



Green Nanocomposites for Energy Storage

Ayesha Kausar 🕩

Nanosciences Division, National Center for Physics, Quaid-i-Azam University Campus, Islamabad 44000, Pakistan; dr.ayeshakausar@yahoo.com

Abstract: The green nanocomposites have elite features of sustainable polymers and eco-friendly nanofillers. The green or eco-friendly nanomaterials are low cost, lightweight, eco-friendly, and highly competent for the range of energy applications. This article initially expresses the notions of eco-polymers, eco-nanofillers, and green nanocomposites. Afterward, the energy-related applications of the green nanocomposites have been specified. The green nanocomposites have been used in various energy devices such as solar cells, batteries, light-emitting diodes, etc. The main focus of this artifact is the energy storage application of green nanocomposites. The capacitors have been recognized as corporate devices for energy storage, particularly electrical energy. In this regard, high-performance supercapacitors have been proposed based on sustainable nanocomposites. Consequently, this article presents various approaches providing key knowledge for the design and development of multifunctional energy storage materials. In addition, the future prospects of the green nanocomposites towards energy storage have been discussed.

Keywords: eco-friendly; nanomaterial; nanofiller; energy; supercapacitor



Citation: Kausar, A. Green Nanocomposites for Energy Storage. *J. Compos. Sci.* 2021, *5*, 202. https:// doi.org/10.3390/jcs5080202

Academic Editors: Patrizia Bocchetta and Domenico Frattini

Received: 5 July 2021 Accepted: 30 July 2021 Published: 2 August 2021

Publisher's Note: MDPI stays neutral with regard to jurisdictional claims in published maps and institutional affiliations.



Copyright: © 2021 by the author. Licensee MDPI, Basel, Switzerland. This article is an open access article distributed under the terms and conditions of the Creative Commons Attribution (CC BY) license (https:// creativecommons.org/licenses/by/ 4.0/).

1. Introduction

Green or eco-friendly or eco-polymers are named due to their environmentally friendly nature and production from some renewable resources [1,2]. These polymers are usually biodegradable or compostable [3–5]. Initial research has focused on the use of green polymers [6–8]. Later research has turned towards the formation of green nanocomposites using eco-polymers [9,10].

Green or eco-friendly nanomaterials have several advantages of sustainability, low cost, eco-friendliness, and high performance [11–13]. Different natural and synthetic green polymers and nanofillers have been used to develop green nanomaterials [14]. In the green polymeric nanocomposites, the features of both the green polymers and econanofillers have been incorporated in the high-performance materials [15]. Use of the natural and synthetic green or eco-polymers or eco-friendly polymers and related materials have extended their use in various industrial fields [16–18]. Eco-friendly nanofillers used with the eco-polymers are also environmentally friendly. The eco-nanofillers include the use of metal nanoparticles, polymer nanoparticles, nanoclays, inorganic nanoparticles, carbon nanoparticles, and a similar range of other nanoparticles. The derived nanocomposites from eco-friendly polymers and eco-nanofillers are currently known as green polymeric nanocomposites. The synergistic effects of the eco-friendly polymers and green nanofillers have resulted in several enhanced physical properties, eco-friendliness, and biodegradability of the resulting green nanocomposites [19,20]. The properties of the green nanocomposites usually rely on the nanofiller content, processing technique, and matrixfiller interactions. The green nanocomposites have found varying solicitations in energy devices, electronics, aerospace, packaging, environmental, and biomedical applications. The energy storage devices are the most demanding expedients in the energy sector due to the environmental glitches [21,22]. These devices provide a solution to the use of unverifiable energy sources such as petroleum or coal. In other words, green nanocomposites are alternatives to the pollution-causing energy sources. The most common type of energy

storage devices are capacitors [23]. Among various types of capacitors, supercapacitors are the most efficient ones. Supercapacitors have been researched for their lightweight, durability, and enrichment of energy storage, energy density, and specific capacitance characteristics. The green nanocomposites have expanded research inquisitiveness due to their inexpensiveness, light weight, sustainability, biodegradability, and recyclability properties [24,25]. The eco-friendly nanocomposites have been utilized in numerous engineering applications in the energy sector especially energy storage devices [26–28]. Green polymeric nanocomposite reveals high-performance energy storage, however, their use in advanced energy applications is still challenging [29,30].

In this review, the use of green nanofillers and green polymers in green nanocomposites has been enlightened. This review has been developed focusing on the energy storage applications of sustainable nanocomposites. This article also presents the future prospects of the multi-functional next-generation green nanocomposites. Since revealing the importance of green polymeric nanocomposite, this review has focused on sustainable polymers, nanofillers, and the resulting nanocomposites. To the best of our knowledge, this review paper is novel in the literature due to the originality of the outline and the included literature compared with the previous literature reviews [31,32]. This review is all-inclusive and aims to include the essential technical and commercial developments of green nanocomposites in the energy sector.

2. Green or Eco-Friendly Nanocomposites

Among renowned eco or green polymers is a range of synthetic and natural polymers such as poly(vinyl alcohol), poly(ethylene glycol), poly(ethylene oxide), poly(lactic acid), polyamide, polycarbonate, polyurethane, cellulose, starch, etc. (Table 1) [33].



Table 1. Some green synthetic and natural polymers.



Table 1. Cont.

The green polymers have been prepared through several green synthesis methodologies [34,35]. The green polymers have found applications in adhesives [36], membranes [2], coatings [37], drug delivery [38], and other biomedical applications [39,40]. Green nanofillers used with the polymers are mostly biodegradable in nature. Figure 1 shows a few green nanofillers. Chitosan has been used as a successful green nanofiller [41,42]. Lignin has also played an important role as a green nanofiller [43,44]. Lignin has been used as a nanofiller in several synthetic and natural polymer matrices such as polystyrene, polyethylene, poly(ethylene oxide), poly(vinyl chloride), polyester, and poly(lactic acid), etc. [45,46]. Phyllosilicate nanoclays such as montmorillonite have gained considerable research attention [47,48]. Montmorillonite is a well-known ecological nanofiller [49–51]. Nanoclays have been used with biodegradable polymers to form green systems [52,53]. Among carbon nanofillers, graphene, graphene oxide, and carbon nanotube nanofillers have been widely used with green polymers [54–56]. These nanofillers have enhanced the heat stability, mechanical features, charge transport, thermal conductivity, flame retardancy, antimicrobial features, and biodegradability of the green nanocomposites.



Figure 1. Green nanofillers.

Polyethylene glycol (PEG) is a water-soluble non-hazardous polymer [57–59]. It is also considered a green polymer. The PEG has been reinforced with nanofillers to form nanocomposites [60]. The glass transition temperature of PEG has been found to alter with the addition of additives and nanofillers [61]. In addition, additives and nanofillers have been used to enhance the mechanical properties of the PEG-based nanocomposites [62]. Cavallaro et al. [63] prepared green nanocomposite based on PEG and halloysite nanotubes. The dispersion properties of halloysite nanotubes in the PEG matrix have been studied. The halloysite nanotubes were found to enhance the mechanical properties of the PEG matrix. Due to barrier properties provided by the halloysite nanotubes, the green nanocomposite was used for packaging purposes. Gopi et al. [64] prepared polyethylene glycol and turmeric nanofibers (TNF)-based nanocomposite. The TNF was used as reinforcement in gum arabic (GA) and maltodextrin (MDX). The nanocomposites have been prepared through a multi-step process. Figure 2 shows the reinforcement effect of the TNF nanofibers in the PEG matrix and GA and MDX matrices. The TNF was loaded in 1–7 wt.% contents in PEG and other matrices using a solution blending method. The PEG-TNF nanocomposite has shown fine nanoparticle dispersion and interfacial adhesion through hydrogen bonding interactions. The TNF nanofiller loading up to wt.% was found to enhance the tensile strength and Young's modulus of the nanocomposites to 5.12 MPa and 49.36 MPa, respectively. The neat matrix has lower tensile strength and Young's modulus of 1.84 and 19.76 MPa, respectively. Moreover, the PEG-TNF nanocomposites revealed fine antibacterial activity against Escherichia coli, Staphylococcus aureus, and other bacterial strains. The TNF nanofillers were found useful for creating a reinforcement effect and interfacial interactions in the nanocomposite matrix, thus increasing the overall mechanical properties. Hence, the PEG-TNF nanocomposites were effective in improving the mechanical properties and antibacterial activity for the relevant uses.



Figure 2. Schematic of the preparation steps for turmeric nanofiber-based nanocomposites [64]. Reproduced with permission from Elsevier.

Poly(lactic acid) (PLA) is also a natural green polymer [65–67]. It is biodegradable polyester gotten from starch. Krikorian et al. [68] developed green nanocomposite based on PLA and nanoclay nanofillers. The green PLA/nanoclay nanocomposites have fine biodegradability, crystallinity, and storage modulus properties. The XRD patterns were used to study the effect on the organoclay crystallization within the PLA matrix. Moreover, the storage modulus of the nanocomposites was found to vary with different nanoclay loadings in the range of 20-150 °C. The storage modulus for 15 wt.% nanoclay loading was enhanced by 61.4%, relative to neat PLA. Wang et al. [69] prepared poly(lactic acid) and nanocellulose crystal (NCC) based poly(lactic acid)/nanocellulose crystal (PLA/NCC) nanocomposite. Neat PLA had a tensile strength of 41.9 MPa, while NCC inclusion enhanced the property up to 53.9 MPa. Table 2 shows the crystallinity results for the PLA/NCC nanocomposite. The XRD analysis of PLA and NCC has shown an increase in the crystallinity of the polymer with the nanofiller loading. The neat PLA had crystallinity of 32.6%. The 2 wt.% NCC has shown the highest crystallinity of 37.8% among the nanocomposite samples. The increase in the crystallinity of the nanocomposite with the NCC loading was due to better alignment and packing of the nanocellulose crystals in the polymer matrix, thus causing the crystallinity.

Table 2. The crystallinity of PLA and PLA/NCC [69]. PLA = poly(lactic acid); PLLA/NCC = poly(lactic acid)/nanocellulose crystal. Reproduced with permission from Elsevier.

	PLA	1 wt.% NCC	2 wt.% NCC	3 wt.% NCC	4 wt.% NCC
% Crystallinity of PLA	32.6	37.3	37.8	35.7	34.1

Li et al. [70] also formed poly(L-lactic acid) (PLLA), PEG, and cellulose nanocrystal (CNC)-based PLLA/PEG/CNC bionanocomposites. The CNC was used to enhance the crystallization behavior of the PLLA/PEG/CNC nanocomposites. Figure 3 shows the crystalline morphology of the pristine PLLA and PLLA/PEG/CNC nanocomposite. The low crystallinity was observed in the neat PLLA matrix with few spherulites. In the PLLA/PEG matrix, the inclusion of CNC enhanced the interfacial interactions between the polymer and the nanofillers and also the crystallinity. Consequently, the crystallinity



promoted the formation of convoluted paths in the nanocomposites. The diffusion of the molecules and permeation through the system were enhanced through the nanocomposites.

Figure 3. Schematic representation showing crystalline morphological features in (**a**) neat PLLA; (**b**) PLLA/PEG/CNC with 2 g nanofiller; and (**c**) PLLA/PEG/CNC with 1 g nanofiller governing oxygen permeation. The lines represent the diffusion paths of O₂ [70]. PLLA = poly(L-lactic acid); PLLA/PEG/CNC = poly(L-lactic acid) (PLLA)/poly(ethylene glycol)/cellulose nanocrystal. Reproduced with permission from Elsevier.

Poly(vinyl alcohol) (PVA) is a green eco-polymer. It has fine biodegradability and water solubility. Ibrahim et al. [71] prepared PVA and carboxymethyl cellulose-based nanocomposites. The green nanocomposites have also been used for wound healing and non-toxicity applications [72]. Zhao et al. [73] developed PVA and carboxymethyl chitosan (CMC)-based green matrix filled with silver nanoparticles. The PVA/CMC has shown antibacterial properties. Morsi et al. [74] also designed a PVA/CMC green matrix filled with Au nanoparticles. The green PVA/CMC nanocomposites have fine electrical conductivity and dielectric permittivity to be employed for microelectronic devices.

Green nanocomposites based on starch and nanofillers have been reported [75]. Starch is also an eco-polymer, which has been used with nanofillers [76,77]. Kaushik et al. [78] formed starch and cellulose nanofibrils-based nanocomposites. These nanofibrils were dispersed in a starch matrix via a Fluko high shear mixer. The reinforcing effect of the nanofillers was observed. Neat matrix has a tensile modulus of 76 MPa, whereas a 15 wt.% loading enhanced the property to 224 MPa. Cheviron et al. [79] prepared starch and silver nanoparticles-based nanocomposites. Such green nanocomposites have been used for packaging, antimicrobial, and sensing applications. Lignin is a significant engineering eco-polymer gained from the natural sources [80–82]. Lignin and lignin fibers both have been used in the green materials [83–85].

3. Energy Applications of Green Nanocomposites

Initially, the eco-nanocomposites have been used in the fabrication of turbines [86–88]. The turbine blades were constructed using natural composite having high strength, low cost, and lightweight. Despite the traditional composites for wind turbine blades such as metal and epoxy materials, natural composites based on green polymers and hybrids have been used [89]. Afterward, bio-polymer-based nanocomposite has been used in the optoelectronics industry [90–92]. The eco-polymer-derived donor-acceptor structures have been prepared for photo energy conversion [93,94]. Eco-polymers have been used in light-emitting diode devices (LED) [95-97]. Chen et al. [98] primed a polydimethylsiloxane (PDMS) and zinc sulfide (ZnS)-derived LED. The ZnS nanoparticles were used as green nanofillers in LED. The PDMS/ZnS-based green nanocomposite has shown a fine luminescence spectrum. The ZnS nanoparticles were also used in solar cells as sustainable nanofiller. Thus, the solar cell devices have also incorporated green nanocomposites [99–101]. Ghosh et al. [102] formed poly(vinylidenefluoride-cohexafluoropropylene) and platinum nanoparticles-based green materials for solar cells. The platinum nanoparticles were used in an optimum amount in the green matrix in solar cells. The poly(vinylidenefluoride-co-hexafluoropropylene)/platinum nanoparticle

showed an open-circuit voltage of 2.7–23 V and short-circuit current of 2.9–24.7 μ A. Zhang et al. [103] designed green nanocomposites-based energy conversion devices i.e., thermoelectric generators. The organic polymers-based thermoelectric materials were used for green energy conversion. Most importantly, polyanilines, polypyroles, polythiophenes, and poly(3,4-ethylenedioxythiophene) have been used. Recently, bio-based conducting polymers and nanocomposites have been adopted in the photovoltaics and optoelectronics industries [104,105]. The inclusion of green nanofillers in p-type conjugated polymers may develop donor-acceptor heterostructures for photo energy conversion [106]. In this regard, Zhuang et al. [107] investigated the photophysical properties of the green conjugated polymer-based materials. Green composites have been useful in these devices to improve eco-friendliness and steadfastness [108]. Concisely, the green nanocomposites have found solicitations in various areas of the energy sector such as optoelectronics, supercapacitor, nanogenerators, and other electrical devices.

4. Energy Storage Using Green Nanocomposites

Research has been turned towards consistent electrical energy storage resolutions [109,110]. There are several electrical storage practices that have been adopted for chemical, magnetic, or electrical energy storage such as batteries, solid oxide fuel cells (SOFCs), superconducting magnetic energy storage (SMES) devices, and electrostatic/electrochemical capacitors [111–113]. Among all these systems, capacitors have been found reliable for a reasonable cost, low operating voltage, high power density, and sweeping applications [114]. To present the comparison of different energy storage devices, a Ragone plot is given in Figure 4. Different energy storage devices have their individual characteristic times [109]. The capacitors have fairly high power density and charge/discharge rates relative to SOFCs and batteries. The capacitors have been applied in electronic circuits, electrical vehicles, power systems, and green energy storage systems. The supercapacitor is a significant type of capacity energy storage device [115–117]. To improve eco-friendliness, green nanocomposites and nanomaterials have been used in supercapacitors. In this regard, green synthesis methods have been adopted to form nanocomposites. However, to develop the purely green nanocomposites, mostly green polymers, and green nanofillers have been used. Several attempts have been made towards the formation of nanocomposites using the green method. Çıplak et al. [118] prepared polyaniline (PANI), graphene oxide (GO), reduced graphene oxide (rGO), and gold (Au) nanoparticle-based GO-Au@PANI and rGO-Au@PANI nanocomposites. Figure 5 shows the formation of rGO-Au@PANI nanocomposite using the green method. Initially, GO was converted to rGO. Then, the Au nanoparticles and aniline monomer was adsorbed on the surface of rGO. The Au@PANI was formed in situ. The polyaniline was deposited consistently on the rGO nanosheet surface through in situ polymerization. The electrostatic interactions existed among the gold nanoparticles and rGO nanosheet. The π - π interactions existed among the PANI and rGO. The pristine PANI, GO-Au@PANI, and rGO-Au@PANI nanocomposite electrodes have a specific capacitance of 17.6, 42.5, and 63.5%, respectively. Figure 6 shows the dependence of the specific capacitance on the scan rate of the nanocomposites. The rGO-Au@PANI nanocomposite had high specific capacitance of 212.8 Fg^{-1} at current density of 1 Ag^{-1} .



Figure 4. Ragone plot of different energy storage devices: electrostatic capacitors, electrochemical capacitors, SMES flywheels, batteries, and SOFCs [119]. SMES = superconducting magnetic energy storage; SOFCs = solid oxide fuel cells. Reproduced with permission from Elsevier.



Figure 5. Schematic representation of the preparation of rGO-Au@PANI nanocomposite [118]. rGO-Au@PANI = reduced graphene oxide-gold nanoparticle@polyaniline. Reproduced with permission from Elsevier.



Figure 6. Dependence of specific capacitance on the scan rate (5–200 mVs⁻¹) for pristine PANI, GO-Au@PANI, and rGO-Au@PANI [118]. PANI = polyaniline; GO-Au@PANI = graphene oxide-gold nanoparticle@polyaniline; rGO-Au@PANI = reduced graphene oxide-gold nanoparticle@polyaniline. Reproduced with permission from Elsevier.

Arthisree et al. [120] prepared graphene quantum dot (GQD) doped polyacrylonitrile (PAN) and polyaniline-based PAN/PANI@G nanocomposite. Figure 7 shows the prototype supercapacitor composed of PAN/PANI@G prepared using the green method. The nanocomposite electrode with 1.5 wt.% GQD was formed by sandwiching the PAN/PANI@G between NaCl solution and aluminium foil. The supercapacitor had 1.4 V output power for 60 min working time. The PAN/PANI@G nanocomposite with 1.5 wt.% GQD has shown high specific capacitance. The specific capacitance was found in the range of 105–587 Fg⁻¹. The capacitance values were found higher than the neat polyaniline-based supercapacitor electrode [121]. Green approaches have been used to incorporate the inorganic nanoparticles in nanocomposite electrodes [122,123]. Chakraborty et al. [124] primed styrene-maleic anhydride copolymer and ZnO nanoparticle-based nanocomposite for a supercapacitor.



Figure 7. Illustration for PAN/PANI@G 1.5 wt.% based supercapacitor and its typical digital photograph of optimal nanocomposite verified for voltage generation [120]. PAN/PANI@G = polyacrylonitrile/polyaniline/graphene quantum dot. Reproduced with permission from Elsevier.

Figure 8 shows the specific capacitance of the nanocomposites with varying current densities. The specific capacitance was increased from 145 Fg^{-1} to 268.5 Fg^{-1} . Ceramic nanofillers like BaTiO₃ have also been used in the nanocomposite electrodes [125]. High-performance supercapacitors have been designed using high electrical conductivity, consistency, and optimum processing parameters. In this regard, novel nanocomposites need to be used to fabricate supercapacitors for integrated circuits and other devices.



Figure 8. Specific capacitance versus current density of nanocomposite at 0.1 Ag⁻¹. (0.1 g ZnO nanoparticles) [124]. Reproduced with permission from Elsevier.

As discussed above, carbon materials (carbon nanotube, graphene, etc.) and conducting polymers have been commonly used for the supercapacitor electrodes [126]. Along with the conducting polymers and carbon nanomaterials, the transition metal oxides or hydroxides such as NiO, MnO₂, Ni(OH)₂ have also been used. To make the electrode materials green, one method is the use of regenerated cellulose aerogel [127]. The regenerated cellulose aerogel has also been prepared in combination with graphene oxide. Thus, the green supercapacitor electrodes consist of both cellulose aerogel and graphene oxide. The supercapacitors with green electrodes of the regenerated cellulose/graphene oxide aerogel have shown the moderate specific capacitance of 71.2 Fg^{-1} . Furthermore, it is essential to incorporate the conducting polymers in the regenerated cellulose/graphene oxide aerogel to improve the specific capacitance of the devices [128]. Thus, the graphene oxide and conducting polymers have been converted into green electrically conductive aerogels [129]. A very common method is to mix the GO solution with the solution of cellulose and conducting polymer. In another study, Tian et al. [130] prepared the green nanocomposites through the in situ polymerization of aniline monomer on porous cellulose scaffolds. Later, the Ag nanoparticles were deposited on the green electrodes using the electrodeposition process. Zu et al. [131] formed green electrodes using high surface area carbon and cellulose aerogels. The pyrolysis method was used. The electrode with porous interconnected nanostructure had revealed a high capacitance of 1873 m² g⁻¹. The aerogel had specific capacitances of 302 Fg^{-1} . Yang et al. [132] used bamboo cellulose fibers with the regenerated cellulose and formed aerogel-based green electrodes. The high specific capacitance of 381 Fg^{-1} was attained. Besides, the cellulose aerogels have been doped with nitrogen or sulfur to enhance the capacitance properties of the green supercapacitor electrodes [133,134]. The cellulose has also interacted with the metal oxides, metal hydroxides, and conducting polymers to form green electrodes [135]. There is a need for the introduction of metal or carbon nanoparticles in cellulose to decrease the rigidity of the aerogels. Several attempts have been made on the use of biomaterials-derived green electrodes for supercapacitor application [136]. Table 3 shows green electrode-based supercapacitors derived from biomaterials. The supercapacitors had high flexibility, cyclability, and good specific capacitance of up to 330 Fg⁻¹. However, the green nanocomposite electrodes need to be further researched to enhance the physical characteristics, capacitance, charge density, and electrochemical properties to meet the necessities of high performance supercapacitor electrodes.

Green Material	Specific Capacitance (Fg ⁻¹)	Reference	
Conjugated polymer	212.8	[118]	
Conjugated polymer	105–587	[120]	
Synthetic co-polymer	145-268.5	[124]	
Cellulose	71.2	[127]	
Conjugated polymer	302	[131]	
Cellulose	381	[132]	
Doped cellulose	>100-300	[133,134]	
Starch	168	[137]	
Starch	304	[138]	
Gelatin	183	[139]	
Cellulose	242	[140]	
Cellulose	330	[141]	
Carbohydrate	213	[142]	
Carbohydrate	300	[143]	

 Table 3. Performance of green supercapacitor electrode materials.

5. Advantages/Disadvantages of Green Nanocomposites in Energy Storage

The use of non-green electrodes in supercapacitors may involve environmental pollution, material degradation, high chemical consumption, high toxicity, and high cost. On the other hand, the benefits of using green nanocomposites in energy storage devices are environmental friendliness, high stability of material such as cellulose, electrode preparation at room temperature, and no use of harmful or toxic solvents. Such nanocomposite electrodes have a high dissolving capacity of green materials, insignificant volatility, structural tunability, non-flammability, recoverability, high recycling rate, high capacitance, and high mechanical properties. However, there are some disadvantages of green nanocomposites in energy storage devices. First of all, the capacitance of pure green nanomaterials is often low. There is a need to increase the capacitance of the green electrodes using the non-green conducting polymers and nanocarbons. Some hybrid nanostructures have been prepared using cellulose and graphene oxide nanomaterials. Fewer combinations of nanocarbons with green nanomaterials have been identified so far. The most successful one is the amalgamation of cellulose with the graphene derivatives and conducting polymers. The essential understanding of the structure-property relationships of various types of green polymers and green nanofillers for supercapacitors is valuable. Research in green supercapacitors is an emerging and promising field awaiting future attention.

6. Future and Summary

Effectual electrical energy storage resolutions are keys to future electricity generation problems. The capacitors or supercapacitors have wide-ranging applications in renewable microwave devices, energy storage devices, electronic circuits, electrical vehicles, telecommunication, and other maneuvers and systems. Application fields of capacitors are given in Figure 9. The green nanocomposites prepared renewable resources and through green strategies have been researched for anticipated physical properties, low cost, and facile processing [144]. For high-performance applications, green nanomaterials must have

fine morphology, crystallinity, electrical conductivity, thermal stability, and mechanical strength.

In this regard, better structural interactions and compatibility of the nanocomposites are essential [145–147]. The use of green nanofibers such as chitosan, lignin, starch, cellulose, and nanocarbon-based nanofillers in the green polymers may improve the nanocomposite performance for future solicitations [148,149]. Green materials have been continuously applied in supercapacitors to enhance reliability and charge storage performance. Several approaches have been applied to advance the performance of supercapacitors. The enhancement of the dielectric properties and capacitance may decrease the strength properties of the green nanocomposites. Thus, the interactions in the nanocomposites need to be improved to enhance the charge storage mechanism and energy density of these materials. Aggregation may cause problems in the nanofillers dispersion, nanocomposite formation, and the extent of the electric field generated in the green matrix. Interactions among the matrix and the nanofiller may be electrostatic, covalent interaction, hydrogen bonding, and other interactions form the homogeneous nanocomposites. The comprehensive attempts on the high-performance of green nanomaterials are desirable to exploit the true energy storage potential of these materials [150,151].



Electronic circuits



Microwave communication



High-power applications





Hybrid vehicles



Renewable energy



Distributed power system

Figure 9. Application fields of capacitors [119]. Reproduced with permission from Elsevier.

This review states the development in the field of green nanocomposites for energy storage applications. The inclusion of eco-nanofillers in eco-polymers has led to highperformance green nanocomposites. The energy performance of green nanocomposites depends on the selection of green polymer matrices, eco-nanofiller, and green synthesis methods. The energy storage properties are reliant on the morphology, crystallinity, matrixfiller interaction, electrical conductivity, dielectric properties, capacitance, charge density, charge/discharge ratio, and several other advanced features. The technical applications of green polymeric nanocomposites have been experiential for energy devices including solar cells, electronics, LED, nanogenerators, and energy storage devices such as capacitors and supercapacitors.

Funding: This research received no external funding.

Conflicts of Interest: The authors declare no conflict of interest.

References

- Gomez, J.G.; Méndez, B.S.; Nikel, P.I.; Pettinari, M.J.; Prieto, M.A.; Silva, L.F. Making green polymers even greener: Towards sustainable production of polyhydroxyalkanoates from agroindustrial by-products. In *Advance Applied Biotechnology*; BoD—Books on Demand: Norderstedt, Germany, 2012; pp. 41–62.
- Iordanskii, A.; Kamaev, P.; Ol'khov, A.; Wasserman, A. Water transport phenomena in 'green' and 'petrochemical' polymers. Differences and similarities. *Desalination* 1999, 126, 139–145. [CrossRef]
- 3. Zhong, Y.; Godwin, P.; Jin, Y.; Xiao, H. Biodegradable polymers and green-based antimicrobial packaging materials: A mini-review. *Adv. Ind. Eng. Polym. Res.* 2020, *3*, 27–35. [CrossRef]
- Popuri, S.R.; Hall, C.; Wang, C.-C.; Chang, C.-Y. Development of green/biodegradable polymers for water scaling applications. *Int. Biodeterior. Biodegrad.* 2014, 95, 225–231. [CrossRef]
- 5. Virkutyte, J.; Varma, R.S. Green synthesis of metal nanoparticles: Biodegradable polymers and enzymes in stabilization and surface functionalization. *Chem. Sci.* 2011, 2, 837–846. [CrossRef]
- 6. De Graaf, L.A.; Kolster, P. Industrial proteins as a green alternative for 'petro'polymers: Potentials and limitations. In *Macromolecular Symposia*; Wiley Online Library: Hoboken, NJ, USA, 1998.
- 7. Romaner, L.; Pogantsch, A.; Scandiucci de Freitas, P.; Scherf, U.; Gaal, M.; Zojer, E.; List, J.W. The origin of green emission in polyfluorene-based conjugated polymers: On-chain defect fluorescence. *Adv. Funct. Mater.* **2003**, *13*, 597–601. [CrossRef]
- 8. Fertier, L.; Koleilat, H.; Stemmelen, M.; Giani, O.; Joly-Duhamel, C.; Lapinte, V.; Robin, J.J. The use of renewable feedstock in UV-curable materials–A new age for polymers and green chemistry. *Prog. Polym. Sci.* **2013**, *38*, 932–962. [CrossRef]
- 9. Tan, N.P.B.; Lee, C.H.; Li, P. Green synthesis of smart metal/polymer nanocomposite particles and their tuneable catalytic activities. *Polymers* **2016**, *8*, 105. [CrossRef]
- 10. Modi, V.K.; Shrives, Y.; Sharma, C.; Sen, P.K.; Bohidar, S.K. Review on green polymer nanocomposite and their applications. *Preservation* **2014**, *3*, 17651–17656.
- 11. Stewart, R. Going green: Eco-friendly materials and recycling on growth paths. Plast. Eng. 2008, 64, 16–24. [CrossRef]
- 12. Moustafa, H.; Youssef, A.M.; Darwish, N.A.; Abou-Kandil, A.I. Eco-friendly polymer composites for green packaging: Future vision and challenges. *Compos. Part B Eng.* **2019**, *172*, 16–25. [CrossRef]
- 13. Kausar, A. Progress in green nanocomposites for high-performance applications. Mater. Res. Innov. 2021, 25, 53-65. [CrossRef]
- 14. Sakthieswaran, N.; Sophia, M. Prosopis juliflora fibre reinforced green building plaster materials—An eco-friendly weed control technique by effective utilization. *Environ. Technol. Innov.* **2020**, *20*, 101158.
- 15. Bakhoum, E.; Garas, G.; Allam, M. Sustainability analysis of conventional and eco-friendly materials: A step towards green building. *ARPN J. Eng. Appl. Sci.* **2015**, *10*, 788–796.
- Chemat, F.; Rombaut, N.; Meullemiestre, A.; Turk, M.; Perino, S.; Fabiano-Tixier, A.-S.; Abert-Vian, M. Review of green food processing techniques. Preservation, transformation, and extraction. *Innov. Food Sci. Emerg. Technol.* 2017, 41, 357–377. [CrossRef]
- 17. Ahmed, J.; Varshney, S.K. Polylactides—chemistry, properties and green packaging technology: A review. *Int. J. Food Prop.* **2011**, 14, 37–58. [CrossRef]
- Liu, J.; Feng, Y.; Zhu, Q.; Sarkis, J. Green supply chain management and the circular economy: Reviewing theory for advancement of both fields. *Int. J. Phys. Distrib. Logist. Manag.* 2018, 48, 794–817. [CrossRef]
- 19. Zhang, L.; Zhang, F.; Liu, M.; Hu, X. Novel sustainable geopolymer based syntactic foams: An eco-friendly alternative to polymer based syntactic foams. *Chem. Eng. J.* 2017, 313, 74–82. [CrossRef]
- 20. Ahmadi., Y.; Ahmad, S. Surface-active antimicrobial and anticorrosive Oleo-Polyurethane/graphene oxide nanocomposite coatings: Synergistic effects of in-situ polymerization and *π*-*π* interaction. *Prog. Org. Coat.* **2019**, *127*, 168–180. [CrossRef]
- 21. Chen, S.; Skordos, A.; Thakur, V.K. Functional nanocomposites for energy storage: Chemistry and new horizons. *Mater. Today Chem.* **2020**, *17*, 100304. [CrossRef]
- 22. Wang, H.; Yao, C.-J.; Nie, H.-J.; Yang, L.; Mei, S.; Zhang, Q. Recent progress in integrated functional electrochromic energy storage devices. *J. Mater. Chem.* C 2020, *8*, 15507–15525. [CrossRef]
- 23. Dell, R.M.; Rand, D.A.J. Energy storage—A key technology for global energy sustainability. J. Power Sources 2001, 100, 2–17. [CrossRef]
- 24. Mohan, T.; Kanny, K. Green Nanofillers for Polymeric MaterialsIn Green Nanomaterials; Springer: Singapore, 2020; pp. 99–138.

- Leung, S.N.; Ghaffari, S.; Naguib, H.E. Development of novel multifunctional biobased polymer composites with tailored conductive network of micro-and-nano-fillers. In *Behavior and Mechanics of Multifunctional Materials and Composites 2013*; International Society for Optics and Photonics: Bellingham, WA, USA, 2013.
- 26. Halley, P.; Avérous, L. Starch Polymers: From Genetic Engineering to Green Applications; Newnes: London, UK, 2014.
- 27. Yang, C.; Wei, H.; Guan, L.; Guo, J.; Wang, Y.; Yan, X.; Zhang, X.; Wei, X.; Guo, Z. Polymer nanocomposites for energy storage, energy saving, and anticorrosion. J. Mater. Chem. A 2015, 3, 14929–14941. [CrossRef]
- 28. Siwal, S.S.; Zhang, Q.; Devi, N.; Thakur, V.K. Carbon-based polymer nanocomposite for high-performance energy storage applications. *Polymers* **2020**, *12*, 505. [CrossRef] [PubMed]
- 29. Aigbodion, V.; Okonkwo, E.; Akinlabi, E. Eco-friendly polymer composite: State-of-arts, opportunities and challenge. In *Sustainable Polymer Composites and Nanocomposites*; Springer: Berlin/Heidelberg, Germany, 2019; pp. 1233–1265.
- Mooney, M.; Nyayachavadi, A.; Rondeau-Gagné, S. Eco-friendly semiconducting polymers: From greener synthesis to greener processability. J. Mater. Chem. C 2020, 8, 14645–14664. [CrossRef]
- 31. Hu, X.; Wei, L.; Chen, R.; Wu, Q.; Li, J. Reviews and Prospectives of Co₃O₄-Based Nanomaterials for Supercapacitor Application. *ChemistrySelect* **2020**, *5*, 5268–5288. [CrossRef]
- 32. Yang, H. A review of supercapacitor-based energy storage systems for microgrid applications. In Proceedings of the 2018 IEEE Power & Energy Society General Meeting (PESGM), Portland, OR, USA, 5–10 August 2018.
- Tabone, M.D.; Cregg, J.J.; Beckman, E.J.; Landis, A.E. Sustainability metrics: Life cycle assessment and green design in polymers. Environ. Sci. Technol. 2010, 44, 8264–8269. [CrossRef]
- 34. Wood, C.D.; Cooper, A.I.; DeSimone, J.M. Green synthesis of polymers using supercritical carbon dioxide. *Curr. Opin. Solid State Mater. Sci.* 2004, *8*, 325–331. [CrossRef]
- Bagheri, A.R.; Arabi, M.; Ghaedi, M.; Ostovan, A.; Wang, X.; Li, J.; Chen, L. Dummy molecularly imprinted polymers based on a green synthesis strategy for magnetic solid-phase extraction of acrylamide in food samples. *Talanta* 2019, 195, 390–400. [CrossRef] [PubMed]
- Yuan, C.; Chen, M.; Luo, J.; Li, X.; Gao, Q.; Li, J. A novel water-based process produces eco-friendly bio-adhesive made from green cross-linked soybean soluble polysaccharide and soy protein. Carbohydrate polymers. *Carbohydr. Polym.* 2017, 169, 417–425. [CrossRef]
- Marti, M.; Molina, L.; Aleman, C.; Armelin, E. Novel epoxy coating based on DMSO as a green solvent, reducing drastically the volatile organic compound content and using conducting polymers as a nontoxic anticorrosive pigment. *ACS Sustain. Chem. Eng.* 2013, 1, 1609–1618. [CrossRef]
- 38. Jahangirian, H.; Lemraski, E.G.; Webster, T.J.; Rafiee-Moghaddam, R.; Abdollahi, Y. A review of drug delivery systems based on nanotechnology and green chemistry: Green nanomedicine. *Int. J. Nanomed.* **2017**, *12*, 2957. [CrossRef]
- Green, J.J.; Elisseeff, J.H. Mimicking biological functionality with polymers for biomedical applications. *Nature* 2016, 540, 386–394. [CrossRef] [PubMed]
- 40. Kalantari, K.; Afifi, A.M.; Jahangirian, H.; Webster, T.J. Biomedical applications of chitosan electrospun nanofibers as a green polymer–Review. *Carbohydr. Polym.* **2019**, 207, 588–600. [CrossRef]
- Sevastyanova, O.; Qin, W.; Kadla, J. Effect of nanofillers as reinforcement agents for lignin composite fibers. J. Appl. Polym. Sci. 2010, 117, 2877–2881. [CrossRef]
- 42. Patanair, B.; Saiter-Fourcin, A.; Thomas, S.; Thomas, M.G.; Parathukkamparambil Pundarikashan, P.; Gopalan Nair, K.; Kumar, V.K.; Maria, H.J.; Delpouve, N. Promoting interfacial interactions with the addition of lignin in poly (lactic acid) hybrid nanocomposites. *Polymers* **2021**, *13*, 272. [CrossRef] [PubMed]
- 43. Tang, C.; Chen, N.; Zhang, Q.; Wang, K.; Fu, Q.; Zhang, X. Preparation and properties of chitosan nanocomposites with nanofillers of different dimensions. *Polym. Degrad. Stab.* **2009**, *94*, 124–131. [CrossRef]
- 44. Fauzi, B.; Nawawi, M.G.M.; Fauzi, R.; Mamauod, S.N.L. Physicochemical characteristics of sago starch-chitosan nanofillers film. *BioResources* **2019**, *14*, 8324–8330.
- 45. Grossman, A.; Vermerris, W. Lignin-based polymers and nanomaterials. *Curr. Opin. Biotechnol.* **2019**, *56*, 112–120. [CrossRef] [PubMed]
- 46. Lancefield, C.S.; Westwood, N.J. The synthesis and analysis of advanced lignin model polymers. *Green Chem.* **2015**, *17*, 4980–4990. [CrossRef]
- 47. Jayrajsinh, S.; Shankar, G.; Agrawal, Y.K.; Bakre, L. Montmorillonite nanoclay as a multifaceted drug-delivery carrier: A review. J. Drug Deliv. Sci. Technol. 2017, 39, 200–209. [CrossRef]
- Nazir, M.S.; Kassim, M.H.M.; Mohapatra, L.; Gilani, M.A.; Raza, M.R.; Majeed, K. Characteristic properties of nanoclays and characterization of nanoparticulates and nanocomposites. In *Nanoclay Reinforced Polymer Composites*; Springer: Berlin/Heidelberg, Germany, 2016; pp. 35–55.
- 49. Hosseini, S.M.S.; Mirzaei, M. Assessment of the colloidal montmorillonite dispersion as a low-cost and eco-friendly nanofluid for improving thermal performance of plate heat exchanger. *SN Appl. Sci.* **2020**, *2*, 1719. [CrossRef]
- 50. Penchah, H.R.; Ghaemi, A.; Godarziani, H. Eco-friendly CO₂ adsorbent by impregnation of diethanolamine in nanoclay montmorillonite. *Environ. Sci. Pollut. Res.* **2021**, 1–17. [CrossRef]
- 51. Ngwabebhoh, F.A.; Erdem, A.; Yildiz, U. Synergistic removal of Cu (II) and nitrazine yellow dye using an eco-friendly chitosanmontmorillonite hydrogel: Optimization by response surface methodology. *J. Appl. Polym. Sci.* **2016**, 133, 43664. [CrossRef]

- 52. Bordes, P.; Pollet, E.; Avérous, L. Nano-biocomposites: Biodegradable polyester/nanoclay systems. *Prog. Polym. Sci.* 2009, 34, 125–155. [CrossRef]
- 53. Mallakpour, S.; Dinari, M. Synthesis and properties of biodegradable poly (vinyl alcohol)/organo-nanoclay bionanocomposites. *J. Polym. Environ.* **2012**, *20*, 732–740. [CrossRef]
- 54. Kim, Y. TEMPO-Oxidized Nanofibrillated Cellulose Film (NFC) Incorporating Graphene Oxide (GO) Nanofillers; Virginia Tech: Blacksburg, VA, USA, 2017.
- 55. Wu, Y.-Y.; Zhang, J.; Liu, C.; Zheng, Z.; Lambert, P. Effect of graphene oxide nanosheets on physical properties of ultra-highperformance concrete with high volume supplementary cementitious materials. *Materials* **2020**, *13*, 1929. [CrossRef] [PubMed]
- 56. Wang, R.; Zhang, J.; Kang, H.; Zhang, L. Design, preparation and properties of bio-based elastomer composites aiming at engineering applications. *Compos. Sci. Technol.* **2016**, *133*, 136–156. [CrossRef]
- 57. Alcantar, N.A.; Aydil, E.S.; Israelachvili, J.N. Polyethylene glycol–coated biocompatible surfaces. J. Biomed. Mater. Res. 2000, 51, 343–351. [CrossRef]
- 58. Shu, S.; Zhang, X.; Teng, D.; Wang, Z.; Li, C. Polyelectrolyte nanoparticles based on water-soluble chitosan–poly (l-aspartic acid)–polyethylene glycol for controlled protein release. *Carbohydr. Res.* **2009**, 344, 1197–1204. [CrossRef]
- Lee, J.H.; Lee, H.B.; Andrade, J.D. Blood compatibility of polyethylene oxide surfaces. *Prog. Polym. Sci.* 1995, 20, 1043–1079. [CrossRef]
- Gunbas, I.D.; Aydemir Sezer, U.; Gülce İz, S.; Deliloğlu Gürhan, I.; Hasirci, N. Semi-IPN chitosan/PEG microspheres and films for biomedical applications: Characterization and sustained release optimization. *Ind. Eng. Chem. Res.* 2012, *51*, 11946–11954. [CrossRef]
- 61. Jayan, J.S.; Deeraj, B.; Saritha, A.; Joseph, K. Theoretical modelling of kinetics of glass transition temperature of PEG toughened epoxy. *Plast. Rubber Compos.* 2020, 49, 237–244. [CrossRef]
- 62. Ljungberg, N.; Wesslen, B. Tributyl citrate oligomers as plasticizers for poly (lactic acid): Thermo-mechanical film properties and aging. *Polymer* **2003**, *44*, 7679–7688. [CrossRef]
- 63. Cavallaro, G.; Lazzara, G.; Milioto, S. Sustainable nanocomposites based on halloysite nanotubes and pectin/polyethylene glycol blend. *Polym. Degrad. Stab.* 2013, *98*, 2529–2536. [CrossRef]
- 64. Gopi, S.; Amalraj, A.; Kalarikkal, N.; Zhang, J.; Thomas, S.; Guo, Q. Preparation and characterization of nanocomposite films based on gum arabic, maltodextrin and polyethylene glycol reinforced with turmeric nanofiber isolated from turmeric spent. *Mater. Sci. Eng. C* 2019, *97*, 723–729. [CrossRef]
- 65. Martin, O.; Avérous, L. Poly (lactic acid): Plasticization and properties of biodegradable multiphase systems. *Polymer* **2001**, *42*, 6209–6219. [CrossRef]
- 66. Yamane, H.; Sasai, K. Effect of the addition of poly (D-lactic acid) on the thermal property of poly (L-lactic acid). *Polymer* **2003**, 44, 2569–2575. [CrossRef]
- 67. Simamora, P.; Chern, W. Poly-L-lactic acid: An overview. J. Drugs Dermatol. JDD 2006, 5, 436–440. [PubMed]
- Krikorian, V.; Pochan, D.J. Poly (L-lactic acid)/layered silicate nanocomposite: Fabrication, characterization, and properties. *Chem. Mater.* 2003, 15, 4317–4324. [CrossRef]
- 69. Wang, K.; Lu, J.; Tusiime, R.; Yang, Y.; Fan, F.; Zhang, H.; Ma, B. Properties of poly (l-lactic acid) reinforced by l-lactic acid grafted nanocellulose crystal. *Int. J. Biol. Macromol.* **2020**, *156*, 314–320. [CrossRef]
- Li, L.; Bao, R.-Y.; Gao, T.; Liu, Z.-Y.; Xie, B.-H.; Yang, M.-B.; Yang, W. Dopamine-induced functionalization of cellulose nanocrystals with polyethylene glycol towards poly (L-lactic acid) bionanocomposites for green packaging. *Carbohydr. Polym.* 2019, 203, 275–284. [CrossRef] [PubMed]
- Ibrahim, M.M.; Koschella, A.; Kadry, G.; Heinze, T. Evaluation of cellulose and carboxymethyl cellulose/poly (vinyl alcohol) membranes. *Carbohydr. Polym.* 2013, 95, 414–420. [CrossRef] [PubMed]
- 72. Joorabloo, A.; Khorasani, M.T.; Adeli, H.; Mansoori-Moghadam, Z.; Moghaddam, A. Fabrication of heparinized nano ZnO/poly (vinylalcohol)/carboxymethyl cellulose bionanocomposite hydrogels using artificial neural network for wound dressing application. *J. Ind. Eng. Chem.* **2019**, *70*, 253–263. [CrossRef]
- 73. Zhao, Y.; Zhou, Y.; Wu, X.; Wang, L.; Xu, L.; Wei, S. A facile method for electrospinning of Ag nanoparticles/poly (vinyl alcohol)/carboxymethyl-chitosan nanofibers. *Appl. Surf. Sci.* 2012, 258, 8867–8873. [CrossRef]
- 74. Morsi, M.; Oraby, A.; Elshahawy, A.; Abd El-Hady, R. Preparation, structural analysis, morphological investigation and electrical properties of gold nanoparticles filled polyvinyl alcohol/carboxymethyl cellulose blend. *J. Mater. Res. Technol.* **2019**, *8*, 5996–6010. [CrossRef]
- 75. Soykeabkaew, N.; Laosat, N.; Ngaokla, A.; Yodsuwan, N.; Tunkasiri, T. Reinforcing potential of micro-and nano-sized fibers in the starch-based biocomposites. *Compos. Sci. Technol.* **2012**, *72*, 845–852. [CrossRef]
- 76. Lu, D.; Xiao, C.; Xu, S. Starch-based completely biodegradable polymer materials. Express Polym. Lett. 2009, 3, 366–375. [CrossRef]
- 77. Dintcheva, N.T.; La Mantia, F. Durability of a starch-based biodegradable polymer. *Polym. Degrad. Stab.* **2007**, *92*, 630–634. [CrossRef]
- 78. Kaushik, A.; Singh, M.; Verma, G. Green nanocomposites based on thermoplastic starch and steam exploded cellulose nanofibrils from wheat straw. *Carbohydr. Polym.* **2010**, *82*, 337–345. [CrossRef]
- 79. Cheviron, P.; Gouanvé, F.; Espuche, E. Green synthesis of colloid silver nanoparticles and resulting biodegradable starch/silver nanocomposites. *Carbohydr. Polym.* 2014, 108, 291–298. [CrossRef]

- Frangville, C.; Rutkevičius, M.; Richter, A.P.; Velev, O.D.; Stoyanov, S.D.; Paunov, V.N. Fabrication of environmentally biodegradable lignin nanoparticles. *ChemPhysChem* 2012, 13, 4235. [CrossRef] [PubMed]
- 81. Vanholme, R.; Morreel, K.; Darrah, C.; Oyarce, P.; Grabber, J.H.; Ralph, J.; Boerjan, W. Metabolic engineering of novel lignin in biomass crops. *New Phytol.* **2012**, *196*, 978–1000. [CrossRef]
- 82. Ragauskas, A.J.; Beckham, G.T.; Biddy, M.J.; Chandra, R.; Chen, F.; Davis, M.F.; Davison, B.H.; Dixon, R.A.; Gilna, P.; Keller, M.; et al. Lignin valorization: Improving lignin processing in the biorefinery. *Science* **2014**, 344, 6185. [CrossRef]
- 83. Norberg, I.; Nordström, Y.; Drougge, R.; Gellerstedt, G.; Sjöholm, E. A new method for stabilizing softwood kraft lignin fibers for carbon fiber production. J. Appl. Polym. Sci. 2013, 128, 3824–3830. [CrossRef]
- 84. Kadla, J.; Kubo, S.; Venditti, R.; Gilbert, R. Compere, A.; Griffith, W. Lignin-based carbon fibers for composite fiber applications. *Carbon* **2002**, *40*, 2913–2920. [CrossRef]
- 85. Awal, A.; Sain, M. Characterization of soda hardwood lignin and the formation of lignin fibers by melt spinning. *J. Appl. Polym. Sci.* **2013**, *129*, 2765–2771. [CrossRef]
- Mann, G.S.; Singh, L.P.; Kumar, P.; Singh, S. Green composites: A review of processing technologies and recent applications. J. Thermoplast. Compos. Mater. 2020, 33, 1145–1171. [CrossRef]
- 87. Georgios, K.; Silva, A.; Furtado, S. Applications of green composite materials. Biodegrad. Green Compos. 2016, 16, 312.
- Debnath, K.; Singh, I.; Dvivedi, A.; Kumar, P. Natural fibre-reinforced polymer composites for wind turbine blades: Challenges and opportunities. In *Recent Advances in Composite Materials for Wind Turbine Blades*; WAP-AMSA: Hong Kong, China, 2013; Volume 25, p. 40.
- 89. Mishnaevsky, L.; Branner, K.; Petersen, H.N.; Beauson, J.; McGugan, M.; Sørensen, B.F. Materials for wind turbine blades: An overview. *Materials* 2017, *10*, 1285. [CrossRef] [PubMed]
- 90. Gu, C.; Huang, N.; Gao, J.; Xu, F.; Xu, Y.; Jiang, D. Controlled synthesis of conjugated microporous polymer films: Versatile platforms for highly sensitive and label-free chemo-and biosensing. *Angew. Chem.* **2014**, *126*, 4950–4955. [CrossRef]
- 91. Sukumaran, N.P.; Gopi, S. Overview of biopolymers: Resources, demands, sustainability, and life cycle assessment modeling and simulation. In *Biopolymers and their Industrial Applications*; Elsevier: Amsterdam, The Netherlands, 2021; pp. 1–19.
- Jebur, Q.M.; Hashim, A.; Habeeb, M.A. Structural, electrical and optical properties for (polyvinyl alcohol–polyethylene oxide– magnesium oxide) nanocomposites for optoelectronics applications. *Trans. Electr. Electron. Mater.* 2019, 20, 334–343. [CrossRef]
- 93. Guo, J.; Xu, Y.; Jin, S.; Chen, L.; Kaji, T.; Honsho, Y.; Addicoat, M.A.; Kim, J.; Saeki, A.; Ihee, H.; et al. Conjugated organic framework with three-dimensionally ordered stable structure and delocalized *π* clouds. *Nat. Commun.* 2013, 4, 1–8. [CrossRef]
- 94. Scharber, M.C.; Mühlbacher, D.; Koppe, M.; Denk, P.; Waldauf, C.; Heeger, A.J.; Brabec, C. Design rules for donors in bulkheterojunction solar cells—Towards 10% energy-conversion efficiency. *Adv. Mater.* **2006**, *18*, 789–794. [CrossRef]
- Perumal, P.; Selvin, P.C.; Selvasekarapandian, S.; Sivaraj, P.; Abhilash, K.; Moniha, V.; Manjula Devi, R. Plasticizer incorporated, novel eco-friendly bio-polymer based solid bio-membrane for electrochemical clean energy applications. *Polym. Degrad. Stab.* 2019, 159, 43–53. [CrossRef]
- Haigh, P.A.; Bausi, F.; Kanesan, T.; Le, S.T.; Rajbhandari, S.; Ghassemlooy, Z.; Papakonstantinou, I.; Popoola, W.O.; Burton, A.; Minh, H.L.; et al. A 20-Mb/s VLC link with a polymer LED and a multilayer perceptron equalizer. *IEEE Photonics Technol. Lett.* 2014, 26, 1975–1978. [CrossRef]
- 97. Brabec, C.J.; Winder, C.; Sariciftci, N.S.; Hummelen, J.C.; Dhanabalan, A.; van Hal, P.A.; Janssen, R. A low-bandgap semiconducting polymer for photovoltaic devices and infrared emitting diodes. *Adv. Funct. Mater.* **2002**, *12*, 709–712. [CrossRef]
- 98. Chen, L.; Wong, M.-C.; Bai, G.; Jie, W.; Hao, J. White and green light emissions of flexible polymer composites under electric field and multiple strains. *Nano Energy* 2015, 14, 372–381. [CrossRef]
- Holmes, N.P.; Marks, M.; Cave, J.M.; Feron, K.; Barr, M.G.; Fahy, A.; Sharma, A.; Pan, X.; Kilcoyne, D.A.L.; Zhou, X.A.; et al. Engineering two-phase and three-phase microstructures from water-based dispersions of nanoparticles for eco-friendly polymer solar cell applications. *Chem. Mater.* 2018, *30*, 6521–6531. [CrossRef]
- Park, G.E.; Choi, S.; Park, S.Y.; Lee, D.H.; Cho, M.J.; Choi, D.H. Eco-Friendly Solvent-Processed Fullerene-Free Polymer Solar Cells with over 9.7% Efficiency and Long-Term Performance Stability. *Adv. Energy Mater.* 2017, 7, 1700566. [CrossRef]
- Lee, S.; Jeong, D.; Kim, C.; Lee, C.; Kang, H.; Woo, H.Y.; Kim, B.J. Eco-Friendly Polymer Solar Cells: Advances in Green-Solvent Processing and Material Design. ACS Nano 2020, 14, 14493–14527. [CrossRef]
- 102. Ghosh, S.K.; Sinha, T.K.; Mahanty, B.; Jana, S.; Mandal, D. Porous polymer composite membrane based nanogenerator: A realization of self-powered wireless green energy source for smart electronics applications. J. Appl. Phys. 2016, 120, 174501. [CrossRef]
- 103. Zhang, Q.; Sun, Y.; Xu, W.; Zhu, D. Organic thermoelectric materials: Emerging green energy materials converting heat to electricity directly and efficiently. *Adv. Mater.* **2014**, *26*, 6829–6851. [CrossRef] [PubMed]
- Cui, L.; Yu, S.; Gao, W.; Zhang, X.; Deng, S.; Zhang, C.-Y. Tetraphenylenthene-based conjugated microporous polymer for aggregation-induced electrochemiluminescence. ACS Appl. Mater. Interfaces 2020, 12, 7966–7973. [CrossRef]
- 105. Yuan, K.; Guo-Wang, P.; Hu, T.; Shi, L.; Zeng, R.; Forster, M.; Pichler, T.; Chen, Y.; Scherf, U. Nanofibrous and graphene-templated conjugated microporous polymer materials for flexible chemosensors and supercapacitors. *Chem. Mater.* 2015, 27, 7403–7411. [CrossRef]
- 106. Feng, W.; Long, P.; Feng, Y.; Li, Y. Two-dimensional fluorinated graphene: Synthesis, structures, properties and applications. *Adv. Sci.* **2016**, *3*, 1500413. [CrossRef] [PubMed]

- 107. Zhuang, X.; Gehrig, D.; Forler, N.; Liang, H.; Wagner, M.; Hansen, M.R.; Laquai, F.; Zhang, F.; Feng, X. Conjugated microporous polymers with dimensionality-controlled heterostructures for green energy devices. *Adv. Mater.* 2015, 27, 3789–3796. [CrossRef]
- 108. Zhao, W.; Jiao, Y.; Li, J.; Wu, L.; Xie, A.; Dong, W. One-pot synthesis of conjugated microporous polymers loaded with superfine nano-palladium and their micropore-confinement effect on heterogeneously catalytic reduction. J. Catal. 2019, 378, 42–50. [CrossRef]
- 109. Christen, T.; Carlen, M.W. Theory of Ragone plots. J. Power Sources 2000, 91, 210-216. [CrossRef]
- 110. Liu, X.; Li, K. Energy storage devices in electrified railway systems: A review. Transp. Saf. Environ. 2020, 2, 183–201. [CrossRef]
- 111. Chatzivasileiadi, A.; Ampatzi, E.; Knight, I. Characteristics of electrical energy storage technologies and their applications in buildings. *Renew. Sustain. Energy Rev.* 2013, 25, 814–830. [CrossRef]
- 112. Koohi-Kamali, S.; Tyagi, V.; Rahim, N.; Panwar, N.; Mokhlis, H. Emergence of energy storage technologies as the solution for reliable operation of smart power systems: A review. *Renew. Sustain. Energy Rev.* **2013**, *25*, 135–165. [CrossRef]
- 113. Dunn, B.; Kamath, H.; Tarascon, J.-M. Electrical energy storage for the grid: A battery of choices. *Science* **2011**, *334*, 928–935. [CrossRef]
- 114. Yao, K.; Chen, S.; Rahimabady, M.; Mirshekarloo, M.S.; Yu, S.; Tay, F.E.H.; Sritharan, T.; Lu, L. Nonlinear dielectric thin films for high-power electric storage with energy density comparable with electrochemical supercapacitors. In *IEEE Transactions on Ultrasonics, Ferroelectrics, and Frequency Control*; IEEE: New York, NY, USA, 2011; Volume 58, pp. 1968–1974.
- 115. Frackowiak, E. Carbon materials for supercapacitor application. Phys. Chem. Chem. Phys. 2007, 9, 1774–1785. [CrossRef] [PubMed]
- Borenstein, A.; Hanna, O.; Attias, R.; Luski, S.; Brousse, T.; Aurbach, D. Carbon-based composite materials for supercapacitor electrodes: A review. J. Mater. Chem. A 2017, 5, 12653–12672. [CrossRef]
- 117. Iro, Z.S.; Subramani, C.; Dash, S. A brief review on electrode materials for supercapacitor. *Int. J. Electrochem. Sci.* 2016, 11, 10628–10643. [CrossRef]
- 118. Çıplak, Z.; Yıldız, A.; Yıldız, N. Green preparation of ternary reduced graphene oxide-au@ polyaniline nanocomposite for supercapacitor application. J. Energy Storage 2020, 32, 101846. [CrossRef]
- Yang, L.; Kong, X.; Li, F.; Hao, H.; Cheng, Z.; Liu, H.; Li, J.; Zhang, S. Perovskite lead-free dielectrics for energy storage applications. *Prog. Mater. Sci.* 2019, 102, 72–108. [CrossRef]
- 120. Arthisree, D.; Madhuri, W. Optically active polymer nanocomposite composed of polyaniline, polyacrylonitrile and greensynthesized graphene quantum dot for supercapacitor application. *Int. J. Hydrog. Energy* **2020**, *45*, 9317–9327. [CrossRef]
- 121. Sumboja, A.; Wang, X.; Yan, J.; Lee, P.S. Nanoarchitectured current collector for high rate capability of polyaniline based supercapacitor electrode. *Electrochim. Acta* 2012, *65*, 190–195. [CrossRef]
- 122. Nayak, A.K.; Das, A.K.; Pradhan, D. High performance solid-state asymmetric supercapacitor using green synthesized graphene–WO₃ nanowires nanocomposite. *ACS Sustain. Chem. Eng.* **2017**, *5*, 10128–10138. [CrossRef]
- 123. Rao, Y.; Ogitani, S.; Kohl, P.; Wong, C. Novel polymer–ceramic nanocomposite based on high dielectric constant epoxy formula for embedded capacitor application. *J. Appl. Polym. Sci.* **2002**, *83*, 1084–1090. [CrossRef]
- Chakraborty, S.; Mary, N. Biocompatible supercapacitor electrodes using green synthesised ZnO/Polymer nanocomposites for efficient energy storage applications. J. Energy Storage 2020, 28, 101275. [CrossRef]
- 125. Hao, Y.; Wang, X.; O'Brien, S.; Lombardi, J.; Li, L. Flexible BaTiO₃/PVDF gradated multilayer nanocomposite film with enhanced dielectric strength and high energy density. *J. Mater. Chem. C* 2015, *3*, 9740–9747. [CrossRef]
- 126. Zhong, C.; Deng, Y.; Hu, W.; Qiao, J.; Zhang, L.; Zhang, J. A review of electrolyte materials and compositions for electrochemical supercapacitors. *Chem. Soc. Rev.* 2015, 44, 7484–7539. [CrossRef] [PubMed]
- 127. Ren, F.; Li, Z.; Tan, W.-Z.; Liu, X.-H.; Sun, Z.-F.; Ren, P.-G.; Yan, D.-X. Facile preparation of 3D regenerated cellulose/graphene oxide composite aerogel with high-efficiency adsorption towards methylene blue. J. Colloid Interface Sci. 2018, 532, 58–67. [CrossRef]
- 128. Ouyang, W.; Sun, J.; Memon, J.; Wang, C.; Geng, J.; Huang, Y. Scalable preparation of three-dimensional porous structures of reduced graphene oxide/cellulose composites and their application in supercapacitors. *Carbon* **2013**, *62*, 501–509. [CrossRef]
- 129. Wan, C.; Jiao, Y.; Liang, D.; Wu, Y.; Li, J. A geologic architecture system-inspired micro-/nano-heterostructure design for high-performance energy storage. *Adv. Energy Mater.* **2018**, *8*, 1802388. [CrossRef]
- Tian, J.; Peng, D.; Wu, X.; Li, W.; Deng, H.; Liu, S. Electrodeposition of Ag nanoparticles on conductive polyaniline/cellulose aerogels with increased synergistic effect for energy storage. *Carbohydr. Polym.* 2017, 156, 19–25. [CrossRef] [PubMed]
- 131. Zu, G.; Shen, J.; Zou, L.; Wang, F.; Wang, X.; Zhang, Y.; Yao, X. Nanocellulose-derived highly porous carbon aerogels for supercapacitors. *Carbon* 2016, 99, 203–211. [CrossRef]
- Yang, X.; Fei, B.; Ma, J.; Liu, X.; Yang, S.; Tian, G.; Jiang, Z. Porous nanoplatelets wrapped carbon aerogels by pyrolysis of regenerated bamboo cellulose aerogels as supercapacitor electrodes. *Carbohydr. Polym.* 2018, 180, 385–392. [CrossRef] [PubMed]
- Tian, W.; Gao, Q.; Zhang, L.; Yang, C.; Li, Z.; Tan, Y.; Qian, W.; Zhang, H. Renewable graphene-like nitrogen-doped carbon nanosheets as supercapacitor electrodes with integrated high energy–power properties. *J. Mater. Chem. A* 2016, *4*, 8690–8699. [CrossRef]
- Li, Y.; Wang, G.; Wei, T.; Fan, Z.; Yan, P. Nitrogen and sulfur co-doped porous carbon nanosheets derived from willow catkin for supercapacitors. *Nano Energy* 2016, 19, 165–175. [CrossRef]

- Chen, L.F.; Huang, Z.H.; Liang, H.W.; Guan, Q.F.; Yu, S.H. Bacterial-cellulose-derived carbon nanofiber@ MnO₂ and nitrogendoped carbon nanofiber electrode materials: An asymmetric supercapacitor with high energy and power density. *Adv. Mater.* 2013, 25, 4746–4752. [CrossRef] [PubMed]
- 136. Mensah-Darkwa, K.; Zequine, C.; Kahol, P.K.; Gupta, R.K. Supercapacitor energy storage device using biowastes: A sustainable approach to green energy. *Sustainability* **2019**, *11*, 414. [CrossRef]
- Cao, W.; Yang, F. Supercapacitors from high fructose corn syrup-derived activated carbons. *Mater. Today Energy* 2018, 9, 406–415. [CrossRef]
- 138. Du, S.-H.; Wang, L.-Q.; Fu, X.-T.; Chen, M.-M.; Wang, C.-Y. Hierarchical porous carbon microspheres derived from porous starch for use in high-rate electrochemical double-layer capacitors. *Bioresour. Technol.* **2013**, *139*, 406–409. [CrossRef]
- 139. Fan, H.; Shen, W. Gelatin-based microporous carbon nanosheets as high performance supercapacitor electrodes. *ACS Sustain. Chem. Eng.* **2016**, *4*, 1328–1337. [CrossRef]
- 140. Li, Y.-T.; Pi, Y.-T.; Lu, L.-M.; Xu, S.-H.; Ren, T.-Z. Hierarchical porous active carbon from fallen leaves by synergy of K₂CO₃ and their supercapacitor performance. *J. Power Sources* **2015**, *299*, 519–528. [CrossRef]
- 141. Peng, C.; Yan, X.-B.; Wang, R.-T.; Lang, J.-W.; Ou, Y.-J.; Xue, Q.-J. Promising activated carbons derived from waste tea-leaves and their application in high performance supercapacitors electrodes. *Electrochim. Acta* **2013**, *87*, 401–408. [CrossRef]
- Liu, J.; Deng, Y.; Li, X.; Wang, L. Promising nitrogen-rich porous carbons derived from one-step calcium chloride activation of biomass-based waste for high performance supercapacitors. ACS Sustain. Chem. Eng. 2016, 4, 177–187. [CrossRef]
- 143. Rufford, T.E.; Hulicova-Jurcakova, D.; Khosla, K.; Zhu, Z.; Lu, G.Q. Microstructure and electrochemical double-layer capacitance of carbon electrodes prepared by zinc chloride activation of sugar cane bagasse. *J. Power Sources* **2010**, *195*, 912–918. [CrossRef]
- 144. Zargar, V.; Asghari, M.; Dashti, A. A review on chitin and chitosan polymers: Structure, chemistry, solubility, derivatives, and applications. *ChemBioEng Rev.* 2015, 2, 204–226. [CrossRef]
- 145. Shi, K.; Yang, X.; Cranston, E.D.; Zhitomirsky, I. Efficient lightweight supercapacitor with compression stability. *Adv. Funct. Mater.* **2016**, *26*, 6437–6445. [CrossRef]
- 146. Wang, K.; Li, L.; Zhang, T.; Liu, Z. Nitrogen-doped graphene for supercapacitor with long-term electrochemical stability. *Energy* **2014**, *70*, 612–617. [CrossRef]
- 147. Chen, X.Y.; Chen, C.; Zhang, Z.J.; Xie, D.H.; Deng, X.; Liu, J.W. Nitrogen-doped porous carbon for supercapacitor with long-term electrochemical stability. *J. Power Sources* 2013, 230, 50–58. [CrossRef]
- 148. Olivetti, E.A.; Cullen, J.M. Toward a sustainable materials system. Science 2018, 360, 1396–1398. [CrossRef]
- 149. Asdrubali, F.; Schiavoni, S.; Horoshenkov, K. A review of sustainable materials for acoustic applications. *Build. Acoust.* **2012**, *19*, 283–311. [CrossRef]
- 150. Sharma, R.; Jafari, S.M.; Sharma, S. Antimicrobial bio-nanocomposites and their potential applications in food packaging. *Food Control* **2020**, *112*, 107086. [CrossRef]
- 151. Díez-Pascual, A.M.; Diez-Vicente, A.L. ZnO-reinforced poly (3-hydroxybutyrate-co-3-hydroxyvalerate) bionanocomposites with antimicrobial function for food packaging. ACS Appl. Mater. Interfaces 2014, 6, 9822–9834. [CrossRef]