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Review article

Selection criteria of polymer nanocomposites for electrical energy storage applications: A concise review

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Abstract. Polymer nanocomposites are drawing considerable interest in electrical energy storage research owing to their distinctive characteristics and promising roles in various devices, such as batteries, supercapacitors, and fuel cells. This review examines the selection criteria of polymer nanocomposites for electrical energy storage applications and the current advancements in developing and producing polymer nanocomposites specifically tailored for electrical energy storage applications. Key topics covered include the selection of polymer matrices, choice of nanofillers, fabrication techniques, characterization methods, and performance evaluation of the resulting nanocomposites. The impact of nanofiller dispersion, interface engineering, and morphology control on electrical storage properties is emphasized. Proper dispersion enhances uniformity and interfacial interactions, improving electrical, mechanical, and thermal properties. Interface engineering boosts polymer-nanofiller compatibility, while morphology control optimizes nanofiller structure and arrangement for better storage efficiency. Emerging trends, challenges, and future research directions are also discussed, providing insights for developing advanced polymer nanocomposites with improved electrical energy storage capabilities.

Keywords: *polymer, nanocomposites, fuel cells, nanofillers, fabrication, characterization*

1. Introduction

The dynamic realm of electrical energy storage technologies is propelling forward innovation in materials science and engineering, particularly in the realm of advanced nanocomposite materials. Polymer nanocomposites, crafted from polymers and nanofillers with finely tuned properties, are garnering attention as *via*ble solutions to meet the escalating need for effective and eco-friendly electrical energy storage solutions. By leveraging the collaborative potential of polymers and nanofillers, these polymer nanocomposites present distinctive avenues to augment the efficiency and capabilities of electrical energy storage devices such as supercapacitors, batteries, and capacitive deionization systems [1, 2]. Electrical energy storage is indispensable in facilitating the widespread adoption of renewable energy sources like solar and wind power. By alleviating intermittency issues and ensuring grid stability, it serves a pivotal role in enabling their widespread integration. However, traditional electrical energy storage materials often face challenges related to limited energy density, slow charge-discharge rates, and poor life cycles. Polymer nanocomposites emerge as an enticing option, providing improved electrical conductivity, mechanical robustness, thermal resilience, and ion transport characteristics. These attributes are pivotal for maximizing the efficacy of electrical energy storage devices [3].

Crafting polymer nanocomposites for electrical energy storage entails meticulous attention to diverse elements such as material choice, manufacturing

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methodologies, characterization approaches, and optimization tactics in both design and fabrication processes [4, 5]. Polymers possessing desirable attributes like a high dielectric constant, mechanical resilience, and chemical durability are selected as the foundational matrix material, while nanofillers such as carbon nanotubes, graphene, metal oxides, and clay nanoparticles are incorporated to enhance conductivity, charge storage capacity, and mechanical properties [6, 7]. Several fabrication techniques, including solution mixing, melt blending, *in-situ* polymerization, and electrospinning, offer versatility in tailoring the morphology and properties of polymer nanocomposites to meet specific application requirements [8, 9]. Methods for characterization, including scanning electron microscopy (SEM), transmission electron microscopy (TEM), X-ray diffraction (XRD), Fourier-transform infrared spectroscopy (FTIR), and various thermal analyses such as differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA), offer invaluable insights into the structural, electrical, thermal, and mechanical attributes of polymer nanocomposites.

This review offers a thorough examination of recent progress in the design and fabrication of polymer nanocomposites tailored for electrical energy storage purposes. The key principles underlying the selection of materials, fabrication techniques, characterization methods, and optimization strategies were discussed. Furthermore, the review explores the integration of polymer nanocomposites into various electrical energy storage devices and discusses their potential implications for advancing sustainable energy technologies.

2. Electric energy density

Electrical energy density is a performance metric that quantifies the amount of electrical energy stored per unit mass or volume of an electrical energy storage system. It is essential for evaluating the efficiency and capacity of various electrical energy storage devices, such as batteries, capacitors, and supercapacitors. In the context of batteries, higher energy density allows for the storage of more energy within a given mass or volume, which directly impacts the device's ability to power applications over extended periods without increasing its size or weight. This is particularly crucial for applications such as electric vehicles, where optimizing energy density can lead to longer driving ranges and reduced battery pack weight [10]. In contrast, capacitors, while capable of faster charge/discharge cycles and higher power delivery, typically exhibit lower energy densities compared to batteries, making them suitable for applications that require rapid energy release but not extended storage. The energy density of a system is governed by several factors, including the electrochemical properties of the materials used, the design of the storage device, and the efficiency of the charge/ discharge processes. As such, improving energy density remains a primary goal in the development of next-generation electrical energy storage technologies, as it enables more compact, efficient, and longer-lasting power solutions for a wide range of applications, from portable electronics to grid-scale electrical energy storage systems.

2.1. Dielectric constant (relative permittivity)

The dielectric constant (or relative permittivity) of polymer nanocomposites is a crucial factor that influences their ability to store electrical energy. When nanomaterials, such as nanoparticles of metals, ceramics, or carbon-based materials, are incorporated into a polymer matrix, they can significantly enhance the dielectric constant of the composite material. [11] reported that semi-conductive SiC nanowires can remarkably improve the dielectric permittivity of polyvinylidene fluoride (PVDF). This is because nanofillers such as graphene, carbon nanotubes, or metal oxide nanoparticles have unique electrical properties that improve the material's ability to polarize in response to an electric field [12].

In electrical energy applications like capacitors, a higher dielectric constant leads to greater electrical energy storage, allowing the device to store more energy in a smaller volume [13]. This is particularly important for devices that require high energy density and compactness. For example, polymer nanocomposites with a high dielectric constant can be used in advanced capacitors, improving their electrical energy storage capacity without increasing the size or weight of the device. The ability to tune the dielectric constant of polymer nanocomposites by varying nanofillers' type, concentration, and dispersion is an essential design parameter for developing efficient electrical energy storage materials.

2.2. Breakdown strength (electric field strength)

The breakdown strength of a material refers to the maximum electric field it can withstand before it loses its insulating properties and becomes conductive. In electrical energy storage systems, especially in high-voltage capacitors, the dielectric material needs to withstand significant electric fields without breaking down, as this would cause a short circuit or catastrophic failure of the device. Polymer nanocomposites are typically used to enhance the breakdown strength of dielectric materials. The introduction of nanofillers such as graphene, carbon nanotubes, or metal oxide particles can significantly improve the breakdown strength of the polymer matrix [14, 15]. This is because the nanofillers help to create a more uniform and ordered structure in the composite, reducing defects and inhomogeneities that can serve as sites for electrical breakdown. Additionally, the nanofillers can increase the material's resistance to electrical discharges by enhancing its structural integrity.

2.3. Dielectric loss (dissipation factor)

Dielectric loss, or the dissipation factor (tan δ), measures the energy dissipated as heat when a dielectric material is subjected to an alternating electric field. A low dielectric loss is desirable for electrical energy storage materials, as it indicates minimal energy wastage and heat generation during operation. High dielectric loss, on the other hand, can lead to efficiency losses and overheating of the electrical energy storage device, reducing its performance and lifespan [5].

In polymer nanocomposites, the incorporation of nanoparticles can influence the dielectric loss, depending on the type of nanoparticle used and its dispersion within the polymer matrix [16]. In general, well-dispersed and functionalized nanoparticles, such as carbon nanotubes or graphene, can enhance the electrical conductivity of the composite without significantly increasing the dielectric loss. This is because these nanofillers provide pathways for charge transport that can enhance electrical energy storage, causing excessive energy dissipation [17].

However, poor dispersion or agglomeration of nanoparticles can lead to increased dielectric losses due to the formation of localized regions of high electrical conductivity or trapped charge. Therefore, achieving an optimal dispersion of nanoparticles within the polymer matrix is essential for minimizing dielectric loss in electrical energy storage applications. Materials with low dielectric loss are particularly important for applications like high-frequency capacitors and other electrical energy storage devices, where efficiency and minimal heat generation are critical.

3. Polymer material selection

Choosing polymers with appropriate properties is essential for the development of effective electrical energy storage materials. Polymers play a critical role in providing structural support, facilitating ion transport, and ensuring overall device stability. One key property to consider is a high dielectric constant, which is crucial for capacitive electrical energy storage devices such as supercapacitors [18]. Polymers with high dielectric constants enable efficient charge storage by enhancing the polarization of the material in response to an applied electric field [19]. Additionally, mechanical strength is important to withstand the mechanical stresses experienced during device operation. Polymers with high mechanical strength can maintain structural integrity, preventing mechanical failure and ensuring long-term durability.

Commonly used polymers in electrical energy applications include polyethylene, polypropylene, polyvinylidene fluoride (PVDF), and polyaniline [20, 21]. An example of such applications is a study conducted by Hameed *et al.* [22]. They examined the high energy density and discharge efficiency attained by chlorinated polyethylene films for applications requiring high-energy storage. A study conducted by Liu *et al.* [23] focused on the utilization of ZnO/CuO nanocomposite-based carboxymethyl cellulose/polyethylene oxide polymer electrolytes for applications in energy storage. Polyethylene and polypropylene are widely employed due to their excellent mechanical properties, chemical resistance, and low cost [24]. These polymers are commonly used as separators and structural components in batteries and capacitors [25]. PVDF, on the other hand, is valued for its high dielectric constant, flexibility, and chemical stability. PVDF-based materials are often used as binder materials and electrolytes in lithium-ion batteries and supercapacitors. Polyaniline, a conducting polymer, offers a unique combination of electrical conductivity and redox activity, making it suitable for electrochemical electrical energy applications [26–28].

The study by Wu *et al*. [28] stated chemical stability as another crucial consideration for polymer selection in electrical energy storage devices aside from high dielectric constant and mechanical strength. Polymers must be chemically stable under the operating conditions of the electrical energy storage device to prevent degradation and maintain performance over time. Exposure to electrolytes, solvents, and high temperatures can lead to chemical breakdown and loss of device functionality. Polymers such as PVDF and polyethylene exhibit excellent chemical stability, making them well-suited for use in harsh environments commonly encountered in electrical energy applications [29, 30].

The selection of polymers for electrical energy applications involves a trade-off between various properties such as dielectric constant, mechanical strength, and chemical stability [31]. Depending on the specific requirements of the application, different polymers may be chosen to optimize device performance. Furthermore, advances in polymer synthesis and processing techniques continue to expand the range of available polymers with tailored properties for electrical energy applications [32]. Overall, choosing polymers with suitable properties is a crucial step in the design and development of high-performance electrical energy storage devices.

3.1. Selection of suitable nanofiller

Incorporating nanofillers into the polymer matrix is a proven strategy to enhance the electrical energy performance of polymer-based materials [33, 34]. Nanofillers, such as carbon nanotubes, graphene, metal oxides (*e.g*., titanium dioxide or zinc oxide), and clay nanoparticles, offer unique properties that can significantly improve the conductivity, charge storage capacity, and mechanical properties of polymer nanocomposites [35].

3.1.1. Carbon-based nanofillers

Carbon-based nanofillers, including carbon nanotubes and graphene, are widely studied for their exceptional electrical conductivity and high surface area [36–38]. When dispersed within the polymer matrix, these nanofillers create conductive pathways that facilitate the rapid transportation of ions and electrons, leading to enhanced charge storage and improved electrical conductivity [39]. Additionally, the high mechanical strength and flexibility of carbon nanotubes and graphene contribute to the mechanical reinforcement of the nanocomposite, improving its structural integrity and durability. Feng *et al*. [40] developed a light cellular carbon nanotube graphene/carbon nanocomposite and achieved 43% flexural strength along with 102.7% in fracture toughness.

3.1.2. Metal oxides

Metal oxides, such as titanium dioxide or zinc oxide nanoparticles, offer a different set of advantages for electrical energy storage applications. These nanofillers exhibit high specific surface area, electrochemical activity, and charge storage capacity, making them suitable candidates for enhancing the performance of capacitor and battery-type electrical energy storage [41]. When incorporated into the polymer matrix, metal oxide nanoparticles provide additional active sites for charge storage, and they promote ion diffusion, leading to improved electrical energy performance [42]. Khedulkar *et al*. [43] showed how the porous structure of biochar enhances ion transport, thereby enhancing electrical energy storage capacities and accelerating charge-discharge rates. Furthermore, the inclusion of metal oxides and biochar enhances the conductivity of the composites, aiding efficient electron transportation. Their research centered on utilizing agricultural waste to produce sustainable, eco-friendly, high-energy supercapacitors.

3.1.3. Clay nanoparticles

Clay nanoparticles, such as montmorillonite or kaolinite, are another class of nanofillers commonly used in polymer nanocomposites. These nanofillers offer advantages such as a high aspect ratio, large surface area, and tunable properties [44]. When dispersed within the polymer matrix, clay nanoparticles can improve mechanical properties, barrier properties, and thermal stability, as reported by Karimi *et al*. [45]. Furthermore, clay nanoparticles can serve as templates for the alignment of polymer chains, leading to enhanced electrical conductivity and charge transport within the nanocomposite [46].

In general, integrating nanofillers into the polymer matrix offers a flexible method to customize the characteristics of polymer nanocomposites for electrical energy storage purposes. By judiciously selecting and dispersing nanofillers within the polymer matrix, it is possible to achieve synergistic effects that enhance conductivity, charge storage capacity, and mechanical properties, ultimately leading to improved performance and efficiency of electrical energy storage devices [10]. Table 1 presents a comparison of the nanofillers discussed.

Carbon-based nanofillers offer outstanding mechanical and conductive properties, making them ideal for high-performance applications. However, their high cost, processing challenges, and potential health risks due to agglomeration and inhalation limit their widespread use. In contrast, metal oxide nanoparticles are more affordable and provide excellent thermal stability and UV-blocking properties, making them suitable for high-temperature applications. However, they offer only moderate mechanical reinforcement and may have toxicity concerns due to metal leaching. Meanwhile, clay nanoparticles are a cost-effective option that significantly enhances barrier properties and flame retardancy, which is valuable in packaging and construction applications. Their main limitation lies in providing only modest improvements in mechanical and electrical properties, making them less suitable for applications that require high levels of reinforcement or conductivity.

4. Composite fabrication

The fabrication of polymer nanocomposites encompasses a range of techniques, each offering unique advantages and suitability for different applications. The four prominent methods include solution mixing, melt blending, *in-situ* polymerization, and electrospinning, each of which influences the dispersion of nanofillers within the polymer matrix and consequently impacts the final properties of the nanocomposite.

4.1. Solution mixing

Solution mixing involves dissolving both the polymer and nanofillers in a common solvent, followed by mixing to achieve a homogenous dispersion, as shown in Figure 1. This method offers precise control over nanofiller distribution and

Table 1. Comparison between nanofillers.

Nanofiller type	Mechanical properties	Electrical / thermal conductivity	Aspect ratio and surface area	Thermal stability	Optical properties / flame retardancy	Barrier properties	Toxicity	Dispersion	Cost
Carbon-based nanofiller	High tensile strength (up to 100 GPa) and Young's modulus (up to 1 TPa) [47, 48]	High electrical conductivity and thermal conductivity ~10^6 S/m for graphene [49, 50] / up to 5000 W/(m·K) [51]	High aspect ratio and large surface area (up to 1000 m^2/g for graphene [1]	Moderate to low compared to metal oxides [49]	None	None	Potential respiratory hazards and environmental concerns, especially during handling and disposal [52]	Prone to agglomeration; requires functionalization or surfactants for uniform distribution [53, 54]	High, costly production processes like chemical vapour deposition (CVD) [55]
Metal oxide nanoparticles	Moderate enhancement (tensile modulus improvement typically up to 30% [3]	Limited improvement in thermal conductivity and moderate to high, suitable for high temperatures (up to 1000 °C [56]	Moderate	High, suitable for high-temperature environments [57]	No flame retardancy / effective UV-blocking and photocatalytic properties (*e.g.*, TiO_2 with a bandgap of ~3.2 eV [58, 59]	None	Potential environmental impact and toxicity due to metal leaching, especially in aquatic environments [60]	Generally well-dispersed, but some agglomeration can occur depending on the specific oxide used [61]	Moderate; more affordable than carbon-based nanofillers, easier to produce and source [62]
Clay nanoparticles	Limited improvement (tensile strength increases typically up to 10–15% [3]	Poor	Low aspect ratio and smaller surface area compared to carbon-based fillers [63]	Moderate	No optical properties/enhances flame retardancy and thermal stability [64]	Significant improvements in gas and moisture barrier properties (up to 80% effectiveness [65]	Minimal toxicity, generally considered safe [66]	Challenges in achieving homogeneous dispersion within the polymer matrix, leading to localized weaknesses [63]	Low; abundant and low-cost due to natural availability [64]

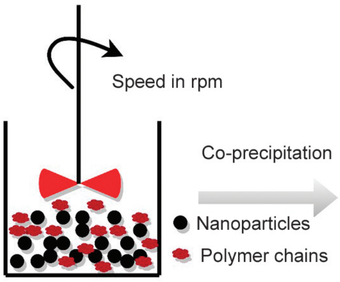

Figure 1. Schematic illustration for the solution mixing method.

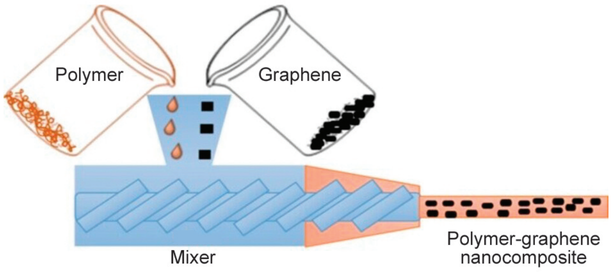

Figure 2. Polymer/graphene manufacture using melt blending process.

allows for the incorporation of a wide range of nanofiller types and concentrations [67]. Solution mixing is particularly well-suited for producing thin films, coatings, and solution-cast membranes used in electrical energy storage devices.

In solution mixing, revolutions per minute [rpm] is a crucial parameter that directly affects the dispersion of nanofillers within the polymer matrix, as illustrated in Figure 1. Higher rpm increases the shear forces applied to the mixture, which helps to break up agglomerates of nanofillers and achieve a more uniform dispersion throughout the solution [68]. This control over dispersion quality is vital for ensuring that the nanofillers are evenly distributed within the polymer chains, which enhances the composite's properties. The optimal rpm depends on factors such as the viscosity of the polymer solution, type of nanofillers used, and the desired application.

In particular, higher rpm is advantageous when incorporating conductive or reinforcing nanofillers like carbon nanotubes, graphene, or metal oxides, as it improves the interaction between nanofillers and the polymer matrix, which is essential for applications in thin films, coatings, and solution-cast membranes used in electrical energy storage devices.

4.2. Melt blending

Melt blending, on the other hand, involves mixing the polymer and nanofillers in a molten state, typically using extrusion or compounding processes [69]. Figure 2 depicts an example of a polymer/ graphene nanocomposite production process. This method is particularly advantageous for high-throughput manufacturing and is commonly used in industries with large-scale production requirements. Melt blending facilitates good interfacial adhesion between the polymer matrix and nanofillers, resulting in improved mechanical properties and ease of processing.

4.3. *In-situ* polymerization

In-situ polymerization involves the simultaneous formation of the polymer matrix and nanofillers within a reaction medium (Figure 3). This method offers excellent control over nanofiller dispersion and interfacial interactions, as well as the ability to tailor the polymer structure and properties. *In-situ* polymerization is commonly used to produce nanocomposites with complex architectures and functional groups, making it suitable for applications requiring precise control over material properties [71, 72].

4.4. Electrospinning

Electrospinning is a versatile technique used to fabricate nanofibrous membranes or mats by electrostatically drawing polymer solutions or melts through a charged spinneret, as depicted in Figure 4. The figure is the schematic diagram of the utilisation of electrical energy to influence the arrangement of the nanofibers.

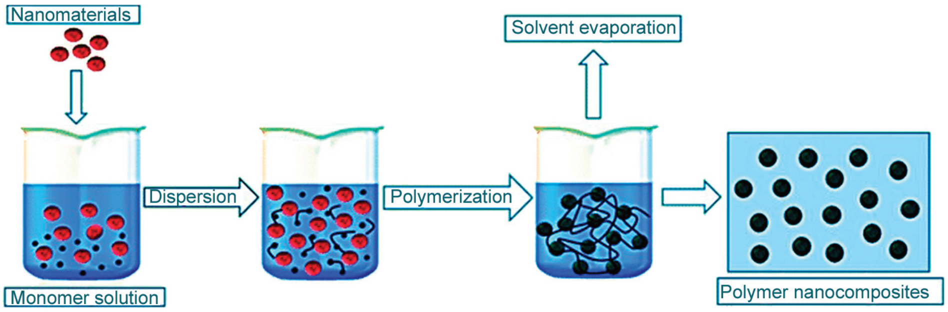

Figure 3. *In-situ* polymerization method [72].

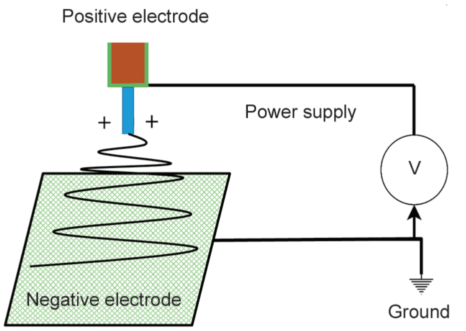

Figure 4. A schematic diagram of the electrospinning system with the spinneret as an anode and the collector as a negative electrode.

A typical electrospinning setup comprises of spinneret containing the polymeric solution, a DC high voltage supply, and a grounded collector. A charged jet of polymer solution is extracted from the spinneret tip (with a typical cone shape in the inset) and reaches the collector with a typical whipping motion [73, 74]. By incorporating nanofillers into a polymer solution or a melt prior to electrospinning, it is possible to produce nanocomposite fibres with enhanced mechanical, electrical, and thermal properties. Electrospun nanocomposites find applications in electrical energy storage devices such as supercapacitor electrodes and separator membranes. The choice of fabrication method depends on various factors, including the desired properties of the nanocomposite, scalability requirements, and specific application needs [72, 75]. Some of the application areas are presented in Table 2, even though Forouzandeh *et al.* [76] concluded in their study that incorporating more than one synthetic approach for fabrication makes it possible to obtain the electrode material for excellent electrochemical properties.

Ongoing research efforts aim to explore novel fabrication methods, like three-dimensional (3D) printing

Table 2. Areas of application for fabrication methods.

S/No.	Fabrication method	Electrical energy storage application	Advantages	Limitations
1.	Solution mixing	Battery electrodes [76, 77], supercapacitors, and solid-state electrolytes	Simple, cost-effective, good for thin films, and suitable for water-soluble polymers	Solvent residue can affect performance, limited to low filler loading, and environmental concerns with solvent disposal
2.	Melt blending	Battery separators, thermal management in batteries and conductive polymers for capacitors [78]	Solvent-free, scalable, and compatible with industrial extrusion processes	High processing temperatures can degrade sensitive fillers and polymers, limited by filler dispersion quality
3.	*In-situ* polymerization	Nanocomposite electrodes, electrolytes in energy devices and fuel cell membranes	Excellent filler dispersion, high compatibility with monomer and filler, and good mechanical properties	Complex processing, longer synthesis times, and challenges in achieving uniform filler distribution
4.	Electrospinning	Nanofiber electrodes, [79, 80] nanostructured separators and electrolytes with enhanced ion transport [81]	High surface area, good control over fiber diameter, and suitable for fibrous applications	Time-consuming, labor-intensive, and difficult to scale for large-area applications
5.	Sol-gel processing I	Used in electrical energy storage devices like supercapacitors and batteries due to high interface control [78]	Precise control over thickness allows multilayer structures and good interfacial adhesion [78]	Time-consuming, labor-intensive, and difficult to scale for large-area applications [78]
6.	Sol-gel processing II	Often used in the production of hybrid nanocomposites for energy conversion and storage [82]	Potential for residual organic content [83]	Often requires further heat treatment and limitations with specific polymer-filler compatibility.
7.	Template synthesis	Utilized in creating nanocomposites with controlled porosity for fuel cells and supercapacitor electrodes [84, 85]	Good control over particle size and morphology, allows design flexibility, and excellent for porous materials	High complexity, template removal can be challenging, and expensive for large-scale production
8.	High shear mixing	Produces nanocomposites for structural energy applications and batteries [86, 87]	Fast, compatible with high filler loadings, and provides good dispersion in high-viscosity polymers	High energy consumption, risk of filler degradation due to mechanical forces, and limited control over final morphology
9.	Ball milling (mechanical milling)	Employed in preparing nanocomposites for electrodes in batteries and supercapacitors [88]	Suitable for high filler content, does not require solvents, and produces homogeneous dispersion	Risk of contamination from milling media, energy-intensive, and potential for particle agglomeration if milling time is not optimized

and improve existing techniques to further enhance the performance and functionality of polymer nanocomposites [89]. Three-dimensional (3D) printing of multifunctional nanocomposites is a novel fabrication area explored recently with the enhancement of 3D printing technologies in recent years. Several methods, including but not limited to micro-stereolithography, extrusion-based direct-write technologies, inkjet-printing techniques, and popular powder-bed technology, are utilised. The 3D nanocomposites may be macro and microstructures manufactured for a wide range of domains. Examples include microelectromechanical systems (MEMS), lab-on-a-chip, microfluidics, engineered materials and composites, microelectronics, tissue engineering, and biosystems [90]. Other novel methods developed are hydrothermal synthesis, microwave-assisted synthesis, supercritical fluid processing, electrochemical deposition and bioinspired and biomimetic synthesis [91].

4.5. Hydrothermal synthesis

Hydrothermal synthesis is a method used to crystallize substances from high-temperature aqueous solutions at high vapor pressures [92]. Typically, the process involves using a sealed vessel called an autoclave (or a hydrothermal bomb (Figure 5)), which allows the containment of water and other reagents at temperatures well above the boiling point of water and under high pressure, without vaporizing the contents. This technique is particularly useful for the synthesis of minerals, ceramics, and various nanomaterials [93, 94].

The conditions within the autoclave, such as temperature, pressure, solvent composition, and time, can be adjusted to optimize the growth of the desired crystalline material. The high pressure and temperature can dissolve materials that are insoluble under normal conditions, and upon cooling, new compounds precipitate [95]. Hydrothermal synthesis is often used to grow synthetic quartz, gems and other single crystals with commercial and scientific applications.

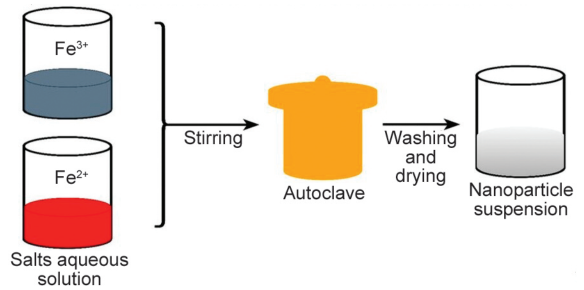

Figure 5. Hydrothermal synthesis of iron oxide nanoparticles.

4.6. Microwave-assisted synthesis

Microwave-assisted synthesis is a technique that uses microwave radiation to heat reactants in a chemical reaction. This method has become popular in both academic research and industrial applications because it can significantly speed up reaction times, enhance product yields, and improve the purity of the final product [96]. The microwave energy directly interacts with molecules, providing rapid, uniform heating, which is often more efficient than conventional heating methods. Microwaves induce dipolar rotation and ionic conduction within the molecules of the reactants, generating heat from inside the material rather than from an external heat source. This can lead to more uniform temperature distribution and reduced thermal gradients throughout the material, minimizing the risk of overheating and degradation of sensitive components [97].

The main advantages of microwave-assisted synthesis are increased reaction speed, energy efficiency, selective heating and control over reaction conditions. Microwave-assisted synthesis is widely used in organic synthesis, material science, and the synthesis of pharmaceuticals, where it contributes to the development of greener chemical processes by reducing reaction times and potentially lowering the energy consumption of chemical processes [97].

4.7. Supercritical fluid processing

Supercritical fluid processing utilizes the unique properties of supercritical fluids, which are substances at temperatures and pressures above their critical points, combining characteristics of both gases and liquids [98, 99]. This technology is prized for its ability to fine-tune solvent power through adjustments in temperature and pressure, enabling highly efficient and selective extraction, reaction, and material processing. Common applications include supercritical fluid extraction (SFE) for extracting compounds from natural products without harmful solvents, supercritical fluid chromatography (SFC) for fast and efficient separation of mixtures, and various particle formation techniques used in pharmaceutical manufacturing [99].

4.8. Electrochemical deposition

Electrochemical deposition, commonly known as electroplating, is a method where a metal is coated onto a conductive surface through the application of an electric current. This process involves two electrodes submerged in an electrolytic solution containing metal salts (Figure 6). The object to be plated serves as the cathode, and the anode is often made of the deposition metal. When electricity flows through the solution, metal ions are reduced at the cathode, adhering as a solid metal layer. This method is crucial in industries like electronics for producing components like circuit boards, automotive and aerospace for improving corrosion resistance and wear properties, and in creating decorative and jewellery items with metals like gold and silver [100].

Electroplating allows for precise control over the thickness of the metal layer, making it a cost-effective means of applying valuable metals and enhancing substrate properties such as durability and conductivity. However, it requires careful handling of potentially harmful chemicals and effective waste management to prevent environmental damage. Despite these challenges, electrochemical deposition remains a fundamental technique in manufacturing, enhancing both the functionality and appearance of metal objects [101].

4.9. Bioinspired and biomimetic synthesis

Bioinspired and biomimetic syntheses involve creating materials and processes that mimic or are inspired by biological systems. These approaches harness the efficient and innovative solutions found in nature to address complex challenges in technology and materials science. For example, surfaces that imitate shark skin prevent biofouling on marine vessels, and synthetic adhesives inspired by gecko feet explore new kinds of non-permanent adhesives. Other applications include catalytic systems modelled after enzymes for more efficient chemical reactions and medical implants that integrate seamlessly with human tissue. Despite their potential, scaling these technologies from laboratory to industrial applications presents significant challenges, including issues of material durability and the need for interdisciplinary collaboration. These novel fabrication techniques hold great promise for advancing the development of polymer nanocomposites tailored for electrical energy storage applications, offering new opportunities to address key challenges such as energy density, cycle life, safety, and sustainability. By integrating innovative synthesis methods with advanced characterization techniques and computational modelling, researchers can accelerate the discovery and optimization of next-generation materials for diverse electrical energy storage technologies [102].

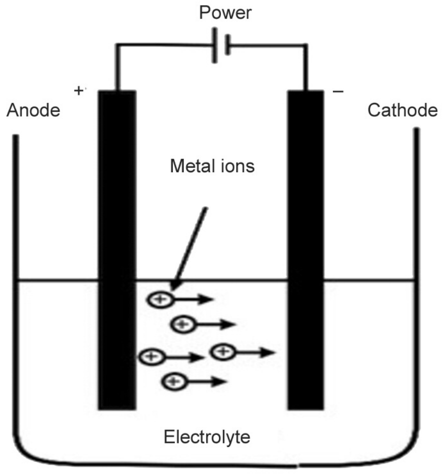

Figure 6. Schematic diagram of the electrodeposition process.

5. Characterization

Characterizing synthesized nanocomposites is essential for understanding their structural, electrical, thermal, and mechanical properties, which are crucial for optimizing their performance in electrical energy storage applications. Various analytical techniques can provide valuable insights into the morphology, composition, and behaviour of nanocomposites.

5.1. Structural characterization

Scanning electron microscopy (SEM) and transmission electron microscopy (TEM) are powerful imaging techniques used to visualize the surface morphology and internal structure of nanocomposites at micro- and nano-scales, respectively. SEM provides high-resolution images of the surface topography, while TEM offers detailed information about the dispersion and distribution of nanofillers within the polymer matrix [103]. Figure 7 shows a distinction between SEM and TEM for a silica/ethylene vinyl alcohol (EVA) composite where the volume fraction of the silica is 2.7%. The bright spots in the SEM micrographs and the dark spots in the TEM micrographs are dispersed primary silica particles on the surfaces of the composite [104].

X-ray diffraction (XRD) analysis is employed to determine the crystalline structure and phase composition

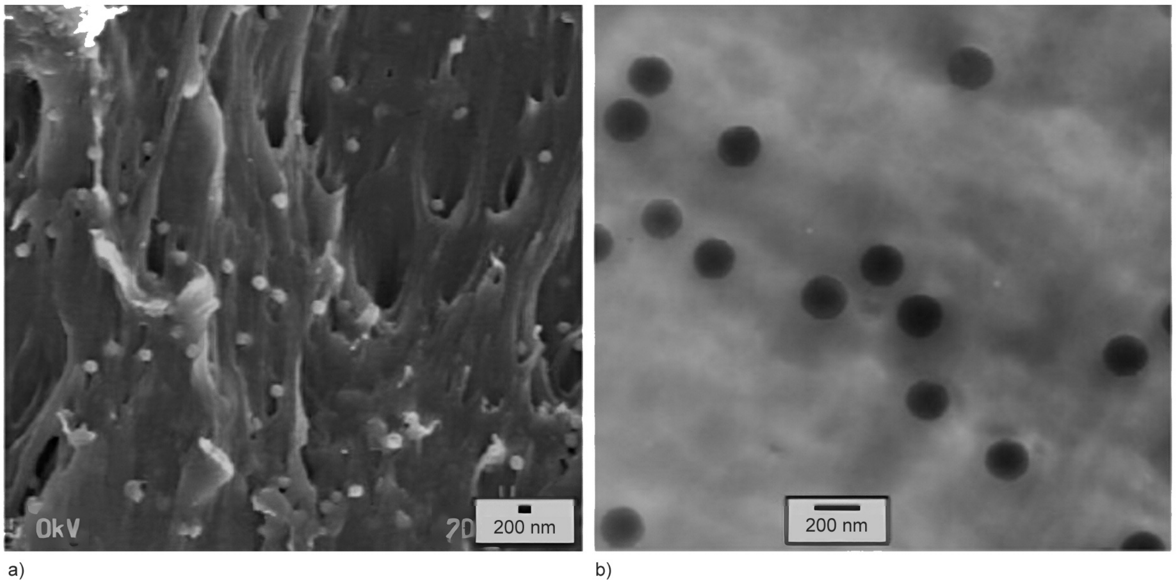

Figure 7. a) and b) are SEM and TEM micrographs showing dispersion states of silica nanoparticles in silica/polymer composites [104].

of nanocomposites. By analyzing the diffraction patterns generated by X-rays interacting with the sample, XRD can identify the presence of crystalline phases, quantify crystallite size, and assess the degree of crystallinity in the nanocomposite [105].

5.2. Electrical characterization

Electrical characterization of composite materials is essential for understanding their suitability in various electrical and electronic applications. This process evaluates key electrical properties such as conductivity, resistivity, dielectric behaviour, and other parameters that govern material performance in applications like energy storage, electronics, and electromagnetic shielding.

5.2.1. Electrical conductivity and resistivity tests

Electrical conductivity and resistivity are foundational electrical properties that determine how easily charge carriers, such as electrons or ions, can move through the composite matrix. Electrical conductivity quantifies the material's ability to conduct electric current, while resistivity measures the opposition to this flow [106]. For composite materials, these properties are heavily influenced by the type, concentration, and distribution of conductive fillers like graphene, carbon nanotubes (CNTs), or metallic particles.

To accurately measure these properties, the four-point probe technique is commonly employed. This method minimizes the contact resistance that can distort measurements, offering precise results even in highly conductive or insulating materials [107]. The concentration and uniformity of conductive fillers are crucial, as they directly affect the percolation threshold, which is the point at which the filler network becomes conductive enough to facilitate electron flow. Through careful tuning of filler concentration and distribution, conductivity can be optimized to meet specific application requirements, such as in capacitors, sensors, or conductive coatings.

5.2.2. Impedance spectroscopy

Impedance spectroscopy is a technique used to analyse the dielectric properties of composite materials, which include the dielectric constant (permittivity) and dielectric loss (dissipation factor). The dielectric constant reflects the material's ability to store electrical energy when subjected to an electric field, while dielectric loss represents the energy lost as heat due to the material's resistance to alternating electric fields.

Impedance spectroscopy involves measuring the material's impedance across a range of frequencies, providing insights into its resistive, capacitive, and inductive behaviors. These measurements can reveal how charge transfer mechanisms operate within the composite and how the conductive and insulating phases are distributed [108]. For electrical energy storage applications, a high dielectric constant is often desirable, while low dielectric loss is critical for minimizing energy wastage in high-frequency devices.

In addition, impedance spectroscopy offers a frequency-dependent profile of the material's behaviour, helping to understand how it responds to varying electric fields and facilitating the optimization of the material for specific applications such as capacitors, supercapacitors, and insulating coatings [109].

5.2.3. Current-voltage (*I–V*) iharacterization

Current-voltage (*I–V*) characterization is used to evaluate the composite's electrical behaviour under different applied voltages. This test determines whether the material exhibits conductive, semi-conductive, or insulative properties. The *I–V* curve provides detailed insights into how the material responds to increasing voltage, revealing any non-linear characteristics [110]. These non-linearities are often observed in composites near the percolation threshold, where a conductive network of fillers begins to form, allowing electron tunnelling or hopping between particles [111]. This behaviour is crucial for composites intended for applications such as sensors, thermoelectric devices, or energy harvesting. Additionally, *I–V* tests are valuable for understanding the electrical breakdown of the material, helping to assess its limits for use in high-voltage environments.

5.2.4. Seebeck coefficient for thermoelectric applications

For composites used in energy harvesting applications, particularly those exploiting thermoelectric effects, measuring the Seebeck coefficient is essential. The Seebeck coefficient indicates how the material generates a voltage in response to a temperature gradient. This property is important for thermoelectric materials used in converting waste heat into electrical energy, such as in thermoelectric generators (TEGs). By determining the Seebeck coefficient, the thermoelectric performance of the composite can be optimized for maximum efficiency in energy conversion [112].

5.3. Thermal characterization

Thermal analysis techniques such as differential scanning calorimetry (DSC) and thermogravimetric analysis (TGA) are employed to assess the thermal properties of nanocomposites. DSC measures the heat flow associated with phase transitions, such as melting and crystallization, providing information about thermal stability and transition temperatures. TGA, on the other hand, measures the changes in sample weight as a function of temperature, enabling the determination of thermal decomposition temperatures and the evaluation of thermal stability [113].

Figure 8 depicts the XRD (Figure 8a), FTIR (Figure 8b) and TGA (Figure 8c) for rice husk-derived nanosilica-reduced graphene oxide@polypyrrole (SiO_2-rGO@PPy) synthesised by Ratsameetammajak *et al*. [114] and this study will be used to elucidate further on the XRD, FTIR and TGA methods.

The XRD patterns of bare polypyrrole (PPy), SiO_2-rGO_2, and SiO_2-rGO2@PPy nanocomposites are

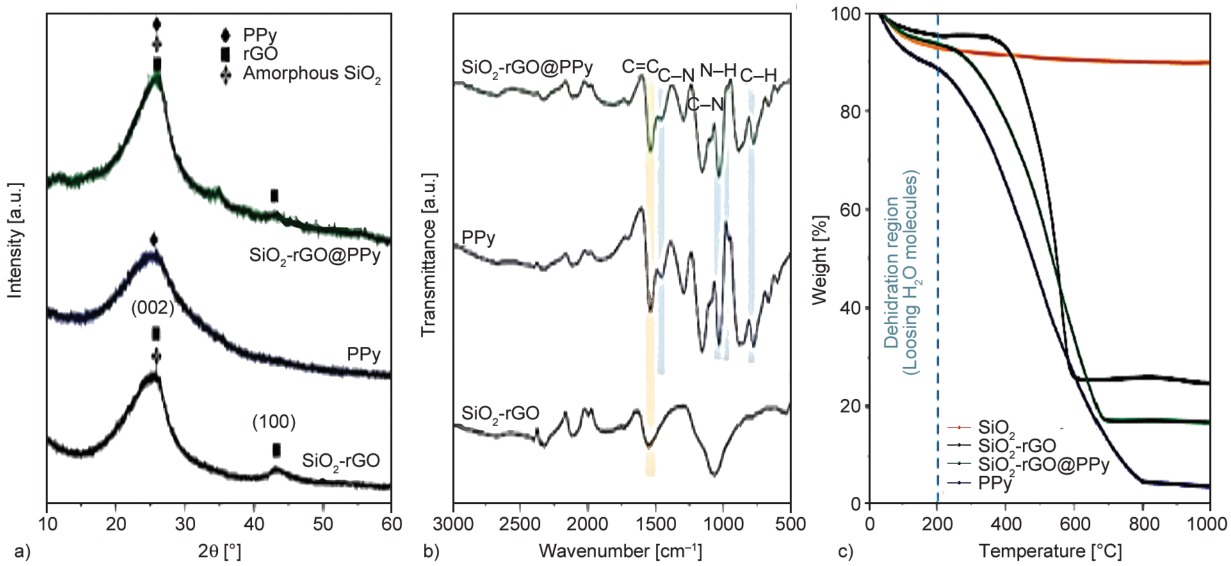

Figure 8. a) XRD, b) FTIR, and c) TGA for rice husk-derived nano silica-reduced graphene oxide@polypyrrole (SiO_2-rGO@PPy) [114].

shown in Figure 8a. The SiO_2-rGO_2 nanocomposite exhibits broad diffraction peaks at 2θ = 25.5 and 43.3°, corresponding to the (002) and (100) planes of graphene. A broad peak at 2θ = 25.4° indicates the presence of amorphous silica. The XRD spectrum of SiO_2-rGO@PPy is similar to that of pure PPy and SiO_2-rGO, with overlapping peaks that necessitated carrying out FTIR analysis for confirmation of phases and chemical structures [114].

Figure 8b, shows the FTIR spectra used to identify functional groups in the nanocomposites. For SiO_2-rGO peaks at 1066, 1541, and 1040 cm^{-1} are attributed to Si–O–C/Si–O–Si, C=C, and C–OH stretching vibrations, respectively. The SiO_2-rGO@PPy spectrum shows reduced peak frequencies, confirming the formation of the nanocomposite. Characteristic peaks for PPy are visible at 1541 cm^{-1} (C=C stretching) and 1456 cm^{-1} (C–N stretching), with additional peaks indicating C–C stretching (1292 cm^{-1}), C–N stretching (1157 cm^{-1}), and N–H deformation (1031 cm^{-1}).

Thermogravimetric analysis (TGA) (Figure 8c) shows the composition of SiO_2-rGO@PPy nanocomposites. The SiO_2 curve shows a 7.5 wt% weight loss around 200 °C due to water evaporation, with 92.5% weight remaining. Pure PPy burns completely by 800 °C, leaving 4.2 wt% residue. For the nanocomposite, two weight loss stages are observed: between 30–200 °C (water evaporation) and 200–680 °C (PPy oxidation). The composition of the nanocomposite is 33.9 wt% PPy, 17.4 wt% SiO_2, and 48.6 wt% rGO.

5.4. Mechanical characterization

Mechanical testing is performed to evaluate the mechanical properties, such as tensile strength, modulus, and elongation at break, of nanocomposites. Techniques such as tensile testing, flexural testing, and nanoindentation are commonly used to characterize the mechanical behaviour and determine the effects of nanofiller incorporation on the mechanical properties of the nanocomposite [115].

By integrating these characterization techniques, researchers can gain comprehensive insights into the structure-property relationships of polymer nanocomposites and optimize their formulation and processing parameters to enhance their performance for electrical energy storage applications.

6. Performance evaluation

For the evaluation of the electrical energy storage performance of polymer nanocomposites, several key parameters need to be assessed, including capacitance, charge-discharge efficiency, cyclic stability, and rate capability. Electrochemical techniques such as cyclic voltammetry (CV), galvanostatic charge-discharge (GCD), and electrochemical impedance spectroscopy (EIS) are commonly employed for this purpose.

6.1. Cyclic voltammetry

Cyclic voltammetry is a versatile electrochemical technique used to investigate the electrochemical behavior of materials over a range of applied potentials. By measuring the current response as a function of the applied potential, CV can provide information about the capacitance of the nanocomposite, as well as its redox activity and charge storage mechanism [116]. The basic setup, as described by Chooto [116], consists of a working electrode (where the redox reaction occurs), a reference electrode (with a stable and well-defined potential), and a counter electrode (to complete the circuit). The working electrode is usually made of a conductive material like platinum, gold, or glassy carbon. Bhattacharya *et al.* [117] stated that the current response in cyclic voltammetry can be described by Equation (1):

$$I = \frac{nFA\sqrt{Dc^*}}{\sqrt{nt}} \tag{1}$$

where I is the current [A], n is the number of electrons transferred in the redox reaction, F is Faraday's constant (96.485 C/mol), A is the electrode area [cm^2], c^* is the bulk concentration of the electroactive species [mol/cm^3], t is the time elapsed since the application of the potential step in [s] and D is the diffusion coefficient of the electroactive species [cm^2/s]. Equation (1) illustrates the dependence of the current on various experimental parameters, such as the concentration of the analyte, the diffusion coefficient, the electrode area, and the scan rate [117].

6.2. Galvanostatic charge-discharge testing

Galvanostatic charge-discharge testing involves applying a constant current to the nanocomposite electrode and monitoring the voltage response over time.

Both GCD and CV experiments typically require a three-electrode setup consisting of a working electrode, a reference electrode, and a counter electrode. This configuration allows for precise control of the electrochemical processes and accurate measurement of the potential or current response [118]. This technique allows for the determination of important performance metrics such as charge-discharge efficiency, charge/discharge time, and specific capacitance. The charge and discharge processes can be described by using Faraday's law of electrolysis. Equations (2) and (3) describe the charge and discharge processes, respectively:

$$Q = \int_0^t I\,\mathrm{d}t \tag{2}$$

where Q is the total charge passed during the charge process [C], I is the constant applied current [A], and t is the time [s].

$$Q = \int_0^t -I\,\mathrm{d}t \tag{3}$$

where Q is the total charge discharged during the discharge process [C], I is the constant applied current [A], and t is the time [s]. Equations (2) and (3) demonstrate that the total charge passed during the charge or discharge process is directly proportional to the applied current and the duration of the process. The charge-discharge efficiency reflects the effectiveness of the nanocomposite in storing and releasing electrical energy without significant losses.

6.3. Cyclic stability assessment

Cyclic stability assessment involves subjecting the nanocomposite to multiple charge-discharge cycles under specific operating conditions. By monitoring the capacitance retention and voltage stability over successive cycles, cyclic stability can be evaluated. A stable and consistent performance over numerous cycles is indicative of good cyclic stability, while degradation in capacitance or voltage indicates poor stability [119]. Capacity retention (CR) is a measure of how well the device maintains its initial charge storage capacity over repeated cycles. It is typically calculated using the following Equation (4) [120].

$$CF = \frac{C_{\text{final}}}{C_{\text{initial}}} \cdot 100\% \tag{4}$$

where C_{initial} is the initial charge storage capacity, C_{final} is the charge storage capacity after a certain number of cycles. Higher capacity retention indicates better cyclic stability [120].

6.4. Rate capability testing

Rate capability testing evaluates the ability of the nanocomposite to maintain its electrical energy storage performance at different charge/discharge rates. By varying the current density during charge-discharge cycles and monitoring the corresponding capacitance and voltage response, rate capability can be assessed. A nanocomposite with high-rate capability can efficiently store and release energy at both low and high charge/discharge rates [121].

Overall, the combination of electrochemical techniques such as cyclic voltammetry, galvanostatic charge-discharge, and electrochemical impedance spectroscopy provides a comprehensive assessment of the electrical energy storage performance of polymer nanocomposites. By analyzing the results obtained from these techniques, researchers can gain valuable insights into the electrochemical behavior, capacitance, efficiency, cyclic stability, and rate capability of the nanocomposite, facilitating the optimization of its design and formulation for specific electrical energy storage applications.

6.5. Optimization

Fine-tuning the composition and processing parameters is a crucial step in optimizing the electrical energy storage performance of polymer nanocomposites. Several key factors can be adjusted to achieve the desired properties and enhance overall performance.

6.5.1. Polymer-to-nanofiller ratio

The ratio of polymer to nanofiller in the composite significantly influences its properties. Increasing the nanofiller content can enhance electrical conductivity, charge storage capacity, and mechanical strength. However, excessive nanofiller loading may lead to aggregation, poor dispersion, and decreased processability. Fine-tuning the polymer-to-nanofiller ratio is essential to balance these factors and optimize performance. Akram *et al.* [122] investigated biomass fly ash (BFA) as a nanofiller to improve the dielectric properties of low-density polyethylene (LDPE) for possible high-voltage applications. The BFA-LDPE sheets were prepared using a conventional solvent mixing and subsequent hot-pressing process, incorporating BFA loadings ranging from 1% to 4 wt%. Among all the nanocomposites, 4 wt% BFA-LDPE exhibited the highest dielectric constant, with a value of 11.58, compared to simple LDPE that had a dielectric constant of 8.33. This improvement is ascribed to the synergistic effects of different inorganic metal oxides (SiO_2, MgO, and Fe_2O_3) present in BFA.

6.5.2. Nanofiller size and morphology

The size, shape, and morphology of nanofillers play a critical role in determining the properties of the nanocomposite. Smaller nanofiller size typically results in higher surface area and improved dispersion within the polymer matrix, leading to enhanced electrical conductivity and charge storage capacity, as demonstrated by Fang and Lafdi [123]. In addition, the morphology of nanofillers (*e.g.*, nanoparticles, nanotubes, or nanosheets) can influence mechanical properties and interfacial interactions with the polymer matrix [124].

6.5.3. Dispersion uniformity

Achieving uniform dispersion of nanofillers within the polymer matrix is essential for maximizing their effectiveness in enhancing electrical energy storage performance. Poor dispersion can lead to localized areas of high filler concentration, reducing mechanical strength and electrical conductivity [125]. Various processing techniques, such as sonication, mechanical mixing, or surface modification of nanofillers, can be employed to improve dispersion uniformity [126].

6.5.4. Processing conditions

Parameters such as temperature, pressure, mixing time, and solvent choice during fabrication significantly affect the morphology and properties of polymer nanocomposites. Optimizing processing conditions can help achieve better dispersion, interfacial adhesion, and overall performance [127]. For instance, controlling the processing temperature and shear rate during melt blending or adjusting the solvent evaporation rate during solution mixing can influence the final properties of the nanocomposite [128].

Adjusting these composition and processing parameters aids in tailoring the properties of polymer nanocomposites to meet specific electrical energy storage requirements. Optimization efforts aim to strike a balance between factors such as conductivity, capacitance, mechanical strength, and processability to achieve the desired performance characteristics for practical applications. In addition, advanced characterization techniques and computational modeling can provide valuable insights into the structure-property relationships and guide optimization efforts effectively.

6.5.5. Scale-up and application

Once an optimized nanocomposite formulation is achieved, scaling up the fabrication process for mass production becomes imperative to meet the increasing demand for electrical energy storage devices in practical applications. Transitioning from laboratory-scale synthesis to industrial-scale manufacturing requires careful consideration of several factors to ensure the reproducibility, efficiency, and quality of the nanocomposite materials. This involves not only upscaling the synthesis protocols but also addressing challenges related to material sourcing, equipment modification, process optimization, and quality control [129]. In scaling up the fabrication process, one of the primary considerations is selecting appropriate equipment and facilities capable of handling larger volumes and throughput rates. This may involve modifying existing laboratory-scale equipment or investing in specialized machinery tailored to the specific requirements of mass production. Equipment selection must take into account factors such as mixing efficiency, reaction kinetics, temperature control, and safety protocols to ensure consistent and uniform production of nanocomposites [130–132].

A crucial aspect of scaling up is optimizing the process parameters to achieve the desired product quality and performance at a larger scale. This may entail fine-tuning parameters such as mixing conditions, reaction times, temperature profiles, and solvent ratios to maintain the properties and functionality of the nanocomposite materials [133]. Process optimization is essential to minimize variability, improve efficiency, and reduce production costs, ultimately enhancing the competitiveness of the fabricated nanocomposites in the market [134].

In addition to process optimization, establishing a reliable supply chain for raw materials is paramount for mass production. Sourcing high-quality polymers, nanofillers, additives, and solvents from reputable suppliers ensures consistency and reliability in the performance of the nanocomposite materials. Moreover, securing a stable supply chain mitigates the risks associated with material shortages, price fluctuations, and quality variability, thereby ensuring uninterrupted production and timely delivery of electrical energy storage devices to the market.

Once the scaled-up fabrication process is established and validated, the optimized nanocomposites can be seamlessly integrated into a variety of electrical energy storage devices for practical applications. These include supercapacitors, batteries, and capacitive deionization systems, where the enhanced properties of the nanocomposites can significantly improve device performance and functionality. By leveraging the high surface area, conductivity, and mechanical strength of the nanocomposites, electrical energy storage devices can achieve higher energy density, faster charging/discharging rate, longer life cycle and improved stability, making them more efficient and reliable for diverse applications ranging from portable electronics to renewable electrical energy storage systems [93]. Overall, scaling up the fabrication process for mass production of optimized nanocomposites represents a critical step towards realizing their commercial potential and widespread adoption in electrical energy storage technologies.

Polymer/graphene nanocomposites are emerging as promising materials for electrical energy storage devices, such as supercapacitors and lithium-ion batteries, due to graphene's excellent electrical conductivity, large surface area, and mechanical strength. Recent studies have demonstrated how adjusting graphene content can optimize these composites for improved flexibility, conductivity, and mechanical properties. In lithium-ion batteries, such composites enhance interfacial stability and ionic conductivity, while in supercapacitors, they increase both energy and power density. These advancements make polymer/graphene nanocomposites ideal for applications in mobile devices, electric vehicles, and renewable energy systems, positioning them as key components in next-generation electrical energy storage technologies [93].

7. Conclusions and future prospects

The future prospects of polymer nanocomposites in electrical energy storage applications are promising, with advancements in material science opening new pathways to significantly enhance device performance. The integration of nanofillers within polymer matrices can substantially improve properties essential for energy storage, such as electrical conductivity, charge retention, mechanical robustness, and thermal stability. These enhancements collectively elevate the efficiency and longevity of electrical energy storage devices. Diverse fabrication techniques, including solution mixing, melt blending, *in-situ* polymerization, and electrospinning, enable precise control over the morphology and properties of the nanocomposites, making it possible to design materials tailored to specific electrical energy storage needs.

The advanced characterization techniques highlighted in this study – encompassing structural, electrical, thermal, and mechanical analyses – are instrumental in evaluating the dispersion, morphology, chemical composition, and thermal stability of nanocomposites. These insights guide the optimization of material formulations, paving the way for high-performance, durable electrical energy storage materials. Once optimized, scaling up fabrication processes is essential for large-scale production, aligning with the growing demand for efficient and sustainable energy solutions. Polymer nanocomposites offer significant potential for integration into a range of electrical energy storage systems, such as supercapacitors, batteries, and capacitive deionization devices. By enhancing core properties, these materials can improve the energy density, cycle stability, and operational flexibility of such devices. Continued collaboration among researchers, engineers, and industry stakeholders will be critical for advancing the commercialization of polymer nanocomposites, driving innovation, and supporting the transition to sustainable energy technologies. This trajectory positions polymer nanocomposites as key contributors to the future landscape of energy storage, with a transformative impact on both efficiency and sustainability in energy applications.

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