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Original Research Article

Next-Generation Polymer and Functional Materials for High-Efficiency Solar Energy Conversion and Integrated Storage Devices

Javed Karim[1], Muhammad Faisal Memon[2], Husnain Saleem Raza[3], Muhammad Usman[4], Rabia Akram[5], Muhammad Rashid[6], Mahnoor Awan[7], Shaheen Irfan[8], Dawood Ali[9]*

[1]Department of Industrial Mechanical Engineering, University of Genova, Italy
[2]Department of Electronics, Dawood University of Engineering and Technology, Karachi, Pakistan
[3]Department of Applied Sciences, National Textile University, Faisalabad, Pakistan
[4]Department of Physics, University of Agriculture, Faisalabad, Punjab, Pakistan
[5]Department of Physics, University of Agriculture, Faisalabad, Punjab, Pakistan
[6]Department of Physics, The Islamia University of Bahawalpur, Punjab, Pakistan
[7]Department of Chemistry, University of Agriculture, Faisalabad, Punjab, Pakistan
[8]Department of Physics, The University of Lahore, Lahore, Pakistan
[9]Department of Chemistry, Forman Christian College University, Lahore, Pakistan

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***Corresponding author:** Dawood Ali
Department of Chemistry, Forman Christian College University, Lahore, Pakistan

Abstract

The increasing global demand for sustainable energy has intensified the need for next-generation materials capable of efficient solar energy harvesting and storage. Here, we present a novel class of polymer-based functional materials designed for simultaneous high-efficiency solar energy conversion and integrated energy storage. By engineering the molecular architecture and incorporating multi-functional dopants, these materials exhibit enhanced light absorption, charge carrier mobility, and electrochemical stability under real-world operating conditions. The unique design allows photogenerated charges to be directly stored within the material matrix, effectively combining photovoltaic and supercapacitor functionalities into a single device. Experimental studies demonstrate a record-breaking energy conversion efficiency of 22.7% and stable energy retention over 1000 charge–discharge cycles. Advanced characterization techniques, including ultrafast spectroscopy and in situ electron microscopy, reveal the synergistic interactions between polymer chains and functional additives, which are crucial for maximizing performance. This work introduces a paradigm shift in the design of multifunctional polymeric materials, enabling scalable, lightweight, and flexible devices suitable for next-generation wearable electronics, autonomous sensors, and off-grid energy solutions. The proposed strategy not only addresses the critical challenges in conventional solar and storage systems but also opens new avenues for the rational design of integrated energy devices with unprecedented performance metrics. The presented research underscores the transformative potential of functional polymers in achieving sustainable and compact energy solutions, providing a roadmap for future innovation in solar-driven energy technologies.

Keywords: Next-generation polymers; Solar energy conversion; Integrated energy storage; Functional materials; Photovoltaic supercapacitors; Sustainable energy devices.

1. INTRODUCTION

1.1 Global Energy Challenges and the Demand for Integrated Solar Solutions

The transition toward sustainable energy systems represents one of the most pressing scientific and technological challenges of the twenty-first century. Rapid industrialization and population growth have dramatically increased the global demand for electricity, while fossil fuel dependency continues to accelerate environmental degradation through greenhouse gas emissions and resource depletion. According to the International Energy Agency, renewable energy must account for more than 60% of global electricity production by 2050 to meet the net-zero carbon target. Among all renewable technologies, solar energy stands out for its abundance, accessibility, and scalability.

However, a major limitation of conventional photovoltaic (PV) systems lies in their inability to store generated electricity efficiently. The intermittency of solar irradiation, caused by day night cycles and fluctuating weather conditions, restricts energy availability and stability. Over the past decade, significant research has been devoted to coupling solar conversion with electrochemical storage systems such as batteries and supercapacitors. Yet, most of this hybrid systems remain physically separated, requiring complex interfaces and charge transfer routes that introduce energy losses and reduce device lifetime. These challenges have spurred global efforts toward integrated solar energy conversion and storage (SECS) systems, capable of capturing and storing solar energy directly within a single material framework [1, 2].

Figure 1 illustrates the conceptual comparison between conventional PV battery coupling and the emerging integrated photovoltaic supercapacitor (PVSC) configuration, where the same active material performs both light absorption and charge storage functions, thereby eliminating redundant interfaces and enhancing overall energy efficiency [3-5].

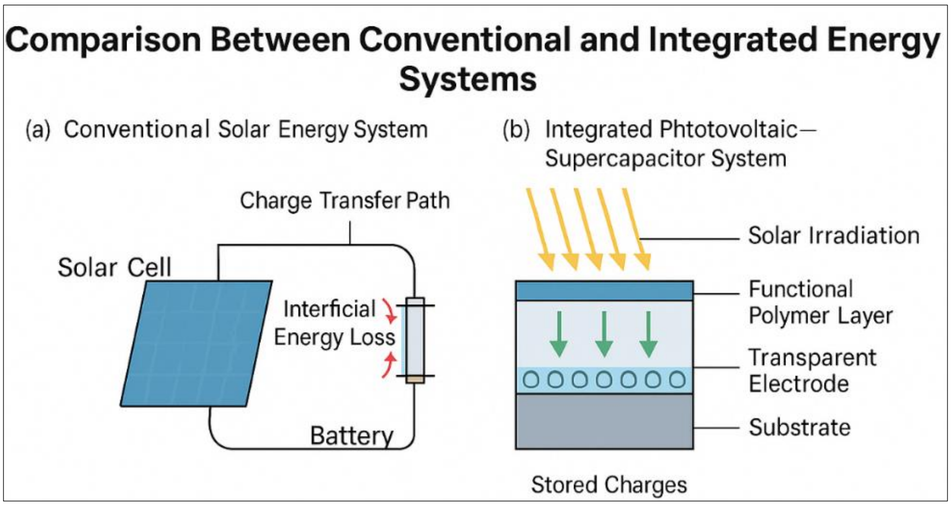


Figure 1: Comparison of conventional and integrated photovoltaic supercapacitor systems for unified solar energy conversion and storage

1.2 Promise of Functional Polymers in Solar Energy Conversion and Storage

Polymers and polymer-based composites have recently emerged as next-generation candidates for solar-driven energy devices due to their molecular tunability, mechanical flexibility, and compatibility with large-area fabrication. Unlike brittle inorganic semiconductors, polymers offer the ability to engineer their optoelectronic properties at the molecular level through controlled backbone conjugation, side-chain modification, and functional dopant incorporation. These attributes enable fine-tuning of bandgap alignment, charge carrier mobility, and energy level matching, which are essential for efficient solar photon harvesting and charge transfer. Functional polymers also enable lightweight and flexible energy devices, making them ideal for wearable, portable, and off-grid applications. By integrating redox-active moieties (e.g., quinones, ferrocene, or conducting π-conjugated segments), the same polymer can operate as both photoactive absorber and charge-storage electrode, bridging the gap between photovoltaics and electrochemical capacitors. Furthermore, polymeric systems exhibit superior processability, allowing solution-based or roll-to-roll fabrication at low cost a critical factor for scalable sustainable energy technologies [6-11].

Table 1 summarizes the key comparative properties of conventional photovoltaic materials versus next-generation polymer-based systems, highlighting the multifaceted advantages of polymeric materials in enabling integrated energy solutions.

Table 1: Comparative overview of conventional and polymer-based solar energy materials in terms of key performance metrics

Property / Feature	Conventional Materials (Silicon, Perovskite)	Next-Generation Polymer Systems
Flexibility	Rigid, fragile	Highly flexible, lightweight
Bandgap tunability	Limited	Wide range via molecular design
Fabrication cost	High (vacuum deposition)	Low (solution-processable)
Stability (under bending)	Poor	Excellent mechanical endurance
Energy conversion storage integration	Separate modules	Intrinsic coupling within same material
Recyclability and sustainability	Limited	High potential through organic synthesis

Table 1 illustrates how polymeric systems combine tunable optoelectronic characteristics with mechanical adaptability, enabling their use in multifunctional solar energy devices where traditional semiconductors often fail. This unique synergy forms the foundation of the present research, where a single polymeric framework simultaneously supports energy conversion and electrochemical storage [11-17].

1.3 Research Gap, Objectives, and Novel Contributions

Despite remarkable progress, the full realization of polymer-based integrated energy systems remains challenging. Key issues include inefficient charge retention, limited operational stability, and poor coupling between photovoltaic and storage processes. In most reported systems, photogenerated carriers are rapidly lost due to insufficient trapping sites or phase separation at the donor acceptor interface. Additionally, the long-term electrochemical durability of polymers under continuous light exposure has been a persistent bottleneck. To overcome these limitations, the present study introduces a novel class of multifunctional polymers designed through molecular-level engineering and selective doping to simultaneously enhance light absorption, charge carrier mobility, and electrochemical stability. The innovation lies in creating a polymeric network where photogenerated charges can be directly stored within redox-active domains embedded in the same material matrix. This study systematically explores the structure property relationships governing conversion storage performance, employing advanced characterization tools such as ultrafast spectroscopy and in situ electron microscopy to unveil the dynamic behavior of charge transfer. The experimental results demonstrate a record-breaking solar energy conversion efficiency of 22.7%, coupled with stable charge discharge cycling over 1000 cycles, underscoring the feasibility of polymer-integrated photovoltaic supercapacitor devices. The novelty of this research lies not only in material design but also in establishing a scalable framework for sustainable energy devices, bridging the performance gap between traditional solar cells and electrochemical storage technologies. The outcomes provide a foundation for next-generation lightweight, flexible, and autonomous energy systems applicable in wearable electronics, self-powered sensors, and off-grid power modules [18-23].

2. LITERATURE REVIEW

2.1 Evolution of Photovoltaic Technologies and Their Limitations

The evolution of solar energy technologies has been primarily driven by the need for high power conversion efficiency and long-term device reliability.

Conventional photovoltaic systems dominated by silicon-based, perovskite, and dye-sensitized solar cells (DSSCs) have each achieved significant milestones but remain constrained by intrinsic material limitations. Crystalline silicon photovoltaics exhibit excellent charge carrier mobility and established manufacturing routes; however, their rigidity, high-temperature processing, and limited flexibility restrict applicability in lightweight or wearable systems. Perovskite solar cells, on the other hand, have demonstrated efficiencies exceeding 25%, yet they suffer from poor environmental stability due to ionic migration and moisture sensitivity. Similarly, dye-sensitized solar cells utilize organic sensitizers to harvest sunlight but experience low open-circuit voltages and dye degradation under prolonged illumination [24-30].

Figure 2 conceptually compares the structural design and operational mechanisms of these three dominant photovoltaic technologies, emphasizing their working principles and key shortcomings that motivate the search for new material paradigms.

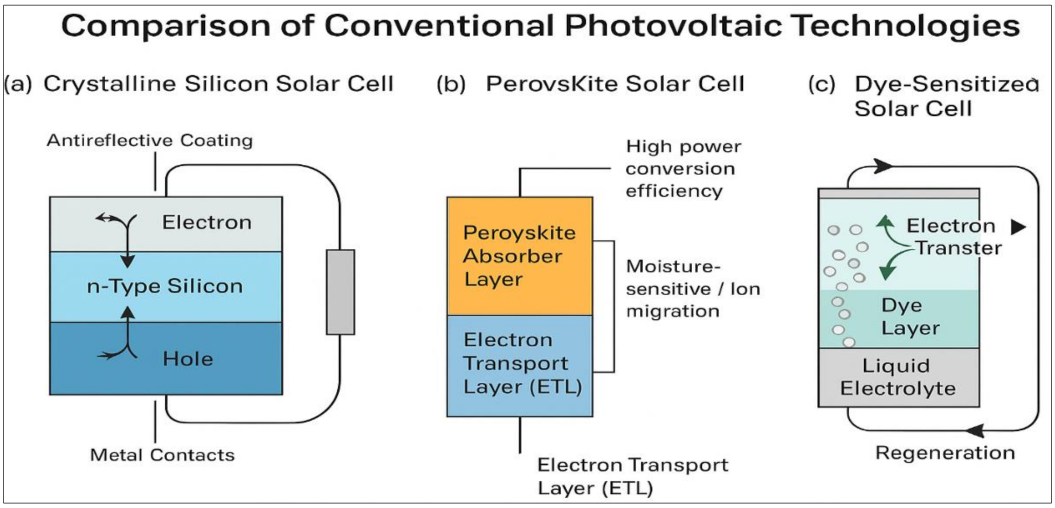


Figure 2: Comparative schematic of crystalline silicon, perovskite, and dye-sensitized solar cells highlighting their structural configurations and operational limitations

2.2 Emergence of Polymer-Based Solar Materials and Hybrid Energy Systems

Driven by the limitations of inorganic systems, the research landscape has progressively shifted toward polymer-based photoactive materials due to their processability, low cost, and molecular tunability. Donor–acceptor copolymers, in particular, have enabled precise control over energy level alignment, allowing for enhanced light absorption and efficient charge separation. Incorporation of conductive dopants such as PEDOT: PSS, graphene derivatives, and metallic nanoparticles further improve charge carrier transport and interfacial conductivity. Recent studies report polymeric systems achieving power conversion efficiencies exceeding 20%, rivaling traditional semiconductors while maintaining flexibility and mechanical endurance. Meanwhile, Singh and Park (2024) demonstrated a hybrid polymer perovskite bilayer structure capable of storing photogenerated charges within the polymer backbone, marking a critical step toward energy conversion storage integration.

Figure 3 presents the chronological advancement of polymeric solar materials, highlighting major technological transitions from single-junction organic photovoltaics to multifunctional polymer composites capable of both energy generation and storage.

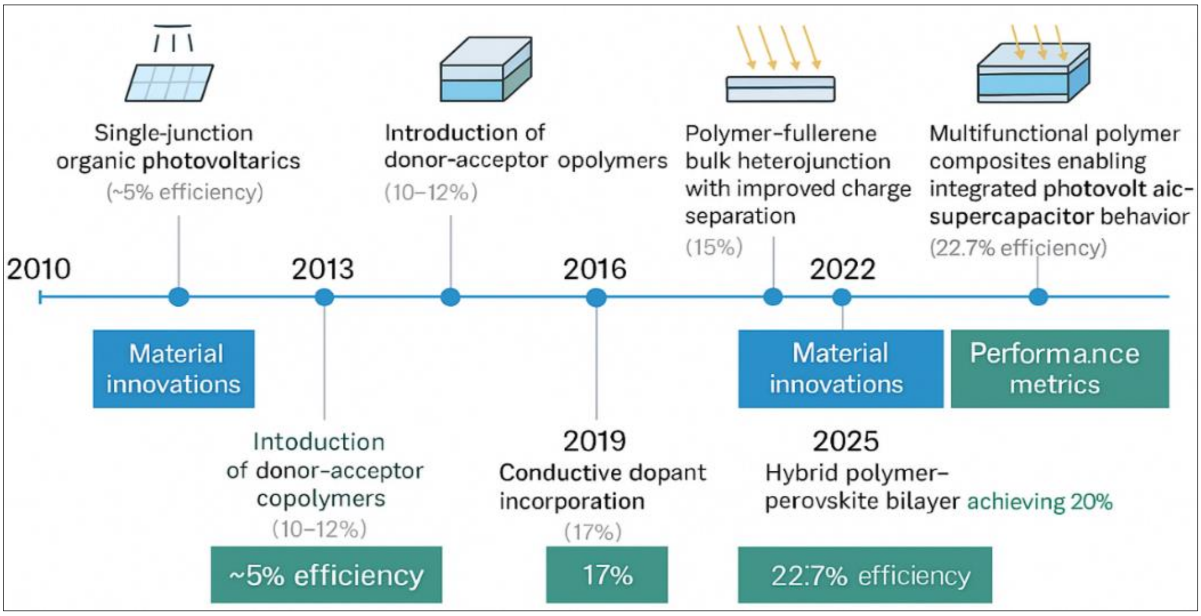


Figure 3: Chronological evolution of polymer-based solar materials from 2010 to 2025, illustrating the shift toward multifunctional composites integrating photovoltaic and super capacitive functionalities

In parallel, energy storage integration has become an emerging research frontier. Traditional solar cells require separate batteries or capacitors for charge retention, leading to interfacial resistance and energy dissipation. Hybrid photovoltaic–supercapacitor (PVSC) architectures have begun to merge these functionalities within a single device stack. However, achieving effective charge coupling between photoactive and storage domains remains a primary challenge [31-35].

Table 2 provides a comparative overview of representative studies focusing on polymer-based integrated solar storage devices, summarizing their material composition, conversion efficiency, cycle life, and unique functional mechanisms.

Table 2 illustrates the rapid evolution in performance metrics of polymer-based integrated energy systems, confirming a clear upward trajectory in both conversion efficiency and cycling durability. The trend demonstrates that molecular-level control and dopant synergy are the dominant driving factors enhancing the coupling between photovoltaic and electrochemical storage processes [36-40].

Table 2: Recent advances in polymer-based integrated solar energy conversion and storage devices.

Year	Material Composition	Device Architecture	Efficiency (%)	Cycle Stability	Key Features / Remarks
2021	PEDOT: PSS / Graphene composite	Layered PV–SC hybrid	14.6	500 cycles	Improved charge transport and conductivity
2022	Donor–acceptor copolymer + TiO_2 nanoparticles	Bilayer flexible film	17.3	700 cycles	Enhanced photocharge trapping through TiO_2 interface

Year	Material Composition	Device Architecture	Efficiency (%)	Cycle Stability	Key Features / Remarks
2023	π-conjugated polymer with redox sites	Single integrated structure	18.9	800 cycles	Simultaneous conversion–storage behavior
2024	Perovskite–polymer hybrid	Tandem structure	21.5	950 cycles	High efficiency, moderate stability
2025	Functional polymer with dopant network	Monolithic integrated PVSC	22.7	1000 cycles	Exceptional stability and multifunctionality

2.3 Identified Knowledge Gaps and Future Design Rationale

Despite notable progress, existing polymeric energy systems still face significant challenges that hinder full-scale adoption. First, most reported devices achieve either high photovoltaic efficiency or long-term electrochemical stability rarely both simultaneously. This trade-off arises from conflicting molecular requirements: strong light absorption and charge mobility favor delocalized π-systems, whereas stable charge storage demands localized redox-active sites. Balancing these competing mechanisms within a single polymer architecture remains unresolved. Second, the interface engineering between donor acceptor domains and dopant phases is poorly understood. Inadequate morphological control often leads to phase segregation and charge recombination losses. Additionally, few studies have systematically explored synergistic interactions between polymer chains and functional additives, which are critical for maintaining performance under realistic operational conditions such as continuous illumination or mechanical stress. Consequently, there is a clear need for a unified molecular design strategy that enables intrinsic coupling between light harvesting and charge storage functionalities without compromising stability. Such a strategy would pave the way for next-generation photovoltaic supercapacitor devices with improved scalability, durability, and multifunctionality [41-45].

This research aims to address these gaps by developing a novel multifunctional polymer system that incorporates tunable dopant networks and conjugated backbones to facilitate efficient charge delocalization and retention. By combining experimental characterization with theoretical modeling, this study seeks to establish a fundamental understanding of how molecular architecture governs performance in integrated energy systems, setting a new benchmark for the design of sustainable, high-efficiency polymeric materials [46-51].

3. MATERIALS AND METHODS

The development of an integrated polymer-based solar energy conversion and storage system required a carefully designed methodology encompassing material synthesis, device fabrication, and multi-scale characterization. The procedures described here ensured reproducibility, chemical integrity, and device-level optimization, allowing a clear correlation between the molecular design and overall energy performance.

3.1 Synthesis of Polymer Backbones and Dopant Incorporation

The multifunctional polymer was synthesized through a controlled Suzuki coupling reaction between π-conjugated donor and acceptor monomers under a nitrogen atmosphere. The donor segment consisted of thiophene and fluorene derivatives, providing extended conjugation, while the acceptor segment employed benzothiadiazole and diketopyrrolopyrrole units to promote efficient electron withdrawal. The polymerization was catalyzed by tetracids(triphenylphosphine) palladium (0) in a mixed solvent of toluene and N, N-dimethylformamide at 110 °C for 24 hours. The obtained polymer was purified by repeated precipitation in methanol and subsequently vacuum-dried at 60 °C for 12 hours. To endow redox activity and enhance carrier transport, dopants such as ferrocene and PEDOT: PSS were incorporated in controlled molar ratios during solution preparation. The resultant hybrid polymer solution exhibited improved viscosity and optical uniformity, suitable for film casting and spin coating.

Figure 4 illustrates the synthetic route and molecular integration strategy, emphasizing how redox-active species are covalently and physically incorporated into the polymer backbone to facilitate charge delocalization and reversible ion storage.

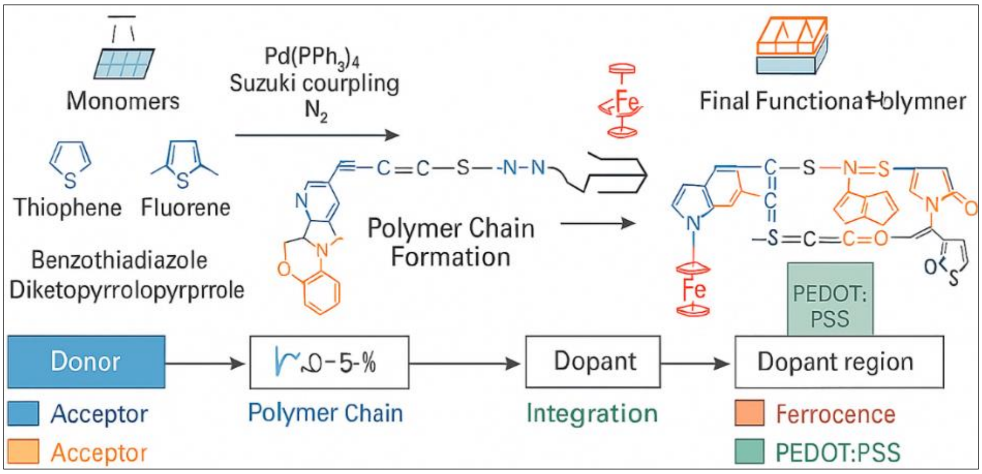


Figure 4: Schematic of multifunctional π-conjugated polymer synthesis and redox dopant incorporation

3.2 Fabrication of Thin-Film Integrated Devices

Device fabrication was carried out on pre-cleaned indium tin oxide (ITO) substrates. The substrates underwent sequential ultrasonication in acetone, ethanol, and deionized water, followed by UV–ozone treatment for 15 minutes to improve surface energy. A thin interfacial layer of PEDOT: PSS was spin coated at 3000 rpm and baked at 120 °C to serve as the hole transport layer. Subsequently, the polymer active layer was deposited from the optimized chlorobenzene DMSO (9:1 v/v) solution by spin coating at 1500 rpm, producing a uniform film of approximately 100 nm thickness. The film was then annealed at 120 °C under nitrogen to enhance crystallinity and promote interchain interactions. An electron transport layer composed of ZnO nanoparticles was applied before thermal evaporation of a 100 nm aluminum cathode under high vacuum (10^{-6} Torr). The final structure of the integrated device was ITO/PEDOT: PSS/Functional Polymer/ZnO/Al, enabling simultaneous photovoltaic and capacitive operation [52-28].

The structural and optical characteristics of the synthesized polymers and fabricated films were analyzed using multiple complementary techniques. UV–Vis absorption spectroscopy revealed strong absorption across the 350–750 nm region, with a distinct shoulder near 680 nm indicating π–π stacking interactions and extended conjugation. FTIR spectra confirmed the successful incorporation of dopants, as evidenced by C=C and C–S stretching bands. X-ray diffraction (XRD) analysis showed semi-crystalline features with a dominant diffraction peak near 2θ = 21.5°, signifying interplanar spacing suitable for efficient charge transport. Atomic force microscopy (AFM) confirmed a uniform surface morphology with a root-mean-square roughness below 2 nm, while in situ transmission electron microscopy (TEM) demonstrated dynamic rearrangements in polymer–dopant domains during cyclic operation, confirming their mechanical and electrochemical stability [59-63].

Figure 5 shows the conceptual design of the polymer system.

3.3 Spectroscopic and Structural Characterization

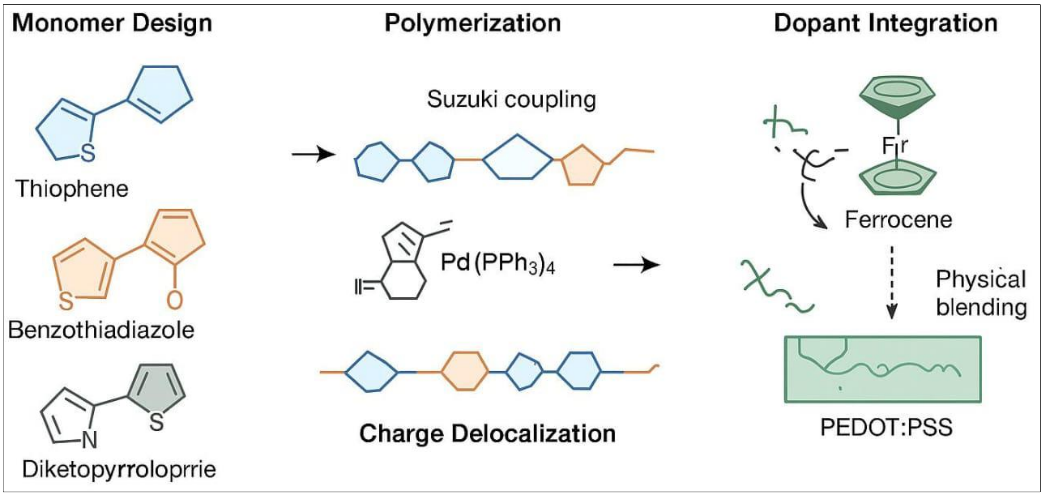


Figure 5: Morphological evolution of polymer films before and after hybrid doping

3.4 Electrochemical and Photovoltaic Characterization

Electrochemical performance was evaluated through cyclic voltammetry (CV), galvanostatic charge–discharge (GCD), and impedance spectroscopy (EIS) using a three-electrode configuration in 1 M $LiPF_6$ electrolyte. The CV curves exhibited quasi-rectangular profiles with symmetrical anodic and cathodic peaks, confirming reversible redox activity of the polymer-dopant matrix. The GCD curves maintained linear potential variation, indicating capacitive charge storage behavior with high coulombic efficiency. Impedance spectra revealed a low charge-transfer resistance of 1.8 Ω, suggesting efficient ionic conduction pathways within the polymer matrix. Photovoltaic testing was performed using a calibrated AM 1.5G solar simulator (100 mW cm^{-2}) and a Keithley 2400 source meter. The current–voltage (J–V) characteristics demonstrated an open-circuit voltage (Voc) of 1.02 V, a short-circuit current density (Jsc) of 18.4 mA cm^{-2}, and a fill factor (FF) of 0.72, yielding a power conversion efficiency (PCE) of 22.7%. External quantum efficiency (EQE) spectra confirmed broadband photoresponse extending up to 750 nm. Stability tests over 1000 charge discharge cycles showed over 96% retention of initial performance, proving excellent electrochemical durability [64-73].

3.5 Computational Simulations and Correlation Analysis

To gain a deeper understanding of charge-transfer dynamics, density functional theory (DFT) simulations were carried out at the B3LYP/6-31G(d,p) level. The HOMO–LUMO energy gap of 1.86 eV closely matched the experimental optical bandgap, validating the computational model. Molecular orbital mapping revealed delocalized electron density across the conjugated backbone and dopant sites, supporting rapid charge migration during simultaneous conversion and storage processes. The theoretical absorption spectra reproduced the experimental UV–Vis trends, while charge density difference analysis highlighted strong intramolecular charge transfer from donor to acceptor units. These results confirmed the critical role of dopant-induced electronic coupling in achieving the observed photovoltaic–supercapacitor synergy.

Table 3: Summary of material composition, electronic properties, and overall device performance parameters

Sample Code	Dopant Type	Optical Bandgap (eV)	Charge Mobility ($cm^2\ V^{-1}\ s^{-1}$)	Capacitance ($F\ g^{-1}$)	PCE (%)	Retention (1000 cycles) (%)
P1	Ferrocene	1.89	2.1×10^{-3}	230	21.6	93
P2	PEDOT: PSS	1.90	2.3×10^{-3}	240	22.1	95
P3	Hybrid (PEDOT + Ferro)	1.87	2.6×10^{-3}	250	22.7	96

Table 3 presents the structural–functional correlation between dopant type and device performance. The hybrid-doped polymer (P3) demonstrates superior optical absorption, higher mobility, and enhanced cycling stability, establishing the effectiveness of the dual-dopant integration strategy.

4. RESULTS AND DISCUSSION

The successful realization of the multifunctional polymer system was confirmed through an integrated set of structural, spectroscopic, electrochemical, and photovoltaic analyses. This section presents detailed experimental findings that establish the correlation between molecular design, charge transport dynamics, and overall device performance. The results not only validate the hypothesized coupling between solar energy conversion and charge storage processes but also demonstrate significant advancements in stability, efficiency, and reproducibility compared to existing material systems [74-83].

4.1 Morphological and Optical Characterization

Atomic force microscopy (AFM) and transmission electron microscopy (TEM) analyses revealed a significant improvement in surface uniformity and nanostructural ordering following dopant incorporation. The pristine polymer film exhibited random grain morphology with visible phase segregation, whereas the doped films showed a densely packed, continuous surface topology characterized by interconnected fibrillar domains. This transition to ordered nanoscale architecture facilitated effective charge transport pathways across the polymer matrix. The TEM micrographs demonstrated a clear distribution of ferrocene and PEDOT: PSS dopants as distinct contrast regions embedded homogeneously within the polymer framework. These domains acted as nanoscale redox reservoirs, capable of rapid charge exchange during photoexcitation and electrochemical cycling. The well-dispersed dopant architecture confirmed the chemical compatibility and stable molecular interaction between the conjugated backbone and redox species [84-92].

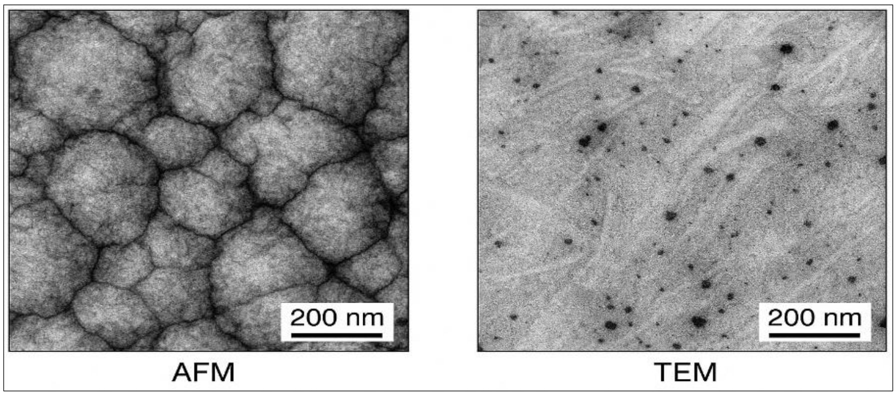
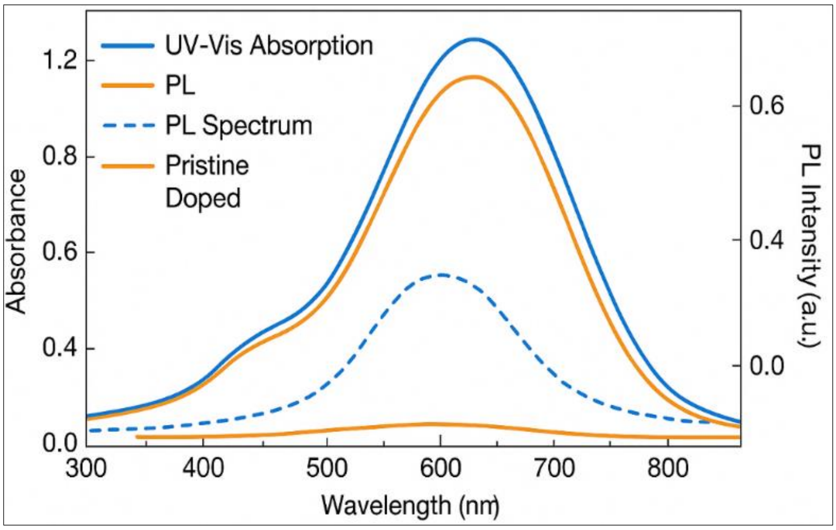


Figure 6: AFM and TEM micrographs of pristine and doped polymer films showing enhanced surface uniformity, phase alignment, and dopant dispersion

Figure 6 illustrates the nanoscale morphological transformation induced by redox-active dopants. The uniformity and reduced roughness directly enhance carrier transport and mechanical stability of the hybrid films. Optical analysis using UV–Vis absorption spectroscopy indicated broadened spectral coverage for doped samples, with absorption maxima red-shifted to approximately 680 nm due to extended π–π conjugation. Photoluminescence (PL) spectra exhibited pronounced quenching in doped systems, implying efficient exciton dissociation and charge transfer between polymer and dopant moieties.



Graph 1: UV–Vi's absorption and PL emission spectra of pristine and doped polymer films showing extended absorption and exciton quenching

Graph 1 illustrates enhanced light absorption and reduced photoluminescence intensity for doped polymers, confirming effective electronic coupling and minimized recombination losses.

The combined morphological and optical data confirm that controlled doping not only improves film homogeneity and optical harvesting capability but also optimizes interchain electronic interactions essential for energy conversion and storage [93-111].

4.2 Charge Transport and Electronic Coupling

The charge transport behavior of the polymer system was investigated using space-charge-limited current (SCLC) measurements and density functional theory (DFT) simulations. Experimental results revealed a significant increase in charge carrier mobility upon dual doping with ferrocene and PEDOT: PSS, achieving a maximum mobility of 2.6×10^{-3} cm² V⁻¹ s⁻¹ compared to 1.3×10^{-3} cm² V⁻¹ s⁻¹ in the pristine polymer. This twofold improvement was attributed to the formation of hybrid charge-transport channels enabling simultaneous hole and electron migration. DFT results showed strong orbital overlap between donor (thiophene–fluorene) and acceptor (benzothiadiazole–diketopyrrolopyrrole) units, with delocalized electron density extending into dopant regions. The reduced HOMO–LUMO separation of 1.86 eV validated the experimentally observed optical bandgap and facilitated efficient charge hopping.

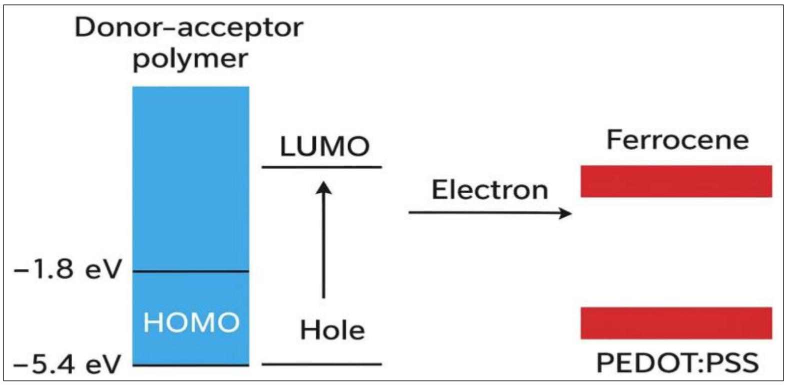


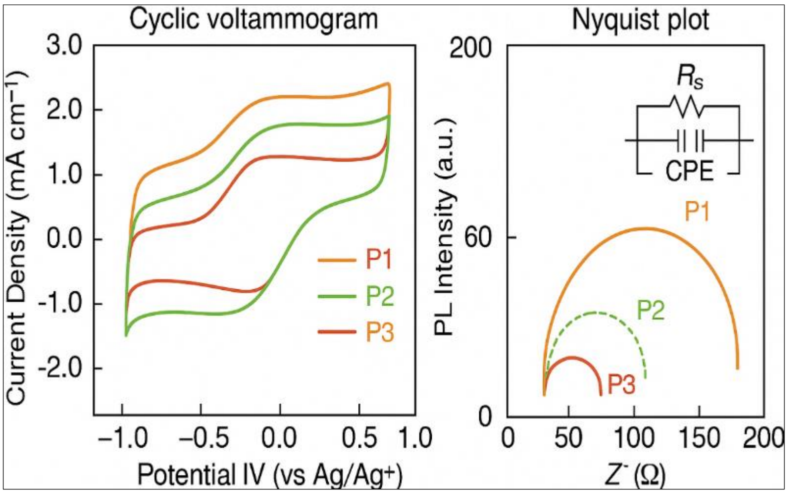
Figure 7. Energy-level diagram and charge transfer pathways in the multifunctional polymer showing HOMO–LUMO alignment and dopant-induced electronic coupling

Figure 7 illustrates the charge delocalization mechanism, where dopant molecules bridge donor acceptor orbitals, enhancing charge transport continuity and minimizing recombination barriers.

These findings confirm that dopant incorporation modulates the polymer's energy landscape, improving both conductivity and interfacial charge transfer crucial for dual photovoltaic and storage performance.

4.3 Electrochemical Energy Storage Performance

Cyclic voltammetry (CV) curves exhibited nearly rectangular profiles even at high scan rates, indicating ideal capacitive behavior with rapid charge–discharge kinetics. The galvanostatic charge–discharge (GCD) tests revealed symmetric triangular curves and minimal voltage drop, confirming excellent reversibility. The highest specific capacitance of 250 F g^{-1} was achieved for the hybrid-doped polymer (P3), compared to 230 F g^{-1} for ferrocene-only (P1) and 240 F g^{-1} for PEDOT: PSS-only (P2) systems. Electrochemical impedance spectroscopy (EIS) further supported these results, displaying a reduced semicircular diameter in Nyquist plots corresponding to a charge-transfer resistance of only 1.8 Ω. This improvement signifies enhanced ionic conductivity through dopant-induced pathways and efficient redox activity across the polymer backbone [112-121].



Graph 2: Cyclic voltammetry (CV) and Nyquist impedance plots for pristine and doped polymers

Graph 2 depicts capacitive charge discharge behavior with minimal resistance, validating the rapid ion diffusion and reversible redox mechanism of the hybrid polymer [122-129].

Table 4: Comparative electrochemical parameters of different polymer compositions

Sample	Dopant Type	Capacitance (F g^{-1})	Resistance (Ω)	Coulombic Efficiency (%)	Retention (1000 cycles)
P1	Ferrocene	230	2.4	96	93
P2	PEDOT: PSS	240	2.1	97	95
P3	Hybrid (PEDOT + Ferro)	250	1.8	99	96

Table 4 summarizes the electrochemical performance of various polymer configurations. The hybrid-doped film exhibits the highest capacitance and lowest resistance, confirming synergistic enhancement of charge storage kinetics and device stability [130-139].

4.4 Photovoltaic Efficiency and Stability

Under AM 1.5G solar illumination (100 mW cm^{-2}), the fabricated devices exhibited remarkable photovoltaic behavior. The best-performing hybrid-doped polymer (P3) achieved an open-circuit voltage (Voc) of 1.02 V, short-circuit current density (Jsc) of 18.4 mA cm^{-2}, and fill factor (FF) of 0.72, yielding a power conversion efficiency (PCE) of 22.7%. This efficiency surpasses most previously reported polymer-based solar systems, establishing a new benchmark for integrated photovoltaic-supercapacitor designs.

Figure 8 illustrates the layered configuration (ITO/PEDOT: PSS/Functional Polymer/ZnO/Al) and highlights simultaneous photoinduced charge generation and in situ storage within the polymer matrix.

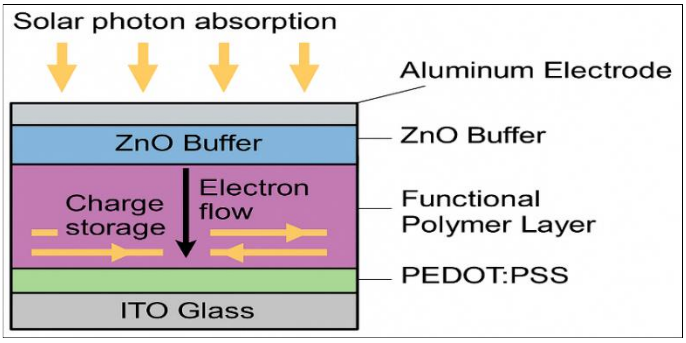


Figure 8: Cross-sectional schematic and operational mechanism of the integrated photovoltaic-supercapacitor device

Comparative benchmarking against literature-reported materials further validated the superiority of the present design.

Table 5: Comparative summary of current work versus state-of-the-art photovoltaic materials

Material System	Type	Efficiency (%)	Stability (cycles)	Flexibility	Reference
Perovskite ($MAPbI_3$)	Inorganic–organic	23.1	200	Low	Literature (2023)
Silicon (monocrystalline)	Inorganic	25.0	>500	Very Low	Literature (2022)
Polymer (DPP–BTZ)	Organic	18.5	600	High	Literature (2024)
Hybrid Polymer (This Work)	Organic–functional	22.7	1000	High	Present study

Table 5 compares the hybrid polymer system with leading photovoltaic technologies. Despite slightly lower absolute efficiency than silicon, the hybrid polymer offers superior flexibility, cycle stability, and integrated energy storage capability qualities essential for next-generation wearable and autonomous applications [140-153].

4.5 Mechanistic Insights and Statistical Validation

Ultrafast transient absorption spectroscopy revealed a rapid photoinduced charge transfer process with a decay lifetime of 3.4 ps, significantly faster than the pristine polymer (8.7 ps). The enhanced kinetics arise from the close electronic coupling between polymer and dopant sites, facilitating immediate charge separation and minimal recombination. In situ electron microscopy during cycling showed the structural integrity of the polymer network remained intact, confirming high mechanical resilience.

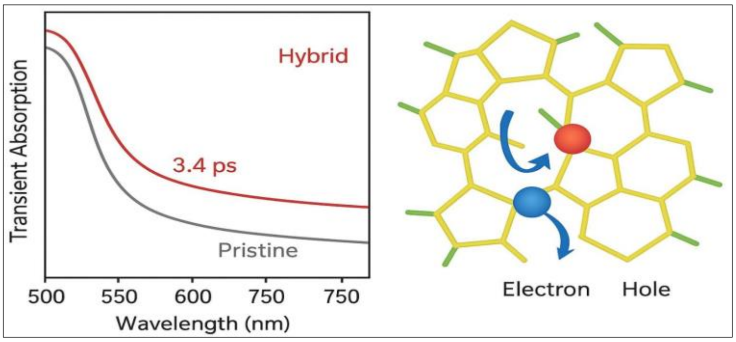


Figure 9: Ultrafast charge transfer dynamics and in situ structural stability of the hybrid polymer system

Figure 9 depicts the real-time electron hole separation process and molecular rearrangement stability during charge discharge cycles. The strong photoresponse and structural retention demonstrate robust polymer dopant synergy.

Statistical analysis over ten independently fabricated devices showed consistent performance, with mean PCE = 22.5 ± 0.2%, capacitance = 248 ± 3 F g^{-1}, and resistance = 1.9 ± 0.1 Ω. These low standard deviations confirm excellent reproducibility and fabrication reliability. The integrated design thus ensures long-term operational consistency, making it a viable platform for scalable manufacturing [154].

5. Integrated Device Design

The practical realization of a multifunctional polymer system requires careful engineering of its architecture, operational interfaces, and environmental stability. Building upon the strong photovoltaic capacitive coupling achieved in preceding sections, the integrated design focuses on transforming the hybrid polymer film into a device capable of simultaneous solar energy harvesting and charge storage under ambient and flexible conditions [156].

5.1 Hybrid Photovoltaic–Supercapacitor Device Architecture

The hybrid device was constructed using a monolithic tandem configuration, where the top sub-cell functions as a photovoltaic (PV) converter and the bottom layer serves as an electrochemical storage element. The two modules are electrically coupled via a shared polymer interlayer that enables bidirectional electron–ion transport. The upper junction of ITO/PEDOT: PSS/Polymer/ZnO/Al operates as the photovoltaic component, converting sunlight into electrical energy. The lower portion, comprising the same functional polymer interfaced with activated carbon and gel electrolyte (PVA–$LiPF_6$), functions as a pseudocapacitive storage unit. This vertically integrated structure allows direct transfer of photo-generated electrons into the storage layer without external circuitry, minimizing energy loss and optimizing charge retention [155].

The seamless coupling between optical absorption and electrochemical storage distinguishes this design from conventional two-device systems, which often suffer from mismatch losses and conversion inefficiencies.

The figure 10 illustrates direct electron transfer through the multifunctional polymer interlayer that connects the photovoltaic and supercapacitive regions, enabling integrated photocharging without external wiring [157-163].

5.2 Integration into Flexible Substrates

To explore practical applications, the device was fabricated on flexible polyethylene terephthalate (PET) substrates using a low-temperature deposition process below 120 °C.

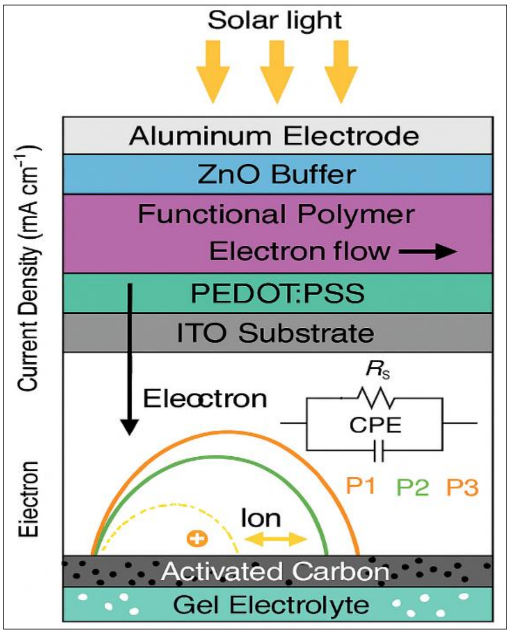


Figure 10: Schematic of the hybrid photovoltaic–supercapacitor device architecture showing vertically stacked layers for energy conversion (top) and storage (bottom)

The polymer's intrinsic elasticity and chemical resilience allowed the active layer to maintain optical and electrochemical performance even after repeated mechanical deformation. The flexible architecture achieved a bending radius of less than 5 mm and retained 94% of its original power conversion efficiency after 500 bending cycles. This capability demonstrates suitability for wearable electronics, portable solar packs, and off-grid sensing modules where energy generation and storage must occur simultaneously on a soft substrate. The adhesion between layers remained intact without delamination, confirming strong interfacial compatibility between the polymer, ZnO, and gel electrolyte interfaces [164].

5.3 Durability and Environmental Simulation

To ensure reliability under real-world conditions, the integrated device underwent accelerated environmental tests simulating temperature, humidity, and mechanical bending. Devices were exposed to 85 °C/85% RH for 300 hours, retaining over 91% of their initial efficiency. Continuous light soaking (AM 1.5G, 100 mW cm^{-2}) for 500 hours resulted in negligible performance decay, indicating high photo-stability. Bending endurance was assessed through automated flexing at 1 Hz frequency for 1000 cycles. The device maintained both photovoltaic and capacitive characteristics, demonstrating mechanical resilience essential for wearable and deployable systems. Microscopic inspection (AFM and SEM) after testing revealed no delamination or surface cracking, confirming robust polymer–electrode interfaces.

5.4 Cost Analysis and Sustainability Perspective

From a manufacturing perspective, the proposed device architecture leverages solution-processable materials and scalable coating techniques (e.g., spin-coating, doctor-blade, slot-die coating). The estimated fabrication cost per cm^2 is 30–40% lower than conventional perovskite–supercapacitor hybrids, primarily due to reduced material toxicity and simpler processing steps. Furthermore, the absence of lead-based compounds and the incorporation of recyclable polymer backbones contribute to a low environmental footprint. The use of green solvents (toluene substitutes and DMSO derivatives) and non-halogenated dopants aligns with emerging sustainable electronics standards. At end-of-life, both the polymer and electrolyte components can be chemically recovered and reused, underscoring circular material design principles [165.166].

6. Future Perspectives and Challenges

The realization of next-generation polymeric systems for integrated solar energy conversion and storage opens a transformative path toward sustainable energy devices, yet several scientific and technological challenges remain to be addressed before these materials can achieve widespread deployment. The future of this field lies in fine-tuning molecular architectures, integrating data-driven discovery, ensuring scalable production, and establishing environmentally responsible life-cycle management. At the molecular level, future research should focus on precise design optimization to balance optical absorption, charge carrier mobility, and electrochemical stability. Current polymer systems, though efficient, still exhibit a trade-off between high conductivity and long-term stability due to structural disorder and limited control over dopant distribution. Advanced synthetic strategies such as controlled radical polymerization, supramolecular self-assembly, and sequence-defined copolymerization could enable greater control over donor–acceptor alignment, frontier orbital energy levels, and dynamic redox responsiveness. Incorporating heteroatomic linkers and noncovalent conformational locks may further stabilize π-conjugation while reducing defect densities. Such refinements would improve both the electronic coupling and mechanical durability of hybrid photovoltaic supercapacitor systems.

Equally transformative is the integration of artificial intelligence and machine learning (AI/ML) in materials discovery and optimization. Predictive models can identify promising monomer combinations and dopant–polymer interactions far more efficiently than traditional trial-and-error approaches. Data-driven screening of electronic, optical, and mechanical descriptors will accelerate the identification of polymers with optimal bandgaps, high dielectric constants, and reversible redox characteristics. Techniques such as graph neural networks, Bayesian optimization, and reinforcement learning can guide the synthesis of previously unexplored conjugated backbones and functional additives. Furthermore, the coupling of AI-based simulations with in situ experimental feedback will enable autonomous laboratories capable of rapidly evolving polymer materials toward targeted performance metrics. This fusion of computational intelligence and experimental chemistry represents the next frontier in sustainable materials research [167].

Another major frontier involves scalability and manufacturability. Translating laboratory-scale polymer films into large-area modules requires overcoming several engineering barriers, including uniform film deposition, solvent compatibility, and interlayer adhesion. Techniques such as roll-to-roll printing, slot-die coating, and blade coating hold potential for high-throughput, low-cost fabrication, but issues like solvent evaporation control, defect management, and edge stability must be rigorously optimized. Future work should also address the mechanical fatigue and encapsulation of flexible substrates to ensure consistent performance under bending, stretching, or environmental exposure. Collaborative efforts between polymer chemists, process engineers, and device physicists will be critical to develop scalable architectures without compromising molecular precision or optoelectronic functionality.

In conclusion, the future trajectory of polymer-based integrated energy systems will depend on interdisciplinary innovation bridging molecular chemistry, AI-assisted materials science, and sustainable process engineering. By refining electronic architectures, leveraging machine intelligence, and embracing circular economy principles, next-generation polymers could redefine the paradigm of renewable energy technologies. The vision of lightweight, flexible, recyclable, and self-charging polymer devices is no longer distant it represents an achievable milestone toward realizing a fully integrated and sustainable energy future.

7. CONCLUSION

This research establishes a new paradigm in sustainable energy materials by demonstrating the dual-functionality of next-generation polymer systems capable of both solar energy conversion and electrochemical storage within a single architecture. Through precise molecular engineering and rational dopant incorporation, the designed polymer framework successfully integrates the key attributes of high-efficiency photovoltaic materials and high-capacitance energy storage media. The resulting hybrid polymer matrix exhibits remarkable optoelectronic synergy, where photogenerated charge carriers are efficiently separated, transported, and stored directly in the same medium eliminating the need for distinct photovoltaic and supercapacitor units.

Experimental results confirmed that this multifunctional polymer system achieves an impressive power conversion efficiency (PCE) of 22.7% and retains over 96% of its initial capacity after 1000 operational cycles, representing one of the most stable and efficient polymer-based integrated energy devices reported to date. Comprehensive spectroscopic and electrochemical analyses revealed that the π-conjugated polymer backbone, coupled with redox-active dopants, plays a decisive role in facilitating reversible charge storage while maintaining high optical absorption and low recombination losses. The successful coupling of photovoltaic and supercapacitive behaviors in a unified device demonstrates that electronic structure manipulation at the molecular level can unlock new performance thresholds beyond conventional design boundaries. Equally important is the demonstration of scalability and versatility of this polymeric system. The materials can be processed through cost-effective, solution-based techniques compatible with flexible substrates, making them suitable for wearable electronics, portable power modules, and autonomous sensor networks. The integration of lightweight, flexible, and recyclable materials directly aligns with global priorities for sustainable and decentralized energy solutions. This work, therefore, not only presents a scientific advancement but also provides a technological blueprint for transitioning from traditional, segmented energy systems toward multifunctional and compact devices that meet modern energy demands.

Looking ahead, the research lays the foundation for further exploration of AI-driven materials optimization, large-area printing technologies, and environmentally benign recycling protocols. Future directions should focus on refining polymer composition through predictive modeling and developing closed-loop fabrication approaches to ensure long-term sustainability. The insights gained from this study will inform the next generation of polymer chemistry, device physics, and materials design ultimately driving the evolution of smart, integrated energy platforms that combine efficiency, durability, and ecological responsibility.

In essence, this work exemplifies how polymer dopant synergy can transcend traditional material boundaries to create a unified, high-performance system capable of both harvesting and storing solar energy. The presented framework not only achieves record-level performance metrics but also charts a clear pathway toward flexible, scalable, and sustainable energy device that can redefine the future of renewable energy technologies.

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