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RESEARCH ARTICLE

High-Loading Waste-Derived Fillers in Poly (Vinylidene Fluoride) Composites: Comparative Effects of Alum Sludge and Wood Dust on Mechanical, Thermal and Dielectric Properties

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Abstract

Poly (vinylidene fluoride) (PVDF) composites with high loadings (50-65 wt%) of waste-derived fillers—alum sludge (AS), an inorganic water treatment residue, and wood dust (WD), a lignocellulosic timber by-product—were prepared via melt compounding and compression moulding without compatibilisers. Mechanical, thermal, and dielectric properties were systematically characterised. Increasing filler loading enhanced Young's modulus but reduced elongation and impact resistance due to restricted chain mobility and stress concentration. Dielectric behaviour differed markedly between the two systems: AS composites exhibited progressive increases in relative permittivity (up to ~70%) attributed to interfacial polarisation, whereas WD composites showed only modest changes due to the low dielectric contrast of lignocellulosic fillers. Differential scanning calorimetry revealed reduced PVDF crystallinity in composites, indicating hindered crystal formation at high filler concentrations. Overall, PVDF can accommodate substantial waste-derived filler fractions while maintaining processability and functional dielectric performance, offering a sustainable pathway for valorising industrial and biomass residues in dielectric materials.

Keywords: alum sludge, PVDF, wood dust, dielectric, composites, high filler loading.

1 Introduction

Poly(vinylidene fluoride) (PVDF) is a semi-crystalline fluoropolymer that has attracted considerable attention in electrical and electronic applications due to its unique combination of dielectric [1], piezoelectric [2], and ferroelectric properties [3]. The polymer exhibits relatively high dielectric permittivity compared to many conventional thermoplastics, together with excellent chemical resistance, thermal

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stability, and good processability [4]. Owing to these characteristics, PVDF has been widely investigated for applications in capacitors, sensors, actuators, energy harvesting devices, and flexible electronic components [5]. In particular, the ability of PVDF to exhibit different crystalline phases with distinct electroactive behaviour makes it an attractive platform material for advanced dielectric and functional composites [6].

In recent years, PVDF-based composites have been extensively developed to tailor dielectric and electromechanical properties for specific applications. By incorporating fillers with different electrical, mechanical, or structural characteristics, the functional performance of PVDF can be modified. For example, Niu et al. [7] incorporated BaTiO3 into PVDF to enhance permittivity for energy storage applications, while Zha et al. [8] demonstrated that the addition of carbon nanotubes as a conductive filler in PVDF-based polymer blends can introduce interfacial polarisation, thereby improving dielectric response at low filler concentrations. Furthermore, it has been reported that nanoparticle fillers can influence the crystallisation behaviour, mechanical stiffness, and thermal stability of the polymer matrix [9].

Alongside these developments, there has been increasing interest in incorporating sustainable or waste-derived materials as functional fillers in polymer composites. The utilisation of industrial by-products and waste streams offers opportunities to reduce material costs, minimise environmental impact, and create value-added applications for materials that would otherwise require disposal. In many cases, such waste materials contain inorganic minerals or lignocellulosic components that can serve as reinforcement phases or functional additives in polymer matrices. However, compared with conventional engineered fillers, the use of waste-derived materials in high-performance polymer composites remains relatively underexplored, particularly for applications involving dielectric or electronic materials.

One such waste material is alum sludge, which is generated as a by-product of drinking water treatment processes in which aluminium-based coagulants, such as aluminium sulphate, are used to remove suspended particles from raw water [10]. During the treatment process, coagulated flocs settle and form a residual sludge that is typically dewatered and disposed of via landfilling. According to data provided by Scottish Water, approximately 10,000 tonnes of alum sludge are generated annually from water treatment works in Scotland alone [11]. The management of this waste stream presents both economic and environmental challenges, motivating the development of more sustainable utilisation pathways [12]. While previous studies have explored applications for alum sludge in aluminium recovery, agriculture, and land reclamation [13], its direct incorporation as a filler in polymer composites remains largely underexplored. Specifically, there is a significant research gap regarding its potential for mechanical reinforcement and its influence on the functional electrical or electronic properties of the resulting materials.

Another abundant waste-derived material with potential for composite applications is wood dust. Wood processing, construction activities, and demolition of timber structures generate large quantities of fine wood residues that are often underutilised or treated as low-value waste [14]. Compared with inorganic fillers, wood dust materials offer distinct advantages, including low density, renewability, and cost-effectiveness when used in composite manufacturing. Their incorporation into polymer matrices can tailor the resulting material properties while contributing to the development of more sustainable material systems. Despite the extensive literature on natural fibre-reinforced polymers, the potential for directly utilising waste wood dust as a filler in functional polymer composites for electrical or dielectric applications has received comparatively limited attention.

An additional challenge associated with the incorporation of waste-derived fillers in polymer composites lies in achieving sufficiently high filler loading while maintaining acceptable processability and material integrity. In many reported systems, filler contents are limited to moderate levels to preserve polymer matrix continuity and ensure adequate dispersion. However, from both environmental and economic perspectives, the ability to incorporate large fractions of waste-derived fillers is highly desirable, as it maximises waste utilisation and reduces the amount of polymer required. Understanding how such high filler concentrations influence the structural and functional properties of polymer composites is therefore important for developing practical materials based on waste-derived reinforcements.

In this context, the present study investigates

PVDF-based composites containing two distinct types of waste-derived fillers: alum sludge (hereafter referred to as AS), representing an inorganic, mineral-rich material originating from water treatment processes, and wood dust (hereafter referred to as WD), representing an organic lignocellulosic by-product from timber processing. By comparing these two fillers within the same polymer matrix, the work aims to explore how differences in chemical composition, particle characteristics, and interfacial interactions influence the resulting composite behaviour. Additionally, a particular focus of this study is the use of unusually high filler loadings of 50, 60 and 65 wt%, which are substantially higher than those typically reported for PVDF composite systems. The composites were prepared through melt compounding followed by compression moulding without the use of compatibilisers, allowing the intrinsic interactions between PVDF and the waste-derived fillers to be evaluated.

The mechanical properties, thermal behaviour, crystallisation characteristics, and dielectric response of the resulting composites were systematically investigated. By comparing the behaviour of AS-filled and WD-filled systems, the study aims to provide insight into how inorganic and organic waste fillers influence the performance of PVDF-based composites at high loading levels. The findings contribute to a better understanding of the structure-property relationships governing highly filled polymer systems and highlight the potential for valorising underutilised waste materials as functional fillers in dielectric composite materials.

2 Experimental Details

2.1 Materials

Three major components were used in this work. PVDF (Kynar Grade 731 from Arkema) was used as the matrix for all composites. WD was used as the organic filler and was derived from a larch wood mixture sourced in Scotland. The wood material was processed through defibration, in which wood chips were mechanically refined into fine fibres using a disc refiner. The resulting WD material was oven-dried at 105 °C for 6h prior to composite fabrication in order to remove absorbed moisture and minimise processing defects associated with water evaporation during melt processing [15]. The second filler investigated in this work was AS, obtained from Scottish Water at the Rosebery Water Treatment Works (Midlothian, Scotland) as a by-product of the municipal water treatment process. The received sludge was dried and subsequently ground and sieved to obtain particles with sizes smaller than 63 μm. This pretreatment procedure followed the protocol reported in previous work [16].

2.2 Composite Fabrication

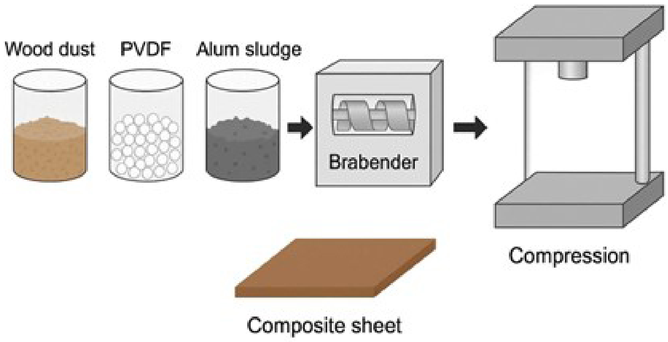

Figure 1. Schematic illustration of the fabrication process for PVDF-based composites containing WD and AS.

The composites were prepared by melt compounding followed by compression moulding, as illustrated in Figure 1. Each formulation was processed in 30 g batches to produce composite sheets with controlled filler-to-matrix ratios. The required components were pre-weighed according to the target formulations (see Table S1 in the Supplementary Information) prior to melt compounding using a laboratory-scale internal mixer (Brabender Plastograph). The mixer was preheated to 185 °C, after which PVDF powder was first introduced into the mixing chamber ($V=50$ cm^3) at a rotor speed of 10 rpm and allowed to melt for 4 min to form a homogeneous molten phase. Subsequently, the WD and AS fillers were gradually added into the melt at the same rotor speed. After complete addition of the fillers, the rotor speed was increased to 60 rpm and compounding was continued at 185 °C for 5min to promote particle dispersion. No compatibilisers or coupling agents were used in order to evaluate the intrinsic interactions between the polymer matrix and the waste-derived fillers. A full list of sample compositions is provided in Table 1.

Following melt compounding, the molten compound was removed from the mixer and immediately transferred into preheated steel moulds for compression moulding. The moulding process was conducted at 190 °C under a pressure of 20 MPa for 5 min using a hydraulic hot press. After the heating stage, the mould was water-cooled while maintaining pressure to solidify the material, producing composite sheets with a thickness of approximately 1 mm.

Table 1. Composition and designation of PVDF composites containing AS and WD.

Sample designation	Filler type	Filler content wt.%
PVDF	n/a	n/a
AS50PVDF50	Alum Sludge	50
AS60PVDF40	Alum Sludge	60
AS65PVDF35	Alum Sludge	65
WD50PVDF50	Wood Dust	50
WD60PVDF40	Wood Dust	60
WD65PVDF35	Wood Dust	65

2.3 Dielectric Characterisation

The dielectric properties of the composites were evaluated using an LCR meter (Keysight E4980A). Measurements were performed over a frequency range of 250 Hz to 1 MHz. Square specimens were cut from the compression-moulded composite sheets and coated with conductive silver paint to form parallel electrodes. With a parallel-plate capacitor configuration, the relative permittivity (εr) and dielectric loss (tanδ) of the materials were measured as a function of frequency. All measurements were conducted at ambient temperature, and the reported dielectric spectra represent the average of multiple measurements for each composite formulation.

2.4 Mechanical Testing

The mechanical performances of the composites were evaluated by tensile and impact testing. Tensile properties were measured using a universal testing machine (Zwick Z030, Germany). Dog-bones specimens were cut from the compression-moulded sheets and tested at room temperature. The tensile tests were performed at a constant crosshead speed of 5 mm min^{-1} in order to determine the tensile strength, Young's modulus, and elongation at maximum force of the materials. Impact resistance was evaluated using a Charpy impact testing machine (Zwick/Roell Pendulum Impact Tester 5113.300). The specimens had dimensions of 80 mm × 10 mm × 1 mm and contained a central notch with a depth of 2 mm. Tests were conducted using a 5 J pendulum, and the absorbed impact energy was recorded. For each formulation, multiple specimens were tested and the average values were reported with their standard deviations to reflect the measurement variability.

2.5 Thermal Analysis

Thermal properties of the samples were analysed using differential scanning calorimetry (DSC) (Q2000, TA Instruments, USA) under a nitrogen atmosphere with a flow rate of $50\,\mathrm{mLmin^{-1}}$. Approximately $10\,\mathrm{mg}$ of sample was sealed in an aluminium pan and scanned over a temperature range from $-50\,°\mathrm{C}$ to $200\,°\mathrm{C}$ at a heating rate of $10\,°\mathrm{Cmin^{-1}}$.

Two heating cycles were performed during the analysis. In the first cycle, the samples were heated to 200 °C to remove any previous thermal history and subsequently cooled to $-50\,°\mathrm{C}$. The second heating scan was then conducted from $-50\,°\mathrm{C}$ to $200\,°\mathrm{C}$, and the resulting thermograms were used for analysis.

The degree of crystallinity of PVDF (X_c) in the composites was calculated using equation (1):

$$X_c(\%) = \frac{\Delta H_m}{\alpha \Delta H_{m0}} \times 100 \tag{1}$$

where ΔH_m is the melting enthalpy of the composite obtained from the second heating cycle ($\mathrm{Jg^{-1}}$), ΔH_{m0} is the melting enthalpy of 100% crystalline PVDF (taken as $104.6\,\mathrm{Jg^{-1}}$) [17], and α is the weight fraction of PVDF in the composite.

3 Results and Discussion

3.1 Composite Morphology and Processability

Photographs of the compression-moulded PVDF composite sheets containing AS and WD fillers are shown in Figure 2. The colour difference between the two composite systems reflects the intrinsic colour of the fillers: AS produces dark brown to nearly black composites, while WD results in lighter brown sheets due to wood's natural colour.

All formulations containing up to 65 wt% filler could be successfully processed into continuous sheets using melt compounding followed by compression moulding. A uniform macroscopic distribution of filler within the polymer matrix was observed, with no obvious large-scale agglomeration or visible defects in the fabricated sheets. Representative SEM micrographs and corresponding EDX spectra are provided in the Supplementary Information document to support these observations (Figures S1, S2). It is evident that PVDF can accommodate relatively high fractions of waste-derived fillers while maintaining sufficient melt processability.

At lower filler loading, the composite sheets exhibit smooth and continuous surfaces with relatively light colour, suggesting that the PVDF matrix remains the dominant continuous phase capable of effectively binding the filler particles. As the

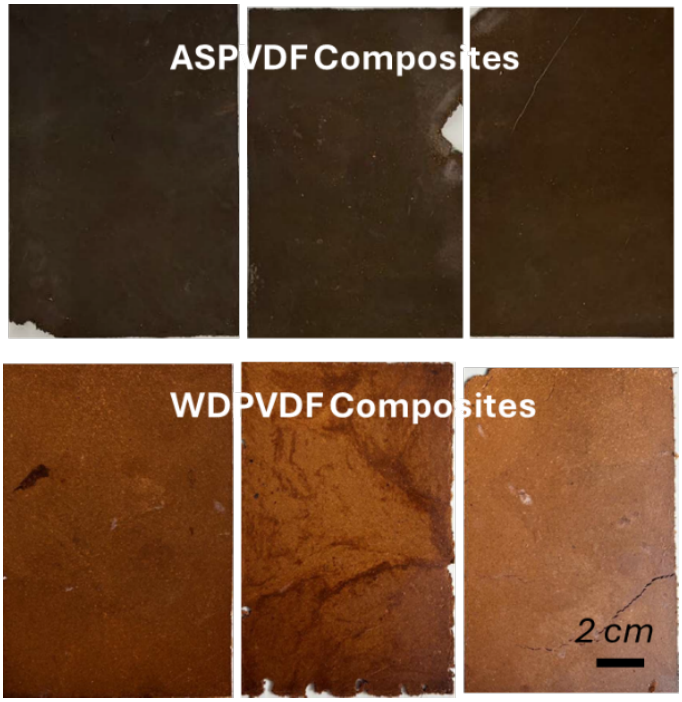

Figure 2. Photographs of compression-moulded PVDF composite sheets containing high loadings of waste-derived fillers: a) AS65PVDF35, b) AS60PVDF40, c) AS50PVDF50, d) WD65PVDF35, e) WD60PVDF40, and f) WD50PVDF50.

filler loading increases, rougher edges and minor surface irregularities become pronounced, and the composite sheets become darker. These features indicate increasing difficulty in maintaining matrix continuity as the available amount of PVDF decreases. It is worth noting that this study intentionally explored very high filler loading in order to identify the practical limits of incorporating waste-derived fillers into PVDF-based composites. During preliminary processing trials, the total filler loading was further increased to approximately 70 wt%. However, under these conditions the compounding process became unstable, with the mixture exhibiting extremely high viscosity, poor flow behaviour, and a dry or crumbly consistency within the internal mixer. As a result, the material could not be processed into continuous sheets. These results define a practical upper processing limit, beyond which the PVDF matrix can no longer maintain a continuous phase capable of supporting the filler network and preserving structural integrity.

3.2 Dielectric Properties

The relative permittivity (εr) and dielectric loss (tanδ) of the two composite systems are presented in Figures 3 and 4 for the AS/PVDF and WD/PVDF composites respectively, together with the neat PVDF for comparison.

At low frequencies, neat PVDF exhibits a relative permittivity of approximately 9, which is consistent with reported literature values [18]. In comparison with the composite systems, distinct differences are observed between the AS-filled and WD-filled materials. For the AS/PVDF composites, the relative permittivity at lower frequencies for AS50PVDF50, AS60PVDF40 and AS65PVDF35 is 12, 15 and 18, respectively, corresponding to an enhancement of roughly 40% to 70% relative to the neat polymer. This behaviour suggests that the incorporation of AS introduces additional polarisation mechanisms within the composite system. AS consists mainly of inorganic constituents such as aluminium hydroxides and metal oxides, which generally possess higher intrinsic permittivity than the PVDF matrix. In addition, the presence of these particles creates numerous heterogeneous filler-matrix interfaces. Under an applied electric field, charge carriers can accumulate at these interfaces, producing interfacial polarisation (also referred to as Maxwell-Wagner-Sillars polarisation) [19]. As the filler loading increases, the interfacial area correspondingly increases, leading to stronger interfacial polarisation and therefore higher effective permittivity.

In contrast, the WD/PVDF composites exhibit a relatively moderate increase in permittivity, remaining within the range of approximately 9–11 even at the highest filler loading, together with a much weaker dependence on filler concentration. This can be explained by the intrinsic characteristics of these fillers. WD consists mainly of cellulose, hemicellulose, and lignin, which possess relatively low dielectric constants compared with inorganic oxide-based fillers; therefore, the direct contribution of the filler to the overall permittivity of the composite is limited. For example, Elloumi et al. [20] investigated different types of wood-filled composites and reported dielectric constants in the range of 1.5-2.5 for systems containing aspen wood, aspen bark, and jack pine wood. Additionally, the interfacial polarisation generated in WD-filled composites is expected to be weaker than that observed in AS-filled systems. The organic nature of WD results in smaller contrasts in electrical conductivity and permittivity between the filler and the PVDF matrix, reducing the driving force for charge accumulation at the interfaces [21].

For all samples, the relative permittivity exhibits a clear dependence on the applied frequency, with higher values observed at low frequencies followed

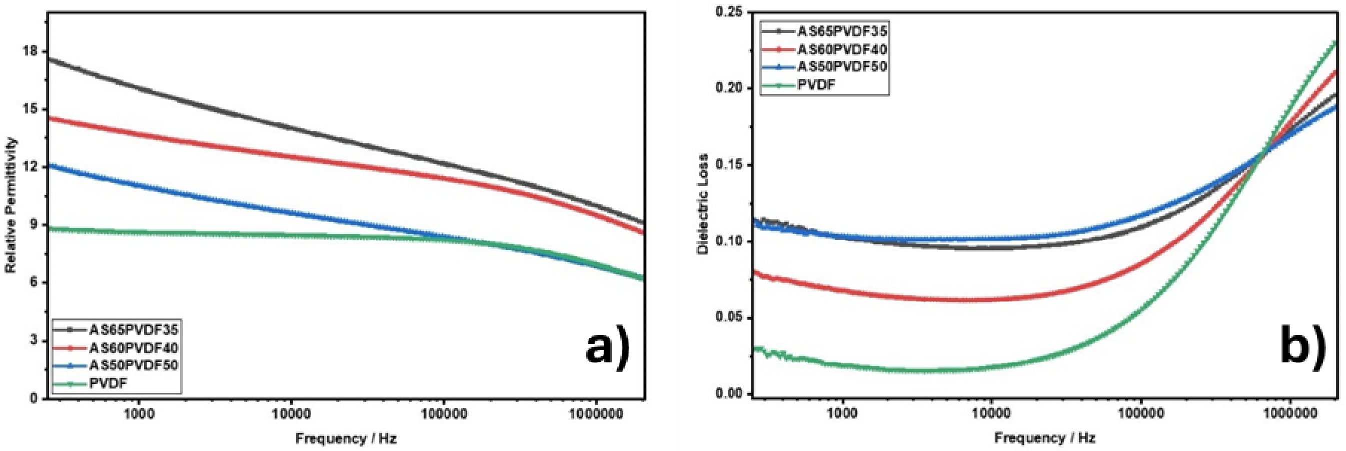

Figure 3. Frequency dependence of a) the relative permittivity and b) dielectric loss for AS/PVDF composites with different filler loadings, compared with neat PVDF.

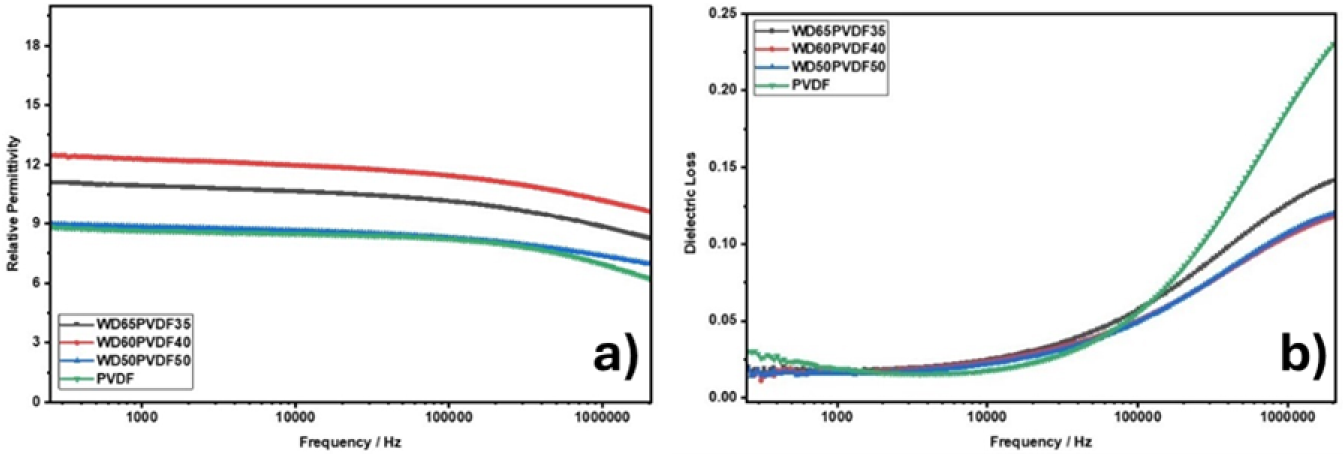

Figure 4. Frequency dependence of a) the relative permittivity and b) dielectric loss for WD/PVDF composites with different filler loadings, compared with neat PVDF.

by a gradual decrease as frequency increases. This frequency-dependent behaviour has been widely reported in dielectric materials [22] and is particularly characteristic of polymer-based dielectric systems, where it arises from multiple polarisation mechanisms operating over different frequency ranges. At low frequencies, dipolar polarisation of the PVDF molecular chains and interfacial polarisation at filler-matrix interfaces can respond effectively to the applied electric field [23]. As the frequency increases, these polarisation processes become progressively unable to follow the rapidly oscillating electric field, leading to a reduction in the measured dielectric permittivity [24]. This frequency-dependent dielectric dispersion is widely observed in heterogeneous dielectric systems and is commonly attributed to space charge polarisation and relaxation processes associated with electrically heterogeneous regions [25].

In addition to the relative permittivity, the dielectric loss behaviour of the composites provides further insight into the polarisation and relaxation processes occurring within the material. Neat PVDF exhibits relatively stable loss values in the low-frequency region, followed by a gradual increase towards higher frequencies. This behaviour is commonly reported for PVDF and is associated with dipolar relaxation of the polymer chains under alternating electric fields [26]. With the incorporation of AS, the dielectric loss values of the composites are higher than those of neat PVDF at low and intermediate frequencies. This increase can be attributed to additional interfacial polarisation arising from the heterogeneous interfaces between the PVDF matrix and the inorganic filler particles. However, as the frequency approaches the higher end of the measured range (around 1 MHz), the dielectric loss of the AS composites becomes slightly lower than that of neat PVDF. This behaviour suggests that the presence of AS particles modifies the relaxation dynamics of the system, possibly due to constrained dipolar motion of PVDF chains in the interfacial regions surrounding the filler particles.

In contrast, the WD composites exhibit dielectric loss values that are broadly comparable to those of neat PVDF in the low-frequency region. At higher frequencies, however, the dielectric loss of the WD composites becomes noticeably lower than that of neat PVDF. This behaviour may be associated with the partial restriction of PVDF chain mobility in the vicinity of the WD particles, which limits the ability of dipoles to respond to rapidly oscillating electric fields [27]. In addition, the relatively lower polarity of lignocellulosic fillers compared with inorganic oxide particles may result in weaker interfacial polarisation, contributing to the reduced dielectric loss observed at higher frequencies.

The combination of enhanced permittivity, (particularly for AS-filled systems), controlled dielectric loss, and high loading of low-cost waste-derived fillers provides advantages in terms of both functional performance and material sustainability. These characteristics make the materials promising candidates for applications such as embedded capacitors, flexible electronics, and insulating components.

3.3 Mechanical Properties

The mechanical performances of the PVDF composites containing AS and WD were evaluated through tensile and impact testing. The tensile properties, including tensile strength, elongation at maximum force, and Young's modulus, are presented in Figure 5 together with representative stress-strain curves.

A clear influence of filler incorporation on the tensile behaviour of PVDF can be observed. Compared with neat PVDF, the addition of both AS and WD results in a noticeable increase in the Young's modulus of the composites (Figure 5(a)). This increase in stiffness is attributed to the presence of rigid filler particles within the polymer matrix, which restrict the deformation of the PVDF chains and enhance the resistance to elastic deformation. As the filler loading increases from 50 wt% to 65 wt%, the modulus of both composite systems increases further, reflecting the progressively larger fraction of rigid phases within the material. Similar behaviour has been widely reported for particle-filled polymer composites, where increasing filler content leads to higher stiffness due to the inherently higher stiffness of the filler phase compared with the polymer matrix [28].

The tensile strength of the composites (Figure 5(b)) does not show significant improvement with increasing filler loading and may slightly decrease compared with neat PVDF. Although rigid fillers can potentially enhance the load-bearing capacity of polymer composites, effective reinforcement depends on efficient stress transfer across the filler–matrix interface, which is governed by the quality of interfacial adhesion between the filler (WD or AS) and the PVDF matrix. Furthermore, the very high filler fractions investigated in this study (50-65 wt.%) substantially reduce the continuity of the polymer matrix. As the matrix content decreases, the ability of PVDF to effectively distribute applied stress throughout the material becomes limited, while filler-filler interactions and interfacial stress concentration become more dominant, ultimately reducing tensile strength. A similar reduction in tensile strength with increasing filler loading was also observed in our previous study [29], where the decrease in strength was attributed to limited interfacial adhesion and increased stress concentration around the filler particles.

The elongation at maximum force (Figure 5(c)) shows a clear decreasing trend with increasing filler content for both composite systems. Neat PVDF exhibits the highest ductility, while the incorporation of either AS or WD significantly reduces the ability of the material to undergo plastic deformation. This reduction in elongation can primarily be attributed to restricted mobility of PVDF chains caused by the presence of filler particles. As the filler loading increases, the continuous polymer matrix becomes progressively interrupted by rigid filler phases, limiting the extent to which the polymer chains can rearrange and deform under tensile loading. In addition, the interfaces between the filler particles and the polymer matrix may act as stress concentration sites, promoting earlier crack initiation and reducing the strain to failure.

The representative stress-strain curves (Figure 5(d)) further illustrate the differences in deformation behaviour among neat PVDF, AS/PVDF composites, and WD/PVDF composites. Neat PVDF exhibits a highly ductile response, characterised by a pronounced deformation region prior to fracture, reflecting its high elongation and relatively low stiffness. In

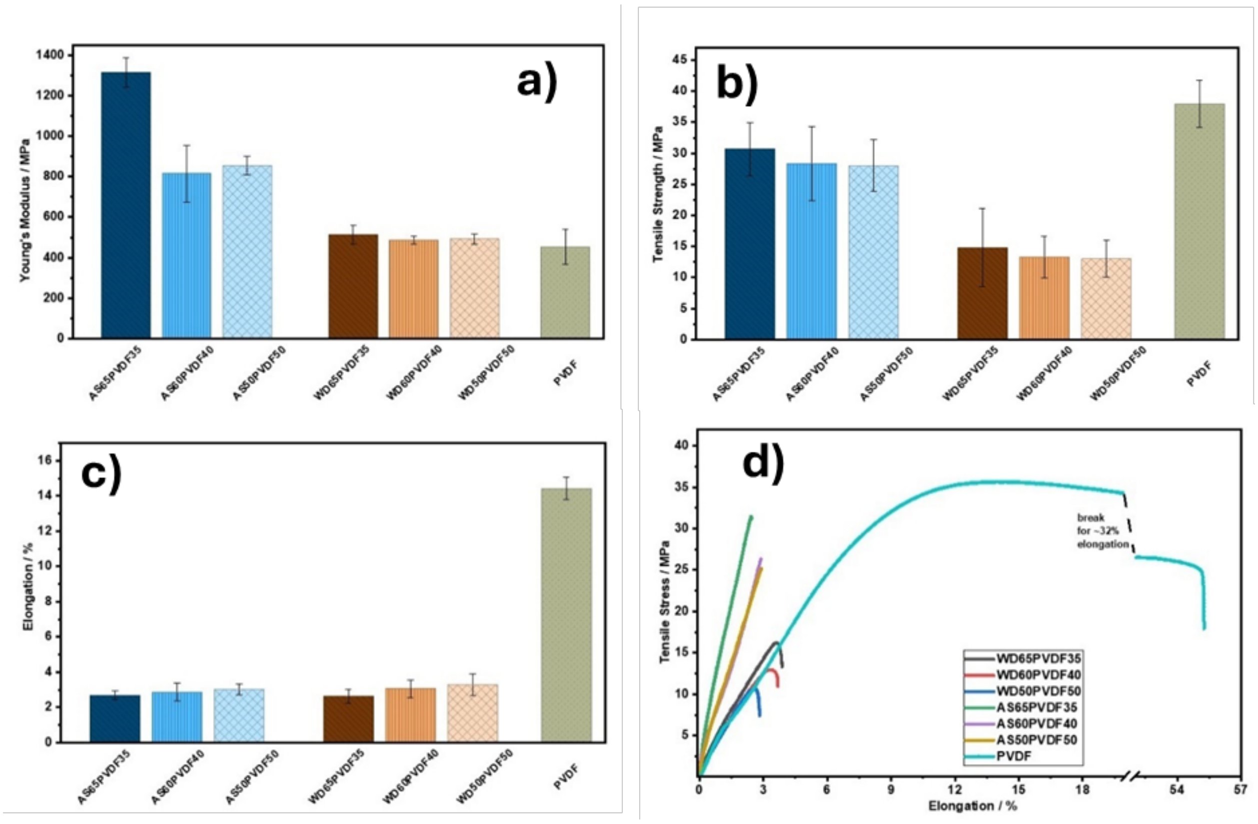

Figure 5. Tensile properties of PVDF composites containing AS and WD at different filler loadings, compared with neat PVDF: a) Young's modulus, b) tensile strength, c) elongation at maximum force and d) representative stress–strain curves.

contrast, both composite systems display steeper initial slopes and substantially reduced strain at failure. As the filler loading increases, the deformation region progressively decreases and fracture occurs at lower strains, indicating a transition from ductile polymer behaviour to a more brittle composite response.

Differences between the two filler types are also evident. Composites containing AS generally exhibit slightly higher stiffness compared with WD-filled composites at similar filler loadings. This behaviour can be attributed to the higher rigidity and mineral nature of AS particles relative to the lignocellulosic WD particles.

The impact resistance results are shown in Figure 6 and follow trends consistent with the tensile deformation behaviour observed in the stress-strain curves. Neat PVDF exhibits the highest absorbed impact energy among all the materials. This behaviour is associated with its high toughness, which corresponds to the total area under the stress-strain curve and represents the energy absorbed prior to fracture. Although PVDF has a relatively moderate modulus, its combination of relatively high tensile strength and large elongation results in substantial toughness and high energy absorption capability during impact.

In contrast, the incorporation of large fractions of rigid fillers significantly reduces the toughness of the composites. As discussed in the tensile results, increasing filler content restricts PVDF chain mobility and disrupts matrix continuity, leading to a pronounced reduction in elongation at failure and consequently a lower overall toughness (as reflected by the reduced area under the stress–strain curves). Additionally, filler particles introduce stress concentration sites that facilitate crack initiation and propagation under impact loading, further decreasing the absorbed impact energy.

3.4 Thermal Analysis

The melting behaviour and crystallinity of neat PVDF and its composites were evaluated and the melting temperature (T_m), melting enthalpy (ΔH_m), and calculated degree of crystallinity (X_c) obtained from the second heating cycle are summarised in Table 2.

Only a single melting peak is observed for all samples at approximately $165\,^{\circ}\mathrm{C}$ to $168\,^{\circ}\mathrm{C}$, corresponding to the melting of the crystalline phase of PVDF. This indicates that the melting behaviour remains

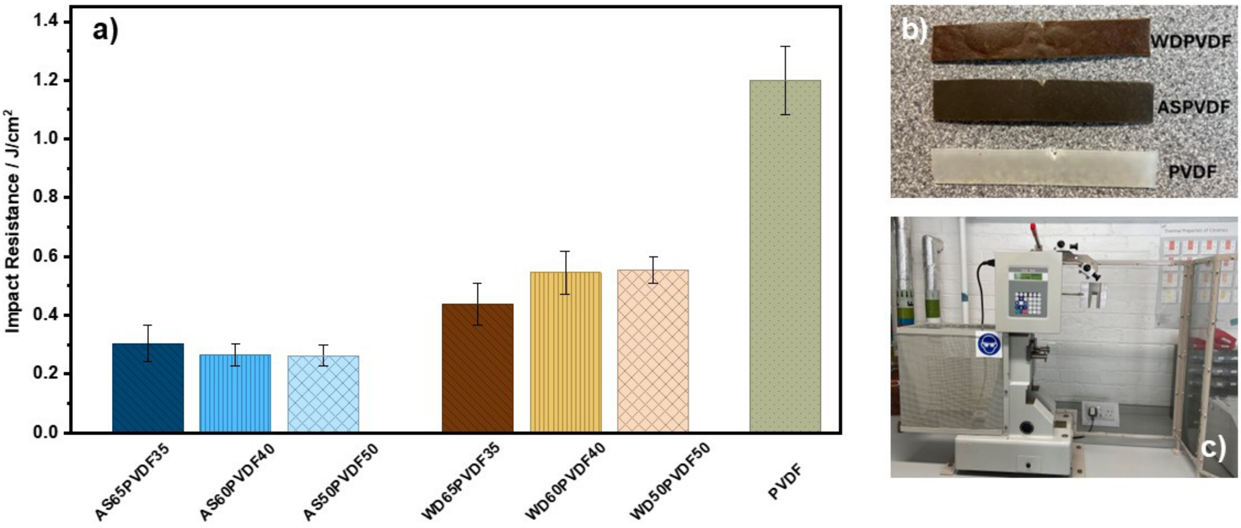

Figure 6. Impact resistance of PVDF composites containing AS and WD at different filler loadings compared with neat PVDF: a) measured impact resistance of the composites; b) representative notched specimens used for the test; c) impact testing instrument.

Table 2. Thermal properties of PVDF, AS/PVDF and WD/PVDF composites obtained from DSC analysis (error based on DSC software).

Sample	T_m (°C) [±1°C]	ΔH_m (J/g) [±1%]	X_c (%) [±1%]
AS65PVDF35	165.9	9.6	26.3
AS60PVDF40	165.4	20.3	32.4
AS50PVDF50	164.9	16.7	31.9
WD65PVDF35	165.7	9.7	26.6
WD60PVDF40	166.1	11.0	26.2
WD50PVDF50	166.8	13.2	28.0
PVDF	168.4	38.7	37.0

dominated by PVDF and that the incorporation of AS or WD does not introduce additional melting transitions within the investigated temperature range.

A slight decrease in the melting temperature is observed for the composite systems compared with neat PVDF. The melting peak of neat PVDF occurs at approximately 168.4 °C, whereas the composites exhibit melting temperatures in the range of 164.9 °C to 166.8 °C. This reduction suggests that the incorporation of high fraction of filler slightly disturbs the crystalline organisation of the PVDF matrix. The presence of particle fillers can interfere with the packing of polymer chains during crystallisation, leading to the formation of less perfect or smaller crystallites that melt at slightly lower temperatures [29].

A more pronounced effect is observed in the degree of crystallinity of PVDF within the composites. Neat PVDF exhibits a crystallinity of approximately 37%, while the filled systems show significantly reduced crystallinity values in the range of approximately 26% to 32%. This decrease in crystallinity is also reflected in the lower melting enthalpy (ΔH_m) values of the composites compared with neat PVDF. The reduction in crystallinity suggests that the presence of large amounts of filler restricts the crystallisation of PVDF during cooling.

Several mechanisms may contribute to this observation. At the very high filler loading investigated in this study, the interparticle spacing becomes progressively smaller, creating a crowded microstructure that constrains polymer chain mobility during crystallisation. Under such conditions, PVDF chains experience increasing difficulty in diffusing toward crystal nuclei and rearranging into well-ordered lamellar structures. The resulting limitation in segmental motion suppresses crystal growth and favours the formation of smaller or less perfect crystallites, which consequently exhibit slightly lower melting temperatures.

Similar behaviour has been widely reported in particle-filled polymer systems. For example, the incorporation of rice husk and wood flour into PA6 significantly reduced the melting temperature and decreased crystallinity from about 47 % in neat W-PA6G to approximately 14 % and 18 % in the corresponding composites, respectively [30]. Likewise, cenosphere-filled HDPE exhibited reduced crystallinity because the filler particles inhibited the close packing of HDPE chains [31]. In LDPE/HIPS composites containing toner waste powder, the crystallinity decreased from 25.4 % to 18.1 %, which was attributed to hindered chain transport, heterogeneous filler distribution, and disruption of nucleation and crystal growth when filler particles were dispersed within amorphous regions [32]. Ming et al. [33] further reported that excessive filler loading can strongly restrict molecular motion due to reduced interparticle spacing and enhanced interfacial constraints, thereby limiting chain-segment diffusion toward crystal nuclei and suppressing crystallisation. These findings indicate that when filler concentration becomes sufficiently high, the particles act primarily as physical barriers that hinder chain folding and crystal development rather than promoting nucleation. This effect becomes particularly pronounced in highly filled systems such as those investigated in the present study.

The reduction in crystallinity also has important implications for the mechanical and dielectric behaviour of the composites. From a mechanical perspective, lower crystallinity generally leads to reduced ductility and toughness, as the crystalline regions in PVDF contribute to load-bearing capacity and resistance to plastic deformation [34]. This is consistent with the observed decrease in elongation at maximum force and impact resistance in the highly filled composites. In principle, a reduction in crystallinity would also be expected to decrease the Young's modulus, as the material becomes more amorphous and thus less stiff [35]. However, in the present study, the opposite trend is observed, with the Young's modulus increasing as crystallinity decreases. This behaviour can be attributed to the dominant influence of the high filler content (50-65 wt%) on the mechanical response. At such high loadings, the stiffness of the composite is primarily governed by the rigid filler phase and the increasing filler-filler interactions, which outweigh the effect of reduced polymer crystallinity. The fillers restrict polymer chain mobility and act as load-bearing components within the composite, leading to an overall increase in stiffness despite the reduction in crystallinity.

From a dielectric standpoint, the amorphous regions of PVDF are more favourable for dipolar orientation under an electric field, whereas crystalline regions tend to restrict dipole mobility. Therefore, a reduction in crystallinity may facilitate dipolar polarisation to some extent; however, in the present system, this effect is likely secondary compared with the dominant influence of interfacial polarisation introduced by the fillers.

4 Conclusions

This study investigated PVDF composites containing high loadings (50–65 wt%) of two waste-derived fillers, AS and WD, prepared by melt compounding followed by compression moulding. The results demonstrate that PVDF can accommodate unusually high fractions of these fillers while maintaining processability and structural integrity. Increasing filler content led to a clear increase in stiffness from approximately $450\,\mathrm{MPa}$ to $1.3\,\mathrm{GPa}$ at 65 wt% AS loading, accompanied by reduced elongation (from $\sim$14% to $\sim$3%) and impact resistance (from $1.2\,\mathrm{J/cm^2}$ to $0.3\,\mathrm{J/cm^2}$) due to restricted polymer chain mobility and stress concentration within the highly filled structure. Distinct differences in dielectric behaviour were observed between the two filler systems: AS-filled composites exhibited increased relative permittivity up to $\sim$70% compared with neat PVDF, which can be attributed to interfacial polarisation associated with the inorganic particles, whereas WD-filled composites showed only minor changes in dielectric response. Thermal analysis further revealed a reduction in PVDF crystallinity from $\sim$37% in neat PVDF to $\sim$26–32% in the composites, indicating that high filler concentrations hinder crystal development within the polymer matrix. Overall, the findings provide insight into the structure–property relationships of highly filled PVDF composites and demonstrate a viable pathway for the utilisation of industrial and biomass waste in sustainable dielectric composite systems.

Data Availability Statement

Data will be made available on request.

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Conflicts of Interest

The authors declare no conflicts of interest.

AI Use Statement

The authors declare that no generative AI was used in the preparation of this manuscript.

Ethical Approval and Consent to Participate

Not applicable.

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Appendix

A Supplementary Information

Table S1. Weight [g] of PVDF and fillers used for compounding mixing.

Sample designation	PVDF [g]	Filler [g]
PVDF	30.00	0.00
AS50PVDF50	15.00	15.00
AS60PVDF40	12.00	18.00
AS65PVDF35	10.50	19.50
WD50PVDF50	15.00	15.00
WD60PVDF40	12.00	18.00
WD65PVDF35	10.50	19.50

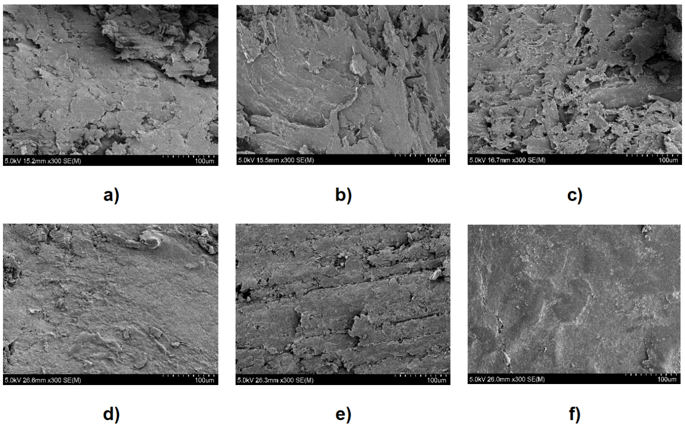
a) b) c)
d) e) f)

Figure S1. Representative SEM micrographs of PVDF-based composites: a) WD65PVDF35, b) WD60PVDF40, c) WD50PVDF50, d) AS65PVDF35, e) AS60PVDF40, f) AS50PVDF50. Samples were sputter-coated with Pd prior to analysis and imaged using 5kV voltage.

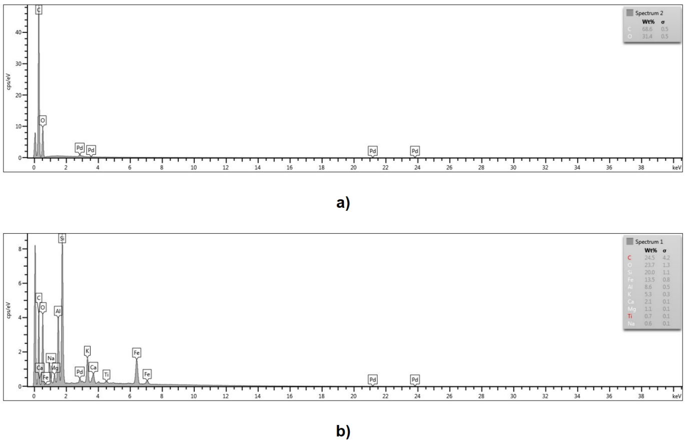
a)
b)

Figure S2. Representative EDX spectra of PVDF-based composites: a) WD50PVDF50 and b) AS50PVDF50. Samples were sputter-coated with Pd prior to analysis and examined using 20kV voltage. The spectrum of the AS-filled composite reveals the presence of O, Si, Fe, Al, K, Ca, Mg, and Na, consistent with the supplier's material specification.

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