans chain formation for lower PMMA percentage around 20-30%. As PMMA is amorphous in nature, with higher percentage it drops the crystallinity of PVDF and strong alpha phase peaks appear and a reduction in beta phase occurs.

3.2 Fourier Transform Infrared Spectroscopy (FTIR)

From Fig.2, it is seen that the peaks around 531, 763, 1495 cm⁻¹ indicates the alpha phase and the peaks around

840, 1070, 1170, 1275 cm^{-1} represents the presence of beta phase. A more prominent peaks around 2873 cm^{-1} and 3016 cm^{-1} is noticed as the PMMA percentage increases which is associated with the C-H stretching vibrations. The peaks at 2873 cm^{-1} and 3016 cm^{-1} describes symmetric and asymmetric vibration mode respectively. Which goes well with other studies where FTIR spectra of PVDF, PMMA from various research works showed that the α -phase of PVDF can be identified by absorption bands around 469, 531, 765, and 1495 cm^{-1} , whereas the β -phase is confirmed by the presence of peaks near 840, 1070, 1170 and 1275 cm^{-1} [33, 34]. The absorption band at 1233 cm^{-1} is attributed to CF_2 groups, while the peaks observed at 2873 cm^{-1} and 3016 cm^{-1} are associated with C-H stretching vibrations. Also, similarly the peak intensity increases with PMMA percentage at 1760 cm^{-1} indicating carbonyl (C=O) group. Similar results were found in a study where the peaks at 1458 cm^{-1} and 1495 cm^{-1} correspond to C-H bending vibrations, while the band at 1759 cm^{-1} arises from the presence of a carbonyl (C=O) group of PMMA [35].

From the analysis the peak intensity for beta phases have comparatively more intensity for 30% PMMA sample, which can be seen for 840, 1170, 1495 cm^{-1} . These findings are consistent with our XRD analysis and the results of other researchers [33-35].

3.2.1 Beta Phase Calculation

The beta content of the blend can be calculated from the FTIR spectrum. The peaks that are used here are 840 cm^{-1} for beta phase content and 763 cm^{-1} for alpha phase content. From the FTIR spectrum the values of A_{840} and A_{763} were taken. Then the beta content was calculated using the Furukawa and Gregorio equation given below as Eq. (1).

$$F(\beta) = \frac{A_{840}}{(K_{840}/K_{763})A_{763} + A_{840}} \times 100 \quad (1)$$

Where, A_{840} and A_{763} represents the absorbance intensities. The value of absorption coefficients at wavenumber 840 cm^{-1} and 763 cm^{-1} are: $K_{840} = 7.7 \times 10^4 \text{ cm}^2 \text{ mol}^{-1}$ and $K_{763} = 6.1 \times 10^4 \text{ cm}^2 \text{ mol}^{-1}$.

It is seen that, with the addition of PMMA there is noticeable changes in the beta content of PVDF. At 20% PMMA the beta content is lower than 30% PMMA sample. After increment in the beta phase content up to 30% PMMA addition, then again slight decrease is noticed in the beta content at 40%. So, at 30% PMMA the beta content reaches optimum value and then decreases. These results shown in Table 2 also align well with the XRD and FTIR spectrum analysis where comparatively stronger beta phase peaks was identified for 30% PMMA sample and also the findings are similar with the research conducted by A. Yousefi [11].

4. Piezoelectric Coefficient (d_{33} value)

Polyvinylidene fluoride (PVDF) is a semicrystalline high-performance thermoplastic polymer distinguished by its excellent piezoelectric characteristics. The average piezoelectric coefficient of neat PVDF is around 24 pC/N [36]. In this study, effect of PMMA blending with PVDF on its piezoelectric performance is analyzed. Here, piezoelectric coefficient was calculated using Eq. (2),

$$d_{33} \text{ value} = \frac{d_{33} \text{ display value } (Q)}{\text{Force}} \quad (2)$$

Here, Force = $250 \times 10^{-3} \text{ N}$

As PMMA is amorphous, non-polar and centrosymmetric, it results in no piezoelectricity. As a result, the crystallinity, dipole moment of the blends cannot reach to the level of PVDF. Also, the non-centrosymmetric β phase is reduced. Thus, with the increment in PMMA percentage the piezoelectric coefficient tends to decrease as shown in Table 3.

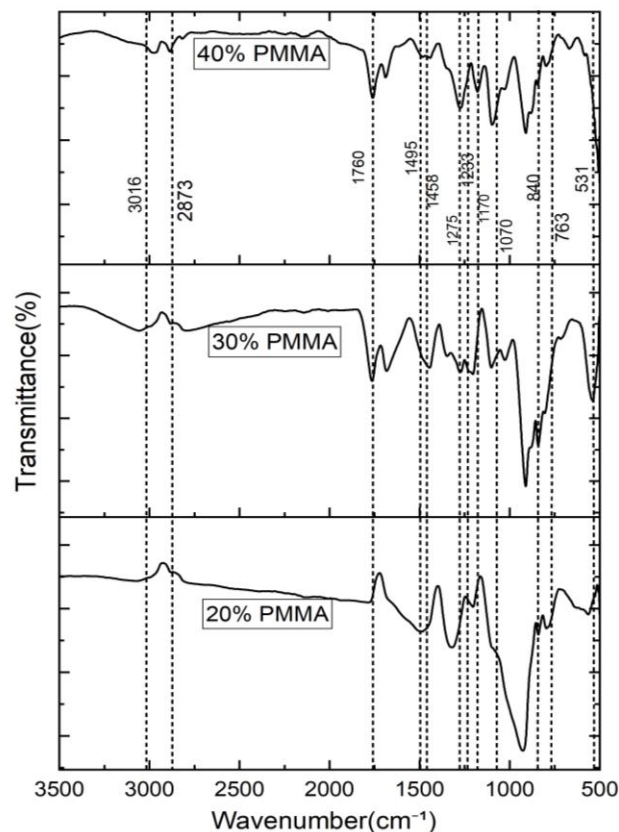


Fig. 2 FTIR spectra of 20%, 30%, 40% PMMA sample

Table 2 Beta phase content of the blends

Blends	F(β) (%)
PVDF-PMMA (80:20)	53.3
PVDF-PMMA (70:30)	63.6
PVDF-PMMA (60:40)	59.5

Table 3 d_{33} value of the blends

Sample	d_{33} value (pC/N)
PVDF-PMMA (80:20)	14.8
PVDF-PMMA (70:30)	14
PVDF-PMMA (60:40)	12.4

5. Dielectric Properties

Dielectric measurements of the three blend samples were taken in the frequency range of 1KHz to 200KHz. From the dielectric analysis, in Fig. 3 it is seen that the maximum dielectric constant at 1KHz was seen for 30% PMMA sample and lowest value was observed for 40% PMMA sample. As pure PVDF having dielectric constant values around 8-12,

there is a reduction in the value for blends due to PMMA which have lower dielectric constant. From the XRD and FTIR spectroscopy, 30% PMMA sample showed more beta phase content and so higher dielectric constant was seen for that sample as beta phase favors high dielectric constant. After 30% PMMA there is a reduction in the dielectric constant, which indicates 30% PMMA addition to be the optimal value for beta phase enhancement and high dielectric constant.

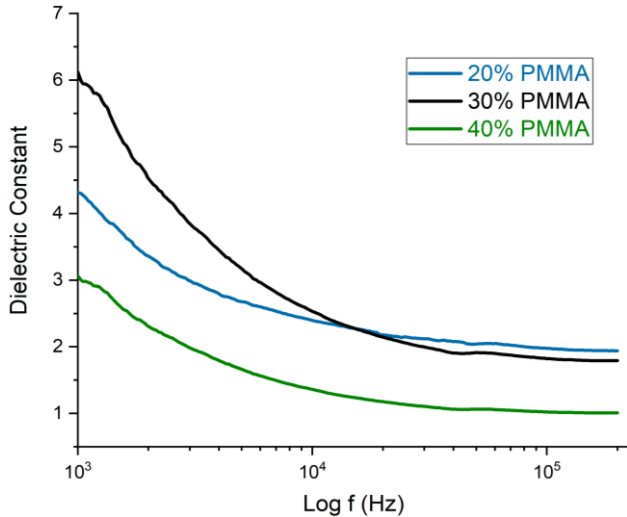


Fig. 3 Dielectric constant of 20%, 30%, 40% PMMA sample

From **Fig. 4**, it is seen that the dielectric loss of the three sample decreases with the increment of PMMA percentage. Lowest dielectric loss was seen for 40% PMMA sample which was 0.08 at 1KHz. Though 20% PMMA sample showed slightly lower loss than 30% PMMA, but with the increment in frequency, lower dielectric loss with high PMMA percentage was observed.

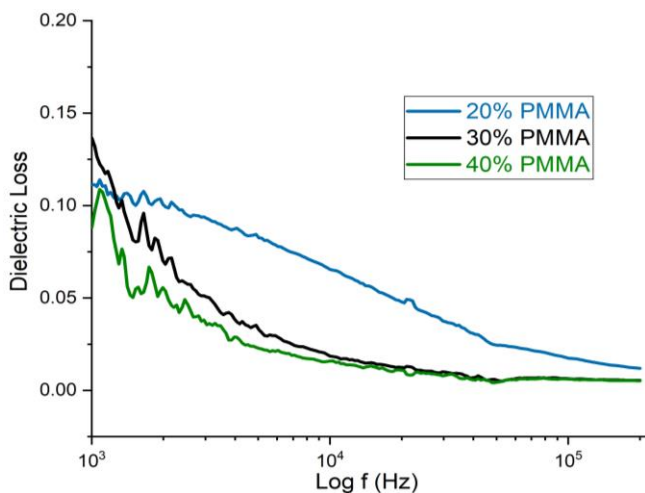


Fig. 4 Dielectric loss of 20%, 30%, 40% PMMA sample

As PVDF is polar in nature, it has strong dipolar motion and domain switching which results in dielectric loss. On the other hand, PMMA is non-ferroelectric, weakly polar and has high band gap, so loss due to polarization and energy dissipation is low. As a result, the dielectric loss in blends reduces. These results shown in **Table 4** go well with the results found by the experiments conducted by F. Deeba et al. [1].

Table 4 Dielectric constant and loss at 1KHz and 100KHz

Sample	Frequency (kHz)	Dielectric Constant	Dielectric Loss
20% PMMA	1	4.31	0.11
	100	1.9	0.01
30% PMMA	1	6.12	0.13
	100	1.82	0.0063
40% PMMA	1	3.06	0.08
	100	1.02	0.006

6. Conclusions

From this research work, the effect on the crystallinity, α and β phase of PVDF, piezoelectric and dielectric properties due to blending of PVDF and PMMA in different compositions is thoroughly studied. Three different compositions were prepared using 20%, 30% and 40% PMMA. The effect of PMMA percentage lower than this didn't show that much prominent effect in the blend properties such as mechanical, optical and electrical properties and with a percentage higher than 40% PMMA makes the composite hard and brittle which loses the required flexibility and is not suitable for practical applications. So, an optimal composition between 20% to 40% were searched in this study analyzing the property changes. The key findings from the experiment are listed below:

- XRD and FTIR ensured the coexistence of the alpha and beta phase of PVDF in the blends.
- Among 20%, 30%, 40% PMMA samples, 30% PMMA shows most prominent beta phase characteristics in both XRD and FTIR spectra.
- The highest beta phase content was 63.6% for 30% PMMA sample and starts to reduce after that.
- The d_{33} value decreases with the increment in PMMA content since PMMA is non-piezoelectric.
- The 20% PMMA sample shows highest d_{33} value of 14.8 pC/N, lower than neat PVDF but with improved mechanical, thermal, and optical properties of the blend, leading to a trade-off between mechanical and electrical properties.
- Maximum dielectric constant was observed for 30% PMMA due to high beta phase content.
- Dielectric loss reduces with the increment of PMMA percentage.

Further studies can be carried out to fabricate nanocomposite using this polymer blend and ceramic nanoparticles with good dielectric properties, which can be a good option for analyzing its effectiveness and sustainability for energy storage applications.

7. Acknowledgement

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Nomenclature

Symbol	Meaning	Unit
α	Alpha phase of PVDF	Dimensionless
β	Beta phase of PVDF	Dimensionless
γ	Gamma phase of PVDF	Dimensionless