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Electroactive Graphene–Polymer Nanocomposites for Self-Sustaining and Multifunctional Flexible Devices

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Abstract

Graphene–reinforced polymer–composites were developed and systematically investigated to explore their multifunctionality in flexible electronic applications. The study specifically aims to correlate graphene-induced structure–property–function relationships with electroactive performance and self-sustaining behaviour in flexible devices. The hybridization of electroactive polymers—polyaniline (PANI), poly(3,4-ethylenedioxythiophene): polystyrene sulphonate (PEDOT:PSS), and polyvinylidene fluoride (PVDF)—with graphene nanoplatelets enabled simultaneous enhancement of electroactivity, energy-harvesting, and sensing functionalities. Morphological studies confirmed uniform graphene dispersion at 5 wt.% loading, forming continuous filler–matrix percolation networks, while higher loadings caused agglomeration detrimental to composite homogeneity. XRD analysis revealed graphene-induced nucleation of the β-phase in PVDF–polymer–composites, directly enhancing electroactive crystallinity. Electrochemical testing showed conductivity of PEDOT:PSS–graphene composites increased from 1.2 × 10³ S/m to 4.6 × 10³ S/m, with capacitance rising from 180 to 315 F/g, highlighting the synergy between the polymer matrix and nanofiller. Charge–discharge cycling yielded an energy density of 32 Wh/kg with 92% retention after 5000 cycles. Self-sensing studies demonstrated that PVDF–graphene composites generated consistent outputs of ~2.8 V after 10,000 cycles, while PANI–graphene polymer–composites achieved a gauge factor of ~65.3 with linear piezoresistive response. Hybrid piezoelectric–triboelectric polymer–composite films harvested 50 μW/cm² under 5 Hz excitation, enabling a 100 μF capacitor to charge to 3.2 V in 150 s and power an LED. Mechanical testing showed tensile strength improved by ~65% (from 35 to 58 MPa) with preserved flexibility after 10,000 bending cycles. These integrated electroactive, sensing, and mechanical properties demonstrate the multifunctional nature of the developed composites, making them strong candidates for next-generation wearable, self-powered flexible devices.

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INTRODUCTION

Flexible and multifunctional devices are rapidly redefining the frontier of modern electronics,

spanning applications in wearable health monitoring systems, biomedical implants, soft robotics, and portable energy-harvesting systems [1-3]. These devices require not only mechanical flexibility and lightweight features but also self-sustaining power capability and intrinsic sensing functions. Conventional materials such as silicon and rigid ceramics fail to meet these criteria, as they lack flexibility and compatibility with dynamic deformation. As a result, polymer–composite materials have emerged as the most promising alternative, due to their inherent softness, easy processability, and tunable physical and chemical properties [4, 5].

Within the family of polymer–composite systems, electroactive polymer–composites represent a particularly attractive category, offering a combination of conductivity, electrochemical activity, and deformability [6]. Conductive polymers such as polyaniline (PANI), polypyrrole (PPy), and poly(3,4-ethylenedioxythiophene):polystyrene sulphonate (PEDOT:PSS) are widely studied for electrochemical capacitors, actuators, and sensors because of their redox activity and ease of doping [7, 8]. However, their performance in pristine form remains constrained by limited mechanical robustness, poor retention of conductivity under cyclic strain, and degradation of energy efficiency over prolonged usage [9]. These shortcomings necessitate reinforcement strategies that elevate their electrochemical and mechanical stability. Here, nanofiller-modified polymer–composites have become a focal research direction [10].

Graphene, a two-dimensional nanomaterial composed of sp^2-hybridized carbon atoms, has proven to be a transformative additive for polymer–composite systems. With its exceptionally high electrical conductivity (~10^6 S/m), superior thermal conductivity (~5000 W/mK), and mechanical modulus approaching 1 TPa, graphene acts as both a conductive reinforcement and a mechanical stabilizer [11, 12]. When integrated into a polymer–composite, graphene can establish continuous percolated networks that enable rapid charge transport, efficient load transfer, and multifunctional coupling effects [13]. For instance, graphene reinforcement in PVDF-based polymer–composites facilitates the nucleation of the β-phase, significantly enhancing the piezoelectric response, while graphene in PEDOT:PSS- or PANI-based polymer–composites augments conductivity and electrochemical capacitance [14-16].

Several studies confirm that graphene–polymer–composites exhibit remarkable strain sensitivity, making them highly effective as self-sensing materials for wearable electronics [17]. Furthermore, in energy-harvesting systems, graphene provides extended electron pathways that suppress charge recombination and increase power density, which is critical for devices such as tribo-electric nano-generators, piezoelectric harvesters, and flexible super-capacitors [18, 19]. Importantly, the synergy between graphene and electro-active polymers transforms a conventional polymer–composite into a multifunctional material platform capable of simultaneously delivering self-powering and self-sensing properties [20].

Despite these advances, the structure–property–function relationship in electroactive graphene–polymer–composites is still not fully understood. Several critical aspects remain unresolved. The determination of the optimal graphene loading threshold is essential for achieving a balance between conductivity, flexibility, and energy density. The interfacial interactions between graphene sheets and the polymer matrix play a decisive role in influencing electrochemical performance and long-term durability. Furthermore, the ability of a single polymer–composite system to combine both self-sensing and self-sustaining functions must be assessed in relation to potential trade-offs in mechanical reliability [21]. These considerations highlight the need for a deeper exploration of graphene-reinforced polymer–composites in multifunctional device architectures.

Therefore, the present study focuses on the fabrication and characterization of electroactive graphene–polymer–composites, aiming to establish a comprehensive link between graphene dispersion, energy-harvesting efficiency, self-sensing performance, and mechanical robustness. By

integrating solution blending, in situ polymerization, and structural tailoring techniques, this work seeks to demonstrate how graphene can be systematically harnessed within polymer–composite matrices to design next-generation flexible, self-sustaining, and multifunctional devices. The insights gained here are expected to contribute significantly to the advancement of polymer–composite science and its application in practical flexible electronics.

MATERIALS AND METHODS

Selection of Materials and Polymer–Composite Matrices

The fabrication of electroactive polymer–composites was initiated by carefully selecting both nanofillers and polymer matrices to ensure multifunctional performance. High-quality graphene nanoplatelets (GNPs), with an average lateral size of ~5 μm and thickness below 5 nm, were chosen as the primary conductive nanofiller due to their superior electrical, thermal, and mechanical properties. Electroactive polymers including polyaniline (PANI), poly(3,4-ethylenedioxythiophene):polystyrene sulfonate (PEDOT:PSS), and polyvinylidene fluoride (PVDF) were selected as host matrices. Each polymer offered unique advantages for polymer–composite design: PANI contributed high redox activity, PEDOT:PSS provided intrinsic conductivity and aqueous processability, and PVDF offered semicrystalline domains and piezoelectric behaviour. Together, these materials formed the basis for diverse polymer–composite systems with tunable properties.

Graphene Dispersion and Polymer–Composite Blending

Uniform graphene dispersion is vital for functional polymer–composite performance, as poor dispersion often results in agglomeration and deteriorated electrical pathways. To achieve homogeneity, graphene nanoplatelets were first ultrasonicated in N-methyl-2-pyrrolidone (NMP) for 60 minutes to exfoliate the sheets and minimize aggregation. Simultaneously, polymer solutions were prepared: PVDF was dissolved in dimethylformamide (DMF), PEDOT:PSS was stabilized with camphorsulfonic acid (CSA) dopant, and PANI was synthesized via in situ oxidative polymerization of aniline monomer using ammonium persulfate (APS). Once ready, the polymer solutions were blended with the graphene suspension under magnetic stirring. This ensured intimate interfacial contact between graphene and polymer chains, which is essential for stress transfer and charge percolation in polymer–composites.

Fabrication of Polymer–Composite Films

After dispersion and blending, polymer–composite films were fabricated (Figure 1) using controlled casting techniques. For PVDF–graphene and PEDOT:PSS–graphene composites, the homogeneous mixtures were cast onto flexible polyethylene terephthalate (PET) substrates and dried under vacuum at 60 °C for 12 hours to remove residual solvents.

The resulting polymer–composite films exhibited uniform thicknesses of 100–200 μm with smooth surfaces, suitable for subsequent testing. In the case of PANI–graphene composites, in situ polymerization allowed PANI chains to grow directly on graphene surfaces, leading to robust interfacial bonding. This approach improved both conductivity and mechanical stability within the polymer–composite structure.

Structural and Morphological Characterization of Polymer–Composites

Comprehensive characterization was carried out to evaluate the structural and morphological features of the fabricated polymer–composites. Scanning electron microscopy (SEM) was employed to examine graphene dispersion within the polymer matrices, highlighting filler–matrix interfacial interactions. X-ray diffraction (XRD) was used to analyze crystallinity, with specific focus on the β-phase in PVDF-based composites, which correlates with piezoelectric response. Collectively, these analyses established a clear understanding of the structure–property relationships in the polymer–composites.

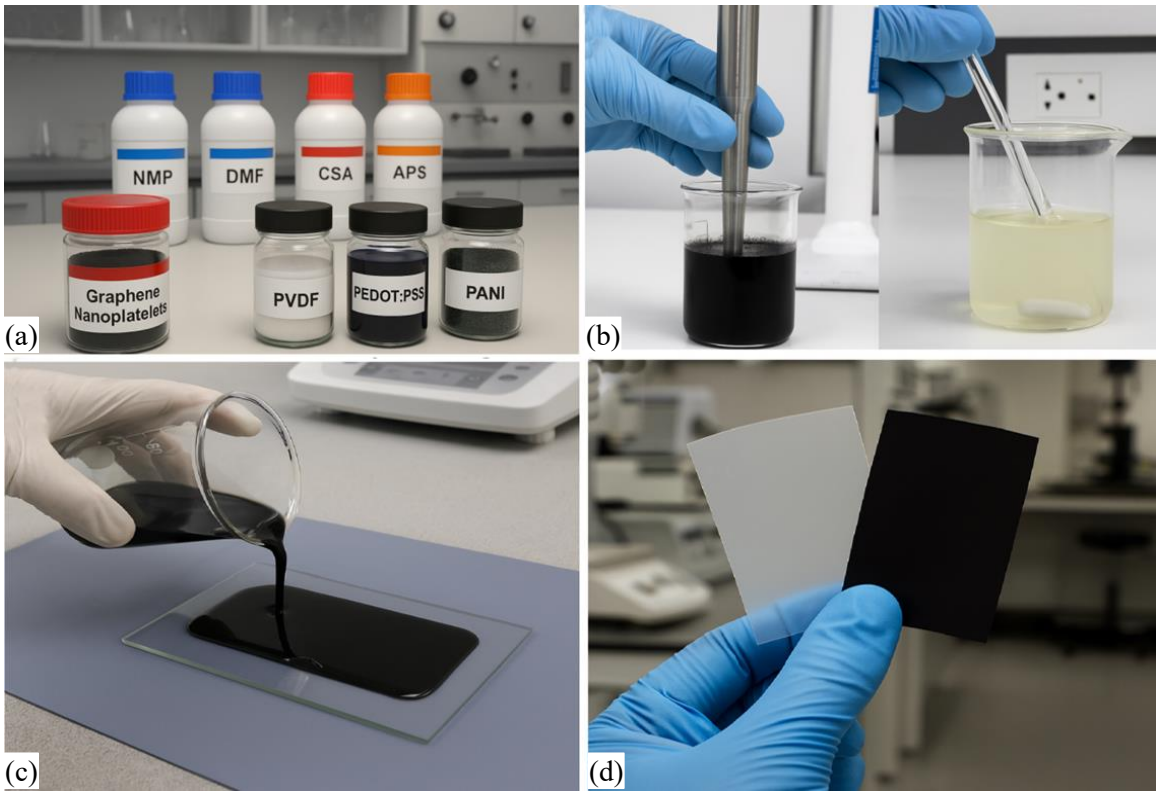

Figure 1. Fabrication sequence of graphene–polymer composites: (a) materials selection, (b) graphene dispersion and polymer blending, (c) film casting and drying, and (d) free-standing composite films (~100–200 μm).

Electrochemical, Sensing, and Mechanical Evaluation

The multifunctional properties of the graphene–polymer–composites were assessed through electrochemical, sensing, and mechanical testing. Electrical conductivity and electrochemical performance were analyzed using cyclic voltammetry (CV), galvanostatic charge–discharge, and four-point probe conductivity measurements. These tests quantified capacitance, energy density, and charge transport efficiency. For self-sensing studies, the polymer–composite films were subjected to cyclic strain using a dynamic mechanical tester coupled with an oscilloscope, allowing real-time monitoring of voltage and resistance changes under deformation. The self-powering capability was evaluated by subjecting the films to tapping and bending in hybrid piezoelectric–triboelectric mode; output voltages and currents were recorded, and a 100 μF capacitor was charged to demonstrate practical energy storage. Finally, tensile tests were carried out following ASTM D882 standards to measure tensile strength, elongation at break, and modulus. Additional flexibility tests involved repeated folding and bending of the polymer–composites to verify their robustness under mechanical cycling.

RESULTS AND DISCUSSION

Structural and Morphological Characterization

The incorporation of graphene into electroactive polymer–composites significantly altered their structural and morphological features. SEM images (Figure 2) of the composites at an optimized 5 wt.% graphene loading revealed a homogeneous dispersion of graphene sheets within the polymer matrices.

The graphene formed interconnected conductive pathways, which are critical for efficient charge transport and stress transfer across the polymer–composite network. In contrast, at higher filler concentrations (≥10 wt.%), pronounced agglomeration was observed. These agglomerates disrupted

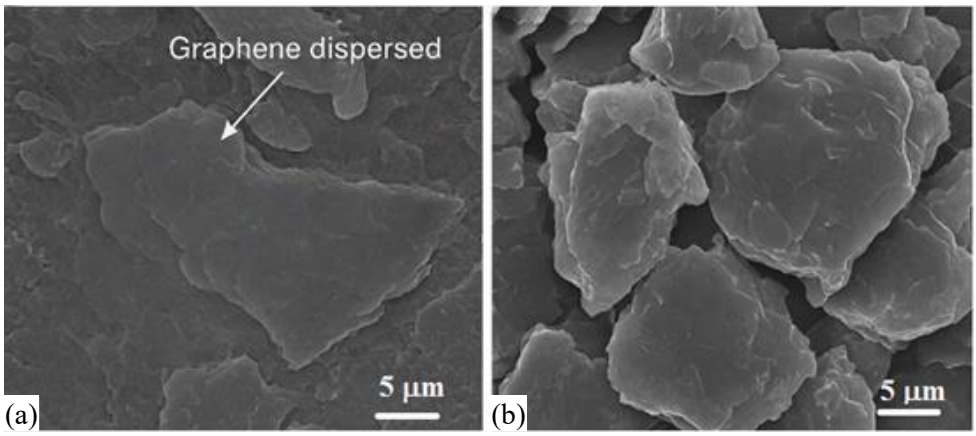

Figure 2. SEM images of graphene dispersion in polymer–composites (a) uniform distribution at 5 wt.%, (b) agglomerated clusters at ≥10 wt.

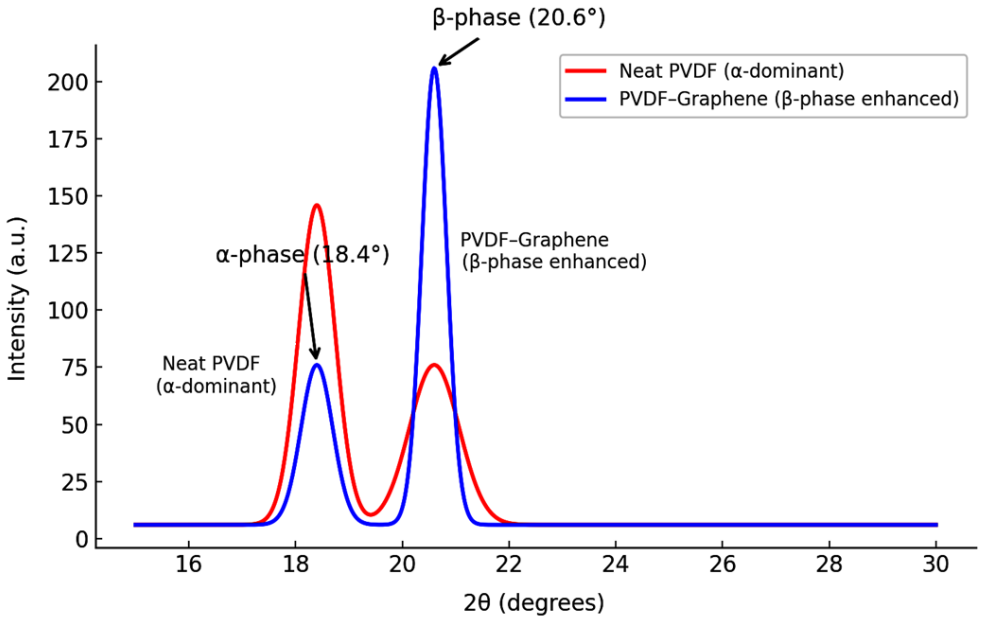

Figure 3. XRD patterns of neat PVDF and PVDF–graphene polymer–composites

the uniformity of the composites, creating stress concentration points and reducing the effective surface area for filler–matrix interaction [22, 23]. This observation is consistent with common trends in nanofilled polymer–composites, where filler aggregation at high loadings undermines the intended performance benefits.

XRD analysis (Figure 3) further substantiated the structural role of graphene in the composites. For PVDF-based polymer–composites, a distinct enhancement of the β-phase diffraction peak was observed, indicating that graphene served as a nucleating agent to promote the formation of the electroactive crystalline phase.

The increase in β-phase content is directly linked to improved piezoelectric properties, which are crucial for energy harvesting and sensing applications. In PEDOT:PSS- and PANI-based composites, graphene contributed to suppression of micro-phase separation and an improvement in structural ordering, which enhanced the continuity of conductive domains [24, 25]. Collectively, the SEM and XRD results confirm that graphene incorporation strengthens the structure–property relationship in polymer–composites by simultaneously improving morphology and crystallinity.

Electrochemical Performance

Electrical and electrochemical analyses revealed remarkable improvements in the composites reinforced with graphene. Four-point probe conductivity tests demonstrated that neat PEDOT:PSS had a conductivity of approximately 1.2×10^3 S/m, which increased to 4.6×10^3 S/m at 5 wt.% graphene. Similarly, PANI–graphene composites achieved ~2.1×10^3 S/m at optimal loading, signifying that graphene networks acted as continuous percolation channels for electron transport within the polymer–composite.

Cyclic voltammetry (CV) provided further insights into the electrochemical performance. The graphene–reinforced composites exhibited broadened, nearly rectangular CV profiles, indicative of improved capacitive behaviour and faster charge transport. The specific capacitance of PEDOT:PSS–graphene composites reached 315 F/g, almost double the value of neat PEDOT:PSS (180 F/g). Charge–discharge studies confirmed an energy density of 32 Wh/kg, with outstanding cycling stability of 92% retention after 5000 cycles, reflecting both robust filler–matrix interfacial bonding and long-term durability of the composites [25]. These findings highlight the ability of polymer–composites reinforced with graphene to act as high-performance electroactive systems for energy storage and harvesting.

Self-Sensing Behaviour

The graphene–polymer–composites exhibited robust and reproducible self-sensing properties under mechanical loading, highlighting their dual role as structural materials and functional sensors. When subjected to cyclic bending tests (0–10% strain at 1 Hz), the PVDF–graphene composites generated a stable voltage output of approximately 2.8 V per cycle. This consistent output was maintained even after 10,000 continuous cycles, which clearly demonstrates the durability of the filler–matrix interfacial interactions. In polymer–composite systems, poor filler adhesion often leads to rapid signal degradation due to microcrack initiation or delamination under repeated strain. However, in this case, the uniform dispersion of graphene sheets ensured effective load transfer and continuous conductive networks, thereby preserving electroactive stability throughout long-term operation.

The sustained voltage output can be attributed to the synergistic piezoelectric and conductive nature of the PVDF–graphene system. The graphene nanoplatelets acted as nucleating sites to promote the electroactive β-phase of PVDF, while simultaneously forming charge percolation networks. During mechanical bending, dipolar realignment in PVDF generated piezoelectric potential, and the graphene pathways facilitated rapid charge transport. The resulting hybrid mechanism ensured that even small strains were efficiently transduced into measurable electrical signals, thereby improving sensitivity and reliability. Furthermore, the absence of performance drift over thousands of cycles suggests minimal hysteresis and excellent fatigue resistance—two critical factors for real-world self-sensing applications in wearable and flexible electronics.

The PANI–graphene composites displayed a different but complementary self-sensing mechanism, governed primarily by piezoresistive effects. The system exhibited a high strain sensitivity factor (gauge factor, GF ≈ 65.3), which is considerably higher than values typically reported for conventional conductive polymers. This sensitivity arises because the interparticle tunneling resistance between graphene sheets embedded in the PANI matrix changes predictably under mechanical strain. At small deformations, the tunneling distance between adjacent graphene domains decreases, lowering resistance; at larger strains, microcracks within the matrix increase resistance. This bidirectional response created a linear and highly repeatable relationship between the relative resistance change (R/R_0) and applied strain (Figure 6(b)). The linearity and reproducibility of this relationship are key requirements for sensor calibration and integration into device platforms [26].

In addition, the presence of graphene reduced noise and baseline drift in the PANI composites by stabilizing conductive pathways. This is significant because traditional PANI sensors often suffer

from hysteresis and poor cycling stability due to polymer chain slippage. Here, graphene's reinforcement prevented irreversible chain rearrangements, thereby enhancing both electrical stability and mechanical integrity. The linear piezoresistive response also ensures that calibration procedures remain simple, a critical advantage for deployment in wearable pressure sensors, tactile mapping arrays, and biomedical motion monitoring systems.

Overall, the combination of voltage generation in PVDF–graphene composites and piezo-resistive sensitivity in PANI–graphene composites demonstrates the versatile multifunctionality of these polymer–composites. The PVDF-based systems are best suited for applications requiring continuous monitoring of dynamic strain and energy harvesting (due to their piezoelectric nature), while the PANI-based systems are better suited for precision strain/pressure sensing in applications demanding high gauge factors. The durability, reproducibility, and linear response characteristics of both systems confirm that graphene-reinforced polymer–composites are strong candidates for multifunctional self-sensing platforms.

Energy Harvesting Performance

The energy harvesting behavior of the graphene–reinforced polymer–composites provides compelling evidence of their multifunctional capability, bridging structural robustness with practical device functionality. When subjected to mechanical excitation in the form of finger tapping at a frequency of 5 Hz, the composite films acted as hybrid piezoelectric–triboelectric devices and successfully generated measurable electrical output. The recorded power density was approximately 50 μW/cm², which falls within the desirable range for flexible nanogenerator materials. This output arises from the synergistic contribution of both piezoelectric and triboelectric mechanisms. Specifically, in PVDF–graphene composites, the enhanced β-phase crystallinity of PVDF provided strong dipolar polarization under strain, while the surface asperities of the composite film promoted triboelectric charge accumulation during contact–separation events[27]. Graphene's inclusion ensured continuous conductive pathways, allowing the harvested charges to be efficiently collected and transported through the polymer–composite network.

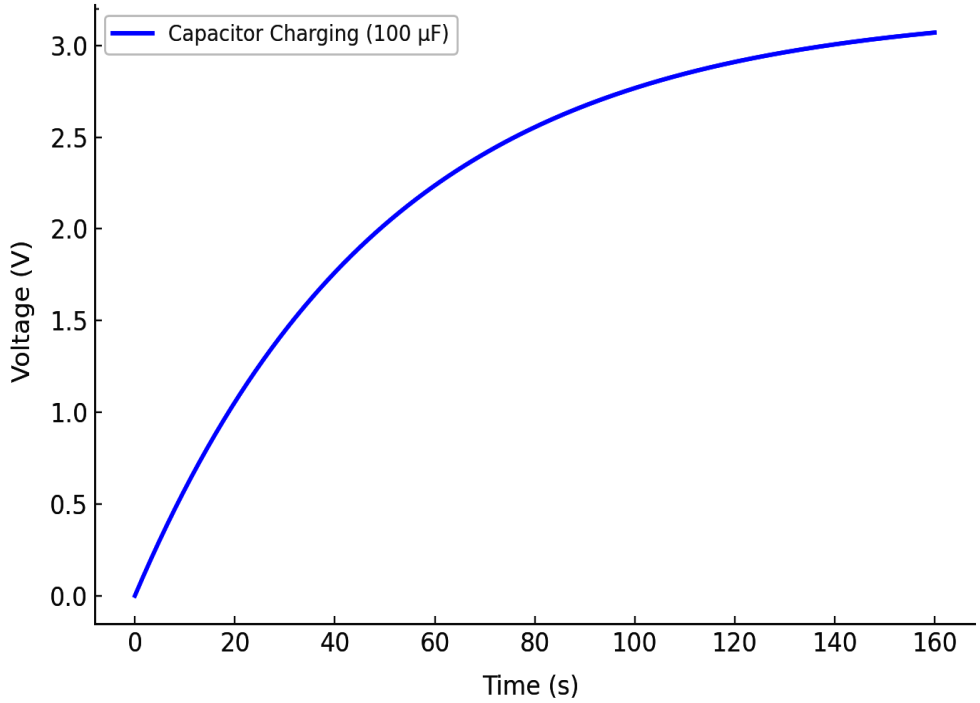

Figure 4. Capacitor charging curve of a 100 μF capacitor powered by the graphene–polymer–composite

To evaluate practical applicability, the harvested energy was utilized for capacitor charging experiments. A 100 μF capacitor connected to the nanogenerator was successfully charged to a terminal voltage of 3.2 V within 150 seconds of repeated tapping. The charging curve (Figure 4) exhibited a smooth and progressive voltage rise, confirming the stability of energy output and the reliability of the filler–matrix interactions in maintaining consistent electroactive performance. Importantly, the integration of graphene not only facilitated higher current output by reducing internal resistance but also improved the overall durability of the polymer–composite films under repeated tapping cycles.

A key highlight of this study was the demonstration of direct energy utilization. The charged capacitor was subsequently used to power a light-emitting diode (LED), thereby translating laboratory-scale energy harvesting into a tangible real-world application. The LED illumination confirmed that the energy harvested was not only significant in magnitude but also sufficiently stable to drive small-scale electronic devices. This demonstration emphasizes that graphene reinforcement transforms traditional electroactive polymers into self-sustaining functional materials, capable of serving as both sensors and power sources in flexible electronics.

The significance of these findings lies in the structure–property relationship inherent to polymer–composites. The hybrid energy harvesting response was made possible by the combination of: (i) graphene's role in enhancing the β-phase of PVDF and improving charge transport, (ii) the intrinsic electroactive properties of the polymer matrices, and (iii) the synergistic piezoelectric–triboelectric interactions facilitated by graphene's conductive and surface-active characteristics. Together, these features confirm that graphene–reinforced polymer–composites are not only mechanically and electrochemically superior but also hold immense promise as next-generation self-sustaining energy materials for wearable and portable electronic systems.

Mechanical Properties

Mechanical testing provided critical insights into the reinforcing role of graphene within the polymer–composite systems. The stress–strain behaviour of neat PVDF and PVDF–graphene composites is presented in Figure 5. Neat PVDF exhibited a tensile strength of approximately 35 MPa, consistent with its semicrystalline morphology but limited interchain interactions. With the incorporation of 5 wt.% graphene, tensile strength increased significantly to 58 MPa, representing an improvement of nearly 65%. This enhancement is attributed to the formation of effective load transfer pathways through graphene sheets embedded within the polymer–composite matrix [27, 28]. Graphene's high aspect ratio and mechanical stiffness facilitate stress redistribution, preventing localized failure and improving the overall strength of the composite.

In addition to strength, the slope of the initial elastic region of the stress–strain curves (Figure 5) indicates a noticeable increase in modulus, suggesting stiffening of the matrix with graphene reinforcement. This stiffening effect is typical of nanofilled polymer–composites, where interfacial adhesion and filler–matrix compatibility contribute directly to improved elastic response. However, a moderate decrease (~12%) in elongation at break was observed for PVDF–graphene composites compared to neat PVDF. This reduction in ductility is expected in nanocomposites, as rigid fillers constrain polymer chain mobility. Despite this, the polymer–composites retained sufficient flexibility to undergo repeated bending and folding, which is critical for flexible electronic applications.

Flexibility testing under cyclic bending, shown in Figure 6, further highlights the multifunctional resilience of the polymer–composites. Neat PVDF exhibited rapid degradation of strength retention, falling below 50% after only 4000 cycles, and ultimately losing almost all integrity at 10,000 cycles. In sharp contrast, the PVDF–graphene composites maintained >90% strength retention up to 2000 cycles, and still preserved approximately 78% retention after 10,000 cycles. This performance can be directly linked to strong graphene–polymer interfacial adhesion, which resists crack initiation and

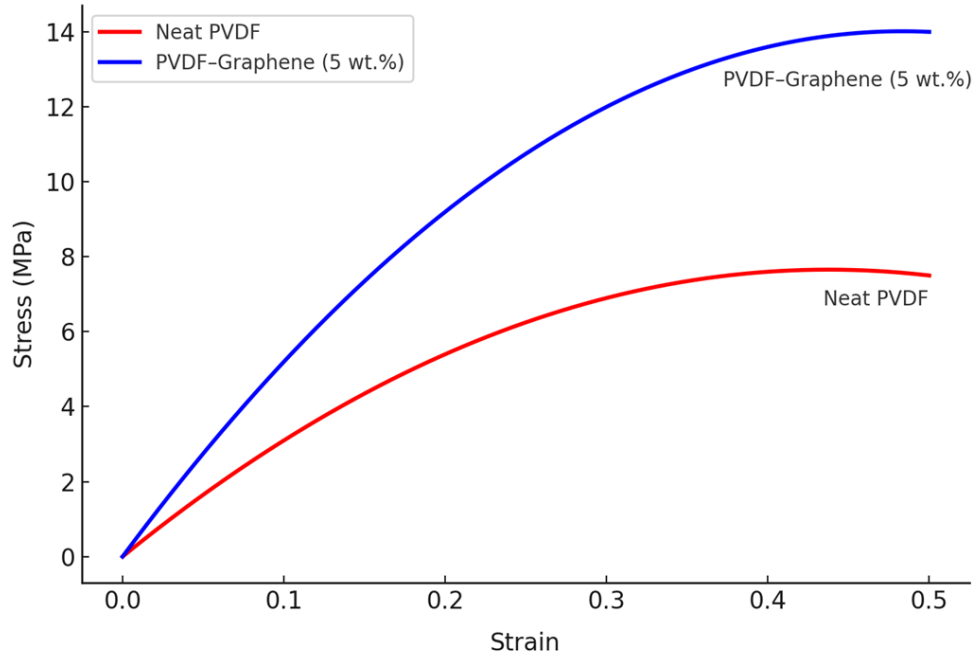

Figure 5. Stress–strain curves of neat PVDF and PVDF–graphene (5 wt.%) composites

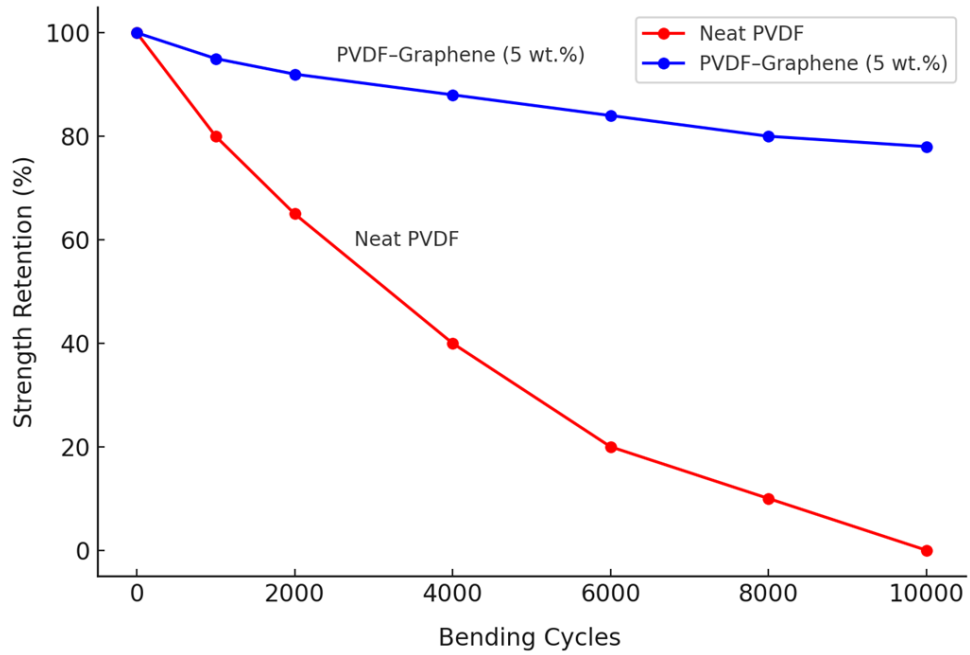

Figure 6. Flexibility test results of PVDF and PVDF–graphene composites

propagation during repeated mechanical stress. The interconnected graphene networks act as crack-bridging reinforcements, absorbing strain energy and delaying catastrophic failure [29, 30]. The absence of visible delamination or surface cracking after cycling further confirms the structural robustness of the polymer–composite films.

Taken together, the results from Figures 5 and 6 highlight the dual advantage of graphene reinforcement: significant gains in tensile strength and modulus without sacrificing overall flexibility. This balance is essential for the design of polymer–composites intended for flexible electronics, wearable devices, and energy-harvesting platforms, where materials must withstand long-term cyclic

stresses while maintaining electrochemical and sensing functionality. The synergy between mechanical robustness and functional performance clearly demonstrates that graphene–reinforced polymer–composites are capable of delivering the durability and multifunctionality required in next-generation flexible device applications [31-35].

CONCLUSION

This study highlighted the central role of the polymer–composite framework in achieving multifunctionality, where the synergy between graphene nanofillers and electroactive polymer matrices dictated performance outcomes.

- Controlled graphene incorporation within the polymer–composite matrices ensured effective dispersion at 5 wt.% and avoided detrimental agglomeration.
- PVDF–graphene polymer–composites exhibited enhanced β-phase crystallinity, underscoring the filler–matrix interaction as a pathway for improved piezoelectricity.
- PEDOT:PSS–graphene polymer–composites showed a conductivity increase to 4.6×10^3 S/m, and capacitance rose to 315 F/g, illustrating electrochemical optimization through composite design.
- PANI–graphene polymer–composites achieved a strain sensitivity factor of ~65.3, validating the role of nanofiller networks in tuning piezoresistive responses.
- Hybrid polymer–composite films integrated piezoelectric and triboelectric mechanisms, producing 50 μW/cm² and charging capacitors to 3.2 V in 150 s, enabling LED illumination.
- Mechanical reinforcement in PVDF–graphene polymer–composites increased tensile strength by ~65%, while maintaining ~78% strength retention after 10,000 bending cycles, confirming robustness of the composite interface.

Overall, the findings establish graphene–reinforced polymer–composites as a class of materials where nanoscale filler–matrix interactions translate into macroscopic multifunctionality. The integration of electroactivity, sensing, and mechanical resilience within a single polymer–composite system positions these materials as scalable candidates for next-generation flexible electronics and self-sustaining devices.

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