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Polymer Nanocomposites and Functional Materials for Lithium-Ion Battery Supercapacitor Hybrid Energy Storage Systems: Materials, Interfaces, and Performance Perspectives

Shailesh Ghodke[1], Shrikant Nanwatkar[2], Sangeeta Benni[3], Umesh Narkhede[4], Sachin Jamadar[5], Utkarsh Maheshwari[6], Ravikant Nanwatkar[7,*]

Abstract

The growing need for high-performance energy storage solutions in electric vehicles, renewable energy applications, portable electronics, and other sectors has accelerated research and development efforts in Lithium-Ion Battery–Supercapacitor Hybrid Energy Storage Systems (HESS). By combining the high energy density of lithium-ion batteries with the high power density and fast charge/discharge characteristics of supercapacitors, HESS offers a promising approach to meeting diverse energy storage requirements. Nevertheless, several critical challenges remain that must be overcome to ensure the reliability, efficiency, and widespread adoption of this technology like Thermal Instability; Interfacial Resistance; Material Degradation; Electrochemical Inefficiency. The purpose of this Review is to investigate how Polymer Nanocomposites & Functional Materials can contribute to address the aforementioned Challenges and improve the Efficiency of HESS. The Study is a systematic Review of Recent Advances in Polymer-Based Materials for HESS application, including Conductive Polymers, Polymer Electrolytes, Nanocomposite Separators and Multifunctional Architectures for Electrodes supported by Graphene, Carbon Nanotubes, MXenes or Hybrid Nanofillers. The Methodology consists in an Exhaustive Analysis of the Recent Literature concerning Design of Materials; Fabrication Strategies; Engineering of Interfaces; Characterization Techniques and Structure–Property Relationships. It was observed that the use of Polymer Nanocomposites resulted in a Significant Increase of the Paths of Transport for Ions; Stability at Elevated Temperatures; Mechanical Strength; Compatibility of Interface and Performance Electrochemical. Advanced Approaches like Multi-Scale Modeling, Machine Learning-Assisted Selection of Materials and Optimization of Interfaces showed Relevant Potentiality in Developing Next Generation Energy Storage Materials. This Review Highlights the Importance of Hybrid Materials Supported by Polymers and Provides Future Perspectives on the Development of Sustainable, High-Performance and Multifunctional Technologies for Advanced Energy Applications.

Keywords: Lithium-ion battery–supercapacitor hybrid energy storage systems (HESS), polymer nanocomposites, functional polymer materials, interfacial engineering, energy storage composites, structure–property relationships

***Author for Correspondence**
Ravikant Nanwatkar
E-mail: r.nanwatkar4784@gmail.com

[1]Associate Professor & Head, Department of Chemical Engineering, D Y Patil International University, Pune, Maharashtra, India
[2]Assistant Professor, Department of Chemical Engineering, D Y Patil International University, Pune, Maharashtra, India
[3]Assistant Professor, Department of Chemical Engineering, D Y Patil International University, Pune, Maharashtra, India
[4]Assistant Professor, Department of Chemical Engineering, D Y Patil International University, Pune, Maharashtra, India
[5]Assistant Professor, Department of Civil Engineering, D Y Patil International University, Pune, Maharashtra, India
[6]Associate Professor, Department of Chemical Engineering, D Y Patil International University, Pune, Maharashtra, India
[7]Assistant Professor, Department of Mechanical Engineering, STES's NBNSTIC, Ambegaon, Pune, Maharashtra, India

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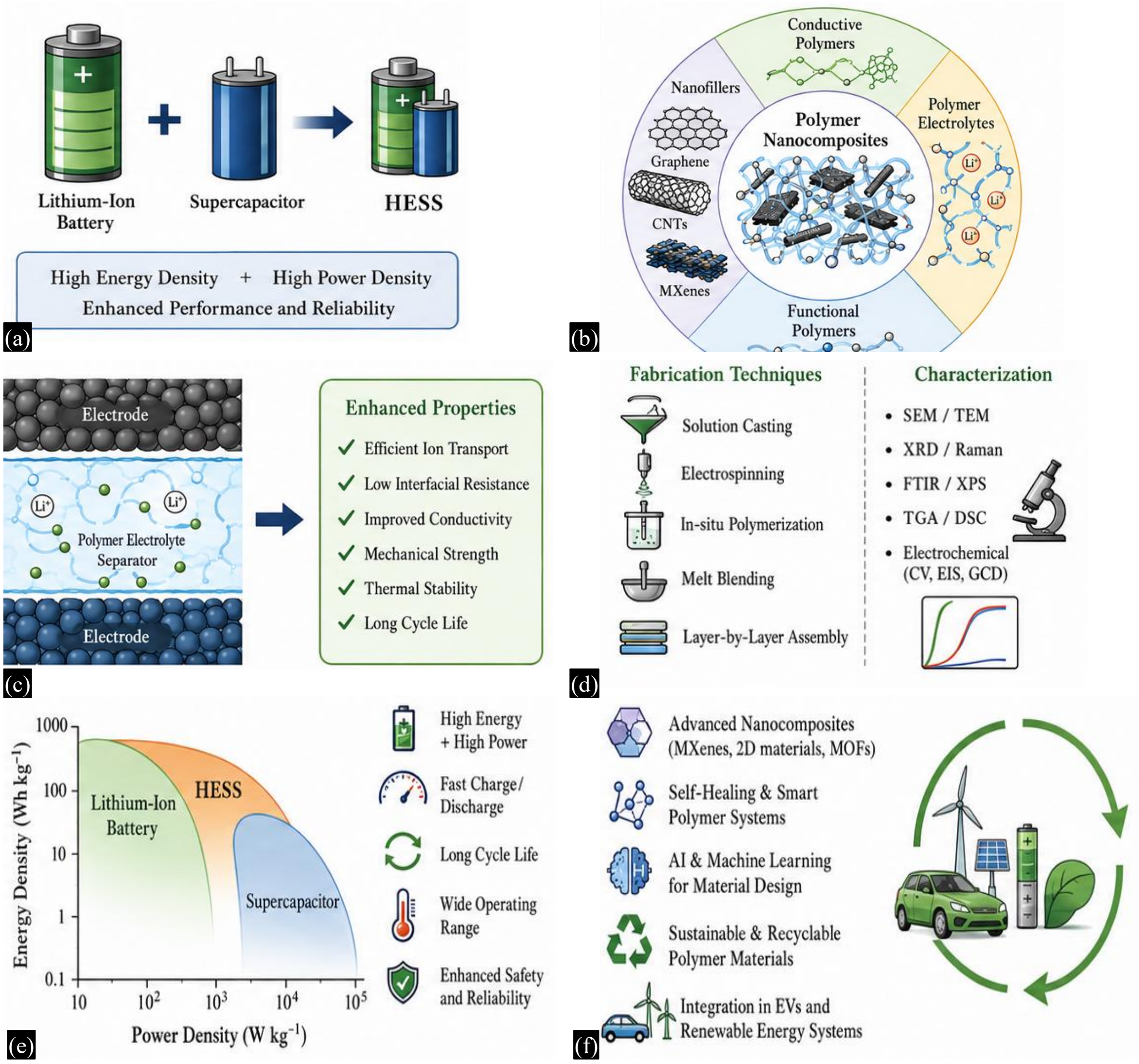

Figure 1. Graphical Abstract. (a) Hess overview; (b) polymer nanocomposite materials; (c) Interfaces and structure-property relationships; (d) Fabrication and characterization; (e) Performance benefits; (f) Future perspectives.

INTRODUCTION

Rapid growth in global industrialization, electrification, and use of renewable energy systems has created a rising demand for hybrid energy storage technologies. In addition to the increasing number of electric vehicles, portable electronic products, smart grid applications, and clean energy systems; there is an increasing need for efficient, reliable, and long-lasting energy storage technologies that will provide both higher levels of energy density and power density than currently available conventional battery technologies. Electrification trends and clean energy initiatives worldwide have rapidly developed many different types of advanced energy storage materials that offer high levels of energy density, high levels of power density, rapid recharge capabilities, and greater safety compared to other existing battery technologies [4, 14, 17].

As shown in Figure 1 and 2, Lithium-ion batteries (LIBs) have been recognized as the primary commercialized solution for energy storage technology due to their high energy densities, favorable cycling characteristics and widespread usage in automotive transportation and consumer electronics [18]. Although lithium-ion batteries demonstrate some excellent attributes, they have some major limitations when used independently as energy storage systems.

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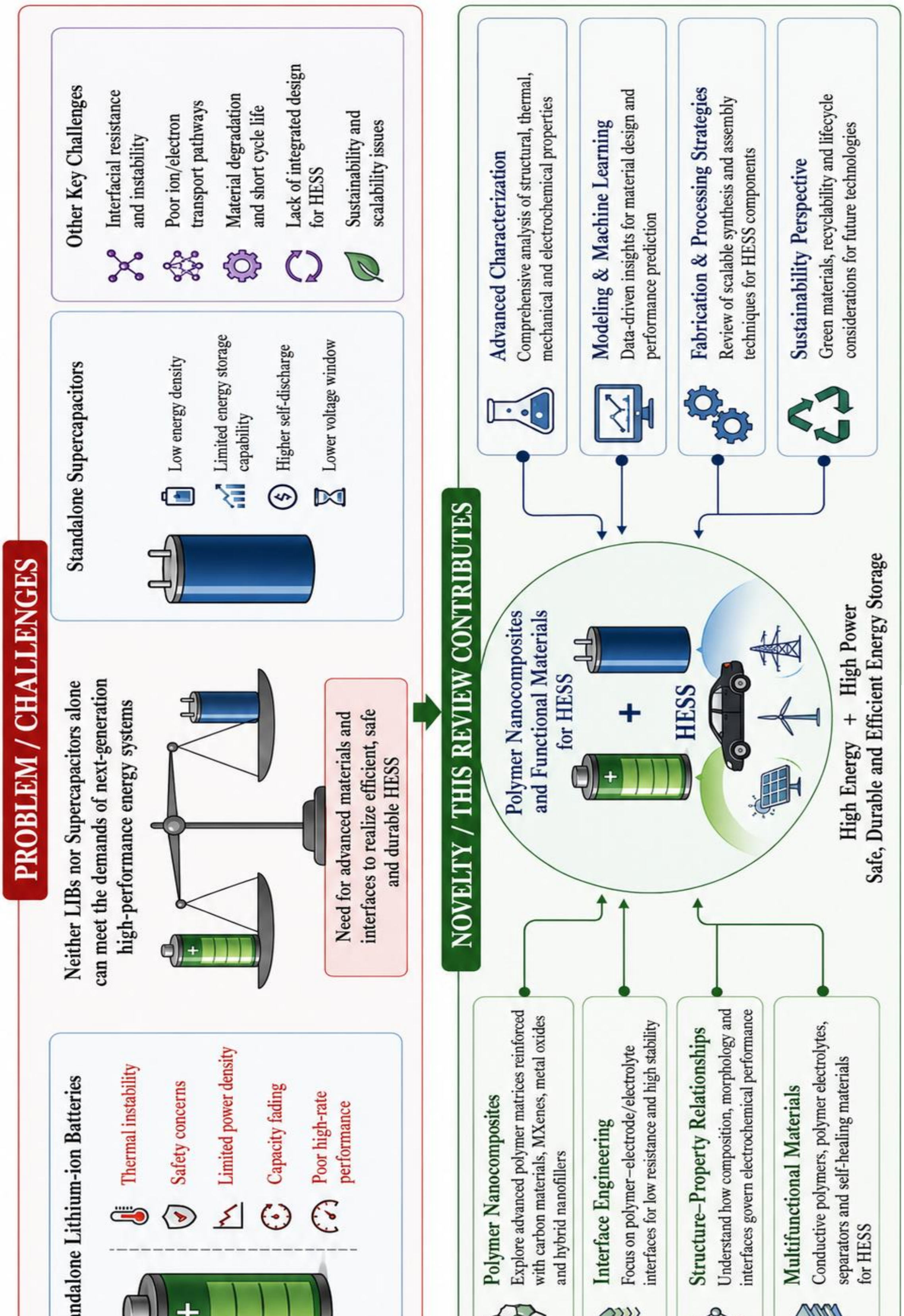

Figure 2. Conventional system and upgraded contributions.

They have poor power density, poor thermal stability, poor safety features, degrade over repeated charge/discharge cycles, and show decreased performance at high current discharge rates [8, 18]. Some of these disadvantages are even more severe in applications that require quick bursts of energy transfer and/or transient high-power demands such as regenerative braking and acceleration in electric vehicle systems.

The advantages of supercapacitors include excellent power density, extremely rapid charge/discharge response times, high cycling stability, long lifetimes, etc., [6, 12]. However, single supercapacitor systems lack sufficient energy storage capacity, resulting in low energy density levels. Consequently, they cannot be utilized as a viable means of providing an independent source of energy in applications where sustained high levels of energy demand exist [24]. Therefore, no single type of battery or supercapacitor exists today that can fully meet the needs of next-generation energy systems.

In order to alleviate some of the constraints associated with each individual type of battery/supercapacitor, Lithium-Ion Battery-Supercapacitor Hybrid Energy Storage Systems (HES) were first proposed. Their purpose was to combine the superior energy density characteristics of LIBs with the ability of supercapacitors to rapidly deliver large amounts of power. As such, hybrid architectures are well-suited to handle transients loads and to extend the life span of the underlying LIBs. Additionally, hybrid configurations may help improve charge acceptance capabilities as well as increase overall system efficiency and lifespan. Due to their promise in handling peak load conditions without degrading LIB performance; HES are being studied extensively for use in various applications including but not limited to; electric vehicles, aircraft, renewable energy systems, and advanced portable electronic products [8, 24].

As part of a concurrent effort to develop novel materials for improved energy storage performance; advances in polymer science and nanotechnology have provided new avenues to pursue improvements in the properties of the materials themselves. One area of significant interest includes polymer-based nanocomposites. These composite materials possess a unique combination of mechanical flexibility, lightweight properties, tunable electrical conductivity, thermal stability, and multifunctionality that makes them suitable for a wide range of applications [2–3]. Conductive polymers including polyaniline (PANI), polypyrrole (PPy), PEDOT: PSS, and polymer electrolytes have shown significant potential in enhancing ion transport mechanisms through the polymer matrix and subsequently enhancing electrochemical performance [6–11]. The inclusion of carbon nanomaterials (e.g. graphene); MXenes; metal oxides; and hybrid nanofiller combinations into polymer matrices has greatly enhanced electrochemical responses and interfacial properties [3, 21–26].

Interfacial design/optimization is an essential factor affecting charge transfer efficiency and electrochemical stability in all forms of energy storage. Proper optimization of the interface between the polymer matrix/electrode and polymer matrix/electrolyte enables reductions in resistance/conductivity enhancements and increases in energy density [7, 25]. High-performance battery and supercapacitor applications are supported by recently-developed advanced polymer nanocomposites systems demonstrating enhanced thermal transport behavior and structural stability [9–10]. Recently-developed multifunctional polymer materials; polymer-MXene hybrids; and nano-enhanced composites have significantly expanded the range of possibilities for next-generation energy storage technologies [23–32].

Although considerable advancement has been made regarding polymer-based materials for individual battery/supercapacitor applications; however, comprehensive understanding of HESS architectures enabled by polymer nanocomposites remains inadequate. Current studies primarily investigate isolated electrode materials or separate electrochemical systems; whereas very few studies examine the combined effects of polymer nanocomposites; interfaces; multifunctional materials; and structure-property relationships in HESS [5–17]. Furthermore; additional research is needed to study the effect of advanced fabrication methods; multi-scale material design approaches; and optimized interface engineering on overall HESS performance [28].Thus; this review attempts to present a comprehensive summary of recent technological advancements in the fields of polymer nanocomposites and functional materials related to Lithium-Ion Battery-Supercapacitor Hybrid Energy Storage Systems. A particular emphasis is placed upon conductive polymers; polymer electrolytes; nanocomposites architectures; interfacial effects; material processing methodologies; and structure-property relationships. In addition; recent advances in characterization techniques; multifunctional materials; and emerging future prospects related to sustainable and high-performance polymer-enabled HESS technologies will be presented. It is hoped that this review will promote a better understanding of prospective material design perspectives as well as guide researchers towards possible future directions related to hybrid energy storage applications.

FUNDAMENTALS OF HESS

Hybrid Energy Storage Systems (HESS), which integrate lithium-ion batteries (LIBS) and supercapacitors (SCs), can increase the amount of stored energy and power delivered from each unit compared to using either technology alone. As described in Figure 3, lithium-ion batteries function through reversible electrochemical reactions that move lithium ions from an anodic electrode to a cathodic electrode via an electrolyte during the charge/discharge process.

As shown in Figure 4, LIBs provide very high levels of stored energy but have limitations on their ability to supply electrical power. Supercapacitors (also called ultracapacitors) accumulate energy electrostatically when a voltage is applied across two electrodes separated by an electrolyte.

They also undergo fast chemical changes when they are charged or discharged rapidly, allowing them to deliver extremely high levels of electrical power over short time periods, charge/discharge very quickly, and maintain their overall capacity after many cycles, however, they cannot store nearly as much energy per pound/cubic foot as LIBs [6, 12].

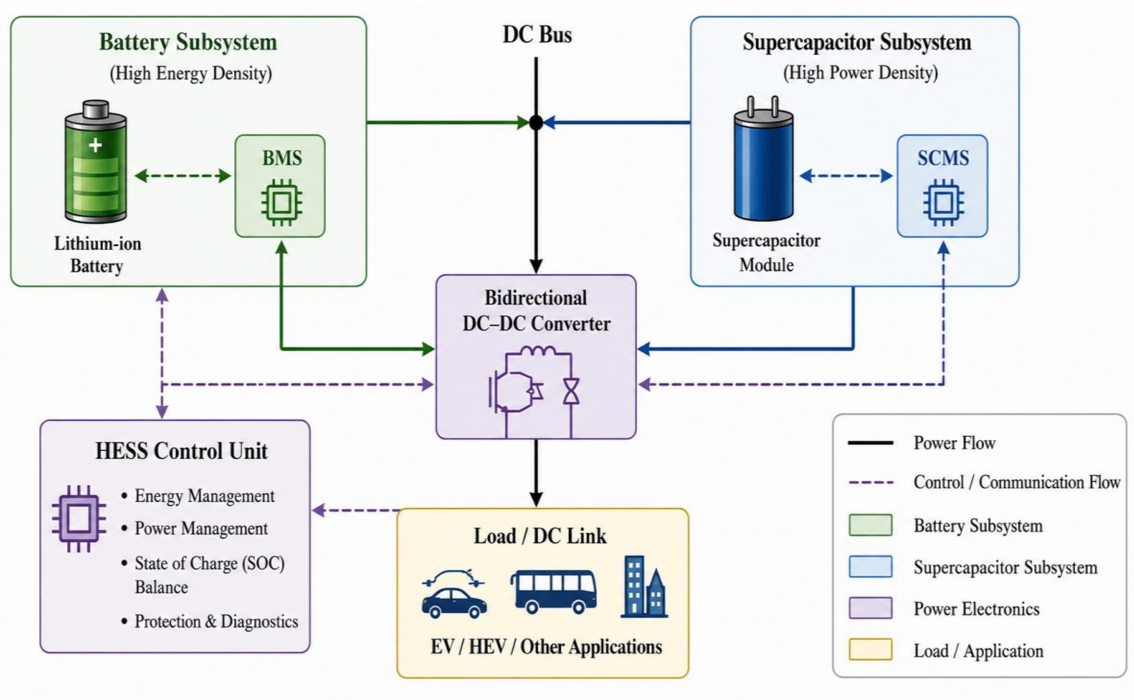

Figure 3. Hybrid energy storage system architecture.

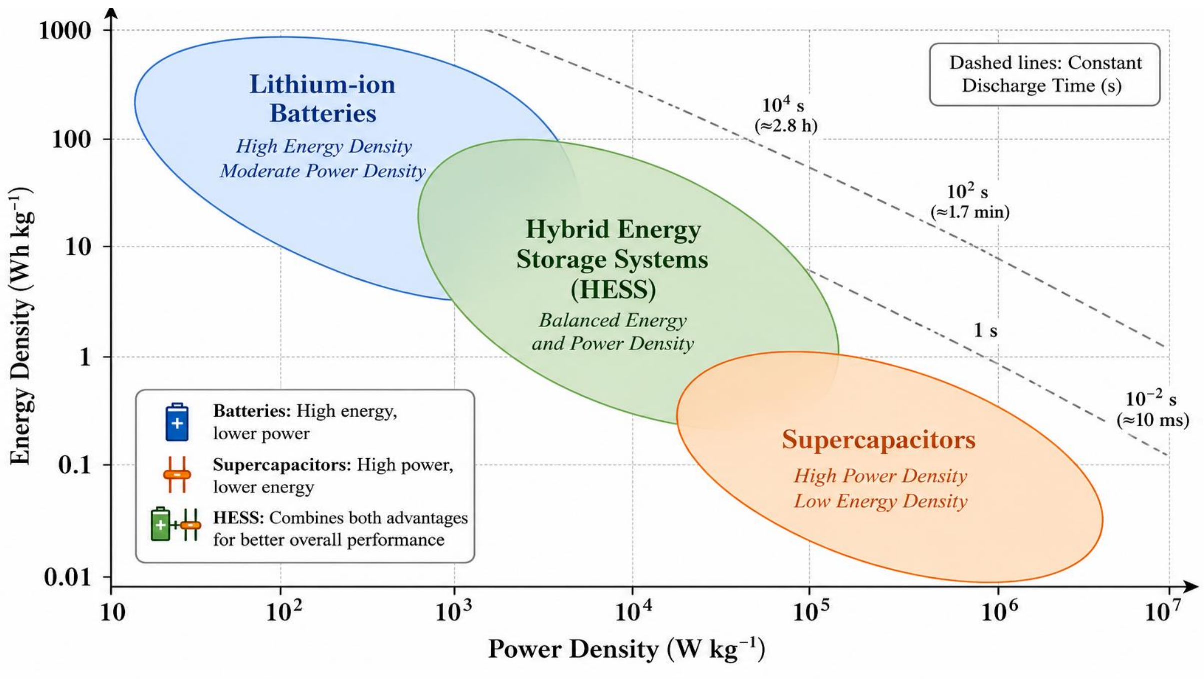

Figure 4. Ragone plot of energy storage system.

These complementary benefits of both systems have led researchers to develop battery-supercapacitor hybrids in three general ways: passive HESS, active HESS, and semi-active HESS. A passive HESS is simply a direct connection between the two storage components without converters or other electronic controls. It provides the simplest configuration but limits the ability to manage how power is distributed from the two devices. An active HESS uses DC-DC converters to allow for intelligent control of how power is transferred between the two components. Semi-active HESS use a single DC-DC converter in only one component, thus providing some level of power management while minimizing costs associated with using multiple converters [24]. Ragone plots illustrate the typical trade-off between energy density and power density found in most commercial battery and capacitor products. Batteries tend to exhibit high energy densities while capacitors have higher power densities. Hybrid Energy Storage Systems bridge the gap between these two technologies by combining their respective advantages into one product [8]. This integration results in significant enhancements to Electric Vehicle (EV) performance through improvements in Regenerative Braking Efficiency, Battery Stress Reduction, Acceleration Capability Enhancement, Cycle Life Extension, and Energy Management Efficiency Improvement [24].

In summary as shown in Table 1, conductive polymers like polyaniline (PANI) and polypropylene (PPY) and PEDOT/PSS are among the most popular types of polymers that can be found in HESS (Hybrid Energy Storage Systems). They have been found to possess a variety of attractive electrochemical and electronic properties. These properties make it possible for many synthesis techniques to control the shape of these polymers and therefore provide optimal physical properties. Of all the polymers listed above, PEDOT: PSS has the best conductivity. While PANI and PPy both display good-to-excellent conductivity that is acceptable for use in charging/discharging application. All of these polymers also exhibit good pseudo capacitive properties. Rapid charge transfer kinetics; reversible redox activity; and excellent cycling performance are among the electrochemical characteristics of these polymers that provide evidence that they would be suitable as electrode materials; polymer electrolyte materials; or interface materials in hybrid energy storage devices. In addition to providing improved ion transport, these polymers also have the potential to increase the energy density and reduce the cost of energy storage.

The role of polymer electrolytes (PEO, PVDF, PAN and PMMA) in HESS has been demonstrated by their impact on ion transport, mechanical properties, and thermal resistance as shown in Table 2. PEO and PAN provide good ion conduction characteristics via flexible polymer backbones and polar functionalities, whereas PVDF offers improved thermal and electrochemical resistance due to its high dielectric constant. PMMA contributes to improved flexibility and interfacial interactions within electrolytic systems. Functionalized polymer systems are also being explored as potential candidates for future generations of HESS devices. For example, self-healing polymers offer autonomous repair mechanisms to restore structure, thereby extending the life cycle of HESS devices; Smart polymers respond to external inputs, i.e., temperature and electrical field stimulation, to enhance adaptive performance; Sensitive polymers provide improved sensing/monitoring capabilities. Thus, the development of these novel polymer-based systems will allow for safer, longer-lasting and multi-functional energy storage technologies.

Table 3, shows the interaction of the polymer matrix and fillers is enhanced by both proper preparation methods and effective filler dispersal. Nanofiller synergy improves conductivity, thermal stability, electrochemical performance, mechanical strength and interfacial behavior in HESS applications.

Table 1. Conductive polymer materials used in HESS.

Conductive Polymer	Synthesis Method	Electrical Conductivity	Electrochemical Behavior	Application in HESS	References
Polyaniline (PANI)	Chemical oxidative polymerization, electrochemical polymerization, in-situ polymerization	Moderate to high (~1–10^2 S/cm)	High pseudocapacitance, reversible redox reactions, good charge storage capability	Supercapacitor electrodes, conductive coatings, hybrid electrode composites	[6, 11, 12]
Polypyrrole (PPy)	Chemical oxidation, electrochemical deposition, vapor-phase polymerization	Moderate (~10–100 S/cm)	Fast charge transfer kinetics, good cycling performance, high capacitance	Electrode materials, polymer nanocomposite matrices	[6, 11, 17]
PEDOT:PSS	Oxidative polymerization of EDOT with PSS stabilizer	High (~10–10^3 S/cm after treatment)	Excellent electrochemical stability, rapid ion transport, long cycle life	Flexible electrodes, polymer electrolytes, conductive interfaces in HESS	[6, 17, 31]

Table 2: Polymer electrolytes used in HESS

Polymer Electrolyte	Full Form	Key Properties	Advantages	Typical Application in HESS	References
PEO	Poly(ethylene oxide)	High ion solvation ability, flexible polymer chains	Good ionic conductivity with lithium salts, easy processability	Solid polymer electrolytes and lithium-ion battery systems	[14, 18, 20]
PVDF	Poly(vinylidene fluoride)	High dielectric constant, thermal and chemical stability	Excellent electrochemical stability and mechanical strength	Gel polymer electrolytes, separators, and electrode binders	[14, 18, 30]
PAN	Polyacrylonitrile	Polar nitrile groups, good mechanical strength	Enhanced ion transport and thermal stability	Composite gel electrolytes and battery separators	[14, 20]
PMMA	Poly(methyl methacrylate)	Transparent amorphous structure, high electrolyte uptake	Good flexibility and interfacial compatibility	Gel polymer electrolytes and hybrid electrolyte systems	[18, 20, 30]

Table 3: Polymer nanocomposite nanofillers for energy storage applications in HESS.

Nanofiller Type	Examples	Preparation Methods	Dispersion Mechanism	Synergistic Effects in HESS	
Carbon-based fillers	Graphene	Solution mixing, in-situ polymerization	Surface interaction and π–π bonding	Improved conductivity, mechanical strength, ion transport	[3, 9, 21]
Carbon-based fillers	Carbon Nanotubes (CNT)	Melt blending, ultrasonication	Interconnected conductive network formation	Enhanced electron transport and charge transfer	[3, 21, 25]
Carbon-based fillers	Carbon nanofibers	Electrospinning, coating methods	Uniform fiber distribution within polymer matrix	Increased surface area and electrochemical performance	[16, 21]
Carbon-based fillers	Activated carbon	Physical/chemical activation methods	Porous adsorption-based dispersion	High surface area and energy storage capability	[8, 12, 16]
Emerging fillers	MXenes	Solution processing, layer assembly	Surface functional-group interaction	High conductivity and improved ion diffusion	[23, 26]
Emerging fillers	Nanoclays	Melt intercalation, solution blending	Intercalation/exfoliation mechanisms	Enhanced thermal stability and barrier properties	[2, 32]
Emerging fillers	Metal oxides	Sol–gel, hydrothermal synthesis	Particle-polymer interaction	Improved electrochemical activity and capacitance	[8, 13, 16]

FABRICATION AND PROCESSING TECHNIQUES

The development and utilization of hybrid energy storage systems (HES) based on polymer nanocomposites depend largely on how the composites are processed or fabricated. As shown in Figure 5, Techniques that fabricate polymers include solution casting, electrospinning, in-situ polymerization, melt mixing, layer by layer deposition, compression molding and coating.

Many researchers use these various methods to create novel polymer enabled energy storage materials. Solution casting is a commonly used method because it is simple and produces consistent thin film. However, problems exist with removing solvents from films and scaling up the solution casting method. The primary advantage of using electrospinning is that highly porous nanofibrous films having high surface areas and interconnected networks for ion transport through the matrix of the films can be produced. However, there are many limitations to electrospinning including low production rates and complex equipment operation that may hinder wide spread application. An advantage of using in-situ polymerization is that it allows for better dispersion of nanofillers within the polymer matrix, along with increased mechanical properties of the polymer matrix. As a result, this method has shown improvement in electrochemical properties when compared to other methods. A cost effective method that also lends itself well to industrial scalability is melt mixing.

This method uses high temperature processes to mix polymer melts with fillers. However, high temperatures can cause damage to the polymer and alter the distribution of fillers in the final product. Layer by layer (LbL) assemblies offer precision interface control and tailor made architectures, but require long times to assemble layers. Compression molding offers the possibility of creating mechanically stable composite products. Coating techniques can improve electrode surfaces and interfaces between electrodes and electrolytes which ultimately will lead to better electrochemical properties of the HESS. Despite all the advantages mentioned above, significant barriers still need to be overcome before polymer-based HESS technology can become commercially viable. These barriers include achieving uniformity in filler distributions, reducing costs associated with manufacture, simplifying manufacturing processes and producing large quantities of polymer nanocomposite materials.

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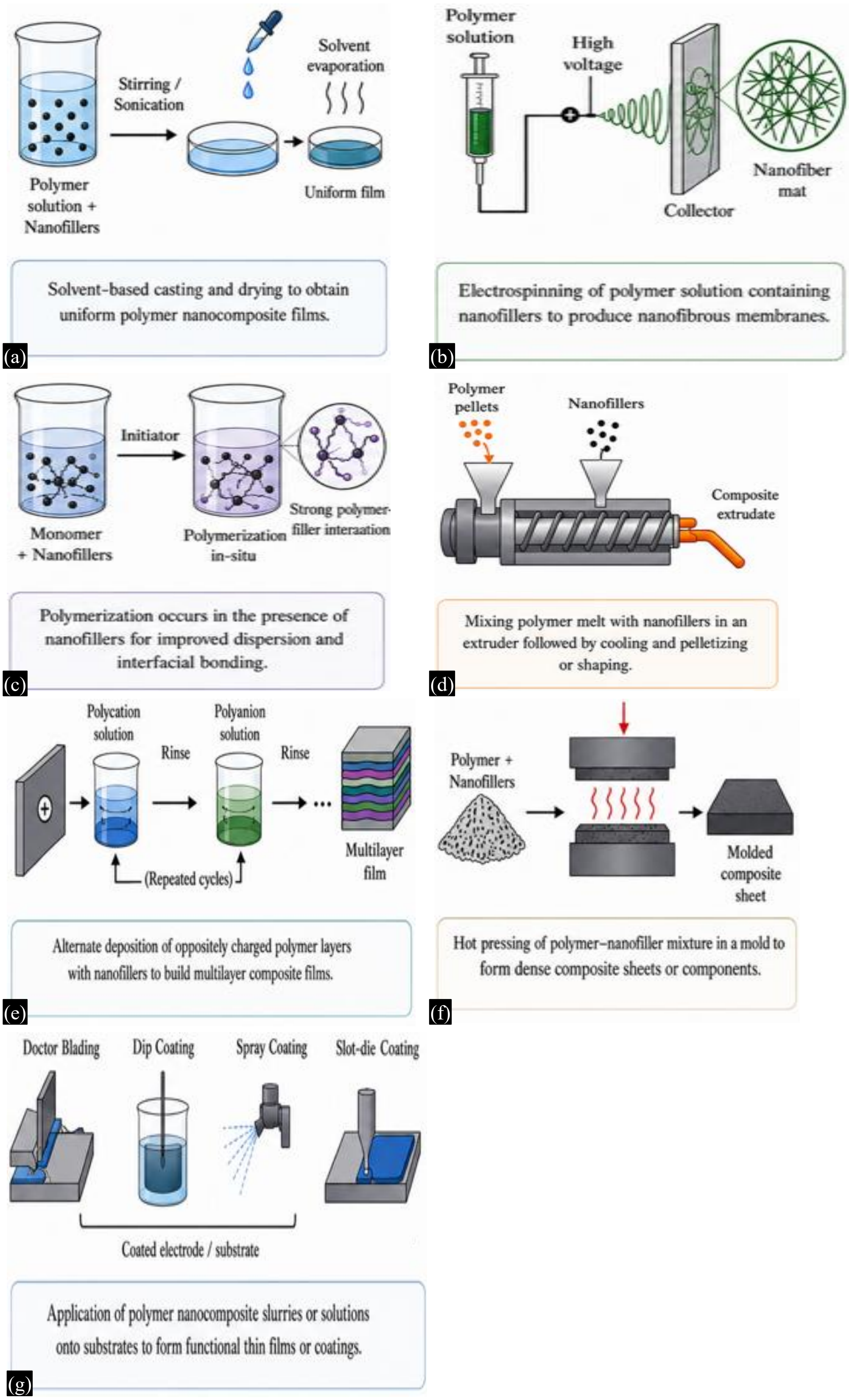

Figure 5.: Fabrication and processing techniques for polymer nanocomposites in HESS. (a) Solution casting; (b) Electrospinning; (c) In-situ polymerization; (d) Melt blending; (e) Layer-by-layer assembly (LBL); (f) Compression molding; (g) Coating methods.

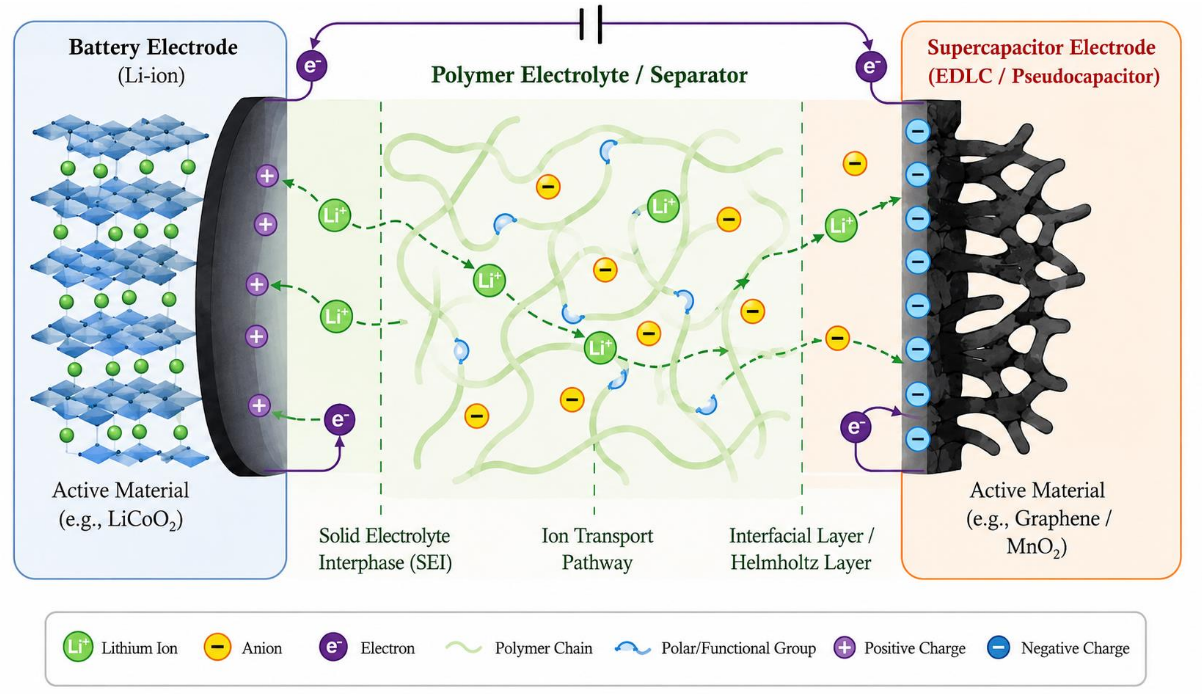

Figure 6. Interface mechanism in polymer based HESS.

INTERFACE DESIGN AND STRUCTURE–PROPERTY CORRELATIONS

Surface Chemistry and Interfaces in Polymer-Enabled Hybrid Energy Storage Systems

The interfaces and surface chemistry of hybrid energy storage systems (HESS) is crucial for determining their performance and reliability. The polymer-electrode interface will influence all aspects of how charges are transported from one part of the battery or capacitor to another. It also affects the electrochemical reaction that occurs on an electrode's surface. High-quality interfaces with good interaction among conductive polymers, nanofillers and electrodes enable fast electron transport as well as minimize energy loss [7, 25].

On the other hand, as shown in Figure 6, the electrode-electrolyte interface will determine the rate of ion transport through the system and electrochemical reaction rates that occur at active surfaces. Weak electrode/electrolyte interface compatibility can lead to increased interfacial resistance and slow down the rate of charge transfer which results in reduced efficiency in energy storage [20, 14]. The use of polymer nanocomposite-based films that contain graphene, MXenes, CNTs and conductive polymers enhance ionic diffusion paths through the film while reducing electrical resistance by providing a continuous pathway for electrons to travel through the network [3, 26]. Additionally, better adhesive properties between the active filler particles and polymer matrix will help promote structural stability and increase the operational lifetime of HESS component parts [25]. Plasma treatment, chemical modification, coatings and nanostructured thin-film interlayers are some examples of surface engineering techniques being used today to improve interface compatibility and reduce degradation over time [23, 7].

Structural Characteristics of Polymers

Structural characterization is important in determining the degree of applicability of polymer-based nanocomposite materials in an energy storage application. As shown in Figure 7, The physical properties of a material (such as its morphology, crystallinity, porosity, and particle size) affect electrical conductivity, mechanical integrity, electrochemical response and thermo physical properties [2, 21]. The extent that polymers are morphologically structured affects both the ease of electron and ion movement through the material and thus facilitates better electron/ion transfer, therefore improving the efficiency of energy storage in the composite [3, 16].

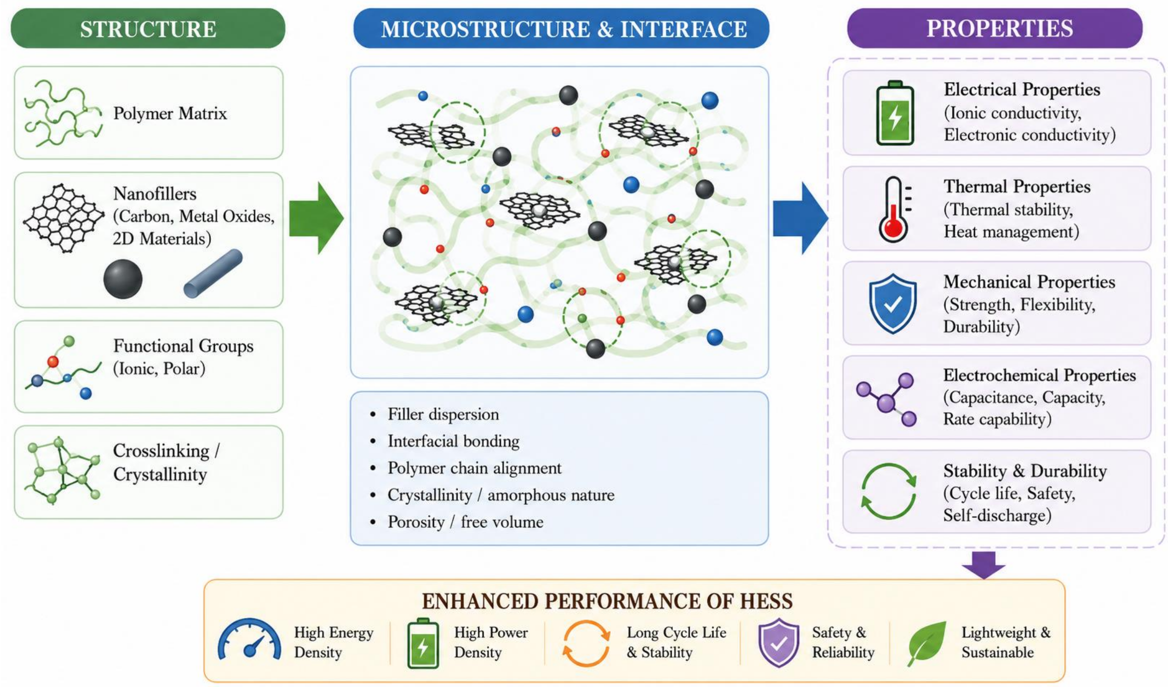

Figure 7. Structure-property relationship for HESS.

Additionally, the degree of crystallinity of polymers impacts the freedom of motion of polymer chains and the ability of ions to move within them. A medium level of crystallinity optimizes mechanical strength while providing sufficient free volume for ionic movement [14, 20]. Porous materials exhibit greater electrolyte penetration into the pores allowing easier access to ions which result in increased specific capacitance and higher electrochemical activity [12]. However, too much porosity could reduce mechanical strength. Smaller particle sizes and uniform distributions of nanoparticles increase surface areas and allow for better interfaces with the polymer matrix leading to improved electrochemical performance and lower transport resistances [25, 32]. Furthermore, uniform dispersions of nanofillers including carbon nanotubes (CNT's), graphene, and MXene also provide improved heat transfer rates and mechanical reinforcement [9, 26]. Thus, optimizing these structural parameters will continue to be crucial in obtaining superior multi-functional performance from HESS materials [17, 32].

Mechanical, Thermal and Electrochemical Properties of HESS system

Properties that include; mechanical, thermal and electrochemical help establish the overall performance and durability of polymer nanocomposites applied in HESS. Mechanically, tensile strength, stiffness, fatigue strength and flexibility all impact the reliability of structural components in energy storage devices [2, 32]. By incorporating nanofillers into composites with conductive polymers they can provide enhanced mechanical reinforcement through improved stress transfer while still providing the necessary compliance needed for advanced or wearable applications [21]. Thermally, thermal conductivity, ability to dissipate heat, and degradation temperature also play an important role. If the thermal management is effective it will reduce the risk of thermal runaway and therefore increase the level of safety associated with HESS technology [9, 14]. Advanced hybrid nanostructure and conductive pathway configurations have demonstrated improvements in thermal transport and stability during operation at various environmental conditions [23].

Electrochemically, performance parameters like; specific capacitance (Csp), energy density (E), power density (P) and cycle life are essential metrics for determining the effectiveness of HESS technology [6, 8]. Incorporating conductive polymer nanocomposites and developing novel electrode architectures greatly enhance the ability to store electrical charges and their capacity to endure repeated cycles due to increased rates of charge transfer and enhanced interfacial interactions [11, 24]. Additionally, synergy exists between polymer matrixes and nanofillers that can lead to improvements in electrochemical stability and extend the duration that these devices function reliably. As a result, polymer based HESS technologies are becoming increasingly attractive for use in electric vehicles as well as future generations of energy storage technologies [1, 17].

As shown in Table 4, structural and morphological characteristics of polymeric nano-composite materials for Hybrid Energy Storage Systems (HESS) can be characterized by many different advanced analytical techniques.

Table 4. Advanced characterization techniques for polymer nanocomposites in HESS.

Characterization category	Technique	Purpose/information obtained	Application in HESS	References
Structural characterization	XRD (X-ray Diffraction)	Crystal structure, phase identification, crystallinity analysis	Study polymer structure, nanofiller dispersion, and phase evolution	[3, 14, 21]
	Raman Spectroscopy	Molecular vibration and bonding characteristics	Analyze graphene/CNT interactions and conductive network formation	[9, 21, 26]
	FTIR (Fourier Transform Infrared Spectroscopy)	Functional groups and chemical bonding	Identify polymer–nanofiller interactions and surface chemistry	[20, 25, 31]
	XPS (X-ray Photoelectron Spectroscopy)	Surface elemental composition and oxidation states	Evaluate interface chemistry and surface modification effects	[7, 23, 27]
Morphological characterization	SEM (Scanning Electron Microscopy)	Surface morphology and particle distribution	Examine porosity, filler dispersion, and electrode morphology	[2, 5, 29]
	TEM (Transmission Electron Microscopy)	Internal structure and nanoscale interfaces	Investigate nanofiller alignment and interfacial structures	[3, 16, 26]
	AFM (Atomic Force Microscopy)	Surface roughness and nanoscale topography	Characterize surface interaction and interface properties	[7, 25]
Thermal characterization	TGA (Thermogravimetric Analysis)	Thermal degradation and material stability	Determine degradation temperature and thermal resistance	[14, 17, 32]
	DSC (Differential Scanning Calorimetry)	Glass transition and melting behavior	Analyze crystallinity and thermal transitions	[14, 20], [30]
Electrochemical characterization	CV (Cyclic Voltammetry)	Redox activity and capacitance behavior	Evaluate electrochemical kinetics and charge storage	[6, 11, 24]
	EIS (Electrochemical Impedance Spectroscopy)	Charge transfer resistance and ion transport	Study conductivity and interfacial resistance	[6, 25, 30]
	Charge–Discharge Analysis (GCD)	Capacity and cycle performance	Assess energy density and cycle stability	[8, 12, 24]
Interface characterization	In-situ characterization	Real-time monitoring of interface evolution	Analyze dynamic electrochemical and structural changes	[7, 25]
	Synchrotron X-ray Techniques	High-resolution structural analysis	Investigate interfacial phenomena and reaction mechanisms	[7, 16]
	Neutron Techniques	Ion transport and internal structural analysis	Study diffusion pathways and interface evolution	[7, 17]

The structural techniques utilized to evaluate crystallinity, chemical bonding and surface composition include X-Ray Diffraction (XRD), Raman Spectroscopy, Fourier Transform Infrared Spectroscopy (FTIR), and X-Ray Photoelectron Spectroscopy (XPS). Evaluation of morphology, distribution of nanofillers and interface topology is possible through the use of Scanning Electron Microscopy (SEM), Transmission Electron Microscopy (TEM), and Atomic Force Microscopy (AFM). Thermogravimetry Analysis (TGA) and Differential Scanning Calorimetry (DSC) are utilized for the assessment of thermal stability and phase transition within these materials. Electrochemical techniques that have been applied for the evaluation of electrochemical behavior include Cyclic Voltammetry (CV), Electrochemical Impedance Spectroscopy (EIS) and Charge/Discharge Analysis. Techniques used to characterize the interfaces of composites for HESS materials in situ during operation utilize synchrotron X-rays and neutrons to analyze ion-transport phenomena and to study interfacial changes. Overall, all of these characterization tools have established the relationship between the structural features of polymeric composites and their respective functional properties, thereby supporting the development of highly performing materials enabled by polymers for Hybrid Energy Storage Systems.

MODELING, SIMULATION AND MACHINE LEARNING APPLICATIONS

Modeling and Simulations

The use of molecular modeling and simulation has been an important component in the study of the performance of polymer nanocomposite materials in Lithium-Ion Battery – Supercapacitor Hybrid Energy Storage Systems (HESS). Using MD simulation methods allows for insight into the interactions of polymer chains, mechanisms of ion transport, and behavior at the interface of polymers and nanofillers. This type of knowledge is very important for understanding the properties of materials and how charges will be transported through a material [7, 25]. FEM is also widely used to determine whether there is sufficient mechanical stress, proper temperature distribution, adequate electrochemistry, or sufficient structural integrity when subjected to normal operating conditions [9, 32]. Multiscale modeling provides a framework that combines the material properties of materials at the nano scale to performance at the macro scale allowing for establishment of relationships between structure and property, and to predict how devices will behave over time [17, 21]. Material scientists utilize constitutive equation models to simulate both mechanical and electrochemical response under different operational conditions which can help guide material development and/or predict performance levels [14]. In addition, the emergence of digital twin technology represents an advance simulation tool set that includes real-time monitoring, predictive analytics and optimization of hybrid energy storage system performance by utilizing data driven approaches [17]. These simulation tools enable researchers to quickly develop new materials and assist in designing future generations of hybrid energy storage systems enabled by polymers.

Application of Machine Learning Techniques

The use of machine learning (ML), as an accelerating mechanism for developing and optimizing polymer nanocomposite-based energy storage devices for Lithium-Ion Battery-Supercapacitor Hybrid Energy Storage Systems (HESS), is growing rapidly. Rapidly-developing materials prediction capabilities using machine learning enables researchers to quickly identify polymers that have good potential as matrices or fillers for nanocomposites, or hybrid combinations of both. Moreover, there exist various property optimization methodologies which allow researchers to predict the conductivity, thermal stability, energy density, and mechanical properties of these materials based on large amounts of experimental data from testing. Additionally, advanced ML techniques enable researchers to extract features from their experimental data to determine the key factors affecting the performance of the materials and/or the HESS device(s) being developed. Further, surrogate models can be utilized to decrease computational costs associated with simulating complex physical processes occurring within the system; therefore, accelerating the process of identifying promising candidates among a wide variety of possible material combinations. Most recently, data-driven material design approaches combine Artificial Intelligence (AI) with Material Informatics to develop new structure-property relationships which will ultimately help guide the design of multifunctional polymer systems for next-generation HESS application. The above-described ML techniques provide significant reductions in the amount of experimentation required and thus accelerate the time it takes to develop high-performance energy storage materials utilizing polymer.

POLYMER COMPOSITE MATERIALS AND GREEN POLYMER APPROACHES

The Advantages of Using Polymer Composites for Next Generation Hybrid Energy Storage Systems Based On Lithium-Ion Batteries and Supercapacitors

Hybrid Energy Storage System (HESS) is a new form of lithium-ion battery-supercapacitor that uses polymer composite material. This type of material allows for combining structural function with energy storage, and it has been considered as one of the most ideal types of materials for use in next generation hybrid lithium-ion battery-supercapacitor energy storage systems. When used in conjunction with polymer composites, light weight structural elements can greatly lower the total mass of the entire system while at the same time retain its mechanical properties; these characteristics make the use of polymer composites extremely desirable for electric vehicles (EV), since lowering vehicle weight will increase the driving range and improve energy efficiency [19–21]. In addition, recent advances in multi-functional composite materials, including composites that incorporate conductive polymers, nanofiller, and structural matrix have made it possible for these materials to perform multiple functions at the same time, including providing mechanical support, electrical conductivity, thermal management, and energy storage [10, 17]. More recently, advancements have been made in the field of structural batteries and now there exist many different types of materials that allow for electrical energy to be stored within the structure of a battery itself, thus allowing for separate energy storage units to be eliminated from the design and also greatly reduce the complexity of the system [17, 22]. Another area that is gaining popularity is the development of load bearing energy storage systems. These types of systems utilize structural components of a vehicle, such as body panels, chassis components, and composite frames to store electrical energy while they continue to bear mechanical loads [19]. The use of this type of multi-functionality offers several advantages when using polymer composites in electric vehicles. Some of these advantages include improved space utilization, fewer number of components needed, increased power-to-weight ratio, and increased efficiency of the vehicle as a whole. Therefore, the polymer composite-based hybrid energy storage system technology is expected to be key to the development of lighter, more compact, and higher performance energy storage systems for future automotive transportation systems [17–19, 32].

Environmental Concerns and Eco-Friendly Technologies

The need for eco-friendly technology, coupled with rising environmental concern is causing growth in the use of sustainability and green polymer technologies for the development of Hybrid Energy Storage Systems (HES) that utilize polymer. The interest in biodegradable polymers; PLA, chitosan, cellulose and starch-based materials for HES is due to the ability of these materials to be used in order to lessen the impact on the environment, but still provide desired mechanical and electro-chemical properties [19, 31]. Also, bio-derived materials made from renewable resources; lignin, natural fibers, biomass derived carbon materials are being developed as substitutes for conventional petroleum based polymers, which will allow for the continued development of green energy storage technologies [19, 21].

Recycling methods for composite materials and battery parts are needed so that there can be recovery of recyclable materials, reduction of waste generation, and less resource depletion [1, 14]. The above mentioned recycling methods help enable the use of a closed-loop system or "circular economy," whereby materials are continuously recycled, regenerated and integrated back into the manufacturing process. This provides for better resource utilization and increased sustainability [1]. Life Cycle Assessment (LCA) has been identified as one of the most effective tools available to determine the environmental impact caused by the production, processing, operation, and end-of-life of products/materials. Researchers have used LCA to determine the overall sustainability of HES systems utilizing polymer [17, 32]. Therefore, combining green polymer technologies with sustainable designs can lead to the creation of high performance and environmentally responsible HES systems.

Current Challenges

Challenges relating to polymer nanocomposite and functional material systems in lithium-ion battery-supercapacitor hybrid energy storage systems (HESS) include:

1. *Interfacial incompatibility:* Poor interactions between the polymer matrix, nanofillers and electrode materials could be expected to increase resistances at interfaces which would negatively affect the overall efficiency of charge transport.
2. *Thermally unstable material properties:* Elevated operating temperatures have been shown to potentially decrease electrochemical performance as well as raise concern regarding both thermal stability and potential fire hazards due to degradation of materials [15–20].
3. *Scale up production concerns:* While many fabrication techniques used to produce advanced polymeric composites face challenges when scaled up, the need for uniform dispersion of nanofillers throughout a composite and consistency within the material itself will likely limit the ability of producers to scale their product up efficiently [29–34].
4. *Increased manufacturing costs:* The use of conductive polymers, advanced nanofillers, etc. along with increased complexity in the processing of these composites is also increasing manufacturing costs and limiting the applicability of HESS's by industry [9, 21].
5. *Degradation over time:* Both long term cycling has resulted in polymer degradation, structural failures, electrode degradation, and decreased overall reliability of HESS's [8, 27].
6. *Challenges associated with recycling:* There are limited recycling options available today for composite materials that are environmentally unfriendly which raises serious concerns about how to recycle or dispose of HESS's in an environmentally responsible manner [2, 23].
7. *Lack of industry wide standards for testing and characterization:* There currently are no universally accepted standards for either testing, evaluating performance, and characterizing composite materials for HESS's making comparisons between research studies nearly impossible [16, 31].

CONCLUSION AND FUTURE SCOPE

Conclusions

- In this paper a comprehensive overview of the latest advancements in the application of polymer nanocomposites and functional materials within Lithium Ion Batteries-Supercapacitors Hybrid Energy Storage Systems (HESS) is provided.
- Advances in the fields of conducting polymers, polymer electrolytes, nanocomposite configurations and processing techniques have greatly improved electrical conductivity, heat dissipation, mechanical integrity and electrochemical efficiency.
- As well, through the implementation of engineered interfaces researchers were able to demonstrate significant effects upon ion transport, charge transfer mechanisms and structural stability of individual HESS elements.
- While many performance enhancements have been made to date; however, issues such as cost-effectiveness, long-term degradation of materials and environmental impacts continue to challenge widespread adoption.
- Through advancements in intelligent material design, multifunctional polymer-based composites and other advanced forms of polymer enabled HESS technologies we anticipate further advancement toward the realization of safe, light weight and high performing hybrid energy storage technologies suitable for automotive applications and emerging energy systems.

Future Directions

- There are a number of areas that will require additional research before the full potential of Polymer-Enabled Hybrid Energy Storage Systems (HESS) can be realized. For example, there

are numerous possibilities for using new types of self-healing polymer systems as they could potentially repair structural damage when it occurs during use or extend the overall life of an energy storage device.
- Researchers also believe that by combining artificial intelligence (AI)-driven material design and data-centric approaches researchers will be able to find new materials much faster than they do today and better understand how to achieve desired properties. Solid state HESS devices exhibit increased safety, higher energy densities and greater thermal stability than their traditional counterparts.
- Additionally, the continued development of multifunctional polymer composite materials, smart interfaces, and next generation nano-composites that exhibit improved interfacial compatibility and adaptable functionalities will help improve both the performance and longevity of HESS devices. Research into these emerging technologies will provide critical insights to identify what obstacles exist currently to achieve next-generation performance from HESS devices.

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